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Improvement of the Ecological Efficiency of Synthetic Motor Fuel Production in Ukraine

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Abstract: Solving the problem of improving energy security is one of Ukraine's challenges in modern conditions. One of the ways to solve this problem is to organize the production of synthetic motor fuel from the available domestic carbon-containing raw materials. The relevance of developing the production of synthetic motor fuel in Ukraine from non-oil raw materials is associated with the shortage of deposits of traditional crude hydrocarbon and the destruction of the last processing capacities due to russian aggression. The article aims to substantiate the possibility of efficiently producing synthetic motor fuels from the available mineral hydrocarbon raw materials. Analyzing the existing deposits of hydrocarbons allowed for determining low-metamorphosed coal as the most expedient raw material base. A comparative analysis of various technologies made it possible to suggest the organization of the production of synthetic motor fuel through indirect hydrogenation, followed by fuel synthesis in the Fischer–Tropsch process. Calculations performed for low-metamorphosed Ukrainian coal showed the technical and environmental efficiency of the hydrogen enrichment of synthesis gas. To enrich synthesis gas with hydrogen, it was proposed to cooperate with producing synthetic motor fuel with coal mines (suppliers of raw materials, including methane for the production of additional hydrogen) or coke ovens and by-product enterprises that produce hydrogen-rich coke oven gas.

Keywords: energy security, alternative fuels, coal, gasification, hydrogen, CO_2 emission.

1. Introduction

In recent decades, interest in producing and using various alternative types of motor fuels, as evidenced by Demibras [1] and Khaustova et al. [2], has been growing all over the globe, primarily meaning synthetic, non-petroleum fuels, which has become the subject of the studies of scientists such by Stranges [3], Willauer and Hardy [4].

Synthetic motor fuels (SMF), like liquefied gas, gasoline, diesel fuel, and kerosene, are a complete analog of petroleum motor fuel and, according to Ram and Salkuti [5], the transition to its use does not require any changes in the design of internal combustion engines.

The interest of some countries in the production of SMF is explained by the desire to ensure their energy security by reducing dependence on the global oil market and focusing on available raw materials suitable for the production of motor fuels. It is characteristic that the production of SMF has been developed in countries that do not have significant oil reserves or oil refining facilities. Thus, according to the data by Kyzym et al. [6], SMF production capacities operate in South Africa, China, Qatar, Malaysia, and others.

In terms of providing the economy with the necessary fuels and lubricants, solving the problem of ensuring energy security is an urgent problem for modern Ukraine. According to the data portal of the extractive industry, the geological reserves of available raw materials for producing traditional petroleum liquid and gaseous fuels are relatively small. The existing capacities for processing domestic and imported raw materials have been destroyed due to russian military aggression. So, according to the computed data [6], the national motor fuel market highly depends on import issues.

Highly industrialized countries have developed national energy programs that study the processes of obtaining SMF in laboratory conditions and at pilot plants, as well as creating powerful industrial complexes for producing these fuels [7].

The article aims to substantiate the possibility of efficient SMF production in Ukraine from the available mineral hydrocarbon raw materials.

2. Literature Review

The global scientific community's attention to the organization of SMF production from raw materials of non-oil origin is explained by many factors, the classification of which is given in Figure 1. All these technologies for obtaining SMF can be divided into industrially developed (processes such as Sasol, Siemens, Prenflo, Texaco [8–10]), close to implementation (for example, oil shale pyrolysis, as proposed by Yang et al. [11]), under development and perspective (for example, gasification of wastes, as proposed by Bilets et al. [12]).

Areas of scientific research on the production of SMF are summarized in Figure 1.

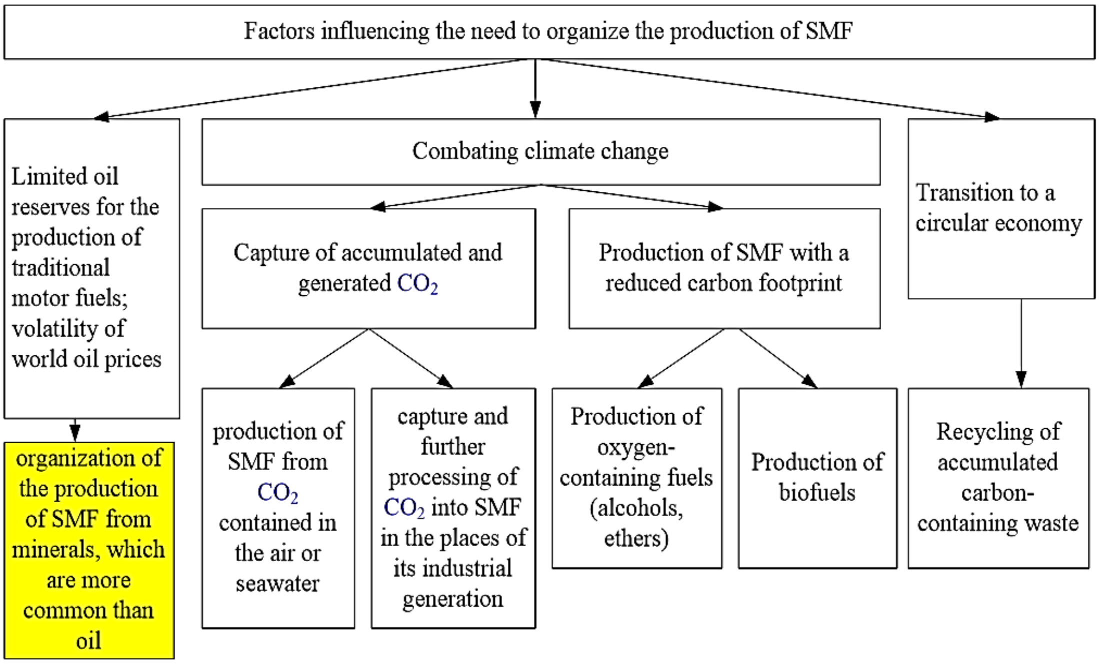

Figure 1 – Areas of scientific research on the production of SMF

Scientific research related to the production of SMF and aimed at combating global warming can be divided into two main areas. The first is the production of SMF, which produces carbon dioxide emissions lower than traditional fuels, namely oxygen-containing fuels (alcohols, ethers), as proposed in [13–16].

The decrease in the carbon footprint of such fuels has usually been explained by oxygen in the composition of fuel molecules, i.e., the proportion of carbon in an individual fuel molecule is less than in fuel molecules of petroleum origin. At this, specific emissions of combustion products (for example, grams per liter) are compared. Still, the calorific value of oxygen-containing fuels is lower than that of hydrocarbon fuels. If we compare carbon dioxide emissions per unit of heat produced, oxygen-containing fuel is more environmentally hazardous than hydrocarbon fuel.

For example, relevant technologies have become industrially widespread in Brazil [17]. The oxygen-containing types of SMF are usually used as additives to traditional fuels. In other words, they cannot wholly replace light petroleum products (including as a result of their low calorific value).

The second direction is the production of the so-called "E-fuels" [18–20] – fuels, the production of which is unrelated to using fossils.

