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POLYMER WASTE AS A PROSPECTIVE RAW MATERIAL FOR THE PRODUCTION OF PETROLEUM PRODUCTS AND CONSTRUCTION MATERIALS

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Abstract. A comprehensive approach to the disposal of solid polymer waste through the technological processing into various types of fuel, lubricants and construction materials is provided. The proposed approach will lead to the improvement of the environmental situation in Ukraine (due to the reduction of the number of landfills). The proposed method of solid polymer waste processing includes their collection, sorting, cleaning and thermal processing.

Keywords: solid household waste; polymers; sorting; technological processing; destruction; compounding; motor fuels; plastic lubricants; building materials.

1. Introduction

The rapid development of the technosphere involves the production of a wide range of materials for industrial and domestic use, characterized by a high level of consumer properties. However, in the absence of certain recycling or reprocessing technology, these materials, after losing their consumer properties, accumulate in landfills and dumps, causing a negative environmental impact. Modern materials used today in households, construction, industry, automotive manufacturing, shipbuilding, electronics, and other fields include polymers. According to data provided in work[1], polymers make up approximately 16 % of municipal solid waste (MSW). It is known that the majority of generated polymer waste (around 93–96 %) does not undergo technological reprocessing or further reuse[2]. Additionally, in the context of the acute shortage of hydrocarbon resources in Ukraine, these materials can be considered as alternative (secondary) raw materials for the production of petroleum products and construction materials, which are currently in high demand.

According to data presented in work[3], approximately 40,000 tons of polymer waste accumulate annually in Ukraine, predominantly consisting of polyolefins in their morphological composition: high-density polyethylene (HDPE) and low-density polyethylene (LDPE) (a total of 35 %) and polypropylene (PP) at 12 %. Consequently, given their processability[4], substantial reserves, and the necessity of processing to reduce environmental impact[5], these polymer wastes represent a promising alternative raw material not only for recycling technologies but also for processing into components or directly into commercial petroleum products and various functional construction materials.

Thus, the aim of this study is to provide a general assessment of the resource and functional potential of secondary polymer raw materials accumulated in Ukraine, as well as to explore the potential for their technological processing into components or domestic commercial products that could reduce Ukraine's import dependence.

To achieve this research goal, it is necessary to formulate an approach to determining polymer raw materials suitable for the production of specific components or commercial products. This approach can be represented in the form of a structural flowchart, as shown in Fig. 1.

Thus, when choosing the polymer raw materials for the production of a component / product, it is essential to thoroughly investigate the technological possibilities of such production, the economic feasibility, and the environmental impact. Moreover, the economic feasibility will largely depend on the processability of the raw material, its ability to be recycled, the complexity of this processing, and the by-products (wastes) generated during production. From a technological standpoint, a key factor influencing the operational properties of the component / commercial product is the functional potential of its properties.

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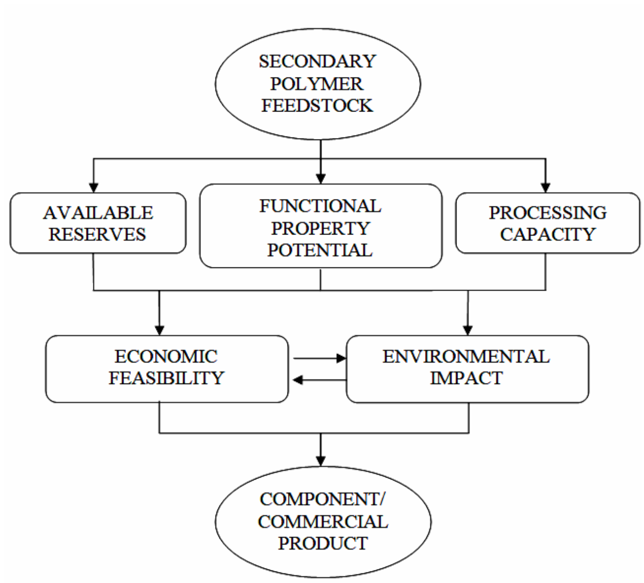


Fig. 1. Approach to determining the origin of polymer raw materials for component / commercial product production

The functional potential of properties can be represented as a set of characteristics of the raw material that forms a certain level of operational (consumer) properties of the component / commercial product. In turn, the level of operational properties of the resulting component or commercial product will consist of the potential of the raw material preserved during its processing and the properties acquired through the application of various technological methods, varying parameters (temperature, pressure, duration of the process) of processing, and the addition of additives (activators).

