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Utilization of Hydrogen-Containing Gas Waste from Deep Oil Refining at a Hybrid Power Plant with a Solid Oxide Fuel Cell †

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Abstract: The article is devoted to the issues of the utilization of hydrogen-containing gas wastes in oil refining deep processing. Gas wastes consist of hydrogen, methane, ethane, propane, butane, other saturated and unsaturated C5-C7 hydrocarbons, sulfur compounds, carbon monoxide, carbon dioxide, nitrogen and oxygen. The use of a hybrid power plant for efficient conversion of the potential energy of the gas mixture into electrical and thermal energy is proposed. It is shown that gas waste from oil production has a net calorific value comparable to the calorific value of natural gas (46 and 49 MJ/kg, respectively). Fuel gas is a valuable product that can be used after desulfurization instead of burning in the atmosphere. The article proposes the developed composition of the adsorbent for hydrogen sulfide capturing, including 40% wt. bentonite, 40% wt. calcium oxide, 10% wt. zinc oxide and 10% wt. manganese oxide. The capture rate was 98.3%. A comparison of various types of fuel for a hybrid power plant with a high-temperature fuel cell and an assessment of the efficiency of using gas waste from oil refineries was carried out. It is shown that fuel gas from oil production waste has a high potential for use in power plants due to its high calorific value and a number of other advantages compared to natural gas.

Keywords: hybrid power plant; solid oxide fuel cell (SOFC); gas turbine; hydrogen energy

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1. Introduction

Every year, society produces millions of tons of solid, liquid and gaseous waste in connection with social, agricultural and industrial production. If a set of organizational measures at the government level for the management and disposal of waste is not implemented, then humanity may face serious problems, such as an impact on health and the environment. At the same time, for the production of electrical energy and heat for the implementation of man-made activities, it is necessary to burn a huge amount of fossil fuels—coal, oil and natural gas. The constant growth in consumption causes excessive and unregulated use of fossil resources, highlighting their limitation, as well as causing environmental pollution due to increased greenhouse gas emissions. In this regard, the scientific community is constantly looking for renewable and alternative fuel sources, on the one hand, and environmentally friendly ways to manage waste, on the other hand [1]. These two big problems currently facing the world community can be combined and solved by the development and implementation of waste disposal technologies in clean renewable energy production cycles.

Recycling technologies offer the conversion of waste into fuel forms such as bioethanol, biobutanol, biogas, biogytan, LNG and synthesis gas through incineration, pyrolysis, gasification or biological treatment processes such as anaerobic digestion and fermentation, as well as a combination of different technologies that can be used to meet the growing demand for energy. While preserving the environment, waste disposal technologies ensure a sustainable supply of fuel. The quality of biofuel is affected by the raw materials from which the final product is made and the processing technology, with the production of a product that meets the needs of consumers to the maximum extent possible while complying with environmental requirements during production. The ways, technologies and methods of disposal and reuse of waste are important for the sustainable development of society and are among the priority areas for the development of science and technology in the world [2].

A modern oil refinery consists of several complex successive technological chains that end with the production of a certain fuel component or other petroleum products. Each link in the chain—a technological installation—has a specific purpose. The final link is the mixing of the components in the required proportions to ensure compliance with all technical and environmental requirements. Another important task in the production of environmentally friendly fuels is to reduce the harmful effects on the environment and humans. The solution to this problem is achieved by investing in the development of technological processes, reducing emissions into the atmosphere and eliminating wastewater discharges into water basins and soils. It would be ideal to create a completely closed production. Figure 1 shows the technological scheme of an oil refinery.

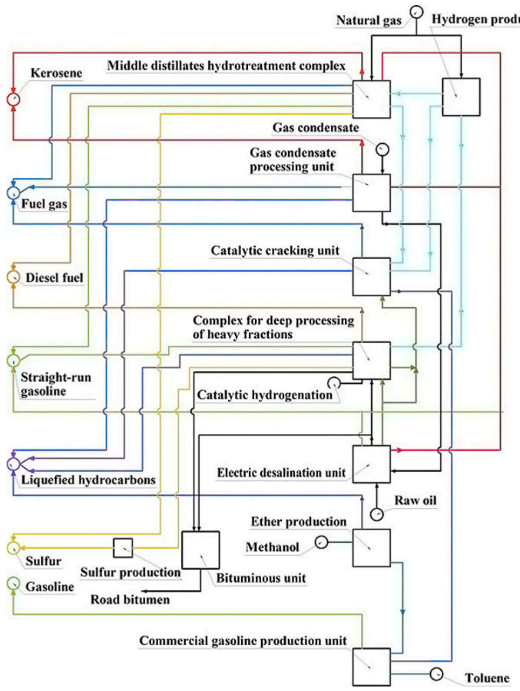

Figure 1. Technological scheme of an oil refinery. The lines in the figure indicate the stages of the oil refining process.