The raw material for such fuel is a mixture of hydrogen and carbon monoxide. Hydrogen should be produced by electrolysis using electricity from alternative energy sources (e.g., wind, solar, and geothermal energy). Carbon monoxide must be obtained from atmosphere dioxide or emissions from industrial plants or thermal power plants. The technology of production of e-fuels involves the following stages: the capture of CO_2 and its subsequent conversion into CO; hydrogen production by electrolysis; the electricity required for electrolysis is obtained at solar (wind) power plants; production of synthesis gas (a mixture of CO and hydrogen) and subsequent synthesis of SMF according to the Fischer–Tropsch process.

This direction appears quite promising because of achieving UN sustainable development goals on climate action. However, the relevant studies are currently at the experimental research stage. The disadvantage of this technology is the high cost of electricity for hydrogen production. It is expected that e-fuels will be of industrial importance when alternative electricity takes a leading position in electricity generation.

Another direction in the organization of SMF production is related to scientific research on processing various accumulated carbon-containing wastes: polymer plastics, tires, and waste from the pulp and paper industry [21]. Usually, such carbon-containing wastes are polymeric or high molecular weight organic compounds consisting mainly of carbon and hydrogen. Such an elemental waste composition allows them to be considered a potential raw material for producing C5–C19 alkanes, which are seen as analogs of petroleum motor fuel.

Processes such as pyrolysis [22–25] are considered for decomposing carbon-containing waste. Relevant technologies are at the stages of development and too far from industrial implementation. The primary attention in scientific research is paid to the technological modes of processing (primarily pressure and temperature) and the selection of catalysts. The researchers hardly pay attention to the circumstances to maximize alkane output from polymeric and macromolecular compounds. It is necessary to add hydrogen artificially. According to our estimates, without solving this problem, the efficiency of the considered processes appears quite problematic.

There are also known papers on waste gasification after preliminary pyrolysis, in which the resulting synthesis gas comes into view as a raw material for further synthesis of SMF [12, 26]. All the processes under consideration have a particular perspective, yet they do not solve the issue of improving the energy security of an individual country in the short term.

In contrast to the aforementioned promising areas of SMF production, replacing oil raw materials with other types of fossil fuels has a history of industrial use.

Generalization of technologies for obtaining SMF, which had industrial development in the world (processes such as Sasol, Siemens, Prenflo, Texaco [8–10]) allowed for building a flowchart of the production from different types of mineral raw materials based on basis Willauer et al. [4] (Figure 2).

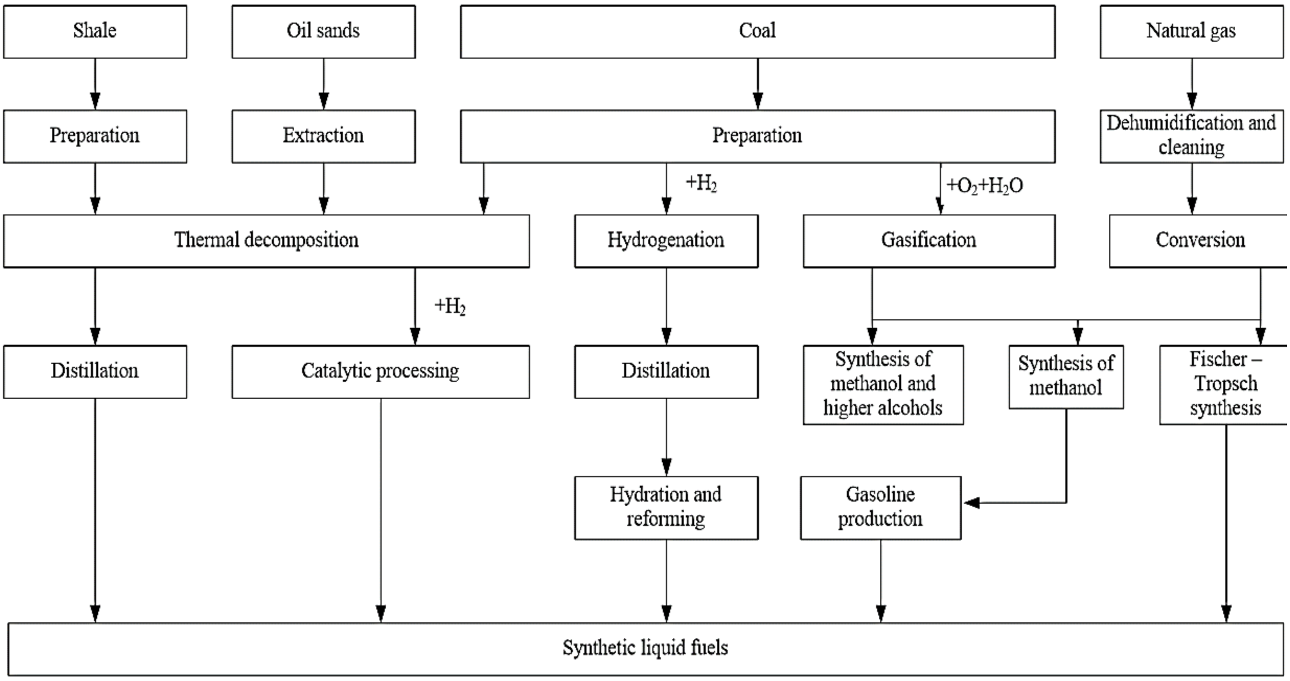

Figure 2 – A flowchart of the production of synthetic liquid fuels

Most of the technologies presented in Figure 2 focus on obtaining synthetic liquid fuels. However, the term SMF is broader. It covers liquid and gaseous fuels (for example, methane or propane-butane fraction, used as fuel for car engines). Obtaining SMF is available only in the Fischer–Tropsch synthesis of those considered technologies.

At present, considering local conditions, the production of SMF can compete with oil refining processes, in particular:

– the production of SMF from oil shale or oil sands by chemical decomposition and subsequent processing [27, 28];

– production of SMF through coal gasification and Fischer–Tropsch synthesis [8];

– synthesis of methanol and higher alcohols from synthesis gas obtained by coal gasification [29, 30];

– conversion of natural gas with the production of synthesis gas and the subsequent production of methanol or higher alcohols, or by synthesis based on the Fischer–Tropsch reaction [31].

Industrially mastered technologies for obtaining SMF from coal have advantages and disadvantages, as shown in Table 1.

Table 1 – Comparison of different industrial technologies and processes to produce SMF from coal

Technology	Disadvantages	Advantages
Thermal decomposition (pyrolysis) [32]	Low output of SMF	Possibility to produce hydrogen from pyrolysis gas to increase the output of SMF
Direct hydrogenation [4, 6]	Ultra-high pressure is required for the process to proceed	Higher output of end products
	Formation of a significant amount of low-reactivity, high-ash finely dispersed tarred sludge, the processing of which is a technically unsolved problem at present	
	The need to organize mass production of hydrogen	
Indirect hydrogenation (through the gasification stage)		
Using the Fischer–Tropsch process [6]	Dependence of output of SMF on the composition of the synthesis gas used	One-stage process of obtaining SMF; synthetic oil obtained in the process of synthesis makes it possible to obtain high-quality SMF
Using the oil process [15]	Dependence of output of SMF on the composition of the synthesis gas used	Possibility of direct use of methanol obtained at the first stage as a component of motor fuel
	The two-stage process of production of SMF	The need for an additional stage to produce a mixture of hydrocarbons

As can be seen, synthetic oil obtained by indirect hydrogenation is characterized by the highest quality.

