The object of this study is coal processing with synthesis gas as the target product. The experimental technology employs a reactor with a liquid high-temperature heat carrier. The technological advancement allows ecological and safe transformation of hard raw material – hard coal.

Hard, brown, and salty coal contains quite a lot of mineral impurities. Thermodynamic analysis of chemical transformations of coal according to its content has been carried out. The analysis was performed under the conditions of the new technology. The technology makes it possible to work simultaneously with solid, liquid, and gas structures. The main technological parameters are considered to be atmospheric pressure, temperature from 1073 to 1373 K, composition and height of the heat carrier in the reactor. The optimal temperature conditions for the process in the melt were revealed. The research was related to the characteristics and properties of liquid high-temperature heat carrier. Two types of laboratory reactors were studied. The composition and volume of the liquid heat carrier contributes to a stable and balanced progress of the target process. The proposed scheme of a reactor with a liquid high-temperature heat carrier for the coal gasification process allows for an environmentally friendly process. The designed scheme contains three zones of the process: conversion, oxidation, and post-oxidation. The reactor scheme is quite simple in structure. The technology in liquid high-temperature heat carrier involves one heat carrier for three reactor zones. The target product of processing is synthesis gas. Synthesis gas can be used directly as a target product with hydrogen being an alternative energy source. Synthesis gas as a raw material can be used to obtain hydrocarbons, separate use of substances for various industries and transformation into various compounds of organic and inorganic synthesis

Keywords: synthesis gas, liquid high-temperature heat carrier, hard coal, three-zone reactor, mineral component

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DEVELOPMENT OF A NEW METHOD FOR STONE COAL CONVERTING INTO A LIQUID HIGH-TEMPERATURE HEAT CARRIER BASED ON ENERGY FACTORS

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1. Introduction

In the 19th century a significant amount of minerals was calculated in the world. The main part is coal seams, which make up hard coal deposits. Such deposits are the property of most countries of the world. The potential of fossil fuels is one of the sources of thermal energy and electricity in a country. To develop the country's industrial sector, it is necessary to learn how to use reserves rationally. Western countries have already learned to use their minerals. One of these countries is the USA. Oil is divided into fractions, which are used for the needs of industry and the population. But oil processing is the main source of environmental pollution. To prevent it, expensive cleaning systems are installed. They use solid minerals as a source of energy due to their relatively low cost. Also, the United States has learned to use its other minerals for the development of the country [1].

If we look at the energy content of the fuel, then 1 kg of hard coal contains 17,200–30,700 kJ, 1 kg of lignite contains 5,600–10,500 kJ [2].

Coal is a solid raw material that is very difficult to transform. Therefore, the process of pyrolysis of coal masses is an important coal processing technology. This process carries large emissions into the environment.

Coal is one of the country's most abundant minerals. This raw material is multifunctional. When processing coal, it is possible to obtain synthesis gas, various types of hydrocarbons. Hydrocarbons are a very good energy and industrial resource. They are subjected to processing to obtain products of organic and inorganic synthesis. The processing of various types of coal is widespread in the industry and research institutes. Processing is a very complex process, so it is necessary to clearly understand its mechanism. There are few research institutes in the world that study the transformation of solid materials. The most common processes with coal are pyrolysis, combustion, and gasification. All of them are related to obtaining thermal energy. However, these processes must take into account an environmental component. These processes were studied using different technologies and in reactors of different designs. Those studies have been known since the last century.

In work [3], the process of coal gasification under the conditions of a liquid high-temperature heat carrier (LHH) is considered. The studies show the probability of the process of gasification of coal with the production of synthesis gas under the conditions of a high-temperature heat carrier as a reaction space. Given the uniqueness of the technology, there is a need for in-depth research into the peculiarities of the progress of processes under the conditions of the technology. There is a possibility of discovering new control parameters of chemical-technological processes. This would make it possible to improve and spread coal processing not only as an energy resource but also to obtain various chemical compounds.

There is a need to restore the chemical, oil refining, and coal industries, especially when using own resources. To this end, this technology in the melt of a liquid high-temperature heat carrier could become an innovative solution.

2. Literature review and problem statement

For the processing of solid raw materials, especially coal, there are pyrolysis, gasification, liquefaction, underground gasification, enabled by the well-known technologies of Lurgi, Winkler, Koppers-Totzek, as well as Texaco method. The known facts of these technologies are that the process takes place during high-temperature processing of solid raw materials (coal, etc.) at high pressure and temperature in an environment of air oxygen, water vapor, carbon dioxide, or hydrogen. Technology gas generators have complex designs. The reaction products are gaseous products. Each technology is specific. Lurgi technology uses a catalytic process of gasification of a dense layer of coarse-grained solid fuel at high pressure. Compounds of alkaline, alkaline soil, and some transition metals (Ni, Fe, Co) are used as a catalyst. Winkler technology employs finely dispersed solid fuel in a fluidized bed. The fraction of solid raw materials (lignite and hard coal, coke, semi-coke) has a size of 10 mm. The Koppers-Totzek technology for the gasification process uses small solid raw materials up to 0.1 mm in size in an oxygen environment with water vapor. Water vapor protects the reactor from slag formation, erosion, and the effects of high temperatures. Ash is released from the bottom of the reactor in a liquid state, cooled and removed as granular slag. The Texaco method is associated with the process of gasification of hydro coal suspension in an oxygen environment at 1350–1500 °C. The technology of underground gasification [4] is an unconventional processing of solid fuel, which takes place directly in the environment of the earth's crust. The principle diagram of the technology uses the activation of the drilling blast.

In [5], it was found that the Koppers-Totzek technology involves partial oxidation of coal with oxygen and steam to obtain synthesis gas and its further processing to obtain hydrogen. Process analysis was studied on a computer model. In this case, the energy efficiency of the process was found to be 59 %. Most of the energy losses are related to cooling water and flue gas emissions.

In work [6], the "hot water drying" technology, which was developed by US scientists, is considered. The technology makes it possible to reduce the moisture content of brown coal, which hinders its processing. The schematic diagram of the coal fuel preparation technology is complex and contains many additional components and containers.