Oil refineries perform desalination, dehydration and the subsequent separation of oil into fractions: hydrocarbon gas, propane–butane fraction, gasoline reaction, kerosene fraction, diesel fraction, vacuum gas oil and tar. Hydrogen, which is obtained by steam reforming of methane and purified by pressure swing adsorption, is used for hydrogenation. The hydrocarbon gas leaving the unit is discharged into the fuel network. By-products of oil refining units are hydrogen-containing and hydrocarbon gases, acidic water and sulfur [3].

Thus, at oil refineries, after atmospheric and vacuum oil processing, as well as installations of catalytic processes, hydrocarbon gases are produced. These gases are of three types: liquefied gases, which are further used as raw materials for petrochemical industries, and fuel gases of two types—those containing a lot of ethane and those containing a lot of hydrogen.

One of the waste products of oil production is fuel gas. Its average composition is presented in Table 1. Fuel gas consists mainly of hydrogen, light hydrocarbons, carbon oxides and sulfur compounds (hydrogen sulfide, mercaptans, etc.). Fuel gas is used partly at the refinery itself to kindle furnaces, most of it is discharged or burned in the atmosphere, which negatively affects the environment and is an irrational use of resources. It is advisable to isolate gas flows with the same type of characteristic properties with their subsequent processing because the beneficial use of these gases is waste disposal and further increases the depth of oil refining. The use of such gases is a cost-effective solution.

Table 1. Composition of gaseous waste used as fuel for SOFC.

Element	Research Fuel
O2	0.588
N2	4.101
H2	3.868
CH4	21.3
C2H6	19.063
C2H4	3.217
C3H8	23.37
C3H6	3.599
C4H10	17.33
C5H12	3.538
Sum of C6	0.92
Sum of C7	0.22
CO2	0.745
H2S	0.3991

Fuel gas is a mixture of methane and C2–C7 hydrocarbons with trace amounts of other compounds. The pollutants are hydrogen sulfide (average content 0.634 g/m^3) and mercaptans (average content 0.1167 g/m^3) according to data from an operating oil refinery. The calorific value of fuel gas is 46 MJ/kg, which is equivalent to natural gas with a calorific value of 49 MJ/kg. Thus, fuel gas is a valuable waste that should be processed after cleaning from sulfur-containing compounds and not discharged into the atmosphere or burned in flares.

Interest in the use of industrial waste as a fuel is due to the high content of methane, constant production, relatively low cost of raw materials and increasing resource potential (due to population growth and increasing demand for electricity), improving the environmental friendliness of the energy industry and waste processing [1].

It is expedient to utilize gas wastes from oil refining by burning fuel gas in gas turbines after purification from sulfur impurities. According to our calculations, with a content of sulfur compounds in the fuel gas of more than 450 parts per million and a natural gas flow rate of more than 900,000 Nm^3 per year without appropriate purification from sulfur, it is possible to add fuel gas to natural gas but in an amount not exceeding 3%, taking into account fuel consumption and the requirements of the rules for power boilers. An increase in the content of unpurified fuel gas leads to corrosion and adversely affects the technical condition of the heating surfaces of the power boiler and burners. Since the fuel gas does not contain polycyclic aromatic and asphaltene-tar compounds, there should be no coking of the fuel gas. With an increased content of sulfur compounds in the fuel, its coking capacity increases.

Sulfur compounds must be removed prior to use on power equipment due to increased corrosivity. In addition, during combustion, it forms sulfur oxides (SOx), which pollute the environment. In SOFC, sulfur compounds bind to the nickel catalyst and block it. The concentration of H_2S in the fuel gas should not exceed 2.82 mg/m^3 for SOFC operation [4]. The most commonly used method for removing hydrogen sulfide is adsorption [5,6]. Depending on the initial concentration of H_2S in the fuel gas, a two-stage desulfurization system may be required, greatly increasing both investment and maintenance costs [7].