In addition, the resulting intermediate products, in some cases, have an independent commodity value, which ensures the flexibility of production in responding to external challenges, in particular, caused by changes in demand conjuncture.

Therefore, the most widespread technologies of indirect coal gasification include the production of synthesis gas and its processing into a mixture of liquid hydrocarbons, the classification of which is presented in Figure 3 based in [4, 32]. Also, analyzing sources [6, 12, 27] allows for allocating the following disadvantages and advantages of the selected gasification technologies (Table 2).

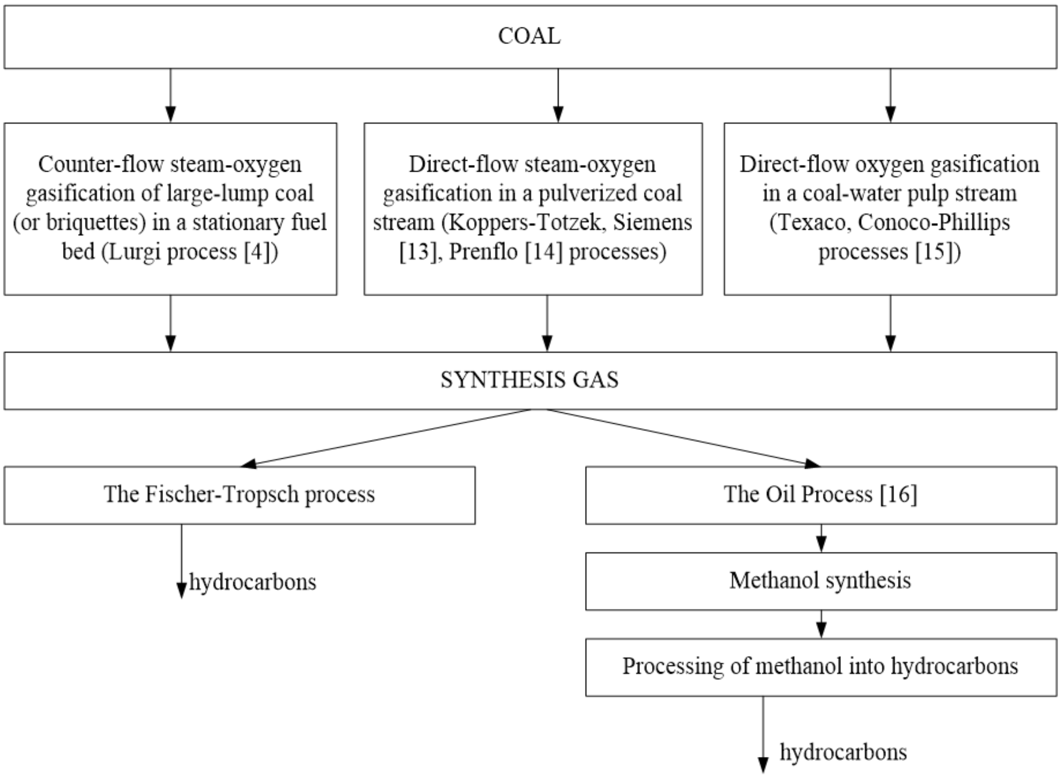

Figure 3 – Technologies of indirect hydrogenation of coal resulting in the production of a mixture of hydrocarbons

Table 2 – Advantages and disadvantages of the selected coal gasification technologies

Gasification technology	Disadvantages	Advantages
Counter-flow steam-oxygen gasification of large-lump coal (or briquettes) in a stationary bed [8]	Use of scarce high-quality coal with a size of more than 3–13 mm	Large-scale industrial implementation plus operational experience
	A large number of by-products	The relative simplicity of equipment design and operating conditions
	A low degree of water vapor conversion and a large amount of generated wastewater	
	High capital and operating costs	High thermal efficiency of processes
Direct-flow steam-oxygen gasification in a pulverized coal stream [8, 9]	large dimensions of the required equipment	The option to process coal with different properties
	The difficulty of transporting crushed coal by a nitrogen stream	
	The intensive abrasive wear of equipment	High speed of the process
	The significant removal of coal with the resulting gas	
	The need to use complex and high-speed	Low amount of contaminants in wastewater
	Automatic control systems	
direct-flow oxygen gasification in the coal-water pulp stream [10]	Low thermal efficiency	The option to process coal with any moisture content
		Absence of steam production stage
	Carrying out the process only at high temperatures and pressures	The possibility of utilization of secondary resources
		High speed of the process
	The complex process automation scheme	Almost complete absence of wastewater
		The possibility of using polluted waters from other industries to prepare pulp

Table 2 shows that the limiting factor for gasification technology in a stationary bed is the need to use the most scarce coarse coal. The risks of using pulverized coal gasification technology in the stream are the large dimensions of the equipment and its intense abrasive wear, which significantly increases both capital and operating costs.

The technology of pulp gasification makes it possible to utilize secondary energy resources as much as possible, compensating for the low thermal efficiency inherent in this process. We consider this technology the most promising technology in Ukraine for the gasification of fine coal of mechanized mining.

Thus, the experience of individual countries shows that the organization of the production of non-oil SMF is focused on those raw materials that are available in sufficient quantities, e.g.:

– the RSA is orientated towards coal [8–10];

– Brazil produces bioethanol from grain crops [14];

– China and Estonia prefer to process oil shale [27, 28].

Such global tendencies in the organization of the production of non-oil sources (the use of various types of raw materials and various technologies for their processing) should be considered when searching for a solution to the problem of improving Ukraine’s energy security.

3. Research Methodology

The research goal is achieved based on the author’s methodology and presented in Figure 4.

