




Investigation of the influence of the physicochemical characteristics of waste on the quality of liquid fuel products from them, obtained by multi-loop recirculation pyrolysis

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ABSTRACT

Purpose: To study the properties of MSW generated in Ukraine in order to ensure their most efficient processing using the multi-loop recirculation pyrolysis technology in order to produce a liquid fuel product.

Design/methodology/approach: The problem of study of MSW accumulation in the world countries was carried out on the basis of scientific databases PubMed, ScienceDirect, Mendeley, ResearchGate, Google Scholar for the corresponding keywords. The MSW properties generated in Ukraine were determined in accordance with the current standards to determine the specific properties of materials. For the experimental study, 3 types of solid organic waste mixtures were compiled, the composition of which characterizes the variable data on the accumulation of household and industrial waste of Ukraine. The quantity and quality of the liquid product obtained was determined using certified equipment.

Findings: The tests revealed that the composition of the solid organic waste mixture when disposing of them by the multi-loop recirculation pyrolysis method has little effect on the composition and properties of the obtained liquid hydrocarbon product; the total time of complete degradation for different mixtures of composite materials (mixtures of polymeric substances) is 180-250 min; the correct selection of the temperature of the circulation system makes it possible to produce a gaseous or liquid product which can then be used as an alternative fuel.

Research limitations/implications: Current study of solid organic waste degradation was carried out by the multi-loop recirculation pyrolysis method. The experimental waste mixtures did not contain very wet waste of small fractions.

Practical implications: The technology of circulating pyrolysis makes it possible to change the qualitative and quantitative composition of the obtained liquid hydrocarbon product, which is a potential fuel, irrespective of the composition of the solid organic waste mixture.

Originality/value: It has been experimentally established that the quality of the liquid hydrocarbon product obtained by the multi-loop recirculation pyrolysis process does not depend on the composition of the solid organic waste mixture, but depends on the process' duration and the third circuit' temperature of the multi-loop recirculation system.

Keywords: Solid organic waste, Multi-loop recirculation pyrolysis, Experimental study, Liquid fuel

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CLEANER PRODUCTION AND BIOTECHNOLOGY

1. Introduction

In urban areas of most countries of the world, the problem of generation and accumulation of municipal solid waste (MSW) has been an urgent problem for many years [1-3]. Some of factors accelerating the waste generation are the increasing urbanization [4,5], uncontrolled consumption [5,6]. In countries with weak economies and in developing countries, a significant part of MSW is thrown away haphazardly or accumulated in open landfills [1,7]. Waste disposal methods at landfills are ineffective, put increasing pressure on the quality of land [7,8], air [7], water [9], and pose serious threats to the quality of the local environment and public health. At the same time, it is known that MSW has an energy potential [10-12] and can be used to obtain alternative fuels [12-14]. In particular, the technology of multi-loop circulation pyrolysis deserves attention [15], since this technology combines in its structure a reactor for the primary (traditional) pyrolysis of organic waste and a multi-loop circulation system (MCS) that ensures the separation of the primary steam-gas mixture (SGM), which comes from the reactor of primary pyrolysis into the MCS circuits. In the MCS, the steam-gas mixture is divided into two pure components: high-energy pyrolysis gas and low-molecular-weight liquid fuel with a molecular weight of 100...200 [14,16].

In order to use the MSW energy properties as efficiently as possible in the recycling process and ensure the environmental friendliness of this process with the production of a high-quality fuel product, it is necessary to study its physical and chemical properties, quantitative and qualitative composition as a raw material.

Thus, the purpose is to study the properties of MSW generated in Ukraine in order to ensure their most efficient processing using the multi-loop recirculation pyrolysis technology in order produce a liquid fuel product.

2. Materials and methods

The problem' study of MSW accumulation in the world countries was carried out on the basis of open information sources, in particular, the search was carried out in the scientific databases PubMed, ScienceDirect, Mendeley, ResearchGate, Google Scholar for the corresponding keywords.

The morphological composition was determined in accordance with the methodological recommendations [17]. The MSW was unloaded from the containers and the individual components were sorted into plastic bags with a capacity of 20 litres. Then the mass of each sorted component and the total mass of solid waste were determined by weighing. Taking the total mass of municipal solid waste as 100%, the percentage of each component was calculated. To determine the fractional composition, the MSW was sieved through a sieve with aperture size of 15x15 mm.

The specific heat capacity of the waste components was determined as:

$$C_{MSW} = 21.9W + 2000, \quad (1)$$

where C_{MSW} is the specific heat, J/(kg·°C); W is MSW humidity, %.

The moisture content of the waste was determined for elementary samples taken from the waste mixture by drying to constant weight at a temperature of 105°C, namely:

- an elementary sample weighing 100 g, weighed (error less than 0.1), dried to constant weight in a drying apparatus, weighed again;
- humidity was calculated using the formula:

$$W = \frac{(m_1 - m_2)}{m_2} 100, \% \quad (2)$$

where m_1 is the mass of the elementary sample before drying; m_2 is the mass of the elementary sample after drying to constant weight.