In [7], the process of coal gasification in a circulating fluidized bed, which operates under a pressure of 8–12 atm and a thermal power of 10 MW, is considered. In this case, it was noted that the technology allows burning high-ash coal of the anthracite type and shows high efficiency of the process. However, the scheme of the technology is complex and contains nodes of ash afterburning, return of coke ash residue and burning of pyrolysis gas. This contributes to additional emissions into the environment.

In [8], the hydrodynamics of a compact gasifier with a double fluidized bed and a bubbling fluidized bed were investigated. The technology is intended for the processing of high-ash coal. Many governing factors have been identified that need to be maintained. The effectiveness of solids recirculation systems has been investigated for their disposal on the bed. Intense back-mixing of solids near the exit from the reactor was observed. A complex flow mechanism was established when using a double fluidized bed and circulation of solids. This complicates the design of the reactor.

Paper [9] discusses the process of two-stage processing of coal using the heat of circulating ash. The schematic diagram of the installation includes the ignition and ash drain chambers, pyro gas discharge and aero fountain remelting. The complexity of the process is also complicated by the scheme with additional solid matter processing units. This causes new emissions into the environment and inclusion in the scheme of ecological cleaning nodes.

Paper [10] describes the process of coal gasification in a fluidized bed in an oxygen environment. The gas generator operates at 700–1000 °C with multidirectional flows of starting materials and liquid flow. The schematic diagram of the installation and operation of the gas generator is complex and contains several flow distribution sections. There are also special conditions regarding the course of the process in a fluidized bed. There is also the possibility of obtaining ash as a by-product, which would be removed together with gaseous products. Ash cleaning and collection, processing or reuse units must be added.

In [11], the productivity of a vertical roller coal mill is considered. In this reactor, it is necessary to control such parameters as uniform mass rate of coal supply, required particle size, and temperature at the output channel. It was found that grinding pressure, primary air temperature, mill motor current tend to increase with an increase in the mass flow rate of supplied coal. The content of volatile substances and moisture in coal improves with an increase in the temperature at the exit from the mill.

The impact on the environment was noted in [12]. It depends on the specific technology used. Coal-fired power generation has a strong impact on environmental emissions. Coal, used in large quantities, has a stronger impact on the environment. But coal could resist these influences.

Study [13] proposed a hybrid oxygen-coal combustion under pressure, which uses fluidized bed combustion and gasification. Both reactors are interconnected by a thermal flow of flue gases. The structural scheme of the reactors is quite complex. The technology makes it possible to use the thermal energy of flue gases under pressure to balance the speed of flows. The hybrid system shows an efficiency of about 34 %. As we can see, this technology also has a complex structure that works under pressure. There is a need to control many parameters to maintain the thermal balance of the system and the environmental friendliness of the scheme.

The reaction products of the considered technologies are hydrogen, synthesis gas, organic compounds, gaseous compounds that could be used in the energy sector.

In work [14], the issue of chemical treatment of coal wastewater is considered. Wastewater contains many mineral impurities. The technology consists of two methods: thermal and membrane method. That is why it is called hybrid membrane purification. The reactor consists of several stages, that is, it undergoes multi-stage cleaning.

Existing coal processing technologies should plan the methods of cleaning the resulting wastewater. This will contribute to the environmental friendliness of technologies.

Research on the processing of oil sludge mixed with hydrocarbon mass is reported in [15]. The product of processing is a type of fuel. The gas generator contained five nozzles, during the operation of which it is necessary to constantly control the flows and their mixing in the reactor for safety and relative completeness of processing. Research was carried out under the influence of basic and special parameters of the process.

From the above analysis, we can conclude that the issues of coal processing are important both for industry and the scientific and research domain. Coal is a complex multicomponent substance that contains many mineral compounds. During the conversion of coal, these compounds are also subject to conversion. And this affects the completeness and mechanism of transformation of the entire process. This point is not considered in existing technologies. Also, the environmental friendliness of the final product and the safety of the process are not considered in existing technologies. The final product will contain residues of these mineral components. All existing technologies have a complex design of reaction nodes and the difficulty of creating an equilibrium of heat and mass exchange in the reaction space.

3. The aim and objectives of the study

The purpose of our work is to devise a new technology of coal gasification in a reactor with a high-temperature heat carrier. This will provide an opportunity to obtain energy from own resources with a rational approach to their use.

To achieve this goal, the following tasks were set:

– to conduct a thermodynamic analysis of possible chemical transformations of coal in LHH;

– to design a scheme of the reactor and check the possibility of the process of converting coal to synthesis gas in a liquid high-temperature heat carrier;

– to develop a basic technological scheme of an experimental and industrial reactor.

4. The study materials and methods

4. 1. The object and hypothesis of the study

The object of our study is the coal processing process. Coal in a country is enough as a mineral to provide energy. However, coal is a non-renewable source of energy. Therefore, it is necessary to invent techniques for its rational processing. This processing should be simple, safe, and environmentally friendly.

Coal is a complex chemical system that contains a large number of minerals. It was assumed that this chemical system has reactivity. The melt of a high-temperature heat carrier was used as a reaction medium. It is also a chemical system, and its components could undergo chemical transformations. To solve this problem, it is necessary to find a technology that would be able to turn this property of matter into a positive and rational fact.

4. 2. Hard and salty coal as raw materials

The basic fact is that most chemical transformations are complex in terms of reaction mechanisms. The best result is thermodynamic confirmation of the course of the corresponding reactions. There are different types of coal: hard, brown, and salted. The main parameter of any coal is ash content, which is expressed by the content of the corresponding minerals. The main content of coal minerals is given in Table 1 [16].