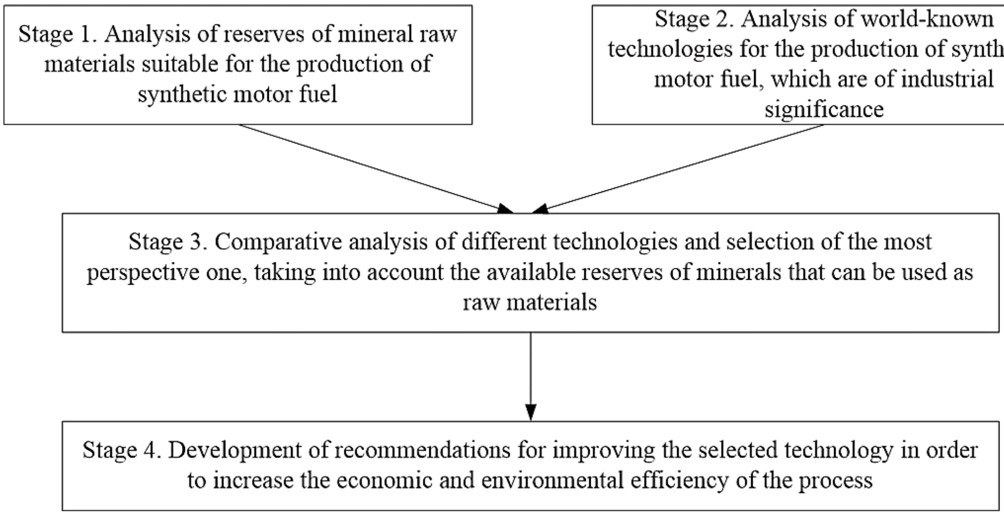

Figure 4 – Methods of selection of mineral raw materials and technology of their processing into SMF

The proposed authors' methodology provides for implementing the following sequence of stages.

In the first stage, while analyzing the reserves of mineral raw materials suitable for the production of SMF, geological reserves of carbonaceous minerals that can be used as raw materials are studied (Figure 1; these are shale, oil sands, coal, and natural gas) (Data portal of the extractive industry of Ukraine). When choosing the most suitable raw materials, the estimated reserve volume is not the only consideration. However, the experience in industrial production and the availability of appropriate infrastructure (e.g., equipment suppliers, availability of qualified personnel) [6].

In the second stage, the study of the main thermochemical processes of transformation of raw materials into either intermediates or finished products is carried out using technologies of industrial importance. At that, the technological parameters of the relevant thermochemical processes are studied, as well as the requirements for the equipment design of the process and the need for the use of catalysts. The considered technologies are compared in terms of the number of stages, the complexity of the equipment design, and the presence of by-products, waste, and harmful impurities.

In the third stage, a comparative analysis of the SMF production technology proposed by the authors is carried out according to the criteria shown in Table 3.

Table 3 – Criteria for the selection of SMF production technologies

Criterion	Selection procedure
The yield of final products from raw materials	The ratio of the amount of SMF obtained from raw materials to the amount of this raw material, t/m^3
	Priority is given to the technology for which this indicator has a higher value
The degree to which the carbon transition of feedstock into final products is possible	The amount of carbon contained in the SMF to its amount in the dry ashless mass of the raw material is calculated in %
	Priority is given to the technology for which this indicator has a higher value
Energy intensity of production	The ratio of the energy spent on production to the amount of SMF obtained, MJ/t
	Priority is given to the technology for which this indicator has the lowest value
The possibility of obtaining the necessary energy resources as by-products of processing	The amount of energy of exothermic reactions utilized to generate electricity concerning the volume of SMF production, MW·h/t
	Priority is given to the technology for which this indicator has a higher value
Emissions of harmful substances into the atmosphere, including greenhouse gases	Emissions of sulfur dioxide, nitrogen oxides, and carbon dioxide into the atmosphere concerning the volume of SMF production
	Priority is given to the technology for which these indicators are lowest

The technology most suitable for processing available raw materials was selected based on the comparison.

In the last stage, recommendations for introduction are developed and substantiated for the selected technology in order to increase the economic and environmental efficiency of the process, in particular in the following directions: maximizing the yield of finished products from raw materials, reducing emissions of harmful substances into the atmosphere (greenhouse gases).

Thus, the consistent realization of the stages of the presented authors' methodology allows for improving the technological, economic, and ecological indicators of the production of synthetic motor fuels in Ukrainian realities, which will significantly contribute to the industrial implementation of the process.

4. Results

4.1. Analysis of mineral reserves of SMF production

Ukraine has reserves of almost all combustible minerals suitable for producing SMF (except for oil sands): oil shale, coal, and natural gas. The probable reserves of menilite oil shale in the C_2 category in Ukraine [33] reach more than $5 \cdot 10^{11}$ tons. However, these reserves are classified as pre-estimated reserves, the amount of which is determined by single probes and samples. In the long run, this mineral can also become a raw material for producing synthetic motor fuels.

This type of mineral is not mined and is not planned for extraction. Issue-related work on clarification and confirmation of reserves has not been carried out. Therefore, within the terms of the present article, this mineral is not considered a raw material for the production of SMF.

The balance reserves of natural gas were estimated at $6.89 \cdot 10^{11}$ m^3 and are constantly decreasing (by an average of 4 % annually), and the related production is constantly complicated (the decline amounts to 1–2 % per year).

The main gas fields of Ukraine have long been exhausted, and industrial production is sufficient only to meet the population's needs and municipal heat and power engineering. The available gas reserves do not allow us to expect a significant increase in production. Accordingly, this fossil fuel cannot be discussed as a possible raw material for the production of SMF in the volumes necessary to increase the country's energy independence.

Thus, the only sources of carbon suitable for the production of SMF are lignite and hard coal, the total actual balance reserves of which in categories A, B, and C1 comprise $4.48 \cdot 10^{10}$ tons. According to the extractive industry data portal, the structure of Ukrainian coal reserves is shown in Figure 5.

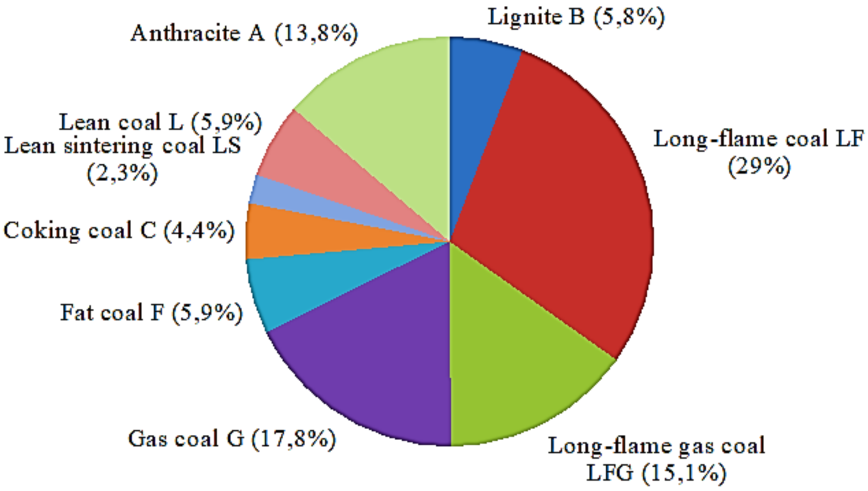

Figure 5 – Grade structure of the balance reserves of coal in Ukraine

Figure 5 illustrates that more than 60 % of the deposits are low-metamorphosed coal (lignite – $2.59 \cdot 10^9$ tons, long-flame – $1.30 \cdot 10^{10}$ tons, long-flame gas – $6.78 \cdot 10^9$ tons, and gas coal – $7.99 \cdot 10^9$ tons). The specific properties of these types of coal, primarily high reactivity, limit its use in traditional areas of coal use – energy and coke chemistry. Simultaneously, high reactivity is considered a desirable property for chemical use (including the production of SMF).