For the experimental study, 3 types of solid organic waste mixtures were compiled, the composition of which characterizes the variable data on the accumulation of household and industrial waste, taking into account the place of their accumulation, the season and other factors. The waste was crushed to a size of $0.05 \times 0.05 \times 0.1$ m. The amount of the obtained pyrolysis gas was measured using a gas meter GSB - 400 GOST 28724 - 90. The absolute error in measuring the volume did not exceed ± 0.5 cm³. During the experiment, an average sample was taken into the gas meter for chromatographic analysis. The control over the content of harmful substances in the air of the working area and the resulting products is also carried out in accordance with GOST 12.1.005. The amount of liquid product withdrawn from each circuit was measured on a Tefal analytical balance with an absolute error of ± 0.01 g. To determine the qualitative composition of the liquid fuel obtained at different temperatures in the experimental plant, samples were taken and analysed on a NeoCHROM chromatograph.

The average molecular weight of the liquid product obtained was calculated according to the formula of B.P. Voinov:

$$M_{av} = 60 + 0.3t_{av} + 0.001t_{av}^2, \quad (3)$$

where t_{av} is average product boiling point.

3. Results and discussion

3.1. Study of the MSW composition

Considering MSW as a raw material for obtaining an alternative fuel, it should be noted that the waste mixture contains components with a high energy potential and a low energy potential (low-calorie). The first group is formed by various organic compounds containing carbon (C), hydrogen (H), nitrogen (N), sulphur (S), oxygen (O), as well as those organic compounds that form volatile substances during destruction. These include, for example, synthetic polymers (all types of plastics, rubber, worn-out car tires, substandard oil products, etc.). The second group should include natural polymers, which are characterized by the presence of oxygen, high humidity and are composed of cellulose. The amount and combination of these two waste groups affects the quality characteristics of the resulting fuel products. Therefore, by ensuring the correct sorting and a different approach to the waste destruction of the first and second groups, can be get better quality fuel products.

The energy properties of MSW are determined by the calorific value of the components. The calorific value of MSW depends on their moisture content, ash content and density, and the MSW mixture density is characterized by morphological and fractional composition. The main factors affecting the morphological composition of MSW are the climatic zone, season, lifestyle of the population and others.

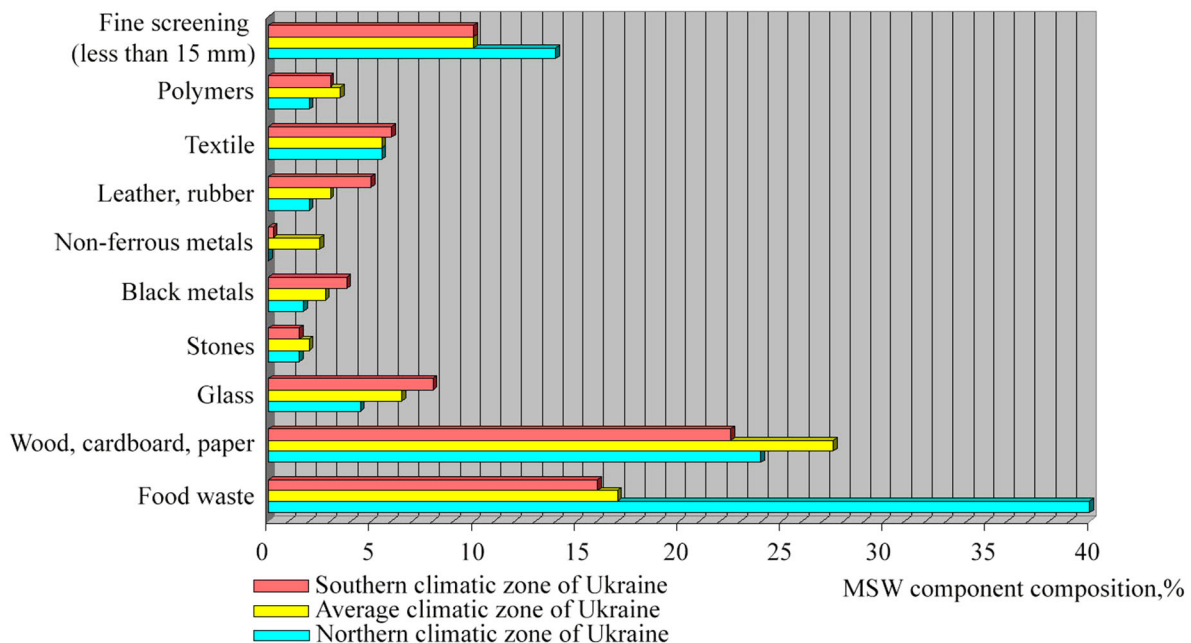


Fig. 1. MSW component composition in different climatic zones of Ukraine (averaged values)

Determination of the MSW morphological composition for different climatic zones of Ukraine was carried out in Poltava (middle zone), Nikolayev (southern) and Kiev (northern). Studies have shown that, as a rule, the waste composition is not the same by season (Figures 1, 2). Similar studies have been obtained by other researchers [18,19].

Observations show that in the spring the food waste content is 20-25% and increases in the fall to 40-55%, which can be explained by the consumption of a large amount of vegetables and fruits in the diet. In winter and autumn, the content of fine screening (less than 15 mm) decreases from 20 to 7% in cities in the southern zone and from 11 to 5% in cities in the middle zone [5]. Consequently, the energy potential of the MSW mixture will also vary depending on the season. This is due to the fact that food waste, as a rule, has a size of less than 50 mm and is characterized by fine fractions in the composition of the so-called "tails". At the same time, packaging materials (paper, cardboard, wood, etc.), as a rule, have a size of more than 150 mm and characterize the coarse fraction in the composition of dry waste. Consequently, a significant predominance of food waste will reduce the energy potential of the MSW mixture, and it is necessary to approach the disposal of such a mixture different from the disposal of a MSW mixture in which a significant amount of packaging materials prevail.