Among the functional potential properties of secondary polyolefin raw materials (HDPE, LDPE, PP), that are useful for the production of petroleum products, are the ability to undergo thermal, thermo-oxidative and thermomechanical degradation, decomposition temperature, and low content or absence of sulfur-containing and nitrogen-containing compounds. In the production of construction materials, the following indicators are the most significant: mechanical strength, water and chemical resistance, melting temperature, and compatibility with mineral fillers.

The technological processing of polymers into various components or commercial motor fuels is fundamentally based on the polymers' ability for thermal degradation. During this process, external thermal energy causes the rupture of C-C bonds, resulting in the formation of polymer macromolecule fragments and monomers[6].

In studies[7–14], it has been proposed to obtain products from polymer raw materials through thermal or thermocatalytic degradation at temperatures of 380–450 °C and pressures ranging from 0.1 MPa to 1.0 MPa, which can subsequently be used as components of motor fuels (automotive gasoline and diesel fuel).

Thermomechanical degradation of polymer raw materials is a way of producing the substance with a lower molecular weight than the original polymer and with different properties[15], which is a benefit in the production of plastic lubricants for various functional purposes[16].

One particularly attractive aspect of polymers when considering them as raw materials for motor fuel production is their low content or the absence of sulfur-containing and nitrogen-containing compounds, especially in polymers used in the food industry (packaging materials, disposable dishes, *etc.*). This is primarily due to the implementation of the Euro-5 environmental safety standard in Ukraine, which regulates sulfur content in motor fuel to no more than 10 ppm[17].

Water resistance and chemical resistance of polyolefin materials[18, 19] are among the most important characteristics considered when formulating recipes for the production of composite construction materials. Such materials are mainly used in industrial construction, particularly in oil refining plants and other enterprises in Ukraine's chemical industry, where acids, alkalis, and other aggressive substances are used in the technological process[20, 21].

The melting point of polymers determines the technological parameters of their processing, the related apparatus design of technological schemes, and production costs[22].

Compatibility of polymers with mineral fillers characterizes the spatial arrangement of particles of different sizes, interrelated voids, pores, and microcracks in the building material, ultimately determining its strength under external loads[23].

It should also be noted that the processing of polymers into commercial products, in most cases, involves their combination with industrial wastes such as used petroleum products, tires (which are produced in large quantities from urban transport), and ash residues from burning fuel (including fossil fuels) at thermal power plants.

It is known that used oils pose a serious ecological threat to the environment and human health, as they are hazardous pollutants of practically all components of the natural environment, surface and groundwater, soil-plant cover, and atmospheric air[24–27]. Used tires are a source of long-term environmental pollution[28]. Moreover, rubber is highly flammable and not biodegradable, and a pile of rubber tires is a convenient habitat for colonies of rodents and insects, many of which are sources of infectious diseases.

2. Experimental

2.1. Materials

Solid shredded (size 2×2 mm) polymers of LDPE, HDPE, PP, and PS types were used as raw materials for the synthesis of fuels, plastic mastics, protective coatings, and construction materials. Waste motor oils SAE10W-40, SAE15W-40, and waste transmission oils with viscosity grade SAE75W-140 were used as based oils for plastic mastics, protective coatings, and one of the components of construction materials. Secondary vulcanized rubber, waste automobile tires (WAT) were used as a filler for plastic mastics, while ash generated at thermal power plants (TPP) during heat energy generation was used for building materials.

The compound fuel samples were obtained based on commercial diesel fuel of the grade DP-3-Euro5-B7 and fuel oil of the grade 100.

2.2. Methodology of fuel fractions synthesis

A crucial stage in any processing technology, after sorting polymer raw materials by type, is their primary preparation. At this stage, polymers are shredded into particles not exceeding 2–3 mm in size, then washed to remove impurities and dried (moisture content reduced to 0.3 %). This stage is necessary both to enhance subsequent processing processes and to reduce the size of equipment dimensions. Next, the prepared raw material undergoes pyrolysis in a reactor (see Fig. 2) at a temperature of 400–450 °C and a pressure of 0.10–0.15 MPa. The prepared feedstock is processed in the pyrolysis reactor (Fig. 2) at a temperature of 400–450 ^{o}C and a pressure of 0.10–0.15 MPa.

