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POLYSTYRENE WASTE PROCESSING BY THE LOW-TEMPERATURE PYROLYSIS METHOD

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The process of low-temperature pyrolysis of polystyrene waste was studied. A laboratory batch reactor was used at a temperature of 400^{0}C. As a result of pyrolysis, 86.8 wt.% of pyrocondensate, 7.0 wt.% of solid residue, and 6.2 wt.% of gaseous products were obtained. The resulting pyrocondensate was fractionated into a gasoline fraction with a boiling range of up to 200^{0}C, a diesel fraction with a boiling range of 200–350^{0}C, and a residual fraction with a boiling point above 350^{0}C. The main physicochemical properties of each fraction were determined to establish possible routes for their practical application. The gasoline fraction of the pyrocondensate is characterized by a high content of unsaturated hydrocarbons and can be used as a commercial gasoline component after hydrogenation and catalytic reforming to increase the octane number. The diesel fraction also contains a significant amount of unsaturated compounds and, after hydrogenation, can be used as a component of diesel fuel. Additionally, it exhibits good low-temperature properties, with a pour point below –20^{0}C. The remaining pyrocondensate after separation can be used as an additive to fuel oil or as a feedstock for other types of fuel compositions.

Keywords: waste, polystyrene, pyrolysis, pyrocondensate, gasoline fraction, diesel fraction, residue.

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Introduction

Every year, a large amount of solid household and industrial waste is generated worldwide, posing a serious environmental threat. Polymer waste is a significant and hazardous component of total waste, accounting for 5–8% of the total volume according to various statistics [1,2]. Polymers are almost non-biodegradable and persist in the environment for decades. They accumulate in landfills and illegal dumps, and also end up in reservoirs, rivers, and oceans, causing significant damage to ecosystems.

The main methods for disposing of polymer waste include incineration [3], recycling or secondary processing [4,5], and pyrolysis, i.e. a thermal decomposition at high or low temperatures to produce liquid and gaseous products [6–8]. Most often, pyrolysis products (particularly pyrocondensate) are used as fuel components for boilers or furnaces, which however reduces the efficiency and economic benefit of such technology [9]. In our opinion, a promising direction is the production of motor fuel components from polymer waste.

Analyzing global trends in the production and consumption of polymeric materials, it can be concluded that polyethylene, polypropylene, and polystyrene reach the largest volumes. Accordingly,

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these polymers make up the bulk of plastic waste. Studies of the pyrolysis of polyethylene and polypropylene have been reviewed in detail in scientific works [10,11], while the thermal decomposition of polystyrene has been studied much less. Therefore, the purpose of our research was to study the features of polystyrene waste pyrolysis.

Experimental

To study the low-temperature pyrolysis of polymer waste, the ground electrical boards were used. Their characteristics are given in Table 1.

A comprehensive thermal analysis of polystyrene waste samples was carried out on Derivatograph Q-1500 of the Paulic, Paulic-Erday system (Hungary), connected to a personal computer. The samples were analyzed using a dynamic mode at a heating rate of 10^0C per minute in an air atmosphere. The sample mass was 100 mg. Aluminum oxide served as the reference substance.

The laboratory setup, consisting of a sealed metal reactor, a water cooler, and a receiving flask, was used for low-temperature pyrolysis of thermoplastic waste. A thermocouple was used to control the reactor temperature. At the initial stage, a portion of the weighed polymer raw material was loaded into the reactor and sealed with a bolted lid. A cooler was connected to the gas outlet tube, then an electric heating element was turned on, and the circulation of cooling water was started. The temperature was gradually raised to the level needed for the pyrolysis process. The pyrocondensate formed during the reaction was condensed in the cooler and fed to the receiving flask. Gaseous products that did not condense (pyrogases) were released into the atmosphere. After the process was completed (the flow of liquid products into the flask stopped), the heating was turned off. The amount of pyrocondensate was determined by weighing the receiving flask before and after the experiment. The reactor was also weighed after cooling to calculate the yield of solid residue.

The pyrocondensate separation into fractions was carried out on a standard laboratory setup used for the distillation of light petroleum products. The setup included a flask, a flask heater, a water cooler, a spider-type fraction collector with a set of receivers for collecting narrow fractions. The sampling temperature was measured with a thermometer, fixing the vapor temperature at the inlet to the cooler.