The MSW bulk, approximately 70-90%, is represented by fractions up to 150 mm, while less than 2% of waste is

represented by fractions of more than 350 mm. Analysis of the morphological composition of individual fractions shows that about 90-95% of food waste from a mixture of solid waste, more than 50% of the total amount of paper and cardboard, as well as about 95% of the total amount of glass, metal, stones and bones belong to the fine fraction, that is, less 150 mm. So, the larger the fraction, the smaller the amount of wet waste and vice versa.

Humidity and ash content reduce the calorific value. Moisture is an indicator that affects not only the calorific value, but also the stability during storage. Ash content is the content of the non-combustible part as a percentage, which is created from mineral impurities of the fuel during its complete combustion. Figure 3 shows the moisture content of the MSW components and their specific heat capacity, and Figure 4 shows elemental composition and calorific value of MSW components.

Mass calorific value characterizes the amount of heat that is released when a unit of mass is completely processed. If the chemical composition of the waste mixture is known, then the mass calorific value can be determined theoretically by the formula of D.I. Mendeleev [20], which has the form:

$$Q = 339(C) + 1030(H) - 109(O - S) - 25(W), \quad (4)$$

where Q is mass calorific value, kJ/kg; C, H, O, S, W is content of carbon, hydrogen, oxygen, sulphur, moisture, %.

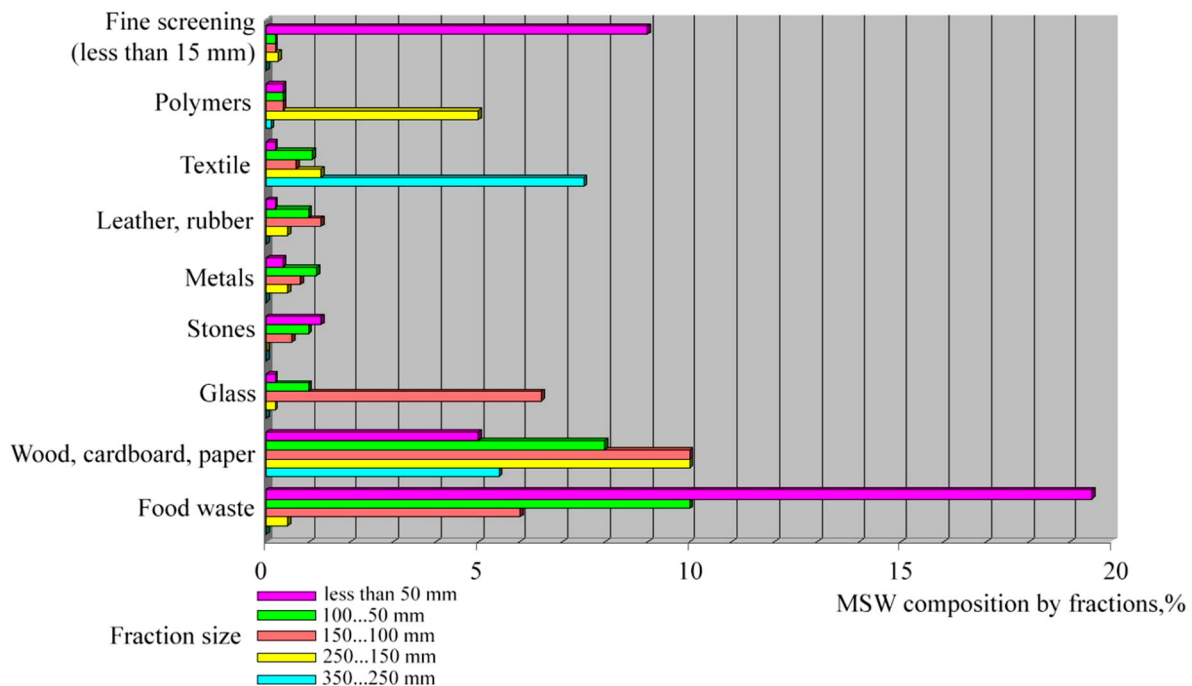


Fig. 2. MSW fractional composition (averaged values)

Thus, solid waste generated in Ukraine can be conditionally divided into two groups: Group I – high-

energy waste; Group II – low-energy waste, the heat of combustion of which does not exceed 30 MJ/kg (Tab. 1).