After pyrolysis, the resulting liquid products were split into fractions at temperatures below 200 °C, 200–300 °C, and 300–360 °C. The compound fuel samples were obtained by adding fractions with boiling point ranges of 200–300 °C and 300–360 °C to commercial motor fuel (DP-3-Euro5-B7) and boiler fuel (mazut 100) in amounts up to 30 %.

The methodology for obtaining plastic lubricant and mastics (protective coatings) involved dispersing the polymer thickener in a dispersing medium using an ultrasonic disperser (UD-22/44, see Fig. 3) operating at a frequency of 44 kHz. Alternatively, dispersion was achieved using a paddle mixer at a speed of 500–700 rpm.

The temperature range for dispersion was maintained between 120–170 °C. After dispersing the thickener and allowing it to stabilize in the dispersing medium for 60 minutes, the process moved to the cooling stage to reach the working temperature of 25 °C. During this stage, the structure of the material starts to form.

The methodology for obtaining building materials involved mixing polymer raw materials, used oils, and ash at a temperature of 200–270 °C (depending on the polymer's properties, particularly its melting temperature) for 30–60 minutes. Subsequently, the resulting mixture was formed under a pressure of 15–30 MPa using a hydraulic press.

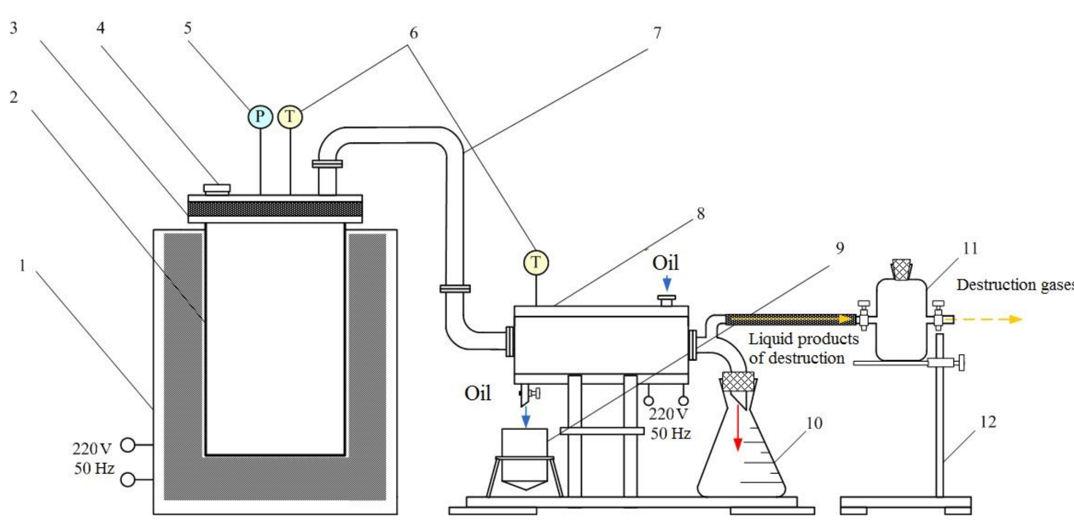


Fig. 2. Pyrolysis reactor: 1 – electric heater; 2 – metal glass; 3 – cap; 4 – safety valve; 5 – pressure controller; 6 – temperature controller; 7 – connecting tube; 8 – condenser-cooler; 9 – glass for oil; 10 – receiver container; 11 – glass-catcher; 12 – tripod

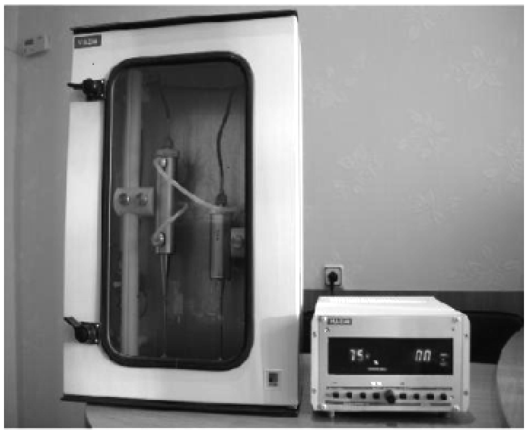


Fig. 3. An ultrasonic disperser UD-22/44

2.3. Research methods

Determination of physical and-chemical quality indicators of fuel fractions obtained from LDPE, HDPE, PP, and PS processing was carried out using standardized methods: density at 15 °C (DSTU IBO 12185); sulfur content (DSTU EN ISO 20884 and ASTM D 1555); ash content (DSTU EN ISO 6245); copper strip corrosion (DSTU EN ISO 2160); kinematic viscosity at 40 °C (DSTU GOST 33); cloud point temperature (DSTU ISO 3015); pour point temperature (ASTM D 97); cetane number (DSTU ISO 5165); fractional composition (ASTM D 86); closed cup flash point (DSTU 4455:2005); open cup flash point (GOST 4333).