Analytical studies were performed on pyrocondensate, its fractions, and solid residue using standard, generally accepted methods. The fractional composition of the light fractions was determined using an ARNS-type apparatus. The iodine number of liquid products was determined according to the Margoshes method. Additionally, the flash point in open and closed cups, freezing and clouding points, density, and refractive index of both pyrocondensate and its individual fractions were determined. The chemical elemental composition of solid and liquid pyrolysis products was determined by X-ray fluorescence spectral analysis on Elvax Light SDD.

Results and discussion

To determine the optimal temperature range for polystyrene waste pyrolysis, derivatographic studies were performed. The obtained thermogram of a polystyrene waste sample (Fig. 1) shows an endothermic effect with a maximum at 139^0C in the temperature range of $120-180^0$C, which is not accompanied by noticeable mass loss. This effect corresponds to the melting process of polystyrene. A further decrease in the sample mass (by 29.23%) at the temperature range of $272-385^0$C is accompanied by a barely noticeable exothermic effect on the DTA curve. This indicates intensive destructive processes in the polymer, as well as thermal oxidation of its decomposition products. The intensive reduction in the mass of a polystyrene waste sample at the temperature range of $385-506^0$C corresponds to the polystyrene pyrolysis stage. In the same temperature range, combustion of products formed during decomposition occurs, accompanied by a pronounced exothermic effect on the DTA curve with a maximum at 461^0C. At this moment, a sharp extremum is recorded on the DTG curve, the maximum of which is at 414^0C.

The results of the thermal analysis were used to select the pyrolysis temperature of polystyrene waste for further experiments, which was chosen at 400^0C. Pyrocondensate, a small amount of solid residue (pyrocarbon) in the reactor, and gaseous decomposition products were formed under these conditions. The material balance of the polystyrene waste pyrolysis process is presented in Table 2.

Table 1

Characteristics of polystyrene waste prepared for pyrolysis

Parameter	Value
color	dark gray
density, kg/m^3	1035
melting point, ^{0}C	174
melt flow index (MFI) (210^0C; 5 kg), g/10 min	5.6
Brinell hardness, HB, MPa	130

The pyrocondensate obtained by pyrolysis of polystyrene waste is a transparent liquid with a hue from light yellow to dark brown, with a characteristic smell typical of thermally decomposed polymer products. The characteristic parameters of pyrocondensate are given in Table 3.

An increased iodine number indicates a significant amount of unsaturated hydrocarbons in the pyrocondensate. The high refractive index, typical of aromatic compounds, indicates the predominance of aromatic structures. This also agrees with the low freezing point of pyrocondensate. In addition, the resulting liquid has a relatively low flash point, which limits its direct use as boiler fuel.

Figure 2 shows the standard distillation curve of pyrocondensate, which reflects its fractional composition. The start of boiling is observed at 72^{0}C, which is significantly higher than the initial boiling of most standard oil samples.

To assess the suitability of pyrocondensate obtained from the pyrolysis of polystyrene waste as a potential raw material for fuel production, it was subjected to fractional separation. During the distillation process, two main distillate fractions were isolated, with boiling points below 200^{0}C and 200–350^{0}C, as well as a residual fraction with a boiling point above 350^{0}C (Table 4). Further studies were carried out separately for each isolated fraction.

According to the research results, it was found that the fraction with boiling points below 200^{0}C contains a small amount of sulfur and a high amount of unsaturated hydrocarbons. The refractive index indicates a significant proportion of aromatic compounds in this fraction, which causes its rather high octane number. The yield of the b.p. 200^{0}C fraction was higher than that of most similar pyrocondensates described in the literature [11].

As can be seen from the data in Table 5, the pyrocondensate fraction boiling between 200–350^{0}C has a high flash point, which agrees with the requirements for diesel fuels. Among its positive parameters are a very low sulfur content and favorable low-temperature properties (low cloud and pour points). The only drawback of this fraction is the increased concentration of unsaturated hydrocarbons, confirmed by the iodine value.