Table 1

Minerals that are part of hard and salt coal

Element	Compound	
	Formula	Name
Cl	$NaCl$	Halit
S	FeS_2	Pyrite
	FeS_2	Pyrite
	$FeSO_4{\cdot}H_2O$	Iron(II) sulphate monohydrate
	$KFe_3(SO_4)_2{\cdot}(OH)_6$	Jarosite
	$Na\ Fe_3(SO_4)_2{\cdot}(OH)_6$	Natrojarosite
	$(K,Na,Fe)_5Fe_3(SO_4)_6(OH)_2{\cdot}9H_2O$	Metavoltine
	$CaSO_4$	Anhydrite
	$CaSO_4{\cdot}0{,}5H_2O$	Hemihydrate
	$CaSO_4{\cdot}2H_2O$	Gypsum
	$Na_2SO_4{\cdot}CaSO_4$	Glauberite
	$MgSO_4{\cdot}6H_2O$	Hexahydrate
	$(Mg,Zn)_8SO_4(OH)_{14}H_2O$	Mooreite
Ca	$CaCO_3$	Calcite
	$CaMg(CO_3)_2$	Dolomite
	$(Ca,Na)Al_2Si_2O_8$	Plagioclase
Al	$Al_4[Si_4O_{10}(OH)_8]$	Kaolinite
	$KAl_2(OH)_2[AlSi_3O_{10}]$	Hydromica
	$Al_2Si_4O_{10}(OH)_2$	Pyrophyllite
	$Mg_2Al_4Si_5O_{18}$	μ – Cordierite
Fe	$FeTiO_3$	Ilmenite
Si	SiO_2	Quartz
Zn	$ZnCO_3$	Smithsonian
P	P_2O_5	Phosphorus anhydride

It is known that some of the primary minerals undergo thermal cracking. Possible processes of thermal cracking of minerals, taking into account the ash content of coal [16], represented by equations (1) to (14), run in the range 1073–2073 K:

$$FeSO_4 \cdot H_2O \longrightarrow FeSO_4 + H_2O, \quad (1)$$

$$2KFe_3(SO_4)_2 \cdot (OH)_6 + 5K_2SO_4 + 5HCl \longrightarrow$$
$$\longrightarrow 3Fe_2(SO_4)_3 + 12KCl + 12H_2O, \quad (2)$$

$$2NaFe_3(SO_4)_2 \cdot (OH)_6 + 5Na_2SO_4 + 5HCl \longrightarrow$$
$$\longrightarrow 3Fe_2(SO_4)_3 + 12NaCl + 12H_2O, \quad (3)$$

$$(K,Na,Fe)_5 Fe_3(SO_4)_6 \cdot (OH)_2 \cdot 9H_2O \longrightarrow$$
$$\longrightarrow (K,Na,Fe)_5 Fe_3(SO_4)_6 \cdot (OH)_2 + 9H_2O, \quad (4)$$

$$CaSO_4 \cdot 0{,}5H_2O \longrightarrow CaSO_4 + 0{,}5H_2O, \quad (5)$$

$$CaSO_4 \cdot 2H_2O \longrightarrow CaSO_4 + 2H_2O, \quad (6)$$

$$MgSO_4 \cdot 6H_2O \longrightarrow MgSO_4 + 6H_2O, \quad (7)$$

$$CaMg(CO_3)_2 \longrightarrow CaCO_3 + MgCO_3, \quad (8)$$

$$FeTiO_3 \longrightarrow TiO_2 + FeO, \quad (9)$$

$$Na_2SO_4 \cdot CaSO_4 \longrightarrow Na_2SO_4 + CaSO_4, \quad (10)$$

$$3Al_4\left[Si_4O_{10}(OH)_8\right] + 4NaCl + 2H_2O \longrightarrow \longrightarrow 4Na\left[AlSi_3O_8\right] + 4HCl + Al_2O_3 + 12H_2O, \quad (11)$$

$$3Al_4\left[Si_4O_{10}(OH)_8\right] + 4Na_2SO_4 \cdot CaSO_4 + 2H_2O \longrightarrow \longrightarrow 4Na\left[AlSi_3O_8\right] + CaSO_4 + 2Al_2(SO_4)_3 + 12H_2O, \quad (12)$$

$$Na_2SO_4 \cdot CaSO_4 + KAl_2(OH)_2\left[AlSi_3O_{10}\right] + 2FeS \longrightarrow \longrightarrow Na_4Al_6SiO_{23}S_4 + CaSO_4 + FeSO_4 + 2H_2O + K_2O, \quad (13)$$

$$Al_4\left[Si_4O_{10}(OH)_8\right] + 4NaCl \longrightarrow \longrightarrow 4Na\left[AlSiO_4\right] + 4HCl + 2H_2O. \quad (14)$$

According to the LHH technology, the target process takes place in the NaCl melt. This is the interaction of coal with water vapor with the formation of synthesis gas. At the same time, chemical transformations of the mineral part of coal and metal oxides falling from the oxidation zone could take place there.

4. 3. Schematic diagram of a laboratory unit for processing coal in a liquid high-temperature heat carrier

A feature of the new technology is the separation of reaction zones in an industrial reactor. The chemical transformation takes place directly in the liquid high-temperature heat carrier (LHH). The process in LHH takes place at atmospheric pressure. Salt was used as a high-temperature heat carrier under laboratory conditions; in the industry there is an opportunity to use slag waste from the coke making process. This gasification technique is promising for the processing of hard, salty, and brown coal. The schematic diagram of the laboratory installation for the coal conversion process was constructed and shown in Fig. 1. The designed laboratory reactor with molten common salt as LHH has passed the workability test [17].

The flow method of research was proposed in the scheme. For this purpose, a bubbling-type reactor located in the furnace was used. The temperature range of the research was maintained at 1273–1373 K. It is possible to use the reaction products in hydrogen processes, having previously separated them from carbon monoxide, as well as directly synthesis gas as a raw material for organic and inorganic synthesis processes.

