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HEAT AND MASS TRANSFER IN HYDROGEN PRODUCTION BY ELECTROLYSIS OF A WATER-COAL SUSPENSION

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The possibility of electrochemical gasification of a water-coal suspension at a temperature of 180–460^{o}*C and a pressure of 7–20 MPa has been proved experimentally. The obtained producer gas contains mainly hydrogen and carbon dioxide as well as traces of methane, propane, and carbon monoxide. After washing carbon dioxide with weak alkaline solutions hydrogen is dispatched for use in power engineering. The liquid phase contains an insignificant amount of hydrocarbons. Heat and mass transfer between reacting media in the electrolyzer under the conditions of phase transitions with partial absorption of the formed gases has been considered.*

Keywords: *hydrogen, carbon, coal waste, water-coal suspension, electrolyzer design, Gibbs energy.*

Introduction. At the present time coal containing 25–30% of an inorganic part is supplied to most thermal electric power plants. After burning, 20% of an inorganic part goes to slurry dumps and 80% to ash dumps. Because of incomplete combustion of coal in furnaces approximately 17–25% of carbon is dumped with ash. Hence it follows that fluxes of dumped carbon are as large as millions of tons per year.

The current study aims at developing an electrochemical technology for the utilization of ash carbon that provides a 2–3% increase in the efficiency of a thermal electric power plant. Here the technological cycle includes processing of the accumulated ash dumps. Supposedly, when slightly augmented, the proposed method will be suited to the utilization of the waste of coal concentration plants and mines.

Experiment, Search for Principles and Optimum Regimes of Operation. A previous investigation indicated the possibility of electrochemical gasification of a water-coal suspension at a temperature of 180–460^{o} and a pressure of 7–20 MPa. The obtained producer gas contains mainly hydrogen and carbon dioxide as well as traces of methane, propane, and carbon monoxide. After washing carbon dioxide with weak alkaline solutions, hydrogen is dispatched for use in power engineering. The liquid phase contains an insignificant amount of hydrocarbons [1].

Conducting the process at atmospheric pressure and a temperature of about 80^{o}C showed that in this case the generated gas contains hydrogen, carbon monoxide, and traces of carbon dioxide. Acetone, alcohol, acids, and ethers are present in the composition of the liquid phase in proportions dependent on the electrode voltage [2].

Electrochemical oxidation of carbon-bearing materials at increased pressures and temperatures is studied using an experimental setup whose schematic is given in Fig. 1. The electrolyzer assembly is shown in Fig. 2. The volume of the test reactor is $0.975 \cdot 10^{-3}$ m^3. The capacity of the reactor as an assembly with electrodes and a holder is $0.89 \cdot 10^{-3}$ m^3. The reactor is designed for a pressure of up to 20 MPa. The amount of a solution poured in the reactor is $0.35 \cdot 10^{-3}$ m^3. The mass of the powder of a carbon-bearing material loaded in the holder is $83.15 \cdot 10^{-3}$ kg. The total volume of the holder with the material is $0.12 \cdot 10^{-3}$ m^3. At the beginning of the process of electrolysis electrodes are immersed in a liquid to a depth of 195 mm. Here the working surface of the electrodes is $3.9 \cdot 10^{-3}$ m^2 and the distance between them varies from 10 to 25 mm. The outside appearance of an electrolysis cell is presented in Fig. 3. Electrodes of various structures (Fig. 4) are used in the current study.

At the beginning of the experiment the needed amount of a granulated carbon-bearing material of a certain fraction is loaded in holder 3 made of glass fabric (Fig. 1). The holder is fitted over electrodes 2 dielectrically sealed in the flange of reactor 1. Thermocouple 18 for measuring the temperature of the reacting mixture is sealed likewise. Reactor 1 is filled with the needed amount of the electrolyte, sealed, evacuated, and flushed by an inert gas (as carbon