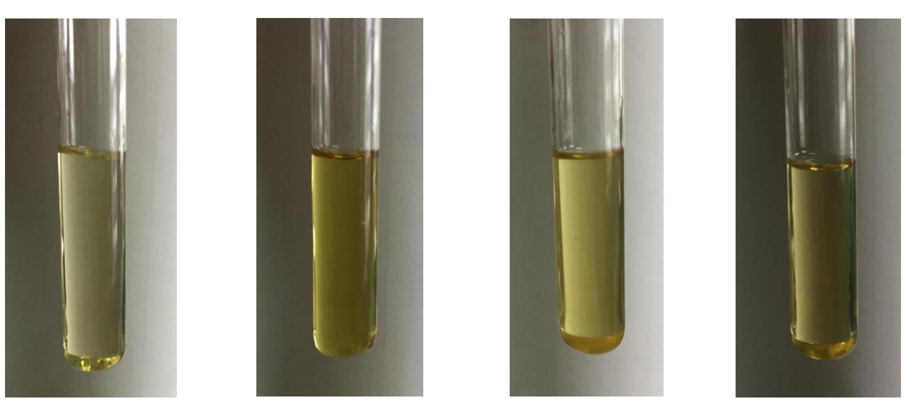
Determination of physical and-chemical quality indicators of plastic lubricants obtained from LDPE, HDPE, PP, and PS processing was conducted using standardized methods: drop point determination method (ASTM D566, GOST 6793); penetration determination method (ASTM D 1403-10(E), GOST 5436); effective dynamic viscosity determination method (ASTM D 1092); colloidal stability determination method (ASTM D 1742, GOST 7142); volatility determination method (ASTM D972, GOST 9566); lubricating properties determination method (DIN 5151 350/4, GOST 6037); water mass fraction determination method (ASTM D 95, GOST 2477); water resistance determination method (DIN 51 807/1); corrosion impact on metals (ASTM D4048, GOST 9.080); corrosion effects on metals (D1743); tribological characteristics using Four-Ball Wear Test (ASTM D 2266).

Determination of physical and-chemical quality indicators of protective coatings (mastics) obtained from LDPE, HDPE, PP, and PS processing was carried out using standardized methods: softening point temperature (ASTM D-36); elongation at 25 °C (ASTM D-113); penetration (ASTM D-5); water absorption after 24 hours (ASTM D570).

Determination of physical and-chemical quality indicators of construction materials obtained from LDPE, HDPE, PP, and PS processing was conducted using standardized methods: average density (ASTM C138 / C138M); compressive strength (ASTM C39 / C39M-21); moisture absorption (ASTM C1585-13); freeze-thaw resistance (ASTM C 666 (17)); coefficient of chemical resistance (ASTM STP 169).

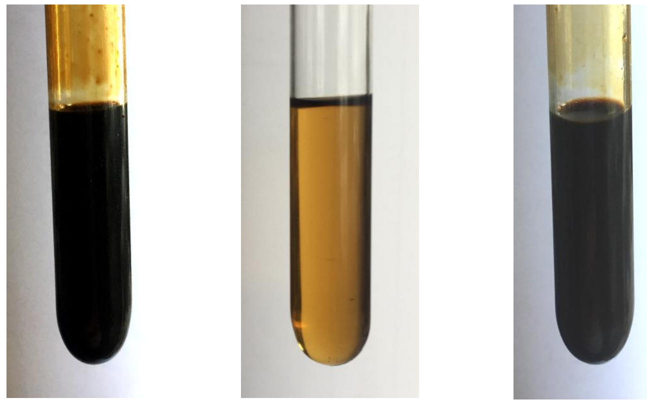
3. Results and Discussion

As a part of the current research, we obtained fuel fractions with boiling point ranges of 200–300 °C and 300–360 °C in laboratory conditions using the methods described above. Subsequently, the 200–300 °C fraction was added in amounts of up to 30 % to the commercial diesel fuel of the grade DF-3-Euro5-B7, while the 300–360 °C fraction was added to fuel oil of the grade 100. The appearance of the compound fuel samples is shown in Figs. 4, 5. The characteristics of the obtained compound fuels are presented in Tables 1, 2.