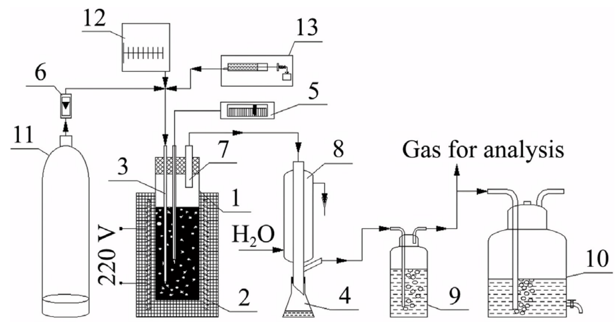

Fig. 1. Schematic laboratory diagram of coal conversion in LHH: 1 – reactor made of quartz or heat-resistant steel; 2 – electric furnace; 3, 7 – quartz tubes; 4 – condensate collector; 5 – thermocouple with a potentiometer; 6 – rotameter; 8 – cooler; 9 – a glass of Drexel; 10 – Marriot capacity; 11 – cylinder with nitrogen or air; 12 – syringe water dispenser; 13 – pneumatic solid material dispenser

5. Results of investigating the coal conversion process in a liquid high-temperature heat carrier

5. 1. Thermodynamic analysis of the coal processing process

First of all, it is necessary to analyze the possibility of the course reaction: thermal cracking, conversion, and oxidation. Thermodynamic analysis will be able to help identify chemical transformations that are likely under the conditions of LHH.

The target reaction is the interaction of coal with water vapor to obtain synthesis gas, which passes through the NaCl melt. In this case, reactions of the mineral part of coal and metal oxides coming from the oxidation zone could take place in the reactor.

Tables 2, 3 give possible reactions of the chemical transformation of carbon. More than 100 reactions have been hypothesized. For each reaction, the equilibrium constant was found in the temperature range of 1073–2073 K. Thermodynamic analysis is given for the processes of conversion and oxidation of carbon and chemical transformations of its mineral component under the conditions of our study.

Table 2

Thermodynamic data on conversion reactions (zone 1) of coal, carbon residue, and metals included in the ash content, and water

Reaction No.	Reactions that may be taking place	ΔH, kJ/mol	Result of reaction course
1	2	3	4
15	$C+H_2O=CO+H_2$	131.28	possible
16	$C+2H_2O=CO_2+2H_2$	90.11	possible
17	$C+CO_2=2CO$	172.45	possible
18	$CO+H_2O=CO_2+H_2$	–41.17	possible (≤1073)
19	$C+2H_2=CH_4$	–74.85	unlikely
20	$2C+2H_2O=CH_4+CO_2$	15.26	possible (>1073)
21	$CO+3H_2=CH_4+H_2O$	–206.13	possible (<1373)
22	$CO_2+4H_2=CH_4+2H_2O$	–164.96	possible (≤1373)
23	$2Fe_2O_3+3C=4Fe+3CO_2$	463.79	possible
24	$Fe_2O_3+C=2FeO+CO$	181.93	possible
25	$2Fe_2O_3+C=4FeO+CO_2$	191.41	possible

Continuation of Table 2

1	2	3	4
26	Fe_2O_3+3C=2Fe+3CO	490.57	possible
27	2Fe+3H_2O=Fe_2O_3+3H_2	–96.73	possible
28	Fe+H_2O=FeO+H_2	–23.04	possible
29	3Fe+4H_2O=Fe_3O_4+4H_2	145.08	unlikely
30	2FeO+H_2O=Fe_2O_3+H_2	–50.65	possible
31	3FeO+H_2O=Fe_3O_4+H_2	–80.77	possible
32	2Al_2O_3+3C=4Al+3CO_2	2170.85	unlikely
33	2Al_2O_3=4Al+3O_2	3351.38	unlikely
34	2Al+3H_2O=Al_2O_3+3H_2	–950.26	possible
35	SiO_2+C=Si+CO_2	517.43	unlikely
36	SiO_2=Si+O_2	910.94	unlikely
37	Si+2H_2O=SiO_2+2H_2	–427.32	possible
38	TiO_2+C=Ti+CO_2	551.24	unlikely
39	TiO_2=Ti+O_2	944.75	unlikely
40	Ti+2H_2O=TiO_2+2H_2	–461.13	possible
41	$CaSO_4$=CaO+SO_3	405.34	unlikely
42	Ca+H_2O=CaO+H_2	–393.28	possible
43	CaO+CO_2=$CaCO_3$	–178.23	possible
44	Mg+H_2O=MgO+H_2	–359.68	possible
45	MgO+CO_2=$MgCO_3$	–100.85	possible
46	$ZnCO_3$=ZnO+CO_2	70.91	possible (<1773)
47	Zn+H_2O=ZnO+H_2	–106.3	possible
48	2P_2O_5=4P+5O_2	2944.6	unlikely
49	2P_2O_5=2P_2+5O_2	3302.1	unlikely
50	2P+5H_2O=P_2O_5+5H_2	–263.25	possible (<1073)
51	P_2+5H_2O=P_2O_5+5H_2	–442	possible (<1073)
52	2H_2O=2H_2+O_2	241.81	unlikely
53	H_2+0,5S_2=H_2S	–84.78	possible
54	CO+0,5S_2=COS	–95.36	possible
55	C+S_2=CS_2	–11.67	possible
56	2SO_3=2SO_2+O_2	197.9	possible
57	2SO_2=S_2+2O_2	722.17	unlikely
58	S_2+4H_2O=2SO_2+4H_2	245.07	unlikely
59	S_2+2H_2O=2H_2S+O_2	314.05	unlikely
60	$CaCO_3$+2HCl=$CaCl_2$+H_2O+CO_2	–39.79	possible
61	CaO+2HCl=$CaCl_2$+H_2O	–218.02	possible
62	2CaO+2Cl_2=2$CaCl_2$+O_2	–321.66	possible
63	4CaO+5Cl_2=4$CaCl_2$+2ClO_2	–434.12	possible
64	$MgCO_3$+2HCl=$MgCl_2$+H_2O+CO_2	0.35	possible
65	MgO+ 2HCl=$MgCl_2$+H_2O	–100.5	possible
66	2MgO+ 2Cl_2=2$MgCl_2$+O_2	–86.62	possible
67	4MgO+5Cl_2=4$MgCl_2$+2ClO_2	35.96	unlikely
68	TiO_2+ 4HCl=$TiCl_4$+2H_2O	67.21	unlikely
69	TiO_2+2Cl_2=$TiCl_4$+O_2	181.59	possible (>1773)
70	2TiO_2+5Cl_2=2$TiCl_4$+2ClO_2	572.38	unlikely
71	Ti+2Cl_2=$TiCl_4$	–763.16	possible
72	Ti+4HCl=$TiCl_4$+2H_2	–393.92	possible
73	SiO_2+4HCl=$SiCl_4$+2H_2O	139.04	unlikely
74	SiO_2+2Cl_2=$SiCl_4$+O_2	253.42	unlikely
75	2SiO_2+5Cl_2=2$SiCl_4$+2ClO_2	716.04	unlikely
76	Si+2Cl_2=$SiCl_4$	–657.52	possible
77	Si+4HCl=$SiCl_4$+2H_2	–288.28	possible
78	Al_2O_3+6HCl=2$AlCl_3$+3H_2O	95.78	unlikely
79	2Al+6HCl=2$AlCl_3$+3H_2	–854.48	possible
80	2Al+3Cl_2=2$AlCl_3$	–1408.34	possible
81	2Al_2O_3+6Cl_2=4$AlCl_3$+3O_2	534.7	unlikely