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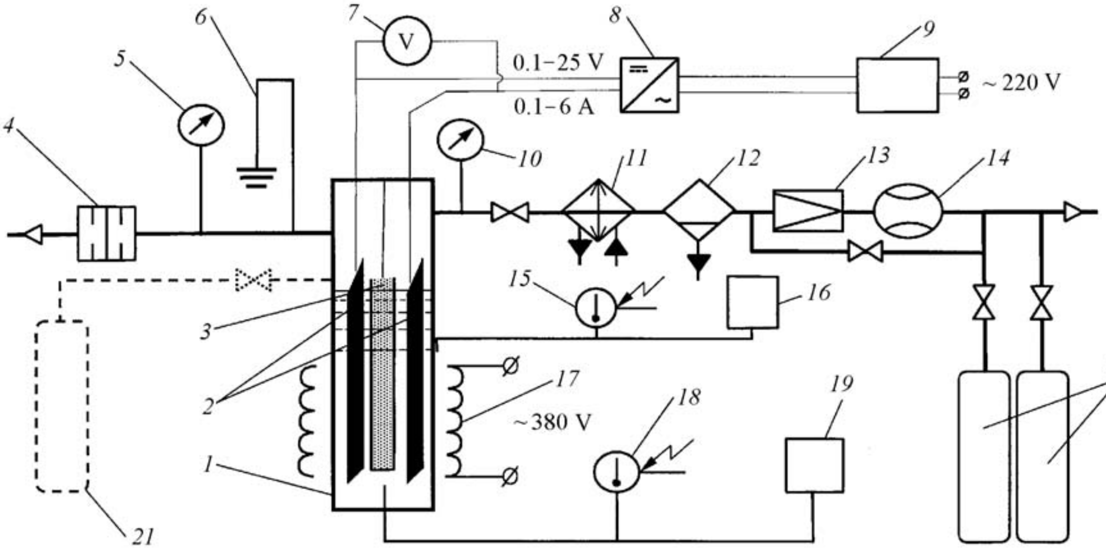

Fig. 1. Schematic of an experimental setup for electrolysis of coal pulp: 1) reactor, 2) electrodes, 3) holder with a carbon material, 4) safety membrane, 5) manometer, 6) ground, 7) universal voltmeter, 8) rectifying-stabilizing unit, 9) laboratory transformer, 10) check manometer, 11) condenser, 12) separator, 13) reducer, 14) flow meter, 15, 18) thermocouples, 17) muffle furnace, 16, 19) secondary devices, 20) containers for the produced gas, 21) container with an inert gas.

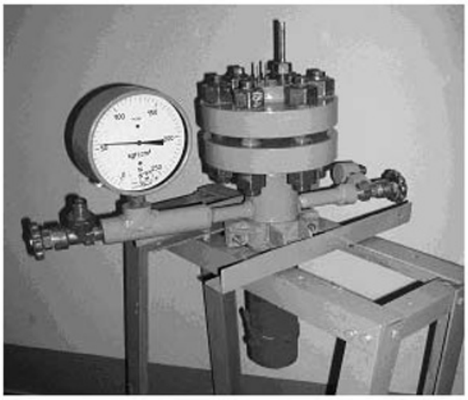

Fig. 2. Electrolyzer as an assembly.

dioxide, nitrogen, argon). Thereafter the reactor is placed in shaft-type muffle furnace 17 and heated to a specified temperature.

When the needed process parameters are attained, an electric current is supplied to electrodes 2 through transformer 9 and rectifying- stabilizing unit 8, and simultaneously the time of onset of the reaction is recorded. Guided by readings of a digital universal voltmeter the electric potential is kept constant over the entire process. The progress of the reaction is tracked relying on an increase in the pressure and temperature and a variation in the electrode potential. The amount of the released gas is calculated from the pressure in the reactor and the temperature. The experiment is assumed to be completed when the pressure does not increase noticeably any longer. On completion of the experiment the reactor is cooled, and the gas is passed through condenser 11 and, after separation of water droplets in separator 12, is pumped into containers 20 for further analysis. The remaining gas is dumped into the atmosphere through reducer 13 and flow meter 14. The reaction products are discharged, the liquid is filtered, its volume is measured, and it is also analyzed. The precipitate and the holder with a carbon material are dried and weighed, and the reactor is thoroughly washed.