a b c d

Fig. 4. The appearance of the studied samples: a – DF-3-Euro5-B7; b – DF-3-Euro5-B7 + 10 % fraction 200–300 °C; c – DF-3-Euro5-B7 + 20 % fraction 200–300 °C; d – DF-3-Euro5-B7 + 30 % fraction 200–300 °C



a b c

Fig. 5. Appearance of the studied samples: a – mazut 100;
b – fraction 300–360 °C; c – mazut 100 + 30 % fraction 300–360 °C

Table 1. Characteristics of fuel compositions based on DF-3-Euro5-B7 and the 200–300 °C fraction

Parameter	Symbol	Units	Amount of the fraction in the fuel composition, %		
			10	20	30
Density at 15 °C	ρ^{15}	kg/m^3	824–826	819–823	815–818
Kinematic viscosity at 40 °C	ν^{40}	mm^2/s	2.71–2.88	2.50–2.70	2.22–2.43
Cetane number	CN	Units	50.5–52.5	49.0–50.0	45.5–48.0
Flash point	t_{fp}	°C	56–59	55–58	51–53
Autoignition point	t_{aip}	°C	295–300	288–296	280–293
Cloud point	t_{cp}	°C	–7÷–5	–11÷–7	–16÷–10
Corrosion of a copper plate (3 h. at 50 °C)	x_c	Class	1a–1b		
Sulfur content	S	ppm	<10		

Table 2. Characteristics of fuel compositions based on mazut 100 and the 300–360 °C fraction

Parameter	Symbol	Units	Amount of the fraction in the fuel composition, %		
			10	20	30
Density at 15 °C	ρ^{15}	kg/m^3	883–886	874–879	865–872
Conventional viscosity at 80 °C	CV^{80}	°E	4.75–5.08	3.60–4.11	2.45–2.70
Flash point	t_{fp}	°C	140–150	128–139	110–120
Autoignition point	t_{aip}	°C	295–300	288–296	280–293
Pour point	t_{pp}	°C	21–25	15–19	6–13
Operating heat of combustion	Q_o	kJ/kg	43300–43500	43550–43700	43600–44000
Sulfur content	S	ppm	400	280	170

Obviously, the commercial diesel fuel DF-3-Euro5-B7 is lighter and clearer compared to the fraction 200–300 °C (Fig. 4, *a*), while the compound fuel samples containing 10–30 % of the 200–300 °C fraction (Fig. 4, *b*, *c*, *d*) also exhibit a more intense color than the diesel fuel DF-3-Euro5-B7. However, in the absence of regulatory documentation that specifies color as a parameter, we believe that this is entirely acceptable.

Considering the appearance photograph (Fig. 5), the 200–300 °C fraction is clear and has a light brown color compared to mazut 100. Therefore, it does not impair or improve the color of the compound boiler fuel.

The conducted research showed that increasing the content of the 200–300 °C fraction in compounded diesel fuel from 10 % to 30 % leads to improvements in some parameters, while simultaneously deteriorating others.

For instance, the addition of the 200–300 °C fraction to diesel fuel reduces the cloud point temperature from –7 to –5 °C down to –16 to –10 °C. At the same time, there is a decrease in values of ρ^{15} (down to 815–818 kg/m³), ν^{40} (down to 2.22–2.43 mm²/s), and t_{aip}. (down to 280–293 °C). However, these changes either remain within acceptable limits for the parameters of commercial diesel fuel or are not normalized at all according to DSTU 7688:2015. The parameter x_c of the compounded fuels does not change with an increase in the content of the 200–300 °C fraction, regardless of the type of polymeric raw material.

Regarding parameters such as CN and t_{fp}, it should be noted that their values decrease to 45.5–48.0 units and 51–53 °C, respectively. These values are low and do not meet the requirements of DSTU 7688:2015. Thus, the aforementioned parameters can be used to determine the rational content of the 200–300 °C fraction in compounded diesel fuel at no more than 20 % by mass.

The addition of the 200–300 °C fraction can be utilized not only to expand the raw material base for the production of commercial diesel fuels but also to improve their ecological properties as well as the operational characteristics of substandard diesel fuels.