According to [34], the high reactivity of low-metamorphosed coal is due to the presence in its structure of a large number of active functional groups, such as carboxyl (–COOH), carbonyl (=C=O), and hydroxyl (–OH), which are cleaved at high speed under thermochemical influences, thus accounting for the formation of hydrocarbons. An increase in the rate of thermochemical reactions increases the productivity of the production plant.

When considering the technologies for individual grades of coal processing into SMF, it is necessary to compare the elemental composition of coal (Table 4) and SMF itself, an analog of fuel of petroleum origin [37].

Table 4 – Average elemental composition of organic mass of the primary grades of fossil coal in Ukraine, %

Grade	Carbon	Hydrogen	Oxygen	Sulfur	Nitrogen
Lignite					
Earthy coal	63–72	5.5–6.5	18–30	1.2–1.5	0.6–0.8
Dense matte coal	67–75	5.0–6.5	15–27	1.0–2.0	0.5–1.2
Hard coal, mined in the Donbas region, Ukraine					
Long-flame coal	76–86	5.0–6.0	10–17	2.0–2.5	1.8
Gas coal	78–89	4.5–5.5	7–16	1.0–1.5	1.7
Fat coal	84–90	4.0–5.4	5–10	1.5–2.0	1.6
Coking coal	87–92	4.0–5.2	3–8	1.5–2.0	1.5
Lean sintering coal	89–94	3.8–4.9	2–5	1.5–2.0	1.4
Lean coal	90–95	3.4–4.4	1.6–4.5	1.5–2.0	1.2
Anthracite	91–96	1.3–3.0	1–2	1.0–1.5	0.1–1.3

The elemental composition of individual grades of coal shows that in the organic mass of brown coal, the amount of carbon exceeds the amount of hydrogen by 13–15 times, and in the organic mass of low-metamorphosed hard coal, the amount of carbon exceeds the amount of hydrogen by 17 times (Table 3).

On the other hand, according to [35], synthetic and petroleum motor fuels are a mixture of saturated hydrocarbons (C5–C19). That is, in particular fuel molecules, the amount of hydrogen is 2 times higher than the amount of carbon. This ratio is characteristic of the elemental composition of conventional oil (petroleum), dominated by kinds of paraffin CnH_{2n+2} [36].

Hence, when processing coal into the SMF using any technology, one of the critical problems is the problem of supplying the required amount of hydrogen. This problem can be solved, in our opinion, either by removing a fraction of carbon from the synthesis process or by supplying additional hydrogen.

4.2. Selection of the technology for conversion of coal into SMF

Possible ways to obtain SMF from coal are the following:

– thermal decomposition – pyrolysis (Figure 6a);

– direct hydrogenation (Figure 6b);

– indirect hydrogenation through the gasification stage (Figure 6c).

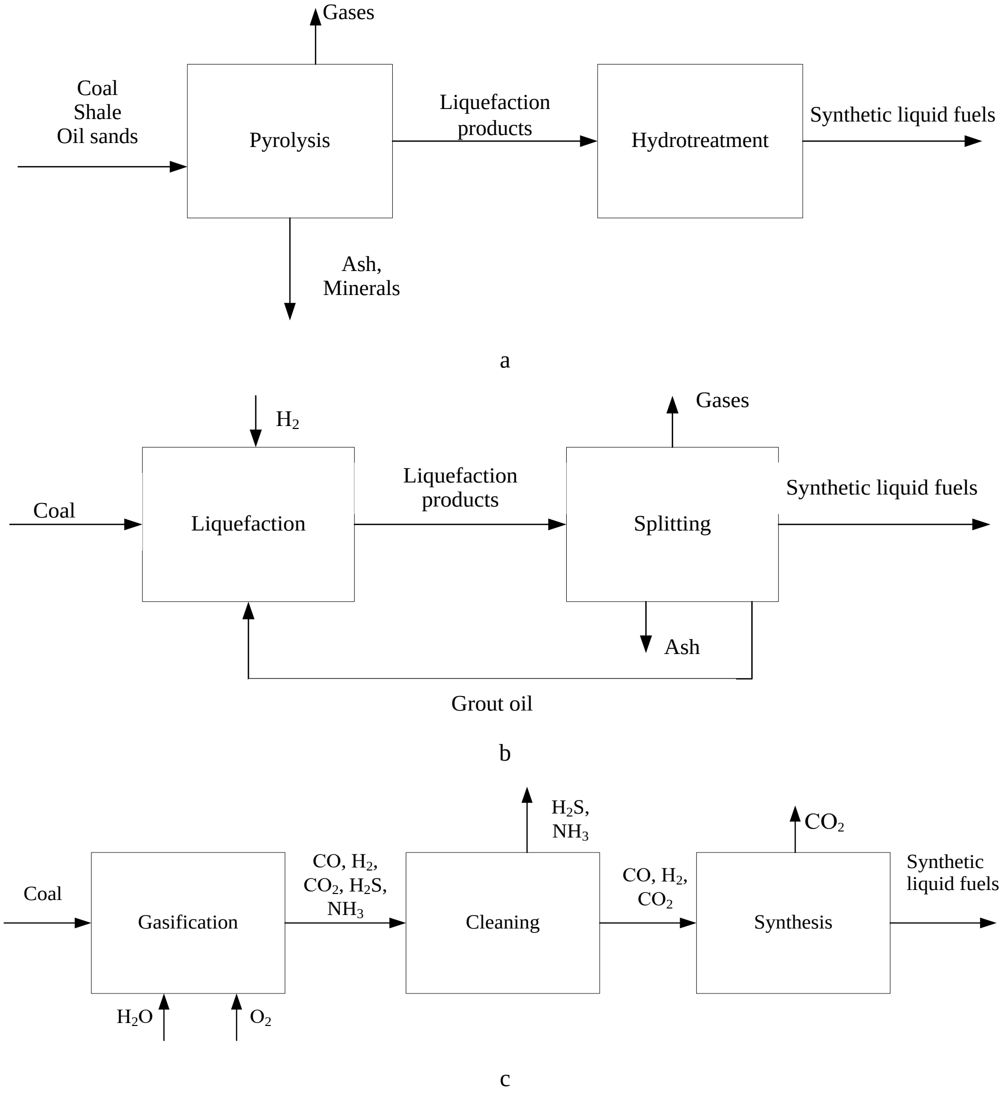

Figure 6 – Technological flowchart of SMF production by different methods: a – SMF production from solid fuels by the pyrolysis method; b, c – coal processing into SMF by direct and indirect hydrogenation, respectively

The authors' comparative chemical analysis results for each SMF production technology is presented below.

Pyrolysis is the thermal decomposition of substances with no access to air. Pyrolysis is the main method of chemical and technological processing of coal in the processes of semi-coking and coking. Modern ideas about the physicochemical essence of pyrolysis of solid combustible minerals were formulated in [34]. In a simplified form, they can be presented using the following sequence [38]:

– thermal decomposition:

$$C \rightarrow G_1\uparrow + L + T; \tag{1}$$

– thermal synthesis:

$$L + T \rightarrow S; S \rightarrow K + G_2, \tag{2}$$

where C – source coal; G_1 – steam-gas products of thermal decomposition; L – liquid-mobile products of thermal decomposition; T – solid residue from thermal decomposition; S – semi-coke; K – coke; G_2 – steam-gas products of thermal synthesis.