Fig. 3. Appearance of an electrolysis cell made of the C–C material.

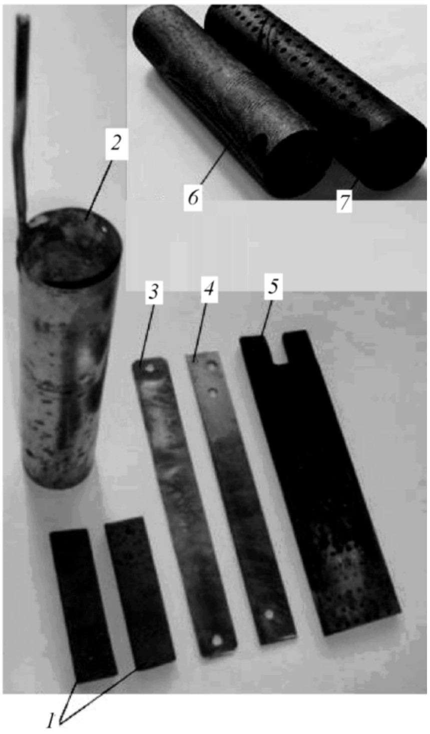

Fig. 4. Structure of electrodes: 1) plane electrode, a solid composite carbon-carbon (C–C) material [3]; 2) cylindrical electrode, Kh18N10T stainless steel; 3) plane electrode, copper; 4) plane electrode, stainless steel; 5) plane electrode with perforation, a composite carbon-carbon material; 6) cylindrical electrode, a solid composite carbon-carbon material; 7) cylindrical electrode with perforation, a composite carbon-carbon material.

In investigations with acid electrolytes the electrolysis cell is placed in a glass beaker to preclude a contact of the solution with the reactor walls. In the case of work with sulfuric and phosphoric acids pressure is set up by argon, and in the case of carbonic acid it is produced by carbon dioxide. In experiments, various materials for manufacturing a cathode were tested, namely, a carbon-carbon (C–C) material [3], a nickel alloy, and copper.

Discussion of Results. An analysis of the gases produced during electrolysis is performed using an LKhM-8MD chromatograph [1]. Experiments are conducted on the following specimens of carbon-bearing materials: two samples of slimes of coal concentration plants, ash of the Zmievsk thermal electric power plant, and coal of the ASh grade (anthracite culm). The elemental composition of these materials is presented in Table 1. Results of the preliminary experimental run are given in Table 2, and generalized results of the investigations are supplied in Table 3.

The current study presents a method to design electrolyzers. The proposed order of evaluating geometric parameters of the device is based on results of the works indicated below.

The mechanism of free convection in the electrolysis of water is given in [4], and the patterns of two-phase flows in air-gas lifts are considered in [5, 6]. The design of circulation circuits of the evaporators and reactors is reported in [7, 8]. A mathematical model of the behavior of bubbles during electrolysis, which incorporats characteristic features of the electrolysis process, is proposed in [9]. In this study the influence of the saturation pressure on the rate of growth of bubbles is noted. The effect of a rise in the process rate with increasing current density is also revealed.

TABLE 1. Proximate Analysis of Carbon-Bearing Materials (data of the Kharkov Institute of Coal Chemistry)

Sample No., material	Humidity	Ash content	Sulfur content	Dry mass, %			Calorific power, kJ/kg (kcal/kg)
	%			C	H	N + O	
1	3.12	38.2	3.65	47.95	3.5	6.7	17555 (4193)
2	2.59	42.9	5.2	44.2	3.0	4.7	15256 (3644)
Anthracite culm (ASh)	1.45	33.44	1.88	59.17	2.41	4.1	20096–21767 (4800–5200)
Ash	1.43	83.95	0.47	12.56	0.36	2.66	—

TABLE 2. Results of Preliminary Experimental Run (ballast argon is indicated in the composition of products)