In addition to H_2S, the fuel gas contains carbon dioxide, which can be removed together with sulfur at the pretreatment stage.

An alternative option for using oil production wastes can be their processing as fuel in the thermodynamic cycle of a gas piston plant, the electrochemical cycle of a high-temperature fuel cell, or in a hybrid fuel cell–gas turbine cycle.

Table 2 provides a comparison of the different technologies that can be used for the disposal of industrial waste [6].

Table 2. Comparison of cogeneration technologies for biogas utilization.

Technologies	Gas Piston Units	Gas Turbines	Fuel Cells	SOFC-GT
Power, kW	110–4400	600–22,000	100–2800	30–250
Electrical efficiency, %	30–42	19–34	36–50+	50–60+
Thermal efficiency, %	35–49	40–52	30–40	30–40
Fuel pressure, kPa	115–653	791–2859	239–308	618–791
Equipment cost ($ kW^{-1})	465–1600	1100–2000	3800–5280	5000-7000
Fuel purification cost ($ kW^{-1})	0–500	0–500	500–3000	500–3000
Operations and maintenance ($ kWh^{-1})	0.01–0.025	0.008–0.01	0.004–0.019	0.012–0.025
Availability, %	90–96	95–97	90–95	75–80
Overhaul (h)	28,000–90,000	30,000–50,000	10,000–80,000	30,000–50,000
NOx (g GJ-1)	6.45–374.1	43.43–120.4	1.29–2.58	40–110.2
CO2 (g GJ-1)	70.09–928.8	52.89–212.4	2.58–6.88	45.8–190.4

Gas piston engines are used to provide electricity to small and medium power consumers. Fuel cells have high electrical efficiency, but a capacity of up to 1 MW. Gas turbines have low inertia, but traditionally are used in large-scale power generation. Gas piston engines are reliable, economical, commercially available and optimized for biogas fuel; therefore, they can also use industrial gaseous waste as fuel. Unlike gas piston plants, gas turbines have lower electrical efficiency and high sensitivity to environmental conditions. Also, gas turbines require high fuel pressure at the inlet, and fuel compression is an expensive and energy-intensive process. In fuel cells, electrochemical conversion of fuel takes place, which, in comparison with combustion, makes it possible to consume less fuel and reduce emissions of greenhouse gases and other harmful substances, such as ash. However, since SOFCs are a relatively new technology, they are very expensive. The overall cost of an oil and gas waste fuel cell system project increases due to the need to purify the fuel, which requires an efficient industrial gas waste pretreatment system. The SOFC-GT hybrid system, combining electrochemical and thermodynamic cycles, makes it possible to achieve an electrical efficiency of more than 60%. But the SOFC-GT system includes a huge number of individual installations and the fuel flows between them, which is associated with the complexity of management and maintenance. Hybrid power plants are presented in the form of pilot industrial samples with low electrical power, not exceeding 250 kW. More powerful systems are theoretically calculated in commercial numerical simulation software packages.

In the literature, researchers study the SOFC processes when using biogas as a fuel, the effect of biofuel on the system performance, the ratio of economic effect to operating costs [8], the conditions for reforming biofuel [9], the effect of poisons (hydrogen sulfide, carbon oxides, etc.) on the activity of the nickel catalyst [10] and low-power hybrid systems [11–15]. Efforts to commercialize SOFCs for biogas applications range from operating a 20 kW pilot plant on landfill gas [16] to a 6 MW plant powering a data center [17–19].

Studies on the performance and technical characteristics of hybrid systems operating on industrial waste from oil refineries as fuel are not as widely represented in the literature.

The purpose of this article is to develop a technology for preparing fuel from oil refining industrial waste and utilizing it in a SOFC-GT hybrid power plant to study the technical and economic advantages of cogeneration using fuel gas, with special attention to the technology of solid oxide fuel cells (SOFCs).

2. Materials and Methods

The proposed concept of the system was calculated in Aspen Plus V.11 according to the laws of conservation of mass, momentum and energy. Electrochemical characteristics were calculated according to Ohm's law, the Nernst equation and the Battler–Volmer equation, similar to the study [6]. The input data for the model on the composition of gaseous wastes of petrochemical industries were obtained from an operating oil refinery.