Continuation of Table 2

1	2	3	4
82	$4Al_2O_3+15Cl_2=8AlCl_3+6ClO_2$	1697	unlikely
83	$ZnCO_3+2HCl=ZnCl_2+H_2O+CO_2$	96.15	possible
84	$ZnO+2HCl=ZnCl_2+H_2O$	25.24	possible
85	$2ZnO+2Cl_2=2ZnCl_2+O_2$	164.86	possible
86	$4ZnO+5Cl_2=4ZnCl_2+2ClO_2$	538.92	possible (>1773)
87	$P_2O_5+10HCl=2PCl_5+5H_2O$	471.47	unlikely
88	$2P_2O_5+10Cl_2=4PCl_5+5O_2$	1514.84	unlikely
89	$4P_2O_5+25Cl_2=8PCl_5+10ClO_2$	4075.68	unlikely
90	$SO_2+Cl_2=SO_2Cl_2$	–66.27	unlikely
91	$SO_3+2HCl=SO_2Cl_2+H_2O$	–24.51	unlikely
92	$CO+Cl_2=COCl_2$	–108.97	unlikely
93	$CO_2+2HCl=COCl_2+H_2O$	116.82	unlikely
94	$2NO+Cl_2=2NOCl$	–77.34	possible (<1373)
95	$NaCl+H_2O=NaOH+HCl$	134.27	possible (>800)
96	$2NaCl+2H_2O=2NaOH+H_2+Cl_2$	453.16	possible (>1773)
97	$2Na^++2H_2O-2e=2NaOH+H_2$	111.52	possible (>1773)
98	$2NaOH+CO_2=Na_2CO_3+H_2O$	–126.40	possible (<1373)
99	$Na_2O+CO_2=Na_2CO_3$	–319.71	possible
100	$2NaCl+H_2O=Na_2O+2HCl$	461.85	unlikely
101	$2NaCl+H_2O=Na_2O+H_2+Cl_2$	646.47	unlikely
102	$2NaCl+3H_2O=Na_2O+2H_2+2HCl+O_2$	945.47	unlikely
103	$2Na^++H_2O-2e=Na_2O+H_2$	304.83	unlikely
104	$2NaOH=Na_2O+H_2O$	193.31	unlikely
105	Na_2CO_3+2HCl«$2NaCl+H_2O+CO_2$	–142.14	possible (≤1773)
106	$2HCl=H_2+Cl_2$	184.62	unlikely
107	$2Cl_2+2H_2O=4HCl+O_2$	114.38	possible

Table 3

Thermodynamic data on air oxidation reactions (zone 2, zone 3) of coal conversion zone products, carbon residue, and metals included in the ash content, and water

Reaction No.	Reactions that may be taking place	ΔH, kJ/mol	Result of reaction course
108	$C+O_2=CO_2$	–393.51	possible
109	$2C+O_2=2CO$	–110.53	possible
110	$2CO+O_2=2CO_2$	–282.98	possible
111	$N_2+2O_2=2NO_2$	68.38	unlikely
112	$N_2+O_2=2NO$	182.58	unlikely
113	$2NO+2O_2=2NO_2$	–114.2	unlikely
114	$FeSO_4=FeO+SO_3$	266.89	possible
115	$4FeS+3O_2=2Fe_2O_3+2S_2$	–985.9	possible (<1773)
116	$4FeS_2+11O_2=2Fe_2O_3+8SO_2$	–3309.92	possible
117	$4FeO+O_2=2Fe_2O_3$	–584.92	possible
118	$2Fe+O_2=2FeO$	–529.7	possible
119	$3Fe_2O_3+CO=2Fe_3O_4+CO_2$	–50.76	possible
120	$Fe_2O_3+CO=2FeO+CO_2$	9.48	possible
121	$FeO+CO=Fe+CO_2$	–18.13	possible
122	$Fe_2O_3+3CO=2Fe+3CO_2$	–26.78	possible
123	$3Fe+4CO_2=Fe_3O_4+4CO$	14.79	possible
124	$3FeO+CO_2=Fe_3O_4+CO$	–39.60	possible
125	$FeO+CO_2=FeCO_3$	–79.79	possible (≥1773)
126	$Al_2O_3+3CO=2Al+3CO_2$	826.75	unlikely
127	$SiO_2+2CO=Si+2CO_2$	344.98	unlikely
128	$TiO_2+2CO=Ti+2CO_2$	378.79	unlikely
129	$CaO+CO=Ca+CO_2$	352.11	unlikely
130	$MgO+CO=Mg+CO_2$	318.51	unlikely
131	$ZnO+CO=Zn+CO_2$	65.13	possible
132	$C+0{,}5N_2+0{,}5H_2=HCN$	132	possible (≥1273)

One of the main components of the mineral part of coal is iron. In the melt, it is possible to undergo reduction reactions of Fe(III) oxide with carbon to Fe and Fe(II). These Fe compounds and Fe(II) oxide when interacting with water could generate hydrogen: equations (27) to (31). These reactions are related to the ancient method of the "iron-steam technique for obtaining hydrogen" [18].