Parameters	Experiment Nos.					
	1	2	3	4	5	6
Carbon-bearing material	Sample No. 2			Sample No. 1	Coal of ash grade (anthracite culm)	Ash of Zmiev thermal electric power plant
Material of anode/cathode electrodes	Carbon/carbon					
Distance between electrodes, mm	25					
Pulp composition	Tap water + carbon-bearing product					
Pressure, in./fin., MPa	7.6/19.1	12.2/18.6	12.4/17.5	12.9/19.3	11.9/18.4	14.1/19.5
Temperature of medium in reactor, in./fin., ^{o}C	310/460	180/285	270	285/325	167/300	225/325
Voltage, V	25	8	2	11–10	11	
Current strength, A	5	1	0.1	2.5–0.4	1.0–0.2	
Current density, A/cm^2	0.128	0.026	0.003	0.064–0.01	0.026–0.005	
Time of experiment	5 h 40 min	12 h 50 min	20 h 45 min	32 h 50 min	81 h 50 min	72 h 10 min
Electrolyzer material	Steel					
Gas composition, % (volume fractions)						
H_2	20.86	9.43	2.26	12.1	13.94	14.47
CO_2	5.36	0.58	0.77	1.15	1.73	2.39
CO	—	—	—	—	—	—
Ar + O_2	73.51	89.62	96.72	86.75	83.31	82.79
CH_2	0.27	0.37	0.25	—	1.02	0.35
Molar ratio of gases (H_2:CO_2)	3.89	16.25	2.94	10.52	8.06	6.05

The circulation of flows in the electrolyzer has features common to those of the circulation of flows in the hydrogen generator.

Previously it was shown that, as a result of the reaction of alloys with water, a substantial quantity of heat is released [10]. The latter circumstance leads to the formation of water vapor, which provides a stable circulation of flows in the device. In the present study the circulation of flows in the electrolyzer is provided by the gases formed near electrodes. Therefore, in a first approximation the lifting channel in the electrolyzer can be considered as such in the reactor or evaporator.

Common to the methods of designing evaporators and reactors are:

a) methods of determining the theoretical velocity of an adiabatic flow of gas and water vapor in the channels of devices with natural circulation,

b) an evaluation of the resistance of the channels to two-phase flows,

c) techniques of using the patterns of symmetry and asymmetry in designing devices and their units.

Differences are observed in the selection of constituent kinetic equations and in the calculation of the velocity of a gas-liquid flow near the reaction front, i.e., near the heat and mass transfer surface that is moving in reactors and immobile in evaporators [11, 12].

The speed of the liquid circulation in hydrogen generators is determined using methods [8, 13]. Here the rate of the hydrogen liberation from water W is found from the Gibbs–Volmer dependence implying, in a first approximation, a linear relation between W and the driving force of the process, which is the chemical potential μ (in the adopted procedure the chemical potential is the value of the Gibbs energy per 1 kg of the active part of an

TABLE 3. Generalized Results of Basic Investigations (initial pressure in the electrolyzer was produced by carbon dioxide, ballast carbon dioxide is not indicated in the composition of products)

Parameter	Experiment Nos.						
	7	8	9	10	11		12
Carbon-bearing material: coal (anthracite)	Without coal		Without holder	Anode in holder	Both electrodes in holder		Partition with perforation
Material of anode/cathode electrodes	Carbon/carbon						
Distance between electrodes, mm	25				10		
Electrolytic solution	H_2SO_4	Distilled water + CO_2 (carbonic acid)					
pH before/at the end of experiment	3/5	1.8*	1.9*		2*		4/6
Pressure, in./fin., MPa	2.4/3.8	3.5/4.0	4.1/4.6	4.3/4.6	3.7/5.1	3.7/8.0	13.5/16.0
Furnace temperature, in./fin., °C	180	100/50			50	50/90	150/200
Voltage, V	11		10	11	10		2.66
Current strength, A	0.5	0.2	1.2–0.8	≤ 0.1	6–5.5		0.46
Current density, A/cm^2	0.02	0.008	0.048/0.032	≤ 0.004	0.24–0.22	0.24– 0.06	0.019
Rate of hydrogen production, m^3/(m^2·sec)	$2.45 \cdot 10^{-6}$	$3.02 \cdot 10^{-6}$	$3.93 \cdot 10^{-6}$	$3.82 \cdot 10^{-6}$	$0.64 \cdot 10^{-6}$	$6.57 \cdot 10^{-6}$	$0.96 \cdot 10^{-6}$
Material of cell frame	Glass						
Glass composition, % (volume fractions)							
H_2	66.67	66.67	66.24	66.67	66.67	64.55	59.46
CO_2	23.72	33.33	20.55	27.47	28.08	32.27	39.57
CO	—	—	—	—	—	2.28	0.97
O_2	9.62	—	12.57	5.87	5.25	—	—
CH_4	—	—	—	—	—	0.90	—
Molar ratio of gases (H_2:CO_2)	2.8	~2	3.2	2.4	2.37	2	1.5