With an increase in the content of the 300–360 °C fraction in compounded heating fuel from 10 % to 30 %, there is an improvement in the quality indicators of the fuel (reduction in values of ρ^{15} (down to 865–872 kg/m³), CV^{80} (down to 2.45–2.70 °E), t_{pp} (down to 6–13 °C), S (down to 170 ppm), and an increase in Q_o (up to 43600–44000 kJ/kg)), exception for the t_{fp} parameter.

The value of the t_{fp} parameter decreases gradually from 0 % to 30 % mass in mazut VPF (300–360 °C) down to 110–120 °C, which is a negative aspect leading to increased fire hazards when using, storing, pumping, and transporting mazut. However, the t_{fp} values remain within acceptable limits according to the requirements of DSTU 4058-2001. Based on the aforementioned quality indicators, the 300–360 °C fraction can be used in quantities of up to 30 % for compounding with mazut grade 100 to obtain heating fuel with improved operational, particularly viscosity-temperature properties.

The use of waste petroleum products and used automotive tires together with secondary polymer raw materials allows for the production of a product with characteristics listed in Table 3.

Table 3. Characteristics of plastic lubricants (antifriction) with filler

Parameter	Units	Value
Base oil	–	Waste oil SAE10W-40, SAE15W-40, SAE75W-140
Polymer content	% mass.	5.0
Filler		Automobile tires
Filler content	% mass	5.0–20.0
NLGI compliance	Class	3/4/5
Dropping point	°C	135–223
Penetration at 25°C	$mm \times 10^{-1}$	125–235
Water-resistance	–	Stable
Colloidal stability	%	1.9–6.9
Corrosion effect on metals	–	Resistant (steel)

The obtained products (Table 3) are promising for use as antifriction plastic lubricants for lubricating friction units (such as rolling and sliding bearings) in industrial equipment and vehicles.

The lubricants produced can serve as substitutes for products like Mobilgrease AA N2, Greasrex D 60, Shell Retinax A, Shell Unedo grease 2,3, and Shell Alvanis grease R3.

Considering the direction of producing building materials, it should be noted that dispersing secondary solid melted polymer materials in waste petroleum products – lubricating oils (industrial, hydraulic, turbine, electrical insulation, motor, transmission) – can yield a product, the characteristics of which are presented in Table 4.

The properties of the product listed in Table 4 allow its use as a building material for protecting underground engineering communications, waterproofing building foundations, and as an adhesive for roofing, wooden and metal structures. The obtained product can serve as a substitute for bitumen-polymer mastics such as IZOFAST and ECOBIT.

Such a material can be applied by hot (140–170 °C) application onto surfaces that require protection from moisture or anticorrosive treatment. The application technology may involve immersing parts in a bath of heated material or using automatic spraying of the heated material onto the surface.

In addition, based on the obtained material, mastics can be produced that are convenient to use and can be applied to the surfaces of components and structures through brushing. For this purpose, stabilizing agents and a solvent need to be added to the obtained material, which, upon drying, will form a strong protective coating layer.

Another building material obtained by melting secondary polymer raw materials with mineral fillers that are impregnated with waste petroleum products is a solid material, the characteristics of which are presented in Table 5.

Table 4. Quality indicators of protective coatings (mastic)

Parameter	Units	Value
Polymer content	%	10–25
Softening point	°C	80–140
Penetration at 25 °C	$mm\times10^{-1}$	10–40
Extensibility at 25 °C	cm	2–6
Water saturation during 24 hours	%	0.02–0.12

Table 5. Characteristics of the obtained product

Parameter	Units	Value	
		Obtained sample	Hard concrete Class B35
Polymer content	%	20–35	–
Ash content	%	55–70	–
Used oil content	%	5–7	–
Dye content	%	1–3	–
Surface appearance	–	Uniform surface, no pores	Uniform surface, no pores
Average density	kg/m^3	1830	2370
Compressive strength	MPa	35	32.1
Water absorption	% mass	0.4	4.9
Frost resistance	cycles	550	300
Coefficient of chemical resistance	–	0.56	0.33

The conducted results showed that the proposed technology for producing polymer-based construction materials is suitable for the recycling of waste plastic lubricants and the production of a commodity product with a wide range of applications (such as paving slabs, drainage systems, covers, and wells for underground utilities, *etc*.).

