THE EFFECT OF STEAM ON AIR GASIFICATION OF MECHANICALLY ACTIVATED COAL IN A FLOW REACTOR

by

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A combined steam-gas plant with in-cycle steam gasification of coal and hot gas purification is considered as a promising technology for increasing the efficiency of energy production with simultaneous reduction in environmental impact. To intensify steam-air gasification, mechanical activation of fuel is used; it consists in high-stress grinding in a mill-disintegrator. The supply of steam to the gasifier allows an increase in reactivity of fuel, suppression of sorbent decomposition in the unit of hot desulfurization, reduction in the steam load on the shift reactor, increase in useful external work of gas turbine expansion, reduction in nitrogen oxide formation, and general increase in the efficiency and ecological compatibility of energy generation. On the other hand, a significant amount of steam can deteriorate the heat balance and efficiency of the gasifier. In this work, the influence of the steam/fuel ratio on steam-air gasification of mechanically activated Kuznetsk coal in a flow reactor was studied experimentally. The excess air coefficient was maintained constant and equal to 0.51, which corresponded to a temperature at the reactor outlet of about 1100 °C. When steam was supplied, the fuel and air-flow rates were adjusted to ensure a constant gas-dynamic regime. To evaluate the obtained regimes, the heat and material balances were compiled. A positive effect of steam on characteristics of the gasification process was revealed. For the studied coal, the maximum degree of coal conversion and the calorific value of synthesis gas are achieved with a steam/fuel ratio of about 0.4 kg/kg.

Key words: *coal gasification, mechanically activated coal, experimental studies*

Introduction

An increase in the Earth population and improvement in the human life quality contribute to an increase in the need for electric energy. Despite the rapid development of RES, basic energy needs are met by fossil fuels. Due to the limited fossil energy resources and global climate changes caused by anthropogenic impacts, the fundamental problem of the energy sector is an increase in the efficiency of energy production from fossil fuels with minimal impact on the environment. Among fossil fuels, the largest energy reserves are concentrated in coal. A promising technology for electricity production from coal is the combined steam-gas plants with in-cycle gasification. In addition to improving the energy part of the steam-gas plants, the

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potential for improving the combined steam-gas plants with in-cycle gasification is increasing the efficiency of the unit for solid fuel gasification.

When turning from wet cold to dry hot gas purification, the efficiency of the cycle of the combined steam-gas plants with in-cycle gasification will increase by 2-3% [1].

Most of existing industrial gasifiers operate with oxygen blowing [2-4], while working with air blowing increases the efficiency of the combined steam-gas plants with in-cycle gasification by 1.5-2%, reduces capital costs, and is also more preferable from the viewpoint of regulating the capacity and operation of a steam-gas plant [5].

However, the use of air blowing instead of oxygen in the gasifier requires additional measures to intensify the gasification of fuel. Fundamentally, there are two possible ways to intensify air gasification of coal. The first method is intensification due to high temperature air heating in the external (additional) boiler, which is characterized by significant capital costs at a limited heating temperature (600-800 °C) [6]. The second method is intensification by increasing the reaction properties of coal by mechanical activation [7]. Mechanical activation consists in fine grinding of coal in a mill-disintegrator. During mechanical activation, the total surface area of fuel particles increases due to a decrease in their size and mechanochemical transformations of the organic part of fuel, associated with a significant energy impact on the fuel particles, occur; the ultimate composition of fuel and the content of the functional groups of organic compounds change [8]. Fuel particles are heated above 100 °C during a short stay time (less than 1 second) as a result of energy impact. During mechanical activation, free radicals are formed on the particle surface [9], which increases the rate of chemical reactions of combustion and gasification and reduces the temperature of coal dust ignition. In [10], stable ignition of a vortex reactor with a capacity of 5 MW (heat) was achieved using mechanically activated coal, without air heating and heat supply from a gas burner. In [11], the regimes of air autothermal gasification of coal were studied at a 5 MW reactor.