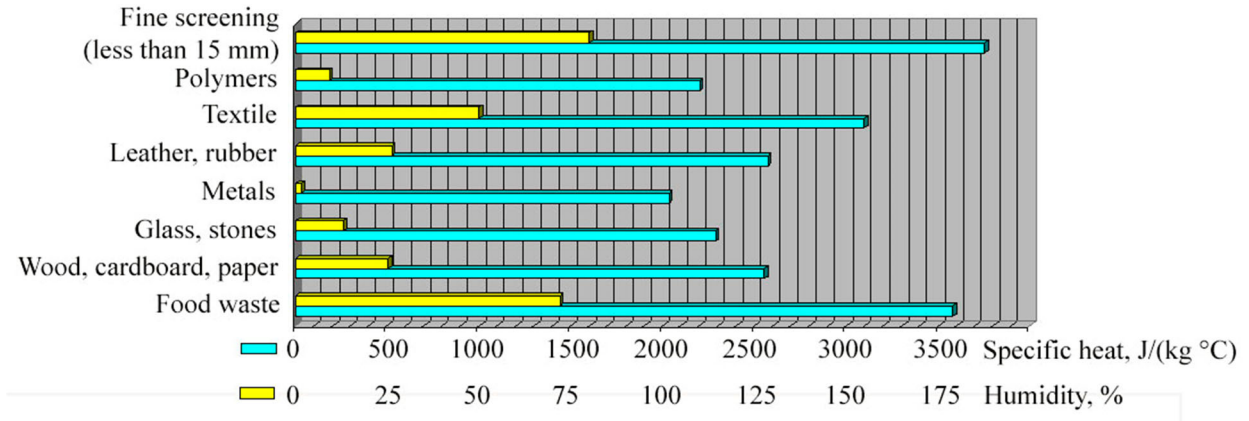


Fig. 3. Specific heat capacity of MSW components

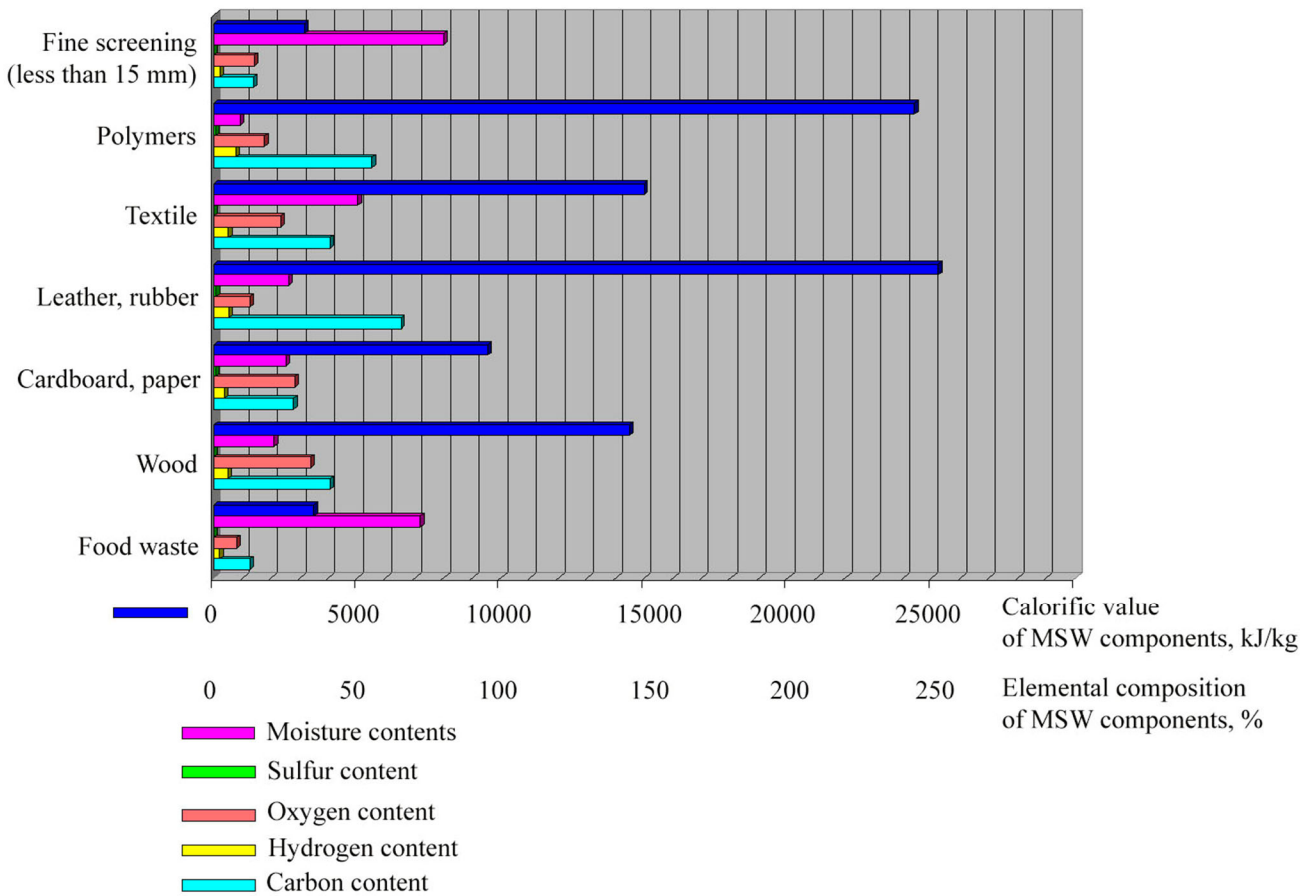


Fig. 4. Elemental composition and calorific value of MSW components

Table 1.
High energy and low energy waste

Group I	Group II
10-20% polyethylene	
3-6% polystyrene	2-7% rubber
5-10% polypropylene	20-25% worn out tires
10-15% polyethylene terephthalate	20-30% paper
up to 10% oil sludge and waste oils	2-5% textiles, rags
up to 2% polyvinyl chloride	

3.2. Description of the recommended technology of multi-circuit recirculation pyrolysis for the MSW processing

At present, the pyrolysis of MSW is the most studied environmentally friendly and demanded technology. For the disposal of organic waste, this process is carried out in a sealed reactor without air oxygen or with a limited amount of it [21-24] that makes it possible to obtain gaseous and liquid fuel products [25,26].