The residual fraction of pyrocondensate with a boiling point above 350^{0}C is a viscous substance visually similar to a lubricant (Table 6). Its composition includes a significant proportion of unsaturated

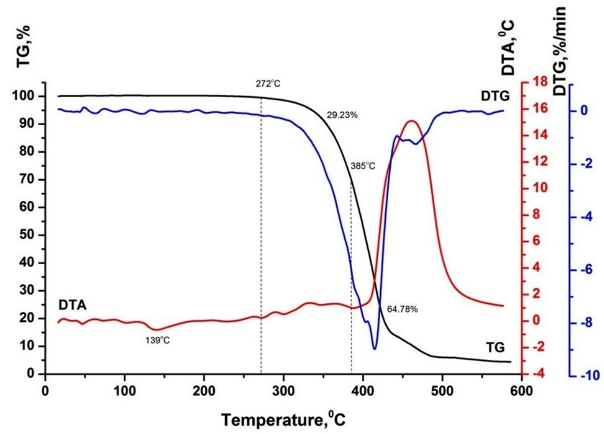

Fig. 1. Thermograms of a polystyrene waste sample

Table 2

Material balance of the polystyrene waste pyrolysis process

Raw materials and products	Amount, wt.%
Uploaded: – polystyrene waste	100.0
Obtained: – gas and losses – pyrocondensate – residue (pyrocarbon)	 6.2 86.8 7.0
Total	100.0

Table 3

Characteristics of pyrocondensate from polystyrene waste pyrolysis

Parameter	Value
density, kg/m^3	835
refractive index, n_D^{20}	1.5397
total sulfur content, wt.%	0.006
iodine number, g I_2/100 g	64.8
freezing point, ^{0}C	<–20
flash point: – open cup, ^{0}C – closed cup, ^{0}C	 66 45

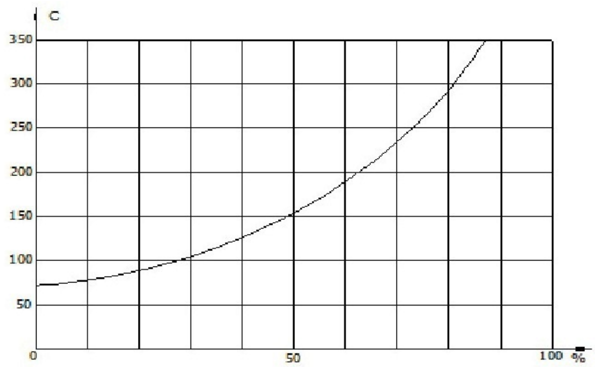

Fig. 2. Standard distillation curve for pyrocondensate from polystyrene waste pyrolysis

Table 4

Characteristics of the b.p. 200⁰C pyrocondensate fraction from the polystyrene waste pyrolysis

Parameter	Value
yield relative to pyrocondensate, wt.%	62.6
density, kg/m^3	783
refractive index, n_D^{20}	1.5275
fractional composition, ^{0}C:	
– initial boiling	72
– 10% distilled	91
– 50% distilled	140
– 90% distilled	187
– end boiling	201
total sulfur content, wt.%	0.004
iodine number, g I_2/100 g	73.4

Table 5

Characteristics of the 200–350⁰C pyrocondensate fraction from the polystyrene waste pyrolysis

Parameter	Value
yield relative to pyrocondensate, wt.%	24.6
density, kg/m^3	842
refractive index, n_D^{20}	1.5324
iodine number, g I_2/100 g	65.1
fractional composition, ^{0}C:	
– initial boiling	195
– 10% distilled	223
– 50% distilled	295
– 90% distilled	340
– 98% distilled	351
total sulfur content, wt.%	0.007
cloud point, ^{0}C	–12
freezing point, ^{0}C	<–20
flash point in closed cup, ^{0}C	56

Table 6

Characteristics of the residue >350⁰C after pyrocondensate distillation from polystyrene waste pyrolysis

Parameter	Value
yield relative to pyrocondensate, wt.%	12.8
density, kg/m^3	937
refractive index, n_D^{20}	1.5479
total sulfur content, wt. %	0.014
iodine number, g I_2/100 g	57.2
freezing point, °C	+20
flash point:	
– open cup, ^{0}C	126
– closed cup, ^{0}C	103

hydrocarbons, confirmed by its high iodine value. However, the low-temperature properties of this fraction, in particular the pour point, limit its use as a fuel oil component.

