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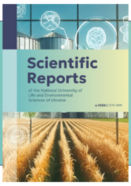

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High-permeability selective polymer membranes for pyrocondensate separation

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Abstract. The aim of the work was to determine the optimal technological parameters for obtaining pure hydrocarbon fractions and extracting pyrocarbon and heavy pyrolysis resin. The work used a method of membrane separation of pyrocondensate using highly permeable selective polymer

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membranes. It was established that the average composition of pyrocondensate contains up to 58.4% of hydrocarbons suitable for obtaining gasoline, kerosene and diesel fractions, as well as 20% of unsaturated hydrocarbons and 20% of pyrocarbon with heavy pyrolysis resin. The effectiveness of membrane technology for separating pyrocondensate with the production of gasoline (8.2%), kerosene and diesel (23.9%) fractions with low sulphur and unsaturated hydrocarbon content has been proven. The characteristics of the fractions obtained met the requirements of standards for their use as finished commercial products. The optimal separation temperatures for each fraction were determined: gasoline – 50°C, kerosene – 70°C, diesel – 85°C. The dependence of membrane permeability on temperature and selectivity for unsaturated hydrocarbons and sulphur was established. The separation process ensured the simultaneous removal of pyrocarbon and heavy pyrolysis resin, which simplifies the technology and increases its efficiency. The proposed catalytic methods for processing pyrocarbon sediment and unsaturated hydrocarbon residues allow for the complete processing of pyrocondensate from 96% to 98%. The possibility of using membrane technology for the efficient processing of pyrocondensate with minimal impact on the environment has been confirmed. Analytical and experimental principles for calculating membrane separation devices and their productivity have been developed. The proposed solutions can be applied to the production of high-quality fuel fractions from waste materials

Keywords: fractional composition; pyrolysis; petroleum polymer resins; catalytic treatment; diesel fraction

Introduction

In the modern world, considerable attention is paid to technologies for processing industrial waste and obtaining marketable products while improving environmental performance. In this context, the processing of polymer waste, in particular worn car tyres, is of great interest. This type of waste ranks first in terms of tonnage compared to other polymer waste. In Ukraine, the annual increase in worn tyres is 250-300 thousand tonnes, and the level of their utilisation does not exceed 10%. The accumulation of such waste creates a significant anthropogenic load on the environment and requires the introduction of effective technologies for comprehensive recycling. One of the most promising areas in this context is thermochemical processing, in particular pyrolysis, which allows valuable fuel and chemical products to be obtained (Boichenko *et al.*, 2024).

Scientist I.Ya. Zakhara (2025) analysed modern approaches to the disposal and processing of worn car tyres, focusing on the environmental and technological aspects of existing methods. The author showed that traditional methods, in particular incineration and mechanical processing, were accompanied by a significant environmental burden, while thermochemical methods, primarily pyrolysis, were considered more promising in terms of obtaining valuable products and reducing harmful emissions. K.A. Ibragimov *et al.* (2020) investigated the composition, properties and uses of liquid pyrolysis products, paying particular attention to catalytic methods of their processing. The authors established the patterns of selective hydrogenation and oligomerisation of unsaturated hydrocarbons and showed the prospects for obtaining petroleum polymer resins and fuel fractions, while noting the technological limitations associated with coking and the loss of part of the hydrocarbon raw materials.

The article by I. Shlonchak & O. Batrachenko (2024) examined modern approaches to the disposal of car tyres in Ukraine, taking into account global experience. The authors analysed the main processing technologies and identified their advantages and disadvantages. They also emphasised the need to develop infrastructure and improve the rubber waste management system in the Ukrainian context. The work by

K. Hrynyshyn *et al*. (2022) showed that low-temperature pyrolysis allows obtaining pyrocondensate with a clearly defined fractional composition, including gasoline and diesel fractions, as well as heavy residue. The authors emphasised the possibility of comprehensive use of tyre processing products and their return to the production cycle.

Experimental studies by A. Khurmamatov & K. Akhmedova (2025) also confirmed the effectiveness of pyrolysis as a tyre processing technology. This article examines in detail the physicochemical properties of pyrolysis oil, gas and solid phase. The authors found that the synthetic fuel obtained could be a promising energy resource, provided that the process is further refined and optimised. The importance of pyrolysis as an environmentally sound way to reduce rubber waste and reuse it was also emphasised. The work of A. Ranskyi & B. Korinenko (2023) considered the prospects of obtaining synthetic oil as an alternative energy resource through the pyrolytic processing of polypropylene waste. The authors showed that thermochemical decomposition of polymers allowed plastic waste to be converted into liquid hydrocarbon products suitable for further use in the fuel and chemical industries. The study emphasised the importance of developing pyrolysis technologies as an effective way to dispose of polymer waste while obtaining valuable products. The article by V. Chernega (2024) analysed the problem of worn car tyre accumulation from an economic and environmental point of view. The author emphasised that used tyres are one of the most common and dangerous types of polymer waste, and their reuse and recycling are essential for reducing the anthropogenic impact on the environment. The work justified the need to introduce modern tyre disposal technologies that would ensure not only an environmental effect but also economic feasibility through the recovery of secondary resources.

The work of W. Jerzak *et al*. (2024) expanded scientific understanding of the pyrolysis process through the use of modern analytical methods and different types of reactors. The authors focused on the influence of heating rate and catalyst introduction method on the formation of aromatic hydrocarbons and raw soot yield. The study also demonstrated the complexity of the thermal decomposition mechanisms of tyre raw materials and the relationship between process conditions and the chemical composition of products. The work of N.H. Zerin *et al*. (2023) stated that pyrolysis is a more environmentally friendly method of processing waste tyres compared to traditional methods, as it allows waste to be converted into oil, gas and solid phase (char) without causing harmful emissions during combustion. The study also summarises the influence of process parameters (temperature, residence time, particle size) on the yield and composition of pyrolysis products, highlighting the technological complexity of this method.

Thus, there is a pressing need to improve technologies for processing pyrocondensate obtained from the pyrolysis of car tyres to increase its efficiency and obtain commercial products with predictable physical and chemical properties. The aim of the work was to determine the optimal technological parameters of the process of separating pyrocondensate obtained by pyrolysis of worn car tyres and extracting pyrocarbon and heavy pyrolysis resin from it to obtain pure hydrocarbon fractions using membrane technology. To achieve this aim, the following objectives were set: to determine the composition of pyrocondensate from the pyrolysis of worn car tyres; to carry out membrane separation of pyrocondensate to obtain commercial products for use in the automotive industry; to determine the optimal conditions for pyrocondensate separation.