The disadvantage of standard pyrolysis technological processes is the difficulty of determining the necessary technological process modes that would ensure the effective destruction of all waste with different physicochemical characteristics in a single stream. Analysing the literature on pyrolysis products of organic waste, it was found that without additional chemical treatment, liquid pyrolysis products cannot be used in industry [27]. As a rule, it is required to install gas cleaning devices to reduce sulphur and ash emissions [28-31], as well as additional processing of liquid pyrolysis products to ensure the required performance of standards [32]. Consequently, this disadvantage prevents the achievement of the required depth of destruction of the entire mixture of high molecular weight organic waste.

The technology of multi-loop recirculation pyrolysis [15] is free from such a disadvantage (Fig. 5). Its essence lies in the achievement of deep destruction of the entire mixture of organic wastes due to recirculation and gradual decomposition of heavy fractions of intermediate products formed during the primary pyrolysis of waste [33].

Products of such waste processing can be light fractions of liquid fuel, pyrolysis gas and solid residue. Carrying out the process with recirculation allows you to get one hundred percent conversion of raw materials is impossible without recirculation through the thermodynamic restrictions imposed on the process. In accordance with the theory of recirculation, the change in the reactor power will occur due to the action of three factors: the amount of recirculated, the concentration of components in the total volume of the reactor and mass transfer [34,35].

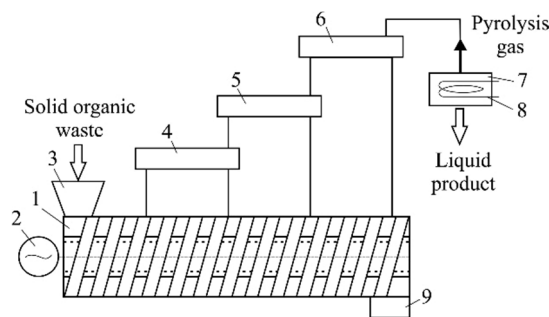


Fig. 5. The multi-loop recirculation pyrolysis scheme: 1 – reactor; 2 – electric drive; 3 – device for loading waste; 4 – the 1st circuit; 5 – the 2nd; 6 – the 3rd circuit; 7 – output capacitor; 8 – condenser water cooling system; 9 – device for unloading pyrocarbon; 10 – screw feed of waste

The difference between multi-loop recirculation pyrolysis from the known pyrolysis technology lies in the presence of several circuits. So at the exit from the pyrolysis reactor, the vapour-gas mixture enters the circuits of the multi-loop recirculation system and is subjected to sequential cooling with decreasing temperature on each circuit, starting from the first. Due to forced cooling, heavy fractions of the vapour-gas mixture condense and flow by gravity into the primary pyrolysis reactor, where they undergo secondary destruction, and the light fractions pass to the next circuit [36].

On each circuit, by cooling, a heavy liquid fraction with a lower molecular weight than in the previous circuit is separated from the vapour-gas mixture, polycondensation reactions proceed, forming a heavy liquid fraction from the heaviest components of the vapour-gas mixture, which is returned to the reactor by gravity.

An important physical phenomenon is the contact of the heavy liquid fraction with the primary and subsequent mixture, which promotes heat and mass transfer. As a result of which the heavy liquid fraction is preheated by hotter outgoing streams. Since the heavy liquid fraction and the vapour-gas mixture are not equilibrated in temperature, an exchange of matter will begin between them to establish equilibrium.

At each such contact, the low boiling component of the heavy liquid fraction passes into a vapour-gas mixture, and from the vapour-gas mixture, the high boiling component passes to the heavy liquid fraction. Such an exchange of components is repeated at each section of the multi-loop circulation system, which contributes to the generation of an almost pure low-boiling component of the vapour-gas mixture, which in the initial water-cooled condenser is separated into pyrolysis gas and a liquid fraction with a molecular weight of 100...200.

Table 2.
Composition of the solid organic waste mixtures for experimental research

No. of organic waste mixture	Type of solid organic waste	Composition	Components content, % wt.*
1	Mix of packaging materials	Disposable dishes, wrappers, boxes, boxes	30% PE 55 % PP 15 % PET
2	Packing tape in various grades	Waste industrial scotch rolls	100 % PE
3	Mix of organic dry household waste	Textile waste, toys, furniture parts, packaging	15% PE 25 % PP 30 % PS 15 % PETE

Note: *PE – polyethylene; PP – polypropylene; PS – polystyrene; PETE – polyethylene terephthalate

Table 3.
The main waste types presented by the mixtures

Solid organic waste	Substance
Detergent bottles, transparent packaging and packages, waterproofing film, electrical insulating shells, device housings, worn and remnants of water and gas pipes, ticket pots, children's toys, textile fibres	PE
Food containers, packaging, car components, waterproofing products, sewer pipes and drainage pipes, textile fibres, shut-off valves, watering hoses	PP
Disposable tableware, packaging materials, children's toys, insulation, soundproofing, filtration and finishing materials, disposable medical instruments, textile fibres	PS
Bottles for soft drinks, electrical insulating materials, car and engine body parts, compressors and pumps, textile fibres, packaging materials, sound and heat insulating materials	PETE, PC

The positive features of this technology, in contrast to other thermal methods of waste disposal, are the following [33]:

- the generated gas has a high calorific value and a high content of useful combustible substances such as methane, propane, butane, etc.;
- almost complete MSW utilization is achieved;
- power autonomy of the entire technological cycle.