In general, the positive parameters of pyrocondensate obtained by pyrolysis of polystyrene waste include a high yield of gasoline fraction at a b.p. of 200⁰C, low sulfur content, and low pour point. The main drawback of this pyrocondensate is the significant content of unsaturated hydrocarbons in its composition.

X-ray fluorescence spectral analysis was used to determine the content of individual chemical elements in pyrocondensate, its isolated fractions, and residue (Table 7). The study results show that the metal content in the pyrocondensate is insignificant. Compared to the pyrocondensate obtained by pyrolysis of polyethylene waste, it is somewhat higher but significantly lower than the pyrocondensate from polypropylene waste pyrolysis. Technologically important impurities include Cr, Cu, Mo, and Pb, while impurities such as Ca and Fe are also fixed in the composition. The content of heavy metals, such as V and Ni, which are particularly harmful to oil refining processes, was minimal. Studies have also shown that during distillation, a significant portion of metals is concentrated in the residue, which is important to consider when choosing the optimal ways for its further use.

Conclusions

Studies have shown the fundamental possibility of processing polystyrene waste by low-temperature pyrolysis to produce liquid and solid products suitable for use in the production of motor fuels and other industrial materials. Pyrolysis of polystyrene waste at a temperature of 400⁰C allows obtaining 86.8 wt.% of pyrocondensate, 7.0 wt.% of solid residue and 6.2 wt.% of pyrolysis gas.

It was found that the light fractions of pyrocondensate, after separation, can be used as raw materials for the production of commercial motor fuels. This requires additional hydrogenation, which reduces the content of unsaturated hydrocarbons and also increases the octane number of the gasoline fraction. The pyrocondensate residue after fractionation can be used as a component of fuel oil or as an additive to plastic lubricants. The solid residue formed during low-temperature pyrolysis of oil sludge is advisable for use in road construction as the lower layers of the coating.

Table 7

Content of individual chemical elements in the pyrocondensate of polystyrene waste pyrolysis and its individual fractions

Element	Element content, ppm			
	pyrocondensate	i.b. 200^0C fraction	$200–350^0C$ fraction	residue $>350^0C$
Ca	20.9	12.6	30.3	43.4
V	0.1	0.1	0.1	0.1
Cr	1.7	1.5	1.8	2.5
Mn	0.1	0.1	0.1	0.1
Fe	1.8	0.8	2.6	5.2
Ni	0.3	0.3	0.3	0.3
Cu	12.6	7.4	10.9	41.3
Zn	1	0.5	1.5	2.5
Ba	0.1	0.1	0.1	0.1
Mo	4.4	3.4	4	10.1
Pb	1	0.7	1.2	2.1

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ПЕРЕРОБКА ВІДХОДІВ ПОЛІСТИРОЛУ МЕТОДОМ НИЗЬКОТЕМПЕРАТУРНОГО ПІРОЛІЗУ

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Було здійснено дослідження процесу низькотемпературного піролізу відходів полістиролу. Експеримент виконували на лабораторній установці періодичної дії при температурі 400^{0}С. У результаті піролізу вдалося одержати 86,8 мас.% піроконденсату, 7,0 мас.% твердого залишку та 6,2 мас.% газоподібних продуктів. Одержаний піроконденсат піддавали фракціонуванню, виділяючи бензинову фракцію з температурним інтервалом кипіння до 200^{0}С, дизельну фракцію 200–350^{0}С та залишкову частину з температурою кипіння понад 350^{0}С. Для кожної з фракцій було визначено основні фізико-хімічні властивості з метою встановлення можливих напрямів їх практичного застосування. Бензинова фракція піроконденсату характеризується підвищеним вмістом ненасичених вуглеводнів і може бути використана як складова частина товарних бензинів після проведення процесів гідрування та каталітичного риформінгу для підвищення октанового числа. Дизельна фракція також містить значну кількість ненасичених сполук і після гідрування може застосовуватись як компонент дизельного палива. Крім того, вона має добрі низькотемпературні властивості – температура застигання становить менше ніж мінус 20^{0}С. Залишкова частина піроконденсату після розділення може бути використана як добавка до мазуту або сировина для інших видів паливних композицій.

Ключові слова: відходи, полістирол, піроліз, піроконденсат, бензинова фракція, дизельна фракція, залишок.

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