Materials and Methods

The object of the research was the process of obtaining pure hydrocarbon fractions from pyrocondensate. The subject of the research was a comprehensive approach to pyrocondensate processing by combining traditional methods and unique membrane pervaporation technologies. The study used pyrocondensate obtained at a low-temperature pyrolysis plant at the Rosava private joint-stock company (Bila Tserkva, Ukraine) as a result of the pyrolysis of worn car tyres. The initial pyrocondensate was characterised by a complex multicomponent structure containing up to 58.4% hydrocarbons (light, medium and heavy fractions), about 20% unsaturated hydrocarbons, and 20% pyrocarbon with heavy pyrolysis resin. The content of sulphur, unsaturated hydrocarbons, water and solid impurities was determined in accordance with DSTU 7687:2015 (2015).

The study used highly permeable selective polymer membranes specially designed for the separation of complex organic mixtures. The membranes were selected taking into account their resistance to organic solvents, process temperature and selectivity to pyrocondensate fractions. The membrane separation process was carried out in an experimental laboratory membrane unit with the ability to regulate temperature (from 30 to 90°C) and pressure (up to 10 bar). For each fraction (gasoline, kerosene, diesel), the optimal process temperatures were set: 50°C, 70°C and 85°C, respectively. The research was conducted in the educational laboratory of membrane technology at the Department of Biotechnics and Engineering, Faculty of Biotechnology and Biotechnics, National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute". All analyses of the pyrocondensate and the separated fractions were performed using certified equipment on a YL6100 chromatograph (YoungIn Chromass) in the laboratories of the research institute of the state fuel company MASMA. The following methods and tests were used to assess the quality and composition of the products.

The fractional composition of the gasoline fraction was determined in accordance with DSTU EN 228:2022 (2022), which provided for the determination of fractional distribution temperatures by distillation; the fractional composition of the kerosene and diesel fractions was determined according to the DSTU EN 590:2022 (2022), which includes measuring distillation points to assess the composition of fractions; the sulphur content in the fractions was determined in accordance with ISO 8754 (2003) using energy-dispersive X-ray fluorescence analysis, which allows the concentration of sulphur in petroleum products to be accurately determined; the iodine number (a characteristic of the degree of unsaturation of organic compounds) was determined according to EN 16300:2012 (2012) using the calculation method for fatty acid methyl esters (FAME) based on gas chromatography data. Gas chromatography-mass spectrometry (GC-MS) was used for detailed determination of the component composition of the pyrocondensate and its fractions, as well as for identifying impurities and target hydrocarbons. Fourier-transform infrared spectroscopy (FTIR) was applied to analyse functional groups in the fractions, enabling assessment of the degree of saturation and the presence of impurities. Water content was determined by coulometric titration using the Karl Fischer method. Viscosity measurements were performed with a rotational viscometer to evaluate the suitability of the fractions for use as fuels. Storage stability was assessed by an accelerated ageing method (24 hours at 80°C) followed by analysis of changes in viscosity and composition.

The choice of these methods was dictated by the need to obtain complete and reliable information on the composition, purity, physicochemical properties and stability of pyrocondensate fractions. The application of international standards ensured the comparability of the results with modern requirements for the quality of fuel products and allowed an objective assessment of the effectiveness of membrane technology for industrial use. The component composition of pyrocondensate, both raw and subjected to catalytic treatment in the presence of KTK-1, was studied by gas-liquid chromatography. Figure 1 presents the schematic technological diagram for the production of petroleum polymer resins in the presence of the KTK-1 catalytic complex.

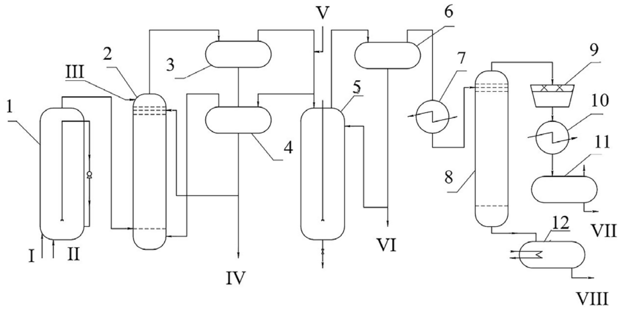

Figure 1. *Schematic process flow diagram for the production of petroleum polymer resins in the presence of the KTK-1 catalytic complex*

Note: 1 – reactor; 2, 5 – reactor-washer; 3, 4 – settler; 6 – separator; 7 – heater; 8 – distillation column; 9 – air cooler; 10 – condenser; 11, 12 – storage tanks. I – pyrocondensate; II – catalytic complex; III – alkali solution; IV – deactivated catalyst; V, VI – water; VII, VIII – finished product

Source: V.T. Dumskyi *et al.* (2014)

Polymerisation was carried out by a continuous circulation method in reactor 1, into which pyrocondensate I and catalytic complex II were fed from below. Reactor 1 was equipped with a special device designed to ensure effective circulation of the reaction mixture and maximum conversion of unsaturated hydrocarbons. In this way, the possibility of accidental breakthrough, monitored by chromatographic analysis, was virtually eliminated. The resulting polymerisate from reactor 1 was fed to reactor-washer 2, where the catalytic complex was decomposed using alkali solution III, which was subsequently discharged from the top of the reactor and, through separators-settlers 3 and 4, directed for water washing V in the reactor-washer. In the separators, the polymerisate was separated from the circulating alkali solution (in separators-settlers 3 and 4), part of which was discharged to a wastewater treatment unit. The deactivated catalyst IV was removed from the bottom of separator-settler 4.

The neutralised polymerisate, via separator 6, where water VI was removed and discharged from the unit, was fed to heater 7 and then to the top of distillation column 8. Unpolymerised hydrocarbons from the top of column 8 were directed to air cooler 9 and then to condenser 10, from which they were transferred to storage tank 11 and subsequently to storage. The resin melt VIII from the bottom of column 8 was collected in tank 12, from which it was pumped out of the unit for dissolution or granulation. The proposed process scheme for pyrocondensate processing made it possible to remove a gasoline fraction purified from unsaturated hydrocarbons, which could be used either for alkylation with ethylene or for the production of high-purity aromatic hydrocarbons, while reducing the number of stages in the financially costly hydrogenation process.

According to the proposed fundamentally new scheme for pyrocondensate processing using KTK-1, targeted synthesis of modified petroleum polymer resins was achieved through the rational utilisation of unsaturated hydrocarbons. These resins possess a set of properties required for their use as components of polymer-bitumen compositions, binding agents in paint and varnish materials, effective plasticisers for rubber compounds, and vulcanising agents for electrical insulation coatings. One of the most reliable and economical methods for improving the quality of road bitumen is protection against ageing and drying. The possibility of using petroleum polymer resins as effective additives to road bitumen is confirmed by research results (Ibragimov *et al.*, 2020).