To increase the SMF yield, it is advisable to carry out secondary pyrolysis of the products formed during thermal decomposition under pressure in a hydrogen environment. By using semi-coking coal with the help of additional hydrotreatment of the obtained steam-gas products, the yield of motor fuels can be increased by up to 40 % of the organic mass of coal. Considering coking, it is necessary to use charges based on well-sintered coal. The output of steam-gas products is up to 25 % of the dry weight of coal.

Technological modes of coking are determined primarily by the need to obtain high-quality coke and do not allow for the high output and quality of steam-gas products suitable for processing into motor fuels (crude benzene and coal tar) [34].

Thus, coal processing by pyrolysis is characterized by a low SMF yield – at the level of 5–6 % of the feedstock (with the possibility of increasing to about 10 %, provided that a sufficient amount of hydrogen is added and all carbon CO and CO_2 are bound into saturated hydrocarbons).

The advantage of this technology is that coke oven gas, a by-product of coking, contains a large amount of hydrogen, which can be used for hydrotreating intermediates (crude benzene and coal tar) to increase the yield of SMF.

More attractive from the standpoint of processing coal into SMF is the technology of direct hydrogenation. Direct hydrogenation (liquefaction) of coal is based on thermal cracking of the side chains of carbon macromolecules and hydrogenation of the formed free radicals [39]. Conventionally, this process can be written down as follows:

$$2xC + yH_2 \rightarrow 2C_xH_y. \quad (3)$$

The process takes place at elevated temperatures (380–450 °C) and considerable pressures (20–70 MPa) in the presence of catalysts (iron, molybdenum, tungsten) and a paste former (grout oil obtained as a by-product in the same technology).

The resulting mixture of liquid products is close to natural oil's composition and properties. The selected fractions of motor fuels for the production of high-quality gasoline and diesel fuel are subjected to further catalytic hydrogenation of aromatic hydrocarbons, sulfur- and nitrogen-containing compounds, as exemplified by the following reactions:

$$\begin{aligned} &C_6H_6 + 3H_2 \rightarrow C_6H_{12}; \\ &C_6H_5N + 4H_2 \rightarrow C_5H_{10} + NH_3; \quad (4) \\ &C_4H_4S + 4H_2 \rightarrow C_4H_{10} + H_2S. \end{aligned}$$

According to laboratory studies and chemical and technological calculations carried out by the authors, the SMF yield from the organic mass of coal can reach 44–54 %, e.g., in the presence of a sufficient amount of hydrogen and the use of hydrogenation processes of aromatic hydrocarbons) [32]. Regarding the dry, i.e., without taking into account moisture) weight of coal, these figures are much lower: 17–19 % for lignite and 40–42 % for hard coal.

The main disadvantages of this technology include the following parameters:

– high pressure (up to 70 MPa), at which thermal cracking takes place, which puts forward increased requirements for equipment design and safety of processes;

– about 50 % of the dry weight of coal is formed by a low-reactive, high-ash finely dispersed tar residue (sludge), the processing of which is a complex and so far technically unresolved problem;

– the need to organize mass production of hydrogen.

However, the disadvantages have led to the limitation of the application of this technology on an industrial scale.

The following technology for the production of SMF, which is most widely used in the world, is the processing of coal by the method of indirect hydrogenation, carried out in two stages: coal gasification and further synthesis of SMF from the resulting gas mixture [8–10].

Coal gasification is a high-temperature process of fuel interaction with the blast's oxidizing components: air, oxygen, water, water vapor, or mixtures. Gasification aims to obtain combustible gases – a mixture of hydrogen and carbon monoxide [40]. The main processes of interaction of fuel carbon with blast components can occur according to the following chemical generalized equations:

$$\alpha C + O_2 \rightarrow 2(\alpha - 1)CO + (2 - \alpha)CO_2; \quad (5)$$

$$\beta C + H_2O \rightarrow (2\beta - 1)CO + (1 - \beta)CO_2 + H_2. \quad (6)$$

The numerical values of the coefficients α and β characterize the ratio of the amounts of CO and CO_2 in the reaction products. They depend on the conditions of the process (primarily the components of the heat balance) and remain within the following limits: $\alpha = 1\ldots2$; $\beta = 0.5\ldots1$.

When using the resulting gas for chemical syntheses in the blast, it is advisable to use oxygen rather than air. Generator gas (or synthesis gas) obtained in gasification is further processed into hydrocarbons. The resulting mixture of hydrocarbons, the so-called "synthetic petroleum", is then divided into separate fractions. These are complete analogs of traditional motor fuels of petroleum (oil and gas) origin: propane-butane fraction, gasoline, diesel and aviation fuel, and other products.

The choice of gasification technology in each specific case is carried out based on the properties of the processed coal, the requirements for the quality and properties of the resulting gas, and the results of comparing the technical and economic indicators of various processes. Two processes, considered below, are of industrial importance for synthesizing the gas into a mixture of hydrocarbons: the Fischer–Tropsch [35] and oil [41] processes.

The Fischer-Tropsch process is carried out at a temperature of 160–320 °C and pressure of about 2.2 MPa, amplified with an iron catalyst [35]. The generalized process equation is as follows:

$$2nCO + (m + 2n)H_2 \rightarrow 2C_nH_m + 2nH_2O. \quad (7)$$

In the oil process [41], hydrocarbons from generator gas production occur in two stages:

1) the synthesis of methanol:

$$CO + 2H_2 \rightarrow H_3COH; \quad (8)$$

2) the direct production of hydrocarbons:

$$xH_3COH \rightarrow C_xH_{2x} + xH_2O. \quad (9)$$

Due to a single stage of generator gas processing, the Fischer–Tropsch process is more attractive than the oil process. The comparison results make it possible to propose indirect hydrogenation of domestic coal with subsequent fuel synthesis in the Fischer–Tropsch process to organize SMF production.

4.3. Ecological indicators of SMF production

An assessment of how the composition of synthesis gas affects specific technical and economic indicators of SMF production can be carried out for the gasification conditions of low-metamorphosed coal, the share of which in the deposits of Ukraine reaches two-thirds.

According to the analysis of the information provided by the Ukrainian State Research Institute for Carbochemistry (UKhIN), quality indicators of typical low-metamorphosed coal are summarized in Table 5.

Table 5 – Characteristics of source coal

Indicator	Value, %
Technical analysis	
The moisture content of the working mass	10.0
Ash content of dry mass	15.0
Release of volatile substances from dry ash-free (combustible) mass	44.1
Sulfur content of total dry weight	1.4
Elemental composition of organic mass	
Carbon	80.9
Hydrogen	5.3
Total sulfur	1.7
Nitrogen	1.1
Oxygen	11.0

Calculations of the gasification process for various technologies and technological parameters allow for the prediction of the characteristics presented in Table 6.