3.3. Experimental study results of the MSW organic components destruction by the multi-loop recirculation pyrolysis method

In previous tests of the technology [37,38], it was found that the process duration and required temperature regime of the multi-loop recirculation system are depending on the raw material composition. Also, the characteristics of different organic waste types affect the fuel products' amount, while for different research waste mixtures the difference in the total fuel products' amount is 7-9%.

To analyse the possibility of joint utilization of an organic wastes mixture with different physicochemical properties, as well as to study the influence of various mixture components on the qualitative and quantitative

composition of the obtained fuel products, model mixtures were drawn up (Tabs. 2-3). Organic waste mixtures prepared for the experimental study correspond to the average statistical data on the formation and accumulation of solid organic domestic and industrial waste in Ukraine (excluding wet waste). The mixtures were compiled on the basis of the variable data on the accumulation and storage of domestic and industrial organic waste, taking into account the places of their formation and collection and accumulation. For the study, we used the formulas of the chemical composition of solid organic waste according to [39, 40] (Tab. 4).

Table 4.
Gross-formula of the investigated components of the organic waste mixture

Fractional composition	Gross-formula
Food waste	C320.3H570.9O188.4N14.9S
Waste paper	C580.6H952.3O440.8N3.49S
Textile	C978.8H1396O416.8N70.2S
Tree	C1321H1904O855.6N4.6S
Polymers	C3.5H5.0O1S
Leather, rubber	C404.4H634.9O58.1N57.2S

Table 5.
Liquid product sampling results*

No. of the organic waste mixture	No. of the liquid product sample	T, °C	t, min	Composition	Liquid product' properties
1	1	100	145	n-alkanes 12.913% Isoalkanes 3.329% Cycloalkanes 6.901% Alkenes 37.485% Dienes 2.501% Arenes 9.793% Others** 27.078%	- average boiling point of hydrocarbons 209.75 °C - curing temperature -39 °C - viscosity at 20 °C 1.4-1.9 cST - cetane number 42 - average molecular weight 185 - light fraction - flash point 24 °C
	2			150	165
2	3	124	225	n-alkanes 22.198% Isoalkanes 0.668% Cycloalkanes 5.1375% Alkenes 13.848% Dienes 2.553% Arenes 3.001% Others** 52.595%	- average boiling point of hydrocarbons 311.37°C - curing temperature -19°C - viscosity at 20 °C 1.4-1.9 cST - cetane number 38 - average molecular weight 250 - flash point 26 °C
	4			165	155
3	5	100	165	n-alkanes 8.337% Isoalkanes 2.622% Cycloalkanes 10.13% Alkenes 24.79% Dienes 8.88% Arenes 6.649% Others** 38.592%	- average boiling point of hydrocarbons 197.05°C - curing temperature -45 - viscosity at 20 °C 1.4-1.9 cST - cetane number 36.8 - average molecular weight 158
	6			160	225

Notes:

*volume and properties of pyrolysis gas obtained were not determined, since only the liquid product has been studied in the present study. Pyrolysis gas was burned on an additional burner.

**chemical components, the amount of which does not exceed 0.001% of the total amount, and which have not been identified

In the first experimental study on the destruction of organic waste, 6 kg of organic waste mixture 1 was loaded. The controlled parameters were: process duration t ; SGM temperature at the outlet from the third circuit to the condenser T . The temperature on the third circuit characterises steady-state operation (stationary operation) of equipment. In the second experimental study, 6 kg of organic waste mixture 2 was loaded into the reactor. After 55 min. after the start of the process a sharp increase in temperature T was observed. In the third experimental study of the degradation of organic waste mixture 3, 6 kg of waste was loaded. The sampling results of obtained liquid products, their qualitative composition and properties are presented in Table 5. Dependence of hydrocarbons' quantity in obtained liquid fuel on temperature change at the outlet (on the third circuit) is presented in Figures 6-8.

3.4. Discussion of the obtained liquid products' properties

The results presented in Table 5 show that the liquid product obtained at $T = 100^{\circ}\text{C}$ and 150°C (liquid product sample 1 and 2 respectively) has low molecular weight corresponding to light fractions. Taking into account that the working temperature in reactor is $580\text{--}590^{\circ}\text{C}$ it may be stated that thermal destruction process is effective enough. A sample of liquid product 3 also has a low molecular weight, but its value greater than 200, which corresponds more to diesel than to gasoline. It is positive that the resulting liquid products contain sufficient light hydrocarbon fractions to provide a low flash point. This indicates that processing of similar waste mixture by multi-loop recirculation pyrolysis removes lighter fractions within 3 hours,