The least likely reactions of metal oxidation with water are reactions with compounds of aluminum (32) to (34), silicon (35) to (37), and titanium (38) to (40). But the result of oxidation is the corresponding oxides (34), (37), (40). The temperature of the study may favor the oxidation of calcium (42) and magnesium (44) by water. As a result, it is possible to form calcium carbonate (43). Decomposition reactions of calcium sulfate are considered unlikely (41). As a result, there is a possibility of formation of magnesium carbonate (45). The most likely reactions in the experimental temperature range of 1073–1773 K are possible reactions of the decomposition of zinc carbonate (46) and the oxidation of zinc by water (47).

It was found that at a temperature of 1173 K, there is a possibility of the process of oxidation of phosphorus with water (50), (51). The reactions of the decomposition of phosphorus oxide (48), (49), and water (52) turned out to be unlikely in the experimental temperature range. It was found that sulfur-containing reaction products could be formed in the experimental temperature range, namely, hydrogen sulfide (53), carbon sulfide (55), and carbonyl sulfide (54). The reaction of reduction of sulfur oxides is possible in the course of reactions (56), (57). The process of oxidation of sulfur by water is unlikely (58), (59).

Thermodynamic analysis revealed the possibility of reactions of titanium (69), silicon (77), and aluminum (79), calcium carbonates (60), magnesium (64), and zinc (83), calcium oxides (61), and zinc (84) with hydrogen chloride in the experimental temperature range. Chlorine has been observed to interact with titanium (71), silicon (76), aluminum (80), calcium oxides (62), (63), and zinc (85) in the temperature range 1073–2073 K. Reactions of chlorine and hydrogen chloride with oxides of phosphorus, sulfur, carbon, and nitrogen turned out to be unlikely in the tested temperature range.

Regarding the transformation of salted coal, specific reactions of sodium chloride (95) to (103) with water were found. These reactions are possible when equilibrium is not reached in the reactor. The most probable are endothermic reactions (95), (96). According to [19], at a temperature of 1073–1223 K, the sodium chloride melt exists in molecular form and in the form of fragments of the original framework structure. At higher temperatures, they exist in ionic form. The reaction of the sodium ion (103) with the formation of an alkali is endothermic and may occur at a temperature above 1373 K. Interaction of the formed sodium hydroxide with carbon dioxide is probable at temperatures up to 1373 K. In this case, a carbonate compound (98) is formed. The interaction of sodium oxide with carbon dioxide (99) is probable in the experimental temperature range. The interaction of sodium chloride with water and carbon dioxide with the formation of hydrogen chloride and carbonate (reverse reaction 105) is likely at a temperature of more than 1773 K. The interaction of chlorine with water (107) is possible in the experimental temperature range.

When working with salted coal, there is a danger of the synthesis of toxic compounds such as dioxins. This is the obtained product of the reaction of HCl during conversion with CO_2, molecular chlorine was found. Chlorine concentration decreases in the temperature range of 673–1173 K.

Carbon and related products, which come with the melt from the conversion zone, enter the process of air oxidation (108) to (110). The temperature of the melt was 1773 K. This increases the temperature of the circulating melt between the zones to 2073 K. In this way, the circulation of the melt could ensure the course of endothermic reactions in the conversion zone. There is also a possibility of other reactions. The reaction of the formation of nitrogen oxides (111) to (113) in the experimental temperature range is unlikely.

Other chemical transformations of metal-containing compounds could take place in the reaction zones of oxidation and post-oxidation. It was found that in the experimental range of temperatures the following reactions are likely:

– decomposition of iron sulfate (114),

– oxidation of pyritic sulfur (116),

– oxidation of iron oxide to oxide (117) and iron to oxide (118),

– reduction of iron oxide with carbon monoxide (119) to (122),

– oxidation of iron and Fe(II) oxide by carbon dioxide (123), (124).

Analysis revealed the following facts:

– the reaction of iron carbonate formation (125) could take place at a temperature above 1773 K;

– the process of reduction of aluminum (126), silicon (127), titanium (128), calcium (129), magnesium (130), and zinc (131) oxides using carbon monoxide is unlikely in the experimental temperature range;

– the reaction of hydrogen cyanide formation (132) is possible at a temperature of more than 1273 K. In this case, the obtained compounds are partially sent to the conversion zone and the post-oxidation zone;

– the process of oxidation of iron sulfide (115) takes place at a temperature below 1773 K.

The results of thermodynamic analysis revealed that from the conversion zone to the oxidation zone, along with the melt, the following also flows in:

– unreacted carbon;

– iron and other metals that have been reduced by carbon and hydrogen;

– NaOH;

– metal oxides and chlorides.

The following facts were discovered:

– chlorine binds to calcium and zinc chlorides;

– sodium compounds are in the form of hydroxide;

– sulfur binds into sulfates or sulfides [16];

– metal oxides and carbonates are part of the slag that goes from the oxidation zone to the conversion zone.

Calculation of the amount of melt in the cycle is determined taking into account the thermal effects of the reactions and consumption of oxidants. In the post-oxidation zone of the reactor, mineral compounds are removed in the form of higher oxides. This makes it possible to obtain slag for further use.

5. 2. Laboratory studies of the coal processing process in a liquid high-temperature heat carrier

The coal conversion process was studied in the reactor, the schematic diagram is shown in Fig. 3. Reagents were fed into the reactor using a dispenser. Water was supplied by a syringe dispenser, solid material was supplied by a pneumatic dispenser, and gaseous reagents were controlled by rotameters. In technology, two types of reactors were investigated (Fig. 2) [20].

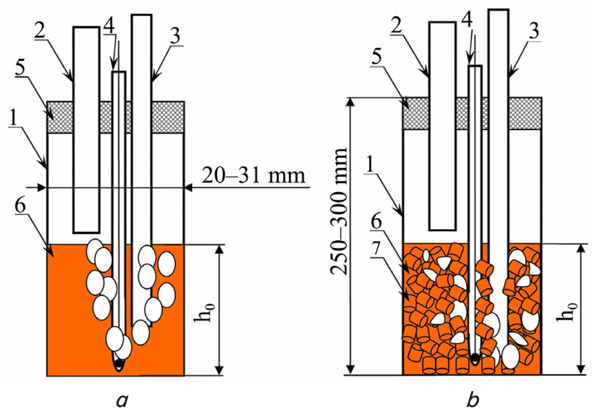

Fig. 2. Schematic diagram of coal conversion reactors in liquid high-temperature heat carrier: *a* — reactor without a nozzle; *b* — reactor with nozzle; h_0 is the height of the melt; 1 — reactor made of quartz or heat-resistant steel; 2 — quartz tube for the removal of reaction products; 3 — quartz tube for the introduction of raw materials and water; 4 — a pocket for a thermocouple; 5 — cap; 6 — molten salt; 7 — quartz nozzle.