Problem statement. The high content of pyrocarbon in pyrocondensate obtained from the pyrolysis of worn car tyres significantly complicated the application of known technologies for its processing. The methods of processing pyrocondensate from polymer waste described in the literature were technologically complex even under conditions of significantly lower pyrocarbon content, which in such systems was on average four times lower than in pyrocondensate from worn car tyres. With an increased pyrocarbon content, the implementation of these technologies was impossible due to the intensive deposition of pyrocarbon and heavy pyrolysis resin on the surface of catalysts in the processes of hydrogenation and catalytic treatment, as well as on the contact elements of equipment during fractionation in rectification columns. Pyrocondensate processing was carried out in stages, involving the preliminary separation of pyrocarbon and heavy pyrolysis resin, followed by the processing of purified pyrocondensate using traditional technologies. Membrane technology using the pervaporation method was used to remove pyrocarbon and heavy pyrolysis resin. The use of pervaporation membranes is due to the fact that they are semi-permeable, have no pores and exhibit anti-adhesion properties with respect to pyrocarbon and resins, so that such contaminants cannot be deposited on their surface. Two processes took place simultaneously in the apparatus with such membranes: the passage of hydrocarbons contained in the pyrocondensate through the membrane wall due to molecular diffusion and the precipitation of pyrocarbon and heavy pyrolysis resin at the bottom of the apparatus during the separation process. The process of separating pyrocondensate into fractions (gasoline, kerosene, diesel) and extracting pyrocarbon and heavy pyrolysis resin from it was carried out on an experimental setup, the diagram of which is shown in Figure 2.

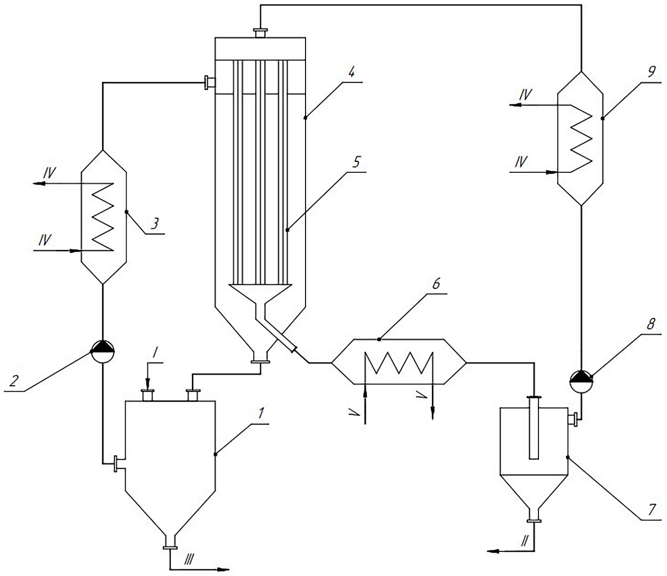

Figure 2. *Schematic diagram of the experimental unit for the separation of pyrocondensate*

Note: 1 – receiving tank, 2 – gear pump, 3 – heat exchanger, 4 – membrane apparatus, 5 – membrane element, 6 – condenser refrigerator, 7 – fraction collector, 8 – steam-gas compressor, 9 – calorifier. I – pyrocondensate, II – fractions, III – pyrocarbon and heavy pyrolysis resin, IV – hot coolant, V – cold coolant

Source: authors’ own work

Before starting work, the devices that were part of the installation were were purged with nitrogen. The output pyrocondensate I was loaded into receiving tank 1, from which it was fed by gear pump 2 through heat exchanger 3, where it was heated to a set temperature, into membrane element 4 on the distribution device. The distribution device ensured uniform irrigation of the outer surface of the tubular membrane elements 5. In the membrane apparatus, the heated pyrocondensate flowed in the form of a thin film over the outer surface of the tubular membrane elements. A heated nitrogen-vapor mixture (a mixture of nitrogen and hydrocarbon vapors) was pumped along the inner surface of the membrane elements using a steam-gas compressor 8 under a slight excess pressure (0.01-0.02 MPa). In this case, certain fractions of hydrocarbons passed through the walls of the membrane elements due to molecular diffusion and mixed in the form of vapour (pervaporation process) with the nitrogen-vapour mixture that was pumped along the inner surface of the membranes. The nitrogen-vapor mixture enriched with the hydrocarbon fraction in a saturated state from the membrane apparatus entered the condenser refrigerator 6, where condensation took place. Condensate II entered the fraction collector 7, from which it was periodically drained, and the nitrogen-steam mixture from the fraction collector was sucked out by a steam-gas compressor 8, heated in a calorifier 9 and fed into the membrane apparatus.

The spent pyrocondensate from the membrane apparatus was drained into a membrane vessel, where pyrocarbon and heavy pyrolysis resin were precipitated from it. The precipitation process is related to the fact that heavy pyrolysis resin exhibits high adhesion to pyrocarbon and dissolves well in hydrocarbon fractions, which are removed from the pyrocondensate by membrane elements during the pervaporation process. Thus, as the content of hydrocarbon fractions in the pyrocondensate decreased, pyrocarbon and heavy pyrolysis resin III were deposited, which were periodically removed from the bottom of the receiving tank. The pyrocondensate from the receiving tank was fed back into the plant's apparatus by a gear pump. Thus, the technological process of pyrocondensate separation at the experimental plant was carried out in a closed cycle.

Results and Discussion

Worn car tyres are a complex multi-component raw material containing a rubber matrix, carbon black, metal cord, textile impurities and a number of organic and inorganic additives. Their accumulation creates a significant anthropogenic load on the environment, and traditional methods of disposal do not ensure the effective extraction and use of the material and energy potential of this type of waste. In this context, chemical processing methods, in particular pyrolysis, are considered a technologically feasible way of comprehensive tyre disposal. Pyrolysis of car tyres allows the organic component of the raw material to be converted into pyrolysis oil and gas, and the inorganic component into a solid carbon phase (carbon black) and metal cord. However, scientific research shows that the yield, fractional composition and physicochemical properties of pyrolysis products depend significantly on the temperature regime, heating rate, process duration, reactor type, and the presence or absence of catalysts. The lack of standardised approaches to assessing process parameters complicates the scaling of technologies and their implementation in industrial practice. In addition, an important engineering and technological task is to evaluate the quality of the pyrolysis products obtained in terms of their further use – pyrolysis oil as a raw material for fuels or chemical synthesis, gas as an energy resource, and solid carbon phase as a filler or functional material. Particular attention should be paid to the analysis of the content of sulphur, nitrogen, oxygen-containing compounds and ash components, which determine the environmental and operational characteristics of the products (Hrynyshyn *et al.,* 2021).