3. Results and Discussion

In the literature, the most often presented architecture of a hybrid system includes a solid oxide fuel cell and a gas turbine. The hybrid system operates under common pressure. A simplified diagram of the Brayton SOFC-GT cycle is shown in Figure 2. At the first stage of fuel treatment, fuel gas passes through a desulphurization stage, and experimental studies on fuel desulfurization were carried out. Based on the results of the analysis of data from the literature, the compositions of adsorbents were developed and their adsorption capacity for hydrogen sulfide was analyzed. The adsorbent compositions were mixed in certain proportions, given a granular form for ease of use and dried at a temperature of 40 °C. Bentonite, CaO, $Ca(OH)_2$, MnO and ZnO in different proportions were used as the main active substances. According to the results of experimental studies, the following composition has the best adsorption capacity, as well as physical and chemical characteristics: bentonite—40%, CaO—40%, ZnO—10% and MnO—10%. The capture rate was 98.3%. The final content of sulfur compounds in the fuel gas was 2.5 mg/m^3.

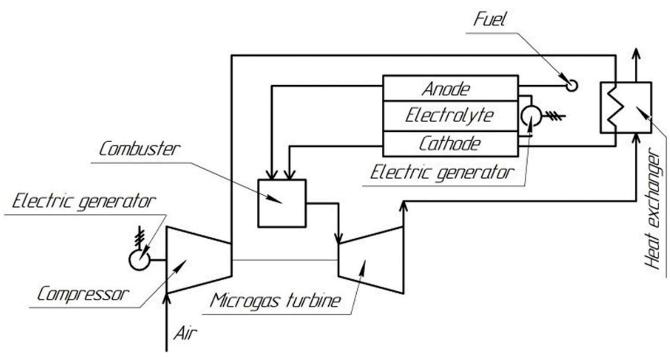

Figure 2. Brighton SOFC-GT hybrid cycle [16].

The desulphurized fuel after preheating is fed into the fuel reforming system. The fuel reformer receives heat from the combustion chamber and steam from the exhaust gases of the fuel cell. The fuel gas contains a significant amount of hydrocarbons up to C7, which must first be converted to synthesis gas by catalytic steam reforming using a nickel catalyst.

With insufficient steam used in the reforming process, the reaction proceeds in the direction of carbon formation rather than carbon dioxide and hydrogen. The resulting carbon is deposited as soot on the surface of the nickel catalyst and leads to its deactivation. Reforming the off-gas without carbon deposits under the proposed operating conditions at an operating temperature of 1073 K and a given fuel gas composition requires a steam-to-carbon ratio of 2.0, which was calculated taking into account chemical equilibria in the reactions using Aspen Plus. The output composition of the synthesis gas is presented in Table 3.

Table 3. Composition of fuel gas after desulfurization and steam reforming.

Element	Mole Fraction
CH4	0.0019
H2	0.594
H2O	0.2
CO	0.163
CO2	0.034
C2H6	6.8×10^{-9}
C3H8	8×10^{-14}
C4H10	3.8×10^{-19}
C5H12	9.8×10^{-24}
C6H14	8.7×10^{-29}
C7H16	6.3×10^{-34}

The exhaust gas heat exchanger serves to pre-heat the air in the heat exchanger. The air is compressed to the inlet working pressure of the SOFC. Then, the heated air is supplied to the SOFC cathode, where it participates in an electrochemical reaction.

The fuel gas is compressed by the fuel compressor to the SOFC operating pressure. The fuel gas then enters the anode of the fuel cell. Here, it undergoes internal reforming and turns into H_2, then participates in an electrochemical reaction with atmospheric oxygen to produce electricity. The unreacted fuel from the anode chamber and the exhaust air from the cathode chamber are combusted in the combustion chamber, which contributes to an increase in the outlet temperature of the exhaust gases, which can be usefully utilized. The exhaust gas stream is expanded in the gas turbine, simultaneously driving an air compressor and an electrical generator to produce additional power. Turbine exhaust gases are used to preheat air in a recuperative heat exchanger. At the last stage, the exhaust gases of the gas turbine can be used to produce steam in a steam generator or demineralized water [18].

According to the proposed scheme, calculations of technical and economical parameters for different types of fuel were carried out to compare and evaluate the efficiency of using gas waste from oil refineries (Table 4).

Table 4. Calculated technical and economical parameters of SOFC-GT on different types of fuel.