Table 6 – Composition of purified synthesis gas under different technological modes of gasification

Indicator	H_2:CO ratio	
	1:1	1:2
Synthesis gas composition after reactor, % vol.		
H_2	45	30
CO	45	60
CO_2	10	10
Amount of synthesis gas per 1 ton of coal, m^3/t	2037	1567
Including		
H_2	940	470
CO	940	940
CO_2	157	157
Amount of synthesis gas for synthesis according to the Fischer–Tropsch process, m^3/t (at the ratio H_2:CO = 2:1)	1410	705
Amount of CO_2 discharged into the atmosphere, kg/t	1231	1693
Amount of carbon of source coal used for the SMF, kg/t	251.8	125.9
Used carbon of source coal, %	40.7	20.3

The presented data characterize the synthesis gas obtained by various gasification technologies purified from harmful impurities, such as hydrogen sulfide and ammonia. Depending on the technological specifics of individual processes, synthesis gas differs in composition (the ratio of hydrogen and carbon monoxide volumes [6]).

Table 6 assumes that 5 % of the original carbon is lost coherently with slag (residues after gasification).

Extrapolating the obtained data to the ratio range H_2:CO from 2:1 to 1:2 allows for obtaining dependencies on this ratio of indicators such as CO_2 emissions and the amount of gas directed to the synthesis of hydrocarbons (Figure 7).

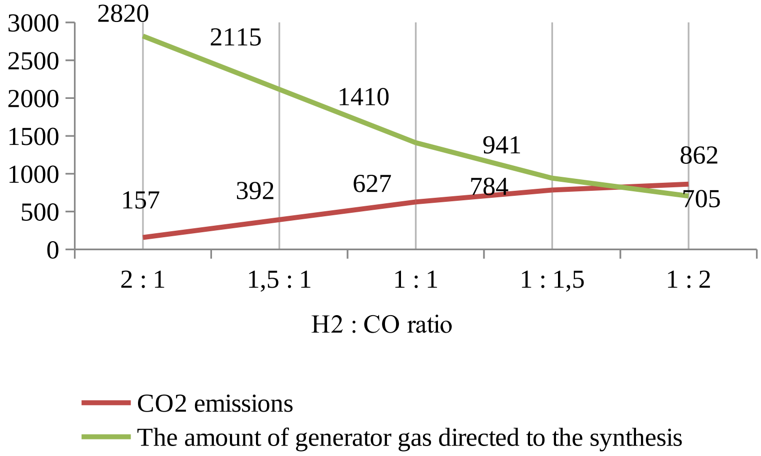

Figure 7 – Dependence of the amount of synthesis gas directed to the production of hydrocarbons and CO_2 emissions from the composition of synthesis gas in terms of the ratio H_2:CO

Therefore, the hydrogen and carbon monoxide ratio determines the technical and economic efficiency of the subsequent synthesis of motor fuels. Figure 7 testify the following: with a ratio of hydrogen to carbon monoxide of 1:2, if 705 m^3 (272.8 kg) of synthesis gas is directed to synthesis according to the Fischer–Tropsch process from 1 ton of coal (with a change in the H_2:CO ratio to 1:1), the amount of synthesis gas increases to 1410 m^3 (440.6 kg).

Also, when the optimal ratio of these components is increased to H_2:CO = 2:1, the amount of synthesis gas increases to 2820 m^3 (881.2 kg).

Simultaneously, the ecological indicators of the process are significantly improved, namely, emissions of the greenhouse gas carbon dioxide are reduced. Considering the molecular weight of individual substances that make up the synthesis gas (Figure 7), the following conclusions can be drawn from the presented data. An increase in the proportion of hydrogen in the composition of synthesis gas from 33.3 % (when the ratio of H_2:CO = 1:2) up to 66.7 % (when this ratio is 2:1), and the amount of carbon dioxide decreases by 5.5 times (from 1693 to 308 kg per 1 ton of coal).

Thus, a significant increase in the technical and economic efficiency and environmental friendliness of SMF production can be achieved by adding hydrogen to synthesis gas in an amount that provides the required ratio of components. Alsunousi and Kayabasi [42] reached the same conclusion.

When considering Table 5 for the ratio H_2:CO = 1:1, to achieve the optimal composition of synthesis gas per 1 ton of coal, the addition of 940 m^3 of hydrogen will be necessary to increase the amount of synthesis gas directed to the synthesis of motor fuel from 1410 to 2350 m^3 (or from 514.3 to 964.3 kg). Accordingly, the volume of SMF will also change.

The following environmental effect is also obtained. The amount of carbon dioxide emissions with the proposed intensification of the process is reduced from 627 to 157 m^3/t (or from 1231 to 308 kg/t) for the processed coal. According to the calculations, the share of usefully used carbon from source coal increases from 40.7 % to 86.4 %.

4.4. Optimization of synthesis gas production

Thus, to increase the efficiency of the process, it is necessary to solve the issue of optimizing the composition of synthesis gas directed to the synthesis of motor fuels. The resulting synthesis gas during coal gasification always contains more carbon monoxide CO than hydrogen due to the need to ensure the heat balance of the process. In part, synthesis gas composition is influenced by the technological mode of gasification (primarily temperature and pressure), which differs in individual processes. This explains why, in modern gasification processes, the ratio of H_2:CO varies from 1:1 to 1:2 [6]. Simultaneously, as proved above, the hydrogen content in the gas must be at least 2 times higher than CO for use in chemical syntheses.

Regarding the H_2:CO ratio, the synthesis gas composition can be optimized in two ways (Figure 8).

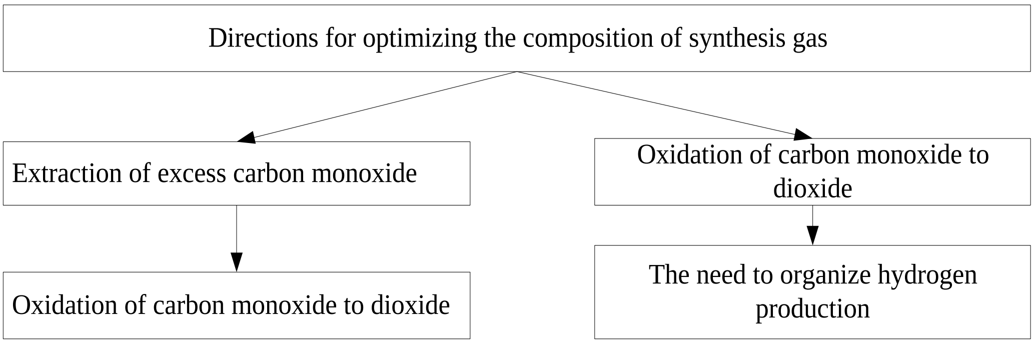

Figure 8 – Ways to optimize the composition of synthesis gas in terms of the ratio H_2:CO

The first way to optimize the composition of synthesis gas is to reduce the concentration of CO. To ensure the desired ratio of H_2:CO, oxidizing part of the carbon contained in the source coal in an exothermic process into carbon dioxide is advisable. This makes it possible to improve the specified ratio in the resulting synthesis gas but leads to irrational consumption of part of the carbon, reduces the yield of synthesis gas, and, ultimately, increases the cost ratio of raw materials for obtaining the final product and worsens the technical and economic indicators of production. Simultaneously, carbon dioxide emissions, known as greenhouse gases, are increasing in the atmosphere. This direction of optimization is typical for all processes of industrial importance. Notably, considering that a significant part of the carbon contained in the feedstock is converted into carbon dioxide and discharged into the atmosphere, this is one of the factors hindering the spread of the technology for producing SMF from coal.