Chromatographic studies show that unsaturated hydrocarbons are almost completely converted during the catalytic treatment of pyrocondensate (98.7-99.8% conversion) and no unsaturated hydrocarbons remain in the pyrocondensate. The optimal conditions obtained formed the basis for the synthesis of petroleum polymer resins for catalytic processing of pyrocondensate. Gas chromatography makes it possible to separate the complex mixtures of hydrocarbons contained in the pyrocondensate fractions into individual components according to their retention times (Table 1). Each fraction is characterised by its own set of peaks corresponding to different organic compounds. The gasoline fraction is characterised by the presence of light hydrocarbons, therefore the main peaks are observed at the beginning of the chromatogram (Fig. 3).

Table 1. *Chromatographic analysis of the obtained liquid fractions of pyrocondensate*

Retention time (min)	Gasoline fraction	Kerosene fraction	Diesel fraction
2-5	High peaks (C5-C7)	Low peaks	Absent
5-10	Medium peaks (C7-C9)	Start of peaks	Absent
10-15	Low peaks (C9-C10)	High peaks (C10-C12)	Start of peaks
15-25	Absent	Medium peaks (C12-C15)	High peaks (C15-C18)
25-40	Absent	Absent	Medium/high peaks (C18-C22)

Source: developed by the authors

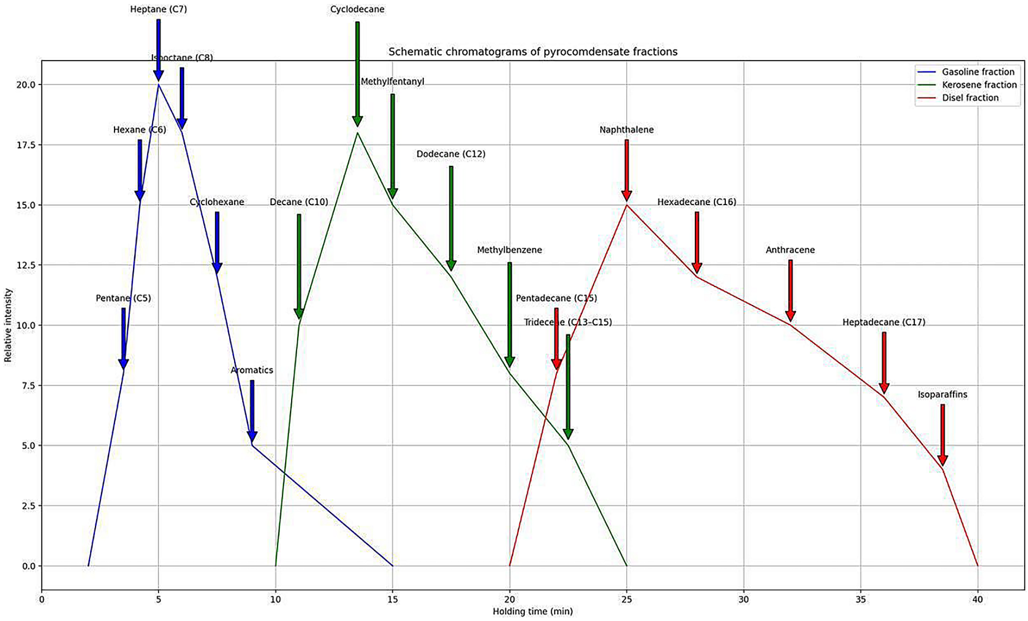

Figure 3. *Chromatogram of gasoline, kerosene and diesel fractions*

Source: developed by the authors

The kerosene fraction contains medium-boiling components, with peaks located in the middle of the chromatogram. The diesel fraction has the longest retention time, corresponding to heavy hydrocarbons. The peak height reflects the relative concentration of the component. The peaks of the gasoline fraction are located closer to the beginning of the chromatogram (short retention time). The peaks of the kerosene and diesel fractions are shifted to the right, corresponding to an increase in molecular weight and retention time. The designation of the peaks (Fig. 3) and their identification (Table 2-4) are presented on the chromatogram.

Table 2. *Gasoline fraction (retention time 2-15 min)*

Peak	Retention time (min)	Identified component	Explanation
1	~3.5	Pentane (C5)	Light alkane
2	~4.2	Hexane (C6)	Main component of the gasoline fraction
3	~5.0	Heptane (C7)	Important for octane rating
4	~6.0	Isooctane (C8, isomer of octane)	Responsible for anti-knock properties
5	~7.5	Cyclohexane	Cycloalkane
6	~9.0	Aromatic hydrocarbons (benzene, toluene)	Small amount of aromatics

Source: developed by the authors

Table 3. *Kerosene fraction (retention time 10-25 min)*

Peak	Retention time (min)	Identified component	Explanation
1	~11.0	Decane (C10)	Medium boiling alkane
2	~13.5	Cyclodecane	Cycloalkane
3	~15.0	Methylnaphthalene	Aromatic hydrocarbon
4	~17.5	Dodecane (C12)	Medium chain alkane
5	~20.0	Methylbenzene	Aromatic component
6	~22.5	Tridecene (C13-C15)	Heavier alkanes

Source: developed by the authors

Table 4. *Diesel fraction (retention time 20-40 min)*

Peak	Retention time (min)	Identified component	Explanation
1	~22.0	Pentadecane (C15)	Heavy alkane
2	~25.0	Polycyclic aromatic hydrocarbon (naphthalene)	Aromatic component
3	~28.0	Hexadecane (C16)	Alkane
4	~32.0	Polycyclic aromatic hydrocarbon (anthracene)	Heavy aromatic
5	~36.0	Heptadecane (C17)	Heavy alkane
6	~38.5	Isoparaffins	Branched hydrocarbons

Source: developed by the authors

Identification of the components was carried out using gas chromatography-mass spectrometry (GC-MS), which provides retention times and mass spectra for accurate substance determination. Comparison with spectral libraries (for example, NIST) made it possible to confirm the identity of the peaks. Standard samples were used to calibrate the holding time. Pyrocondensate obtained as a result of pyrolysis of worn car tyres is an opaque black liquid with a characteristic odour. Its composition is shown in Table 5.

Table 5. *Composition of pyrocondensate from the pyrolysis of worn car tyres*

Parameter	Value
Density	0.921 g/cm^3
Sulphur content	1.6% by mass
Unsaturated hydrocarbon content	20% by mass

Table 5. *Continued*

Parameter	Value
Pyrocarbon content	9% by mass
Heavy pyrolysis resin content	11% by mass

Source: developed by the authors

As can be seen from Table 5, the pyrocondensate contains a large amount of pyrocarbon and heavy pyrolysis resin, a significant quantity of unsaturated hydrocarbons and sulphur, and has a density close to that of heavy crude oil. It was established that the pyrocondensate comprises paraffinic-naphthenic, cyclic aromatic, polycyclic and olefinic hydrocarbons, as well as oxygen-containing compounds. The gasoline fraction isolated from the pyrocondensate using the experimental unit is a clear, colourless liquid with a characteristic gasoline odour. Its fractional composition and properties are presented in Table 6.