*Design value at beginning of experiment.

alloy). The coefficient of proportionality L between W and μ is found in kinetic laboratory studies. With increase in the diameter of the industrial device the coefficient of proportionality takes variable values dependent on the Reynolds number of the circulating flow and the alloy density ρ. In a second approximation the coefficient L is determined more accurately from the Re number and ρ [7, 8, 14]. Further on the real rate of hydrogen liberation is calculated from the Gibbs–Volmer equation followed by computing the real volumetric capacity of the device. Individual periods of the process should be calculated using Erofeev's equation [15]. The solution temperature is determined from the heat balance taking account of heating of the reaction products and device walls. Separate details of the calculation are discussed in [10].

We now come back to considering results of the current study. In this case, the dominant driving force of the investigated process of electrolysis of coal pulp is the electromotive force (emf) E^{o}. The design of the circulation circuit is preceded by the evaluation of E^{o}. The hydrogen interaction with oxygen proceeds following the equation [16]

$$\mathrm{C(graphite)} + \mathrm{O_2} = \mathrm{CO_2}\,. \tag{1}$$

The variation in the Gibbs energy is $\Delta Gi^{o} = -394{,}949$ J. The water solubility of CO_2 at 25°C and a partial pressure of 1 atm is 0.0338 moles per liter. Dissolution proceeds by the reaction

$$\mathrm{CO_2(gas)} = \mathrm{CO_2(liquid)}\,. \tag{2}$$

In this case $\Delta Gi^{o} = 8422$ J. According to the condition pertaining to hydrated solutions,

$$\mathrm{CO_2(liquid)} + \mathrm{H_2O} = \mathrm{H_2CO_3(liquid)}\,,\quad \Delta Gi^{o} = 0\,.$$

Combining Eqs. (1) and (2) with the equation of water decomposition

$$\mathrm{2H_2O} = \mathrm{2H_2} + \mathrm{O_2}\,,\quad \Delta Gi^{o} = 473{,}973\ \mathrm{J}\,, \tag{3}$$

we obtain

$$C(\text{graphite}) + 3H_2O = H_2CO_3(\text{liquid}) + 2H_2\,, \quad \Delta Gi^{\circ} = 87{,}445 \text{ J}\,. \tag{4}$$

Dissociation of carbonic acid proceeds following the equation

$$H_2CO_3(\text{liquid}) = H^+ + HCO_3^-\,, \quad \Delta Gi^{\circ} = 36{,}914 \text{ J}\,.$$

From the above equation in conjunction with Eq. (4) we get

$$C(\text{graphite}) + 3H_2O + e = HCO_3^- + 2.5H_2\,, \quad \Delta Gi^{\circ} = 124{,}359 \text{ J}\,. \tag{5}$$

The emf value for this reaction is determined from the equation [17]

$$\Delta Gi^{\circ} = nFE^{\circ}\,.$$

For carrying out the reaction of water decomposition (3), 4 F had to be expended. Thus, the conducting of reaction (5) necessitates a total of 5 F, whence

$$E = \Delta Gi^{\circ} / (nF) = 0.257 \text{ V}\,.$$

Since the Gibbs energy is a function of temperature and pressure, in subsequent approximations the value of E^{o} should be refined [18].

From the above equations it is seen that with increase in the amount of carbon dioxide dissolved in water the energy consumption for dissociation of coal dust becomes larger. This fact makes it necessary to set values of voltages on the electrode terminals above the theoretical level. A decrease in the rate of gas liberation is also caused by the blocking (as hydrogen is generated) of the reaction surface by ferric, aluminum, and silicon oxides contained in an inorganic part of the coal waste.