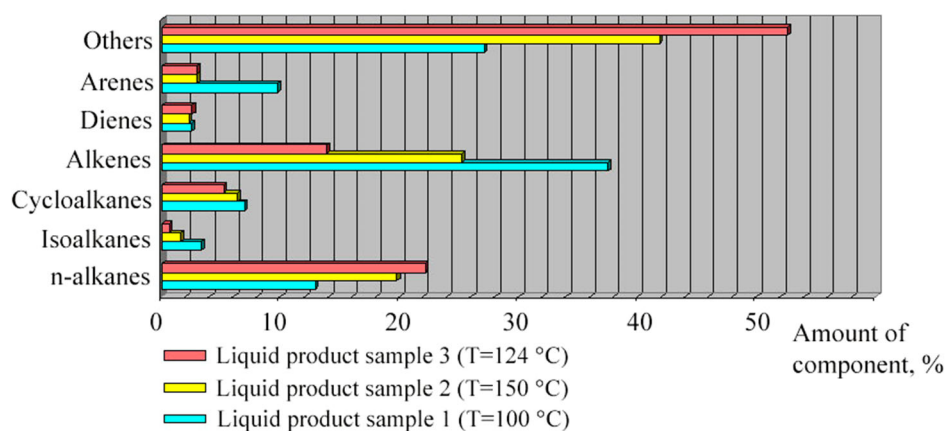


Fig. 6. Organic waste mixture 1. Dependence of quantitative and qualitative yield of hydrocarbons on temperature on the multi-loop recirculation system' third circuit

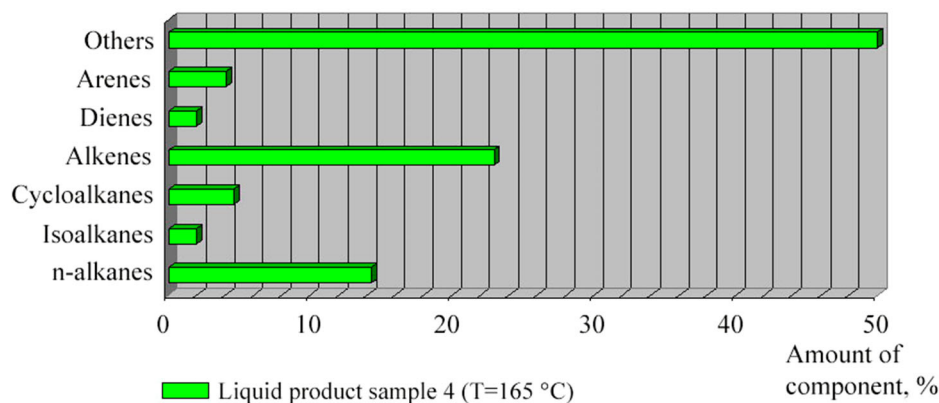


Fig. 7. Organic waste mixture 2. Dependence of quantitative and qualitative yield of hydrocarbons on temperature on the multi-loop recirculation system' third circuit

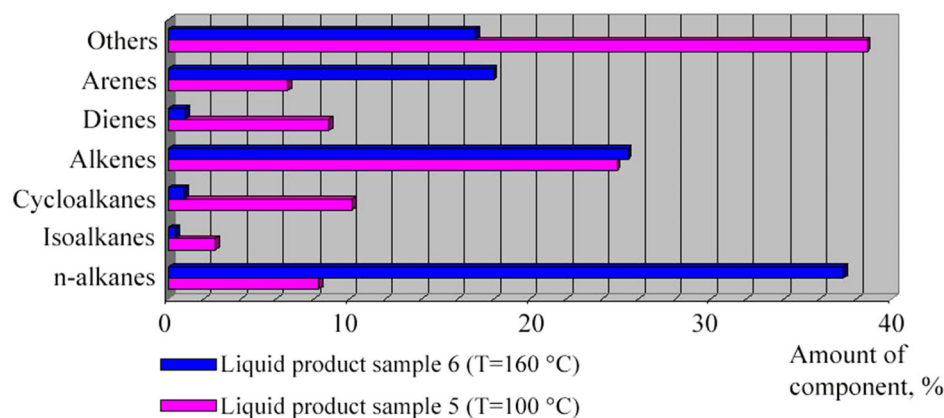


Fig. 8. Organic waste mixture 3. Dependence of quantitative and qualitative yield of hydrocarbons on temperature on the multi-loop recirculation system' third circuit

the amount of which depends on the temperature on the third circuit T and the duration of the process. Consequently, if the recirculation process continues, a liquid fuel with a molecular weight greater than 250 is produced, corresponding to the heavier fractions. In the study of mixture 1, it was found that there was no increase in the molecular weight of the resulting liquid product after the reactor reached steady-state mode. A change in the degree of waste degradation, and consequently in the molecular weight of the resulting liquid fuel, can be achieved by changing the temperature on the circuits of the multi-loop recirculation pyrolysis system.

Figure 6 shows that the highest yield of normal paraffins at process temperature 124°C ; isoalkanes, cycloalkanes, unsaturated and aromatic hydrocarbons at 100°C ; the average amount of all formed hydrocarbons at temperature 150°C . At the same time, Figure 7 shows that at 165°C unsaturated hydrocarbons predominate in the liquid product, and the content of aromatic hydrocarbons exceeds the norm for alternative fuel by 3%. Figure 8 shows that with increasing temperature T and process duration t the amount of formed normal paraffins, saturated and aromatic hydrocarbons increases, while at temperature of the third circuit equal to 100°C a greater yield of isoalkanes, cycloalkanes and dienes is observed.

The results are plotted (Figs. 9 and 10), which show the dependence of the amount of carbohydrates on the temperature on the third circuit and the duration of the process.