Table 6. *Composition and characteristics of the gasoline fraction of pyrocondensate obtained from the pyrolysis of worn car tyres*

Parameter	Value
Pyrocondensate output	26.3% by mass
Density	0.786 g/cm³
Fractional composition:	
Initial boiling point	50℃
10% boiling point	90℃
50% boiling point	151℃
90% boiling point	190℃
Final boiling point	210℃
Sulphur content	0.05% by mass
Unsaturated hydrocarbon content	0.6% by mass

Source: developed by the authors

According to Table 6, it has been established that the fractional composition and density of the gasoline fraction are very close to the standard indicators for gasoline. It contains insignificant amounts of sulfur and unsaturated hydrocarbons, but according to the distillation curve, it has a boiling point that is 20-25℃ higher than that of automotive gasoline. The kerosene fraction obtained from pyrocondensate in the experimental setup is a transparent, colourless liquid with a characteristic kerosene odour. Its fractional composition and indicators are given in Table 7.

Table 7. *Composition and characteristics of the kerosene fraction of pyrocondensate from the pyrolysis of worn car tires*

Parameter	Value
Pyrocondensate output	8.2% by mass
Density	0.852 g/cm³
Fractional composition:	
Initial boiling point	180℃
10% boiling point	198℃
50% boiling point	221℃
90% boiling point	232℃

Table 7. *Continued*

Parameter	Value
Final boiling point	240℃
Sulphur content	0.08% by mass
Unsaturated hydrocarbon content	0.8% by mass

Source: developed by the authors

According to the main indicators (density, acceleration curve, sulphur content), the kerosene fraction obtained meets the requirements of DSTU 7687:2015 (2015). The kerosene fraction of pyrocondensate obtained from the pyrolysis of worn car tyres demonstrates a set of characteristics that make it possible to assess it as potentially suitable for use as a fuel component or as technical kerosene, subject to appropriate finishing treatment. The yield of the kerosene fraction is 8.2% by mass of the pyrocondensate. This is a relatively small but typical proportion for pyrolysis products of tyres, where a significant part is accounted for by the gasoline fraction, heavy distillates and resins. This yield indicates that the kerosene fraction is not the main target product of the process, but can be considered a valuable by-product that increases the overall energy and economic efficiency of pyrocondensate processing. The density of 0.852 g/cm^3 is within the range typical for commercial kerosene (approximately 0.78-0.86 g/cm^3 depending on the application – aviation, technical, lighting, etc.). This indicates: a sufficiently balanced hydrocarbon composition (no excessive amount of heavy components); potential compatibility with existing fuel systems in terms of density. This density level indirectly indicates a predominance of C10_{10}10–C14_{14}14 hydrocarbons characteristic of kerosene in the composition of the fraction. Distillation behaviour is key to assessing the suitability of a fraction for use as fuel. Boiling point: 180°C: this corresponds to the lower limit of the kerosene fraction and indicates the absence of excess volatile gasoline-type components that could lower the flash point and impair safety characteristics. 10% boiling off at 198℃ and 50% at 221℃: these values correlate well with the typical distillation characteristics of kerosene, reflecting a fairly narrow and technologically convenient boiling range. 90% evaporation at 232°C and end of boiling at 240°C: the small gap between 90% and the end of boiling indicates: relative homogeneity of the fraction, a small amount of heavy "tail" components, which has a positive effect on the completeness of combustion and reduces the formation of carbon deposits. Overall, the distillation curve shows that the distillation characteristics of this fraction are close to those of commercial kerosene, which is regulated by the relevant DSTU EN and ISO standards.

The sulphur content of 0.08% by mass is relatively low for pyrolysis products of tyres, which are traditionally characterised by an increased content of sulphur-containing compounds due to the composition of rubber compounds. This indicator is acceptable for most technical applications of kerosene; it may be close to the requirements of certain fuel standards or require minimal additional desulphurisation depending on the intended use (especially for environmentally sensitive aviation or energy fuels). Low sulphur content reduces the corrosive activity of the product and SOx_xx emissions during combustion. The unsaturated hydrocarbon content of 0.8% by mass is low, which has several important consequences: storage stability is improved (unsaturated compounds are prone to oxidation and the formation of resinous products); the risk of varnish and carbon deposits in fuel systems is reduced; this characteristic brings the obtained fraction closer to hydrotreated petroleum kerosene. The fact that the pyrolysis fraction has such a low unsaturated hydrocarbon content indicates the effectiveness of the membrane separation technology and the previous purification stages. Thus, in terms of key indicators (density, distillation curve, sulphur content), the kerosene fraction obtained meets the requirements (implicitly – the current standards for kerosene fuel). The analysed parameters confirm this: the density corresponds to the typical range for kerosene; the distillation characteristics indicate the formation of a "correct" boiling range without an excess of light or heavy impurities; the sulphur and unsaturated hydrocarbon content is acceptably low for technical and energy fuels.

Thus, based on the characteristics given, the kerosene fraction of pyrocondensate from the pyrolysis of worn car tyres can be considered potentially suitable for use: as a component of blended kerosene fuels after minimal refinement (if necessary, additional purification or mixing with petroleum fractions); as technical kerosene for power plants, boilers, combustion processes, etc. In addition, the results obtained demonstrate the promise of membrane technology for pyrolysis condensate processing as a tool for the maximum possible utilisation of tyre pyrolysis products in the fuel and energy cycle, which has both economic and environmental benefits (reduction of tyre waste and replacement of part of traditional petroleum resources). The diesel fraction is a transparent light-yellow liquid with a characteristic odour. Its fractional composition and quality indicators are presented in Table 8.