From the beginning, it is necessary to prepare the melt for the coal conversion reaction in LHH. It was noted that after the course of the process, a melt with residual carbon materials remains in the reactor. Checking the chemical composition of the ash showed that it contains various metal oxides (Table 4).

Table 4

Chemical content of ash

Ash content, %wt.±0,05									
SiO_2	Fe_2O_3	CaO	MgO	SO_3	Al_2O_3	TiO_2	P_2O_5	K_2O	Na_2O*
29.8	31.8	4.45	1.52	3.14	21.06	0.55	0.17	1.25	6.5

*Note: * – in the ash of salt coal from the Novomoskovsk deposit.*

The processes of thermal pyrolysis, conversion, and oxidation of coal were also investigated. In the laboratory reactor, the conversion and oxidation processes were studied separately. The results of the research showed the reality of processes in the melt of a liquid high-temperature heat carrier for different types of coal (Fig. 3).

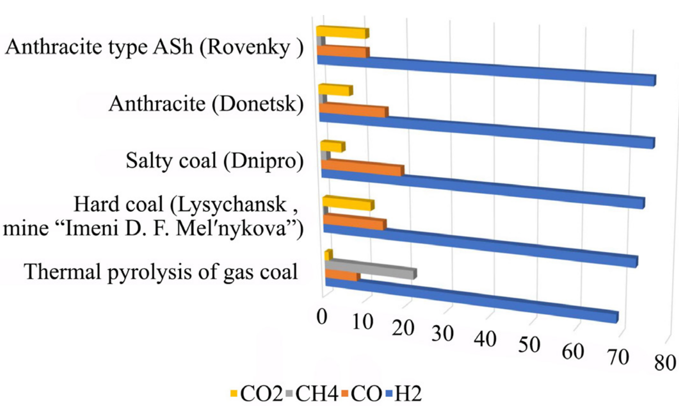

Fig. 3. Results of experimental studies of coal processing in liquid high-temperature NaCl melt

Hard coal and salt coal are classified as young coal, while anthracite is categorized as mature coal. The products of the reaction are synthesis gas in the ratio CO/H_2=1/4,7. It was found that the main indicators of coal are coal 00, gas content, and ash content. The degree of coal maturity characterizes the thermal stress of the process. The gas content and ash content of coal characterize the reaction rate. These parameters make it possible to choose the design of the reactor and the optimal mode of its operation. The processes were carried out on pre-degassed coal of different degrees of maturity and content of mineral impurities.

Analysis of the technological modes of operation of the "solid body – evaporating liquid – melt" system was carried out. The reactor using the bubbling mode, which excludes the removal of gaseous reaction products, turned out to be the best. The products that could be removed from the reactor are melt, ash, coal, and carbon [20].

5. 3. Basic technological scheme of the experimental installation for obtaining synthesis gas in the melt of a liquid high-temperature heat carrier

The conversion process was investigated by continuously feeding coal and water into the sodium chloride melt. Research was carried out on gas and salt coal. The main characteristics of the course of the coal conversion process in a liquid high-temperature heat carrier made it possible to design a three-zone reactor scheme. A liquid high-temperature heat carrier is a slag melt. The schematic diagram of the reactor is shown in Fig. 4 [21].

The peculiarity of operation of the installation with a three-zone reactor is the use of the heat of one reaction at the expense of another. The endothermic reaction of the conversion of coal with water is in equilibrium with the process of oxidation of part of the coal with air:

$$C+H_2O=CO+H_2-131.3\text{kJ/mol},$$

$$C+O_2=CO_2+393.5\text{ kJ/mol}.$$

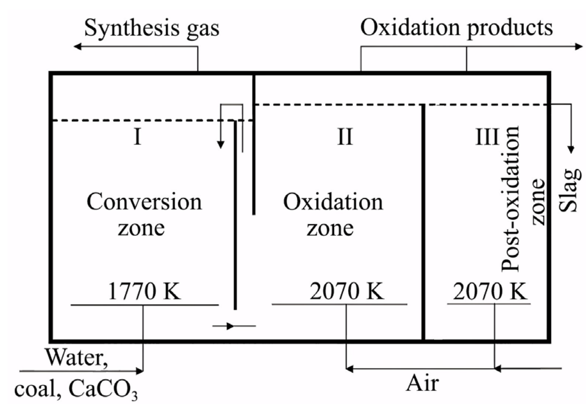

Fig. 4. Schematic diagram of the coal processing reaction unit in liquid high-temperature heat carrier under atmospheric pressure: I — conversion zone (1770 K); II — oxidation zone (2070 K); III — post-oxidation zone (2070 K); Flows: hydrocarbon mixture and $CaCO_3$; air; synthesis gas; oxidation products; slag

The conversion zone and the oxidation zone are separated by the gas phase. This makes it possible to ensure explosion safety of the process. The slag circulates between these zones with the appropriate multiplicity. This maintains the temperature regime. Circulation of slag occurs when there is a difference in the density of gas-liquid mixtures in the reaction zone. Ash is removed from zone III of the reactor (content up to 1 % by weight of carbon) as a slag melt.

Gaseous products of each zone are high-potential heat carriers. They are used for heating flows in the reactor and electricity. Synthesis gas is purified from the sulfur compound from which the commercial product (sulfur or sulfuric acid) is obtained. Flue gases are directed to catalytic purification of toxins.