At the given stage of investigations the enumerated factors made it impossible to decrease the voltage to the theoretical level and thus to attain the rate of gas generation needed for industrial application. The voltage can be decreased somewhat if the cathode is made of copper or nickel-bearing alloys.

In the coal pulp electrolyzer the reaction surface is determined by the working area of electrodes and the specific area of the powder of a carbon-bearing material. In the proposed technology, electrolysis of coal pulp is generally conducted at a pressure precluding water boiling. In this case, the hydrodynamic conditions in the device are determined by a hydrogen flow. The improvement of the circulation of flows in the electrolyzer after the replacement of plane electrodes with cylindrical ones facilitates a washing of deposits from the particle surface, which also leads to a certain decrease in the overvoltage.

Further perfecting of this method of hydrogen production involves the application of gas-absorbing liquids. For this purpose use is made, specifically, of fluorocarbon compounds [19].

An analysis of the conditions of conducting reactions by a patent [20] indicates that an intense removal of carbon dioxide and hydrogen from the system provides the approach to the values of heat transfer coefficients calculated using an equation [8]

$$Nu = 1.85 \cdot 10^{-6} Re^{1.6} Pr^{0.33}\,.$$

The equation is valid in the Reynolds-number range $4 \cdot 10^4 < Re < 2 \cdot 10^5$. The significant influence of the Reynolds number is accounted for by high turbulization of a liquid in the formation of gases on the particle surface.

Economic indices are calculated proceeding from the fact that the development of the process of electrolysis of a coal suspension radically changes the technologies of coal extraction and utilization alike.

Another essential feature of the developed process is the absence of an unstable compound — carbon monoxide (CO) — in the gasification products. Electrolysis of the coal pulp results in the formation of hydrogen and stable carbon dioxide (CO_2). Carbon dioxide is readily washed from the mixture by a weak alkaline solution, which makes it possible to dispatch the produced pure hydrogen directly to an electrochemical generator. The efficiency of a thermal power complex can thus be raised from 35 to 70% [19].

As known, the theoretical consumption of electric energy in generating 1 m^3 of hydrogen in the electrolysis of water at a temperature of 298 K is 2.93 kW·h, while the energy consumption in the electrolysis of coal pulp is no higher than 0.49 kW·h. Taking the optimum efficiency of the electrolyzer to be at a level of 65%, we arrive at the energy expenditure on producing 1 m^3 of hydrogen to be equal to 4.5 and 0.7 kW·h, respectively.

At the present time technologies of nontraditional hydrogen production by electrolysis of water-coal pulp are also actively developed abroad. Thus, in the USA, with a consumption of electric energy of 5.16 kW·h/kg of hydrogen, its price of 0.04 USD/(kW·h), and a price of 32 USD for 1 ton of coal, the cost of 1 ton of hydrogen is 1700 USD, which is 50% lower than the cost of hydrogen produced by the water electrolysis [21, 22].

Conclusions. Coal-pulp electrolyzers should be designed with account for hydrodynamic laws coupled with kinetic and thermodynamic ones. Judging by the tentative evaluation, the rate of interphase heat transfer does not restrict the reaction rate.

Further perfecting of the considered process may lead to a serious change in the energy balance of countries disposing of sizable coal reserves. In other words, prospects for converting industry from the utilization of natural gas to the use of hydrogen will open up.

NOTATION

E, electromotive force, V; e, electron; F, Faraday number (96,484 absolute coulombs); Gi, Gibbs energy, J; L, coefficient of proportionality, $m^3/(m^2 \cdot sec \cdot J)$; n, number of F units; W, rate of hydrogen liberation, $m^3/(kg \cdot sec)$ or $m^3/(m^2 \cdot sec)$; Δ, perturbation of a finite quantity; μ, chemical potential, J/kg; ρ, alloy density, kg/m^3. Superscript: ^{o}, value of a quantity under normal conditions ($P = 0.1$ MPa, $T = 298$ K).

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