Additional intensification of the coal gasification process is possible due to the steam supply to the gasifier. With an increase in the share of steam in the steam-air blowing, concentration of ballast nitrogen decreases, and concentration of an oxidizing agent entering the reaction of coal gasification increases, therefore, the rate of gasification increases. On the other hand, due to the higher heat capacity in comparison with nitrogen and oxygen, the steam supply worsens the heat balance of gasification and lowers the process temperature. A decrease in temperature slows down gasification reactions.

Various data on the effect of steam on coal gasification are presented in the literature. Most studies on the effect of steam on flow gasification of coal deal with steam-oxygen gasification. It is shown in [12] that steam accelerates the ignition of coal dust particles at steam-oxygen blowing, and the effect of steam is more pronounced when using low-grade coals (brown). The influence of steam supply in a Shell gasifier is considered in [13]. It has been found that the addition of steam increases the efficiency of gasification of high-grade coals; the effect of steam addition on gasification of brown coals is negligible. The effect of steam supply on air gasification of biocoal in a flow reactor is investigated in [14]. Steam slightly increases the degree of biocoal conversion; however, this study does not take into account a decrease in residence time as a result of additional steam supply to the air blowing.

The research of steam-air gasification of mechanically activated fuel performed earlier at the IT SB RAS on a set-up with a capacity of up to 1 MW proved the fundamental possibility of the autothermal process. However, due to the axial supply of steam to the reactor, a significant part of steam passed through the reactor without interacting with coal; the heat of synthesis gas combustion was low. With an increase in the steam/fuel ratio of more than

0.5 kg/kg, the time of coal particle stay in the reactor decreased significantly and the gasification process stopped.

In this work, we study the effect of steam supply on gasification of mechanically activated coal in a modernized set-up with tangential steam supply to the reactor. The aim of the study is to obtain new fundamental knowledge about the influence of steam on the dynamics of gas formation and change in the reaction characteristics of coal along the flow gasifier, and to select the optimal flow gasification regime for the prospective integrated gasification combined cycle power plants on steam-air blowing with hot desulfurization.

Materials and methods

Material

The experiments were carried out with oxidized Kuznetsk coal of grade D. The results of the proximate and ultimate analysis of coal are given in tab. 1.

Table 1. Proximate and ultimate analysis of fuel

W [%]	A^d [%]	V^{daf} [%]	C^{daf} [%]	H^{daf} [%]	O^{daf} [%]	N^{daf} [%]	S^{daf} [%]
2	21	41	74.5	4.9	17.7	2.2	0.6

Coal was preliminarily ground in a hammer mill. Then it was mechanically activated in a disintegrator. The power of the disintegrator was 60 kW, disintegrator revolution rate is 3000 rpm, blades length is 200 mm. The fuel supply to the disintegrator was 170 kg/h. Mechanical activation was performed immediately before the experiment. The time between activation and the supply of coal to the reactor was up to 30 minutes. Granulometric compositions of coal, fig. 1, were calculated by the Rosin-Rammler equation according to analysis on 1000, 200, 100, 63, and 40 μm sieves. The *standard* particle size distribution obtained at Novosibirsk TPP-2 after grinding in a ball drum mill is given for comparison.

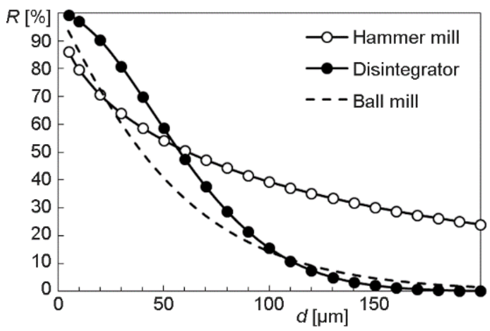

Figure 1. Granulometric composition of coal

Experimental set-up

The scheme of the modernized experimental reactor is shown in fig. 2. The modernization consists in using the tangential steam supply into the reactor instead of axial, which increases the time of particle stay in the reactor and, accordingly, the degree of fuel conversion.