The obtained product can be used in some cases as a substitute for heavy concrete B35.To enhance the appearance of the final product during the mixing stage of the prepared components into the composition, colorants can be added, and for increased strength, reinforcing materials (metal or fiberglass) can be used.

4. Conclusions

The conducted studies confirmed the perspective of using polymer waste (products made from HDPE, LDPE, and PP that have lost their consumer properties) as secondary raw materials capable of ensuring industrial volumes of production of commercial petroleum products and construction materials of various functional purposes. A comprehensive approach has been proposed to determine the feasibility of using polymer raw materials in the production of specific products.

It has been substantiated that the functional potential of properties, which is a combination of the raw material's characteristics that match certain operational (consumer) requirements, is a key factor in determining the raw materials for the production of petroleum products and construction materials. This approach, which is based on defining the resource and functional potential of polymer waste properties, aligns with the principles of the circular economy, where waste is seen as a potential input material for new manufacturing processes rather than being discarded.

The practical implementation of the proposed comprehensive approach to the recycling of solid polymer waste that continuously arises in Ukraine will increase the recycling rate of solid household waste while improving the overall environmental situation. The properties of polymer raw materials that determine the methods and modes of their technological processing have been considered, based on which a methodology for producing components for motor and boiler fuels, anti-friction plastic lubricants, waterproofing and anti-corrosion mastic, and solid composite building material has been proposed.

Experimental studies are implemented in the production and analysis of the characteristics of products obtained from both the thermodestructive processing of polymer waste and the joint thermodestructive processing of polymer waste with waste lubricating oils and used tires. It was defined that increasing the content of the 200–300 °C fraction in the compounded diesel fuel from 10 % to 30 % contributes to a decrease in the cloud point from –7 to –5 °C, down to –16 to –10 °C, which is a positive aspect. However, this leads to a reduction in the cetane number (CN) to 45.5–48.0 and the flash point (t_{fp}) to 51–53 °C, which, according to the requirements of DSTU 7688:2015, limits the content of the 200–300 °C fraction to no more than 20 %.

Increasing the content of the 300–360 °C fraction in the compounded boiler fuel from 10 % to 30 % improves its physicochemical properties: the density (ρ^{15}) decreases to 865–872 kg/m³, the viscosity at 80 °C (CV^{80}) reduces to 2.45–2.70 mm^2/s, the pour point (t_{pp}) decreases to 6–13 °C, the sulfur content (S) decreases to 170 ppm, and the calorific value (Q_o) increases to 43,600–44,000 kJ/kg. Therefore, according to the requirements of DSTU 4058-2001, the recommended content of the 300–360 °C fraction will be up to 30 %.

During the experimental research, anti-friction (NLGI 00/0/1/2/3) plastic lubricants were obtained that can replace lubricants such as NYCO 65 VASELINE (TECHNICAL PETROLATUM (GREASE)), Mobilgrease AA N2, Greasrex D 60, Shell Retinax A, Shell Unedo grease 2,3, and Shell Alvanis grease R3; a protective waterproofing material with a softening point of 80–130 °C and water saturation of 0.02–0.12 %, serving as a substitute for bitumen-polymer mastics of IZOFAST and ECOBIT brands; a building material that has a density 1.3–1.5 times lower, 4–5 % less water absorption, and a compressive strength 2.0–3.0 MPa higher compared to heavy concrete (class B35), and a chemical resistance coefficient (0.56), which allows its use as a material for containers for storing used lubricating oils, oil residues, oil sludges, and acidic asphalts.

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ПОЛІМЕРНІ ВІДХОДИ ЯК ПЕРСПЕКТИВНА СИРОВИНА ДЛЯ ВИРОБНИЦТВА НАФТОПРОДУКТІВ І БУДІВЕЛЬНИХ МАТЕРІАЛІВ

Анотація. *Розглянуто комплексний підхід до утилізації твердих полімерних відходів через їхню технологічну переробку на різні види паливно-мастильних і будівельних матеріалів. Запропонований підхід сприятиме покращенню екологічної ситуації в Україні (завдяки зменшенню кількості полігонів і сміттєзвалищ). Цей комплексний підхід щодо поводження із твердими полімерними відходами полягає в їх збиранні, сортуванні, очищенні та термічному переробленні.*

Ключові слова: *тверді побутові відходи, полімери, сортування, технологічна переробка, деструкція, компаундування, моторні палива, пластичні мастила, будівельні матеріали.*