Figure 9 shows that the concentration of isoalkanes, cycloalkanes and dienes does not change significantly in the temperature range $100\text{--}165^{\circ}\text{C}$. The concentration of normal paraffins (n-alkanes) increases sharply with increasing temperature up to 160°C . The largest number of unsaturated hydrocarbons (alkenes) is formed at 100°C , with increasing

temperature up to 124°C their concentration sharply decreases, and when reaching $T = 150^{\circ}\text{C}$, the number of alkenes increases and stabilizes. The formation of aromatic hydrocarbons (arenes) does not change significantly, and at $T = 160^{\circ}\text{C}$ their concentration sharply increases and then gradually decreases.

The hydrocarbon concentration in the decomposition of solid organic waste by multi-loop recirculation pyrolysis is directly influenced by the duration of the process. Figure 10 shows that the amount of normal paraffins varies with temperature, the lowest yield of n-alkanes is observed between 100-145 minutes, then their amount starts to increase. Isoalkanes are formed by polymerization, they tend to increase octane number, with time of BHP process their quantity slowly decreases, the highest yield of isoalkanes is recorded at the beginning of reactor reaching steady-state condition. Fluctuations in the number of cycloalkanes are also observed: increasing at 145 minutes of the process but decreasing after three hours of running the process. The number of alkenes remains stable after the reactor reaches a steady state at 22-25%. The increase of dienes is observed at 145 min of the process and decreases with time. At 95 minutes into the process, the liquid product contains a large amount of aromatics, but as the process duration increases to 200 minutes, the number of aromatics decreases and then increases again.

Considering that the technology of circulating pyrolysis makes it possible to change the qualitative and quantitative composition of the obtained liquid hydrocarbon product, which is a potential fuel, by changing the temperature on the third circuit T , further search for the optimum equipment regime for obtaining a liquid hydrocarbon product whose properties are close to the properties of conventional fuel for internal combustion engines or other needs is required.

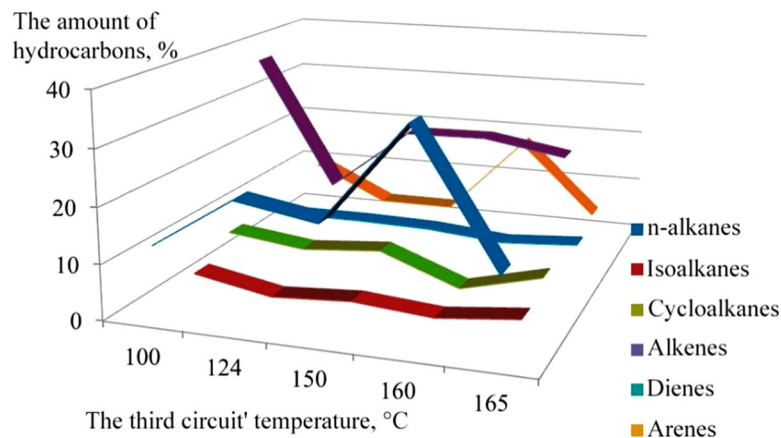


Fig. 9. Dependence of the hydrocarbons' amount in the resulting liquid product on the third circuit' temperature

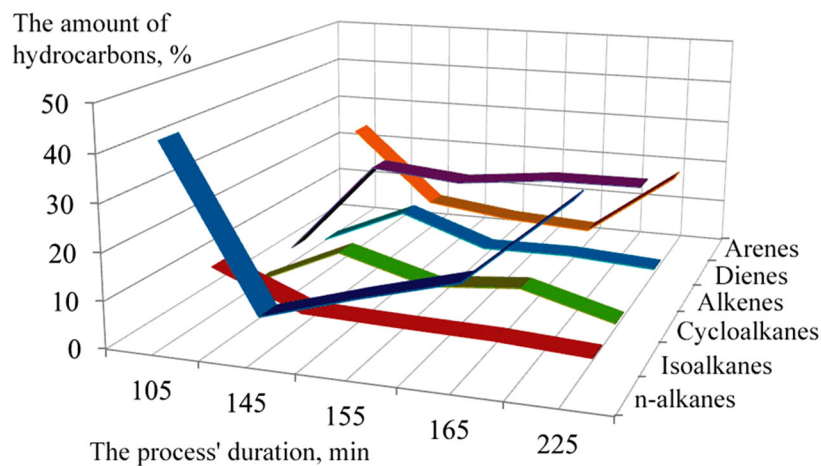


Fig. 10. Dependence of the hydrocarbons' amount in the resulting liquid product on the process' duration

4. Conclusions

1. The current experimental study demonstrates that the composition of the solid organic waste mixture when disposing of them by the multi-loop recirculation pyrolysis method has little effect the composition and properties of the obtained liquid hydrocarbon product. The exception is very wet wastes of small fractions.
2. The tests revealed that the composition of the raw materials influences the time to steady-state operation. Moreover, the steady-state time increases by up to 40% if the amount of PE in the waste material increases.
3. The total time of complete degradation for different mixtures of composite materials (mixtures of polymeric substances) is 180-250 min.

4. The highest gas yield was recorded when processing packaging adhesive tape (PP with impurities), the highest liquid product yield was for a mixture of packaging materials.
5. The correct selection of the temperature of the circulation system makes it possible to produce a gaseous or liquid product which can then be used as an alternative fuel.

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