Table 8. *Fractional composition and indicators of the diesel fraction*

Parameter	Value
Pyrocondensate output	23.9% by mass
Density	0.889 g/cm³
Fractional composition:	
Initial boiling point	240℃
10% boiling point	264℃
50% boiling point	294℃
90% boiling point	327℃
Final boiling point	350℃
Sulphur content	0.12% by mass
Unsaturated hydrocarbon content	1.05% by mass

Source: developed by the authors

According to the main indicators (density, acceleration curve, sulphur content), the obtained diesel fraction meets the requirements of the standards. The sediment deposited in the receiving tank during the pyrocondensate separation process had a density of 1.242 g/cm^3 and a sulphur content of 4.47% by mass. It consisted of 45% pyrocarbon by mass (with a pyrocondensate yield of 9% by mass) and 55% heavy pyrolysis resin by mass (with a pyrocondensate yield of 11% by mass). The residue formed after the pyrocondensate separation process consisted of unsaturated hydrocarbons (with a pyrocondensate yield of 19.51% by mass), had a density of 0.85 g/cm^3 and contained 3.4% by mass of sulphur. As can be seen from Tables 6-8, the gasoline, kerosene and diesel fractions obtained as a result of membrane separation of pyrocondensate have a low content of sulfur and unsaturated hydrocarbons, which negatively affect fuel quality. To reduce the content of these components in fuel, technologically complex processes are used, such as hydrotreating and hydrogenation on highly active catalysts. The use of membrane pervaporation technology allows fractions with a low content of these ingredients to be obtained. These fractions can be used to produce high-quality automotive fuels. Most importantly, used car tyres are used in its production. In addition, thanks to complex mass transfer and diffusion processes that occur at the molecular level during pervaporation, pyrocarbon and heavy pyrolysis resin are removed from the pyrocondensate.

In membrane separation, an important characteristic of the membrane is its permeability. Knowing the permeability of the membrane, it is possible to calculate the productivity of the membrane apparatus. The permeability of the membrane during pervaporation depends on the temperature of the separation process; it increases with increasing temperature. At a certain temperature, the selectivity of the membrane φ (separation quality) begins to decrease. Thus, the calculated permeability of the membrane is taken to be the permeability it has at the highest temperature, provided that $\varphi = 1$. Figure 3 shows the dependence of membrane permeability on the temperature of the separation process of different fractions at $\varphi = 1$. As can be seen from the graphs in Figure 4, the optimal separation temperature is: 50°C for the gasoline fraction, 70°C for the kerosene fraction, and 85°C for the diesel fraction. The permeability at these temperatures is: for gasoline $m = 4.9$ kg/(m^2·h), kerosene fraction $m = 3.8$ kg/(m^2·h), diesel fraction $m = 2.5$ kg/(m^2·h).

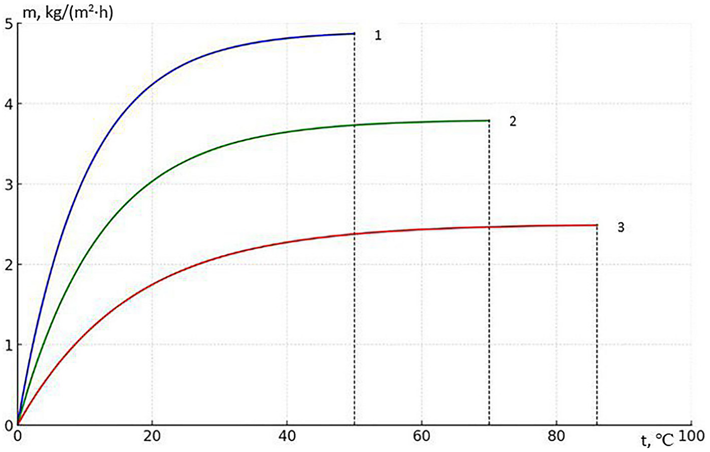

Figure 4. *Dependence of membrane permeability (m) on temperature for different fractions*

Note: 1 – gasoline fraction, 2 – kerosene fraction, 3 – diesel fraction

Source: developed by the authors

The sulphur content in fractions obtained during pyrocondensate separation is crucial when using them as commercial products. The main factor determining the sulphur content in fractions during the separation of pyrocondensate on pervaporation membranes is the temperature at which the process is carried out. Thus, studies were conducted on an experimental setup to investigate the effect of temperature on the sulphur content in fractions. Figure 5 shows the dependence of the sulphur content in the fractions on the temperature of the separation process, assuming that the selectivity is maintained at $\varphi = 0.9988 - 0.9995$ (relative to sulphur) and the maximum permeability of the membranes. As can be seen from the graphs, the sulphur content in the fractions obtained at a selectivity of 0.9988 to 0.9995 at maximum membrane permeability is: for the gasoline fraction 0.05% by mass; for the kerosene fraction 0.08% by mass; for the diesel fraction 0.12% by mass. When the optimum temperature of the separation process is reduced, the sulphur content in the fractions decreases slightly, but the permeability of the membranes decreases significantly. The content of unsaturated hydrocarbons in pyrocondensate reaches 20% by mass, which leads to the formation of resins. Thus, one of the main tasks in the membrane separation of pyrocondensate is to obtain fractions with a low content of unsaturated hydrocarbons. As studies show, the selectivity of membranes (in relation to unsaturated hydrocarbons) at maximum permeability is: for the gasoline fraction φ = 0.994; for the kerosene fraction φ = 0.9992; for the diesel fraction φ = 0.9897. Figure 6 shows the dependence of membrane permeability on temperature for unsaturated hydrocarbons.

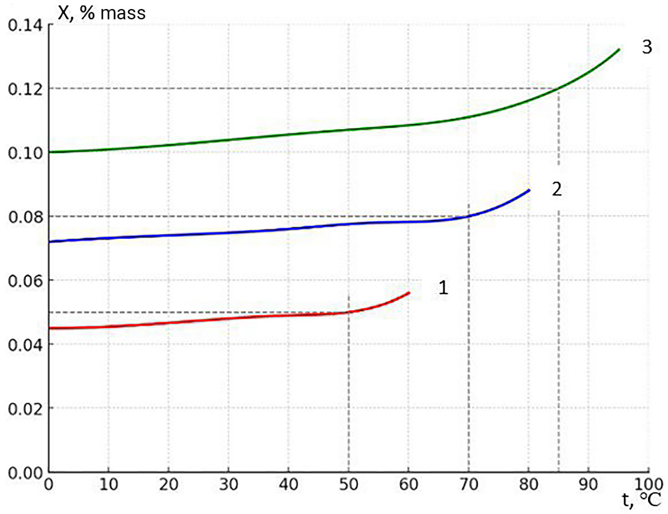

Figure 5. *Dependence of sulphur content in fractions on the temperature of the pyrocondensate separation process*