	Hydrogen	Natural Gas	Biogas from Wastewater	Fuel Gas
Fuel consumption per 1 W of power, mol/hW	0.003	0.012	0.11	0.04
Net calorific value, MJ/kg	140	49	19,4	46
Electrical efficiency, %	71.3	62	51.6	59.1
Total efficiency, %	91	89.7	87.5	89
CO2 emissions, g/MJ	0	144.375	0	0
Fuel cost	2 $/kg	0.79 $/kg	0	0
System cost per 1 kW of power, $/kW	6900	6900	6900	6900
Fuel post-treatment cost, $/kW	500–3000	0–500	500–3000	500–3000

Calculations of electrical efficiency were carried out, taking into account the fuel utilization factor of 0.8, the air utilization factor of 1.4, the ratio of steam to carbon of 2:1 and the SOFC operating temperature of 1073 K. The efficiency of the fuel compressor was taken into account as 71.3%, the efficiency of the air compressor was 66.5% and the efficiency of the DC/AC converter was 96% [20].

The total heat loss of the SOFC-GT system is 4.5% with the following distribution: SOFC—1.5%, combustion chamber—1%, pre-reformer—0.5% and GT—1.5%.

The calculated indicators correspond to the literature data. It can be noted that all types of fuel for the SOFC-GT hybrid system have a number of advantages and disadvantages.

Thus, the lowest molar fuel consumption (per watt of energy produced) can be obtained using hydrogen, due to its maximum net calorific value compared to other fuels. However, the cost of pure hydrogen per kilogram is still relatively high. Fuel from waste (biogas from sewage, fuel gas from oil production waste) is free, but its consumption is much higher. This should be considered to prevent limiting of the production of fuel from waste.

Electrical and total efficiency is also highest when using hydrogen as fuel. The thermal and electrical efficiency of natural gas is comparable to industrial waste fuel gas due to the comparable net calorific value of these fuels. Therefore, fuel gas from oil production is a valuable product that should be reused.

Carbon dioxide emissions can only be taken into account when natural gas is used as a fuel. Carbon dioxide emissions from hydrogen production, as well as from the reuse of waste, are not taken into account in this study. In comparison with the operation of a gas turbine on natural gas, according to the literature (Tables 2 and 3), carbon dioxide emissions in a hybrid system are slightly lower but exceed emissions during fuel cell operation.

The cost of SOFC system installation was determined based on the cost of individual components, according to the literature data, or was estimated using the Aspen Plus program for petrochemical calculations. The cost of a hybrid system significantly exceeds the cost of mono-variant power plants (Tables 2 and 3) due to the complexity of the layout, the large number of plants and connections between them, control sensors and automation devices.

Separate costs are taken into account for additional purification and conditioning of the fuel. For hydrogen, the most commonly used and effective method is pressure swing adsorption. For natural gas and reusable fuel resources, desulfurization and reforming of the fuel is required.

4. Conclusions

The article proposes a technology and a feasibility study for the processing of hydrogen-containing gas waste from oil production at a hybrid power plant with an SOFC. The composition of gas waste from oil refining is presented. The gas mixture contains mainly saturated and unsaturated hydrocarbons C1–C7, carbon oxides and hydrogen sulfide. It is shown that the gas waste (fuel gas) has a net calorific value comparable to the calorific value of natural gas (46 and 49 MJ/kg, respectively). Therefore, fuel gas is a valuable product that can be used as a fuel in hybrid power plants rather than flaring. The main disadvantage of fuel gas is the presence in its composition of sulfur compounds (hydrogen sulfide, mercaptans), which are corrosive active agents. The article proposes the developed composition of the adsorbent for hydrogen sulfide absorption, including 40% wt. bentonite, 40% wt. calcium oxide, 10% wt. zinc oxide and 10% wt. manganese oxide. The capture rate was 98.3%. A comparison of the technical and economical parameters of the hybrid power plant with the SOFC and GT using various types of fuel (hydrogen, natural gas, wastewater biogas, fuel gas from oil production) was made. It is shown that the highest electrical efficiency at the lowest fuel consumption can be obtained by operating on hydrogen fuel. Fuel gas from petrochemical industries has similar characteristics to natural gas, and since it is a waste product, it is free of charge and does not emit greenhouse gases. Additional purification and preparation of fuel require high additional costs, which increases the cost of the entire hybrid system, which includes a large amount of high-tech equipment, automation and control systems.

In conclusion, it should be noted that fuel gas from oil production waste has a high potential for use in power plants due to its high calorific value and a number of other undeniable advantages compared to natural gas.

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