6. Discussion of results based on investigating the process of processing coal in a liquid high-temperature heat carrier

It is a well-known fact that solid material is difficult to undergo any transformation. Scientists of the world have constantly researched and engaged in the development of technologies for its transformation. The world's most famous technologies (Lurgi, Koppers-Totzek, Winkler, Texaco) are associated with different modes of coal processing: stationary, moving, and fluidized beds. Each of them [5–15] has its problematic issues: in stationary and moving layers, it is difficult to create equilibrium of the reaction system in the reactor and complete conversion; the fluidized bed has a special mode of creation and constant management, there is a probability of removal of ash and carbon from the reactor, the difficulty of their separation. More often, these are catalytic technologies that use alkaline, alkaline earth metals as catalysts. Also, these technologies have a complex scheme of the reactor and conversion plant. Almost all schemes work according to the same principle. This is a combination of the reaction zone and the processing zone in one gas generator. In this case, the structural, technological, and economic vision of the scheme and the reactor is complicated.

The procedure for carrying out the process is complex, both the creation of a technological scheme and its maintenance. Most of the processes take place under pressure, there is a need for a preliminary stage of coal or coal mixture preparation. In some cases, there is a need not only for desulfurization but also for deashing and dehydration of raw materials. There is a need to control the parameters of heat and mass exchange, to create technological and environmental safety systems. According to existing technologies, wastewater containing mineral impurities that cannot be regenerated is obtained.

The reactor with a liquid high-temperature heat carrier proposed in this study has a simple structure (Fig. 2). As a heat carrier, it is possible to use both common salt and slag of a coke-making enterprise. The heat carrier must create a melt of the substance at a high temperature, which is the reaction temperature. Thermodynamic analysis proved that there is a possibility of the course of reactions related to the mineral content of the coal. It was found that a large number of chemical transformations of hard coal could take place in the sodium chloride melt (Table 2, equations (27) to (31), (43), (45) to (47), (50), (51) , (53) to (57), (60) to (64), (69), (71), (76), (77), (79), (80), (83) to (85), of salted coal (95) to (103). Therefore, it is necessary to take into account that the metal-rich components could be regenerated (Table 3). These compounds become part of the melt, are transported from one zone of the reactor to another and could be further used in the technology compounds or halogen-containing compounds become part of the slag, are removed from the reactor, but in the technology, it is necessary to control the slag content for its further use.

The LHH technology can be considered environmentally safe, the melt is used further; reaction products are compounds that could be used as an energy resource or raw material for the synthesis of organic products. If possible, fine cleaning and recovery systems are provided.

A feature of the LHH technology is that it uses three aggregate states (Fig. 2), which is not found in existing technologies. The solid phase is coal used as raw material. The gas phase is water vapor used as a reagent. The liquid phase, which is directly the melt of the heat carrier, is used as an equilibrium creator for heat and mass exchange in the reactor. A three-zone reactor with one LHH melt between the zones has been proposed (Fig. 4).

A special fact during the study of the process of gasification of coal in a liquid high-temperature heat carrier was that a balanced system of mass and heat exchange is created in the reactor. This contributes to the uniform distribution of starting substances in the volume of the reactor. In existing technologies, such a fact is not observed, as it is difficult to detect and investigate.

Two types of reactors were designed for research (Fig. 2). This made it possible to determine the influence of reaction parameters on the course of the conversion reaction. It was found that gasification of coal in LHH proceeds with satisfactory speed, high yield, and selectivity. As a result, synthesis gas is obtained with the ratio (Fig. 3) $CO/H_2=1/4\div1/7$. It is more often used as a target product in the production of gasoline and diesel fuel, other hydrocarbons, etc. There is a possibility of selective separation of substances for use both in the recovery processes of organic and inorganic synthesis, and for energy conversion as an alternative source of energy. Each chemical process has its own course parameters: temperature, pressure, concentration, ratio, etc. Therefore, the research technology should be planned in such a way that it is optimally profitable and rational for the further use of the product. It is necessary to be very sure about the purity of the product from the remains of slag or coal that did not react. When using hydrogen as an alternative fuel, it is necessary to clearly approach the issue of separating it from carbon monoxide. It is also necessary to invent a solution for the use of carbon monoxide that has been separated.

The technology in liquid high-temperature heat carrier turned out to be very interesting. It could combine substances in three aggregate states for one chemical transformation in one reactor. For a better understanding of the mechanism of the simultaneous course of chemical transformations during the interaction of the gas-liquid-solid system. There is no such system in existing technologies. They work more with two-phase systems. For research technology in LHH, there is a need to check the progress of other processes with various complex starting substances. Research of the mathematical component of the course of processes in a liquid high-temperature heat carrier is the next task. This will make it possible to predict and, if possible, automate processes. Also, further research may involve searching for other types of heat carriers that would allow processes to proceed at lower temperatures and atmospheric pressure.

In general, the technology with a liquid high-temperature heat carrier has technological, energy, and environmental potential in the development of the chemical, oil refining industry, etc.

7. Conclusions

1. Possible chemical transformations of coal in molten sodium chloride have been determined. Thermodynamic analysis revealed that the reactions take place in the temperature range of 1073–2073 K. According to the mineral composition of hard coal in the melt, it could undergo up to 30 chemical transformations, salty coal up to 10 chemical transformations. It was found that the mineral component of coal, which is recovered in the melt, does not affect the course of the target reaction. It is part of the liquid high-temperature heat carrier and is removed from the reactor together with the slag. It is possible to use the slag further.

2. A principle scheme for processing coal under laboratory conditions has been developed. Two types of reactors were investigated and the optimal mode for the coal processing process was determined. The optimality and guarantee of the course of the coal gasification process was investigated and its validity was proved. It was also analyzed that the technology in LHH could become a promising technology. It has technological, environmental, and energy potential for industrial development, especially in coal processing. It is noted that when processing any type of coal, synthesis gas is obtained in the ratio $CO/H_2=1/4\div1/7$.

3. Based on our research results, the technological scheme of the reactor with LHH was designed and proposed. The reactor contains three zones: conversion, oxidation, and post-oxidation. All zones are connected by the same principle through a liquid high-temperature heat carrier. It maintains the equilibrium of heat and mass exchange between reactor zones. In this reactor, the process takes place simultaneously in the gas-liquid-solid system, that is, in one reactor, the interaction of three aggregate states at once.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study, as well as the results reported in this paper.

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Data availability

All data are available, either in numerical or graphical form, in the main text of the manuscript.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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