Note: 1 – gasoline fraction; 2 – kerosene fraction; 3 – diesel fraction

Source: developed by the authors

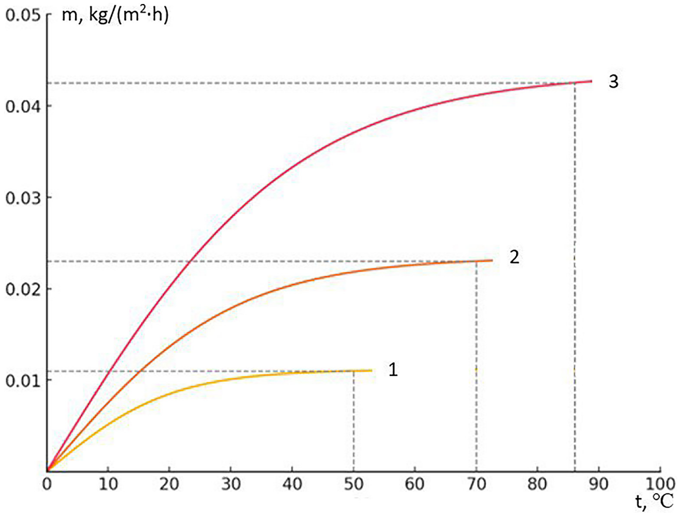

Figure 6. *Dependence of membrane permeability on temperature*

Note: 1 – membranes for gasoline fraction; 2 – membranes for kerosene fraction; 3 – membranes for diesel fraction

Source: developed by the authors

As can be seen from the graphs, the maximum permeability of membranes at optimal temperatures for unsaturated hydrocarbons is: for gasoline fraction membranes m = 0.0114 kg/(m^2·h); for kerosene fraction membranes m = 0.0224 kg/(m^2·h); for diesel fraction membranes m = 0.043 kg/(m^2·h). If the membrane permeability for the fractions is compared with the permeability for unsaturated hydrocarbons, it is 430 times higher for the gasoline fraction and 60 times higher for the diesel fraction than the permeability for unsaturated hydrocarbons. Such a large difference in permeability is due to the structure of the pervaporation membranes, on which their selectivity depends. This allows fractions with a very low content of unsaturated hydrocarbons to be obtained. The sulphur content in gasoline, kerosene and diesel fractions is influenced by the sulphur concentration in the pyrocondensate fed to the membrane apparatus for separation. The more sulphur in the pyrocondensate, the more sulphur in the fractions obtained by membrane separation. In this case, a "plasticising effect" occurs, which is associated with the fact that during the diffusion of saturated hydrocarbon molecules through the membrane, a very small amount of molecules of substances containing sulphur and sulphur compounds dissolved in these hydrocarbons can penetrate. Figure 7 shows the dependence of the sulphur content in the gasoline, kerosene and diesel fractions obtained by membrane separation of pyrocondensate on the sulphur content in the pyrocondensate fed to the membrane apparatus for separation.

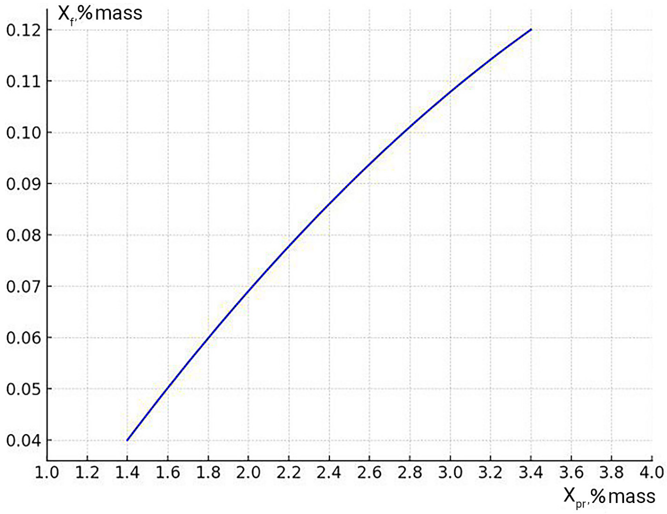

Figure 7. *Dependence of the sulphur content in the fractions X_f on the sulphur content in the pyrocondensate X_{pr}, fed to membrane separation*

Source: developed by the authors

During the separation of pyrocondensate in an experimental setup, in addition to gasoline, kerosene and diesel fractions, pyrocarbon sediment with heavy pyrolysis resin is also formed, as well as a residue containing unsaturated hydrocarbons. The sediment can be briquetted and used as fuel. The residue is a mixture of unsaturated hydrocarbons with sulphur-containing compounds, which can be hydrogenated on highly active catalysts to obtain saturated and aromatic hydrocarbons.

The results obtained in this work regarding the composition, properties and processing directions of pyrocondensate obtained in the process of pyrolysis of worn car tyres are generally consistent with the data presented in the works of other authors, but have a number of fundamental differences that determine the scientific novelty and practical value of the research. In the studies by K. Hrynyshyn *et al.* (2021), the complex multicomponent nature of pyrocondensate is noted, which contains aliphatic saturated and unsaturated hydrocarbons, aromatic and polycyclic aromatic compounds, sulphur-containing components, as well as pyrocarbon and resinous impurities. Similar conclusions are presented in the works of S. Magagula *et al.* (2023), which emphasise that it is the high proportion of unsaturated and aromatic hydrocarbons that causes the instability of pyrocondensate and limits its direct use as motor fuel. The data obtained in this work from the chromatographic analysis of gasoline, kerosene and diesel fractions confirm these patterns and indicate the presence of a wide range of C_5-C_{22} hydrocarbons with an increase in retention time as the molecular weight of the components increases.

The influence of temperature conditions and process parameters on the composition of pyrolysis products has been extensively studied in the works of L.M. Markina & M.S. Kryva (2018). In particular, it has been established that an increase in pyrolysis temperature contributes to the intensification of the destruction of the polymer matrix of tyres and an increase in the proportion of light aromatic hydrocarbons, but can lead to secondary condensation reactions and the formation of resins. Similar trends were found in this work: it was shown that with an increase in temperature and processing time, the content of unsaturated hydrocarbons decreases, with the most intense transformations occurring in the first two hours of the process, and a further increase in temperature does not significantly improve the characteristics of the final product. In a thorough review, S. You *et al.* (2018) analysed the current state and prospects of waste gasification from the perspective of obtaining synthesis gas and biochar, with a focus on high-temperature processes and their energy efficiency. The authors emphasised that the practical implementation of gasification is accompanied by the complexity of equipment design and limited possibilities for selective control of the composition of liquid by-products. In contrast to such approaches, the membrane separation of pyrocondensate proposed in this work is oriented towards mild process conditions and targeted fractionation of liquid products without the need for extreme temperatures.