The flow reactor consists of a scroll swirler, an insert for tangential steam supply and a reaction chamber. The internal diameter of the swirler varies from 340 mm to 240 mm, the diameter of the insert and reaction chamber is 140 mm. The swirler length is 50 mm, the length of the insert and reaction chamber is 1100 mm. The reactor is lined by refractory concrete. The fuel and air are supplied to the swirler through a slot nozzle of 23×45 mm. The steam is supplied tangentially in accordance with the direction of a vortex from the swirler. Large particles of coke-ash residue falling out from the gas flow form a layer in the lower part of the afterburner. The capture of fine particles entrained by the flow and combustion products cooling take place in a centrifugal bubbling apparatus. Purified combustion products are sent to the smoke pipe using a smoke exhaust. The reactor is ignited using a propane burner. Air supply during ignition is carried out by a lighting blower; the flow rate is measured by a rotameter.

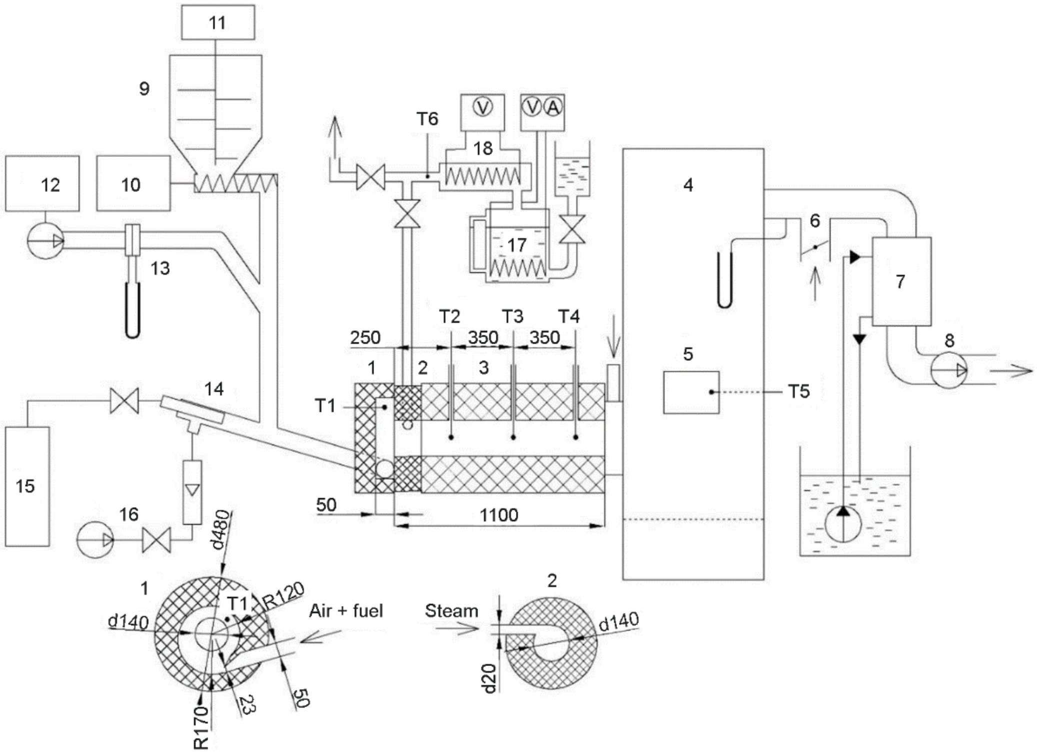

Figure 2. Scheme of experimental set-up; *1 – scroll swirler, 2 – steam supply insert, 3 – reaction chamber, 4 – afterburner, 5 – inspection window, 6 – vacuum controller, 7 – centrifugal bubbling apparatus, 8 – smoke exhaust, 9 – fuel hopper, 10 – screw feeder, 11 – mixer, 12 – frequency-driven air blower, 13 – diaphragm for measuring air-flow rate, 14 – pilot burner, 15 – propane tank, 16 – lighting blower, 17 – steam generator, 18 – steam superheater*

The vacuum is measured by a liquid manometer and regulated by the gate due to the additional air intake. The temperature is measured by T1-T6 thermocouples connected to an automated data acquisition system. Thermocouples in the reaction chamber (T2-T4) are placed along the chamber axis. The gas composition is measured with a TEST-1 gas analyzer. The concentrations of O_2 (0-25% vol.) and NO (0-1000 ppm) are measured by electrochemical sensors, H_2 concentration (0-40% vol.) is measured by a polarographic sensor, and CO (0-100% vol.), and CO_2 (0-30% vol.) concentrations are measured by the optical sensors. The relative measurement error for all components is 5%. The gas sampling tube is placed in a connecting pipe together with a thermocouple, *i. e.* the gas sampling point at the end of the reactor coincides with the location of thermocouple T3. The gas composition in the stationary regimes is measured at three points along the reactor diameter (0.15, 0.5, and 0.85 diameters). For further analysis of regimes, the average value of gas composition over the outlet cross-section of the reactor is calculated.

Coal is fed from the hopper by a screw feeder with an adjustable rotational speed. The uniformity of hopper emptying is provided by a mixer. To measure the fuel-flow rate, the dependence of fuel-flow rate on the speed of feeder rotation is determined previously by weighing.

Air is supplied by a blower with a variable frequency drive. The air-flow rate is measured by a diaphragm.

The steam generator is heated by direct electric current from the rectifier. The power is measured by an ammeter and a voltmeter and is regulated in the range from 0 kW to 30 kW. The steam superheater is heated by the current from an autotransformer, the power is regulated from 0 kW to 3 kW. The temperature of superheated steam is measured by a thermocouple connected to an automated data acquisition system. Before stabilization of the steam flow rate, the steam is sent to the bypass line. The water level in the steam generator is measured by a water measuring tube. The volume of steam generator is sufficient for stable operation for 20-30 minutes, the steam generator is periodically fed from tank 6. To measure the steam flow rate, the dependence of steam flow rate on DC voltage of the rectifier is preliminarily determined.

Experimental program

The reactor is ignited by a propane burner. In 2-3 minutes after ignition, the temperature in the scroll reaches 800-1000 °C and coal is supplied. Then, within 30 minutes, the walls of the reactor are heated in the regime of incomplete combustion of fuel with an excess air coefficient of 0.8.

To study the effect of steam supply, a stationary regime of air gasification of mechanically activated coal was selected as a result of search experiments. The parameters of regime are given in tab. 2. To maintain a constant gas-dynamic regime and time of coal stay in the reactor, the fuel and air-flow rates are reduced when steam is supplied. The experimental program is shown in tab. 3.

Table 2. Parameters of stationary regime of air gasification

Fuel-flow rate [kgh^{-1}]	Air-flow rate [m^{3}h^{-1}]	Temperature at the reactor outlet [°C]	Air excess
13.0	43.7	112	0.51

Table 3. Experimental program

No. of regime	Coal flow rate [kgh^{-1}]	Air-flow rate [m^{3}h^{-1}]	Steam flow rate [kgh^{-1}]	Steam/fuel [kgkg^{-1}]	Air excess (by the input)	Estimated blowing volume (at 1100 °C) [m^{3}h^{-1}]
1	13.0	43.7	0.0	0.0	0.51	230
2	12.0	40.4	2.7	0.2	0.51	230
3	11.2	37.6	4.7	0.4	0.51	230
4	10.5	35.2	6.3	0.6	0.51	230
5	9.8	33.0	7.9	0.8	0.51	230

Processing the experimental results

The excess air coefficient by the fuel and air-flow rates (by the input) is:

$$\alpha_{\text{in}} = \frac{g_{\text{air}}}{b_{\text{fuel}} g_{\text{stoich}}} \tag{1}$$

The excess air coefficient by the synthesis gas composition (by the output) is:

$$\alpha_{\text{out}} = \frac{1}{1 - \dfrac{78}{21}\dfrac{\text{O}_2 - 0.5(\text{CO} + \text{H}_2)}{\text{N}_2}} \tag{2}$$

The discrepancy between the excess air coefficient by the output (actual) and by the input (theoretical) shows the completeness of gasification process.

Evaluation of the material and heat balance allows determination of the measurement reliability and identification of the best regime of gasification.

The flow-rate of synthesis gas is determined from the balance of nitrogen. The degree of coal and carbon conversion is determined from the carbon balance. Synthesis gas humidity is determined from the balance of hydrogen. The convergence of the material balance is determined by oxygen:

$$\text{N(coal)} + \text{N(air)} = \text{N(gas)} \tag{3}$$

$$\text{C(coal)} = \text{C(CO)} + \text{C(CO}_2) + \text{C(res)} \tag{4}$$

$$\text{H}(\mathit{coal}) + \text{H}(\mathit{steam}) = \text{H(H}_2) + \text{H(H}_2\text{O)} \tag{5}$$

$$\text{O(coal)} + \text{O(air)} + \text{O(steam)} = \text{O(CO)} + \text{O(CO}_2) + \text{O(H}_2\text{O)} + \Delta\text{O} \tag{6}$$

The conversion degree of fixed carbon is:

$$X_{\text{c}} = \frac{\text{C(coal)} - \text{C(res)}}{\text{C(coal)}} \tag{7}$$

The conversion degree of fixed hydrogen is:

$$X_{\text{H}} = \frac{\text{H(H}_2)}{\text{H(coal)} + \text{H(steam)}} \tag{8}$$

The degree of coal conversion is:

$$X_{\text{coal}} = \frac{m_{\text{coal}} - m_{\text{c\_res}} - m_{\text{ash}}}{m_{\text{coal}} - m_{\text{ash}}} \tag{9}$$

The input part of the heat balance consists of the chemical energy of coal and the physical energy of coal, air, and steam. Since coal and air are fed into the reactor at a room temperature, only the physical heat of steam is taken into account. The heat balance output consists of chemical energy (calorific value) of synthesis gas, physical heat of synthesis gas, chemical energy of carbon in the coke-ash residue and physical heat of the coke-ash residue. Unbalance is registered as heat losses:

$$Q(\text{coal}) + Q(\text{steam}) = Q(\text{ch\_g}) + Q(\text{ph\_g}) + Q(\text{ch\_ res}) + \text{Q}(\text{ph\_res}) + \Delta Q \tag{10}$$

The efficiency (chemical efficiency) of gasification is:

$$CCE = \frac{Q(\text{chem})}{Q(\text{coal})} \tag{11}$$

Results

The results of measuring the temperature and composition of the synthesis gas in different regimes are presented in figs. 3 and 4. Periodic fluctuations in concentrations of synthesis gas components are associated with disassembling and cleaning the gas sampling tube from the coke-ash residue and moving this tube from the end of the reaction chamber to its middle and beginning in order to obtain data on distribution of gas composition along the reactor. During the first 30 minutes, the reactor was heated; then regime testing was started. Each regime was maintained for approximately 15 minutes. The average temperatures at various points and gas compositions at the reactor outlet are shown in tabs. 4 and 5. It can be seen that the temperatures along the reactor do not change with increasing steam/fuel ratio.

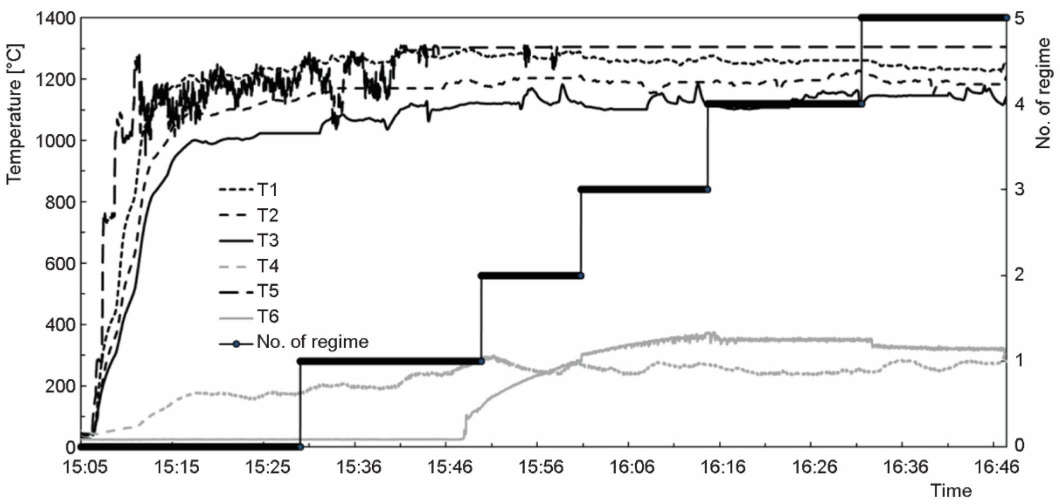

Figure 3. Results of temperature measurements during regime variation (thermocouple numbers correspond to fig. 2)

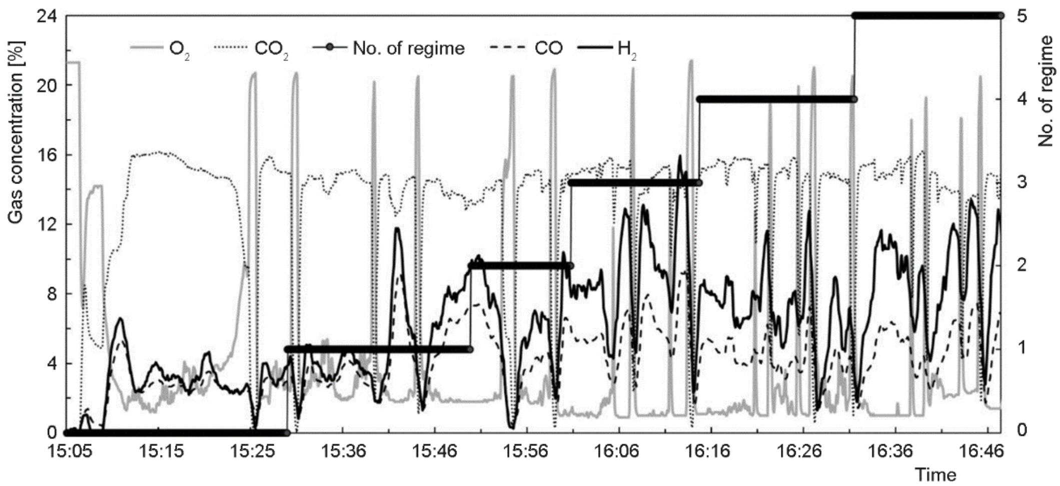

Figure 4. Results of measuring the synthesis gas composition during regime variation

Table 4. The average temperatures at different points of the reactor with varying regimes (thermocouple numbers correspond to fig. 2)

No. of regime	Temperature [°C]					
	T1	T2	T3	T4	T5	T6
1	1260	1270	1180	1120	193	-
2	1308	1273	1188	1117	250	250
3	1300	1260	1187	1123	266	349
4	1300	1261	1213	1155	348	238
5	1300	1230	1182	1156	318	267

Table 5. The composition of dry synthesis gas at the reactor outlet with varying regimes

No. of regime	Concentration [% vol.]				Q, [MJm^{-3}]	H_2/CO
	CO	CO_2	H_2	O_2		
1	2.7	14.5	3.3	4.5	0.70	1.22
2	7.7	13.1	8.8	1.8	1.92	1.14
3	6.9	15.2	12.0	1.0	2.17	1.74
4	6.2	15.6	10.8	1.0	1.95	1.74
5	6.0	15.1	9.7	1.0	1.80	1.62

The results of calculation of the material and heat balances of the regimes are given in tabs. 6 and 7. The dependence of composition of the wet synthesis gas at the reactor outlet on the steam/fuel ratio is shown in fig. 5. The concentration of H_2O in the wet synthesis gas was calculated from