The second way is based on the addition of hydrogen. Enrichment of synthesis gas with hydrogen until the optimal composition is achieved makes it possible to solve the problems that characterize the previous direction, namely, to increase the amount of carbon of feedstock, which passes into the composition of the SMF with a corresponding decrease in the formation of the greenhouse gas CO_2. This conclusion can be drawn from the data presented in Figure 7.

Therefore, from the point of view of improving the efficiency of the production process of SMF from coal, it is essential to find ways to obtain additional hydrogen resources that will be involved in the synthesis of SMF. Overcoming hydrogen deficiency concerning the production of this component is possible in one of the following ways, which are currently used (or can be used) on an industrial scale.

The most widespread is the conversion of natural gas methane or its homologs, followed by CO conversion [43]. Methane conversion proceeds in several stages according to a generalized reaction:

$$CH_4 + 2H_2O \leftrightarrow CO_2 + 4H_2. \qquad (10)$$

This process of hydrogen production has become the most widespread in the world. In Ukraine, where there are no free natural gas resources, it can potentially be used in the case of processing methane obtained from sources other than natural gas (e.g., coal mine methane, biogas). Nevertheless, alternative types of raw materials are currently unavailable.

Another technology for hydrogen production is the conversion of carbon monoxide obtained by the gasification of solid or liquid fuels [44]. It is possible to combine this method with coal gasification technology. Still, to obtain the necessary additional amount of hydrogen, another component of synthesis gas (that is, carbon monoxide) will be used. As a result, carbon dioxide emissions will lose a fraction of the original carbon.

Also, the method of obtaining hydrogen by separating coke by short-cycle adsorption or sequential liquefaction of all gas mixture components, except hydrogen [45], is possible for implementation in the case of cooperation between the production of SMF and a coke plant. Currently, this process is of minor use due to the crisis state of Ukrainian coke and by-product enterprises, characterized by the absence of surplus coke oven gas that is viable to produce hydrogen [46, 47].

So, if we consider the organization of the production of SMF from domestic coal to ensure the state's energy security, then one of the priority tasks is the search for effective industrial hydrogen production methods. Such methods should focus on potentially available resources and consider the current state of the gas, coal, and coke industries.

5. Discussion

Increasing the state's energy security through the organization of SMF production is, in our opinion, a realistic scenario, as confirmed by the following:

First, gas is converted into various hydrocarbons in the Fischer–Tropsch synthesis process, with the number of carbon atoms in molecules ranging from 1 to 35 [35]. Such a composition of hydrocarbons makes it possible to obtain from them the entire range of motor fuels (liquefied gas, gasoline, diesel fuel, kerosene), paraffins, and technical lubricants.

Secondly, using SMF does not require structural changes in car engines, and significantly lower emissions of harmful substances accompany its combustion in internal combustion engines into the atmosphere [5].

This study hypothesizes that ensuring the state's energy security, without sufficient oil production and refining capacities, should be based on using non-traditional technologies to produce motor fuels. As such a technology, our study proposes the production of SMF from synthesis gas (Fischer-Tropsch process), which can be obtained by processing various raw materials that contain hydrogen and carbon, e.g., coal, natural gas, plastic waste, and biomass (Figure 1).

The existing industrial complexes for producing SMF in different countries confirm the commercial attractiveness of the synthesis of motor fuels from non-petroleum raw materials. Nevertheless, for the conditions of Ukraine, the only type of raw material for the mass production of synthesis gas and the subsequent production of SMF can be considered only coal and lignite, the elemental composition of which does not allow obtaining synthesis gas of optimal composition (in terms of the H_2:CO ratio).

According to our calculations (Table 6, Figure 7), a significant increase in the efficiency of SMF production can be achieved if the synthesis gas obtained by coal gasification is additionally enriched with hydrogen.

The specific features of certain hydrogen production methods allow us to formulate preliminary requirements for the first time for the location of SMF production facilities in Ukraine. The following two options look promising.

The first option involves the location of the SMF production near a coal mine, which is a source of methane in the coal mine. The prospects of this option are explained by the fact that the deposits of Ukrainian coal are characterized by high gas content [48].

The coal mine methane can be combined with the methane-ethane fraction (one of the products of SMF synthesis in the Fischer-Tropsch process [35].

It is also possible to locate SMF production near the industrial site of the coke oven and by-product plant, where there are sufficient resources for excess coke oven gas, which contains about 60 % hydrogen [45]. The implementation of this option is possible only after the Ukrainian coke industry recovers from the current crisis.

The obtained results provide grounds for continuing research in detailing the raw material base of the SPM production process, choosing a gasification technology, and finding an effective way to obtain the required amount of hydrogen.

In the future, implementing the project for the production of SMF from domestic coal on an industrial scale will increase the country's energy security (by reducing imports of petroleum products) and load the production capacities of the coal industry and domestic engineering.

6. Conclusions

The organization of the production of SMF from hard coal is an essential way of improving the state's energy security and can be considered an alternative to the traditional production of motor fuel from oil or the import of petroleum products. SMF, in its quality, is a complete analog of traditional motor fuel, and its use does not require any changes in the design of internal combustion engines.

The most prospective method of obtaining SMF from coal in Ukraine is indirect hydrogenation by coal gasification, production of synthesis gas, and its processing into a mixture of hydrocarbons. Indirect coal gasification technology has become widespread in different countries (primarily South Africa and China).

The choice of technology for the first stage of the process (coal gasification) should be carried out based on the properties of the processed coal and the results of comparing technical and economic indicators for different processes. The study showed that the single-stage Fischer-Tropsch process is the most effective for producing hydrocarbons from synthesis gas.

For use in chemical syntheses, the hydrogen content of the gas must be at least twice that of CO. This is not achieved by any of the industrial stages of gasification. Therefore, it is essential to introduce effective industrial methods for hydrogen production and their combination with the production of SMF. A perspective direction for improving the efficiency of SMF production is its cooperation with coke or coal mining enterprises, which are considered a potential source of necessary hydrogen.

The implementation of a large-scale project for the production of SMF from hydrogen-enriched synthesis gas will provide many advantages: reducing the country's import dependence (due to the use of domestic coal), ensuring the sustainable operation of domestic coal mining enterprises, and giving impetus to the development of domestic mechanical engineering. We expect that the creation of SMF production capacities will have a significant social and economic effect.

Creating new and increasing the workload of existing (coal mining and machine-building) enterprises will create new jobs and increase tax revenues to local and state budgets.

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