At the same time, the majority of published studies are focused primarily on optimising pyrolysis conditions or on the subsequent catalytic treatment of pyrocondensate. In the works of S. Papari *et al.* (2021) and M. Mello *et al.* (2023), the main focus is on the processes of hydrotreating, desulphurisation and catalytic upgrading of liquid pyrolysis products. In a thorough review, S. Papari *et al.* (2021) analysed modern approaches to the pyrolytic processing of polymer waste to obtain fuels and chemical products, with a focus on the selection of temperature regimes, reactor types and catalysts. The authors showed that the improvement in the quality of liquid products is achieved mainly through catalytic effects and the intensification of thermochemical processes, while the issue of physical separation of pyrocondensate is practically not considered. In their work, M. Mello *et al.* (2023) conducted a detailed analysis of the pyrolysis processes of worn car tyres and the subsequent desulphurisation of pyrolysis oil. The authors focused on methods of hydrotreating, extraction and catalytic treatment aimed at reducing the content of sulfur-containing compounds, while noting the complexity of such technologies and their energy intensity.

D. Zheng *et al.* (2023) considered the targeted production of valuable aromatic compounds of the BTX fraction through control of pyrolysis conditions, whereas N. Lee *et al.* (2021) investigated the catalytic pyrolysis of polymer waste with emphasis on the influence of zeolite catalysts. The fundamental distinction of the present work lies in the application of membrane pervaporation technology for the separation of pyrocondensate into marketable fuel fractions with the simultaneous removal of pyrocarbon and heavy pyrolysis resin. In contrast to the conventional approaches described in the cited sources, membrane separation enables selective fractionation based on differences in molecular size and diffusion capacity of the components, thereby reducing the content of unsaturated and sulphur-containing compounds in the final fractions without the use of severe thermocatalytic conditions.

In addition, this work implements a comprehensive approach that combines membrane separation with subsequent catalytic oligomerisation of residual unsaturated hydrocarbons in the presence of the KTK-1 catalytic complex. Similar oligomerisation processes are described in the works of H. Ibragimov *et al*. (2013) and S. Boichenko *et al*. (2025), however, these studies do not consider preliminary membrane purification of pyrocondensate, which limits the selectivity of the process and control of the product composition. The results obtained in the current work indicate the high efficiency of combining catalytic treatment and membrane pervaporation technology for processing pyrocondensate from the pyrolysis of worn car tyres. The almost complete conversion of unsaturated hydrocarbons confirms the feasibility of using the catalytic stage as a preliminary purification stage, which minimises the formation of resinous products and increases the stability of the fuel fractions obtained.

Thus, the results obtained confirm the promise of membrane pervaporation technology as an effective tool for incorporating pyrolysis products from worn car tyres into the fuel and energy cycle, which has both economic and environmental advantages. Thus, the results of this work not only confirm the basic patterns established by other authors regarding the composition and properties of pyrocondensate, but also complement them by proposing an effective combined approach to its deep processing. The use of membrane technologies in combination with catalytic processes creates the conditions for obtaining stable fuel fractions with predictable physicochemical properties and increases the overall efficiency of waste tyre recycling.

Conclusions

The study proposes and experimentally confirms an effective method for processing pyrocondensate obtained from low-temperature pyrolysis of worn car tyres using highly permeable selective polymer membranes of the pervaporation type. It has been established that the average composition of pyrocondensate includes 58.4% by mass of hydrocarbons suitable for obtaining fuel fractions, about 20% of unsaturated hydrocarbons, approximately 20% of pyrocarbon together with heavy pyrolysis resin, and 1.6% by mass of sulphur. Membrane separation allows simultaneous separation of commercial fractions and precipitation of pyrocarbon with heavy resin directly in the apparatus, which greatly simplifies the technological scheme. The average yields of target products are 8.2% by mass of the gasoline fraction and 23.9% by mass of the total kerosene and diesel fractions, giving a total yield of fuel fractions of about 32.1% of the initial pyrocondensate.

The optimal process temperatures for achieving maximum membrane permeability with high selectivity are 50°C for the gasoline fraction with a permeability of 4.9 kg/(m^2·h), 70°C for the kerosene fraction with a permeability of 3.8 kg/(m^2·h) and 85°C for the diesel fraction with a permeability of 2.5 kg/(m^2·h). Under these conditions, the residual sulphur content in the fractions is 0.05% in the gasoline fraction, 0.08% in the kerosene fraction and 0.12% by mass in the diesel fraction, and the content of unsaturated hydrocarbons is significantly reduced due to the high selectivity of the membranes for these compounds. The fractions obtained in terms of fractional composition, density, distillation curve, sulphur content and stability meet the requirements of standards DSTU EN 228:2022 and DSTU EN 590:2022 and can be used as commercial fuel products or their components. The total degree of pyrocondensate processing, taking into account the catalytic refinement of unsaturated hydrocarbon residues and the use of sediment as fuel, reaches 96-98%. The process takes place under mild conditions, without aggressive reagents, with low energy consumption and minimal impact on the environment. Further research should focus on scaling up the technology, increasing the durability of the membranes and optimising their composition for industrial implementation.

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Conflict of Interest

None.

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Високопроникні селективні полімерні мембрани для розділення піроконденсату

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Анотація. Метою роботи було визначення оптимальних технологічних параметрів для отримання чистих фракцій вуглеводнів та вилучення пірокарбону та важкої піролізної смоли. В роботі використовувалася методика мембранного розділення піроконденсату за допомогою високопроникних селективних полімерних мембран. Встановлено, що середній склад піроконденсату, який містить до 58,4 % вуглеводнів, придатних для отримання бензинової, керосинової та дизельної фракцій, а також 20 % ненасичених вуглеводнів та 20 % пірокарбону з важкою піролізною смолою. Доведена ефективність мембранної технології для розділення піроконденсату з отриманням бензинової (8,2 %), керосинової та дизельної (23,9 %) фракцій з низьким вмістом сірки та ненасичених вуглеводнів. Характеристики одержаних фракцій відповідали вимогам стандартів для використання їх як готових товарних продуктів. Визначено оптимальні температури процесу розділення для кожної фракції: бензинової – 50 °C, керосинової – 70 °C, дизельної – 85 °C. Встановлено залежність проникності мембран від температури та селективності щодо ненасичених вуглеводнів та сірки. Процес розділення забезпечив одночасне висадження пірокарбону та важкої піролізної смоли, що спрощує технологію та підвищує її ефективність. Запропоновані каталітичні методи переробки осаду пірокарбону та залишку ненасичених вуглеводнів, що дозволяє досягти повної переробки піроконденсату від 96 % до 98 %. Підтверджено можливість використання мембранної технології для ефективної переробки піроконденсату з мінімальним впливом на навколишнє середовище. Розроблені аналітичні та експериментальні засади для розрахунку апаратів мембранного розділення та їхньої продуктивності. Запропоновані рішення можуть бути застосовані для виробництва високоякісних паливних фракцій з відпрацьованих матеріалів

Ключові слова: фракційний склад; піроліз; нафтополімерні смоли; каталітична обробка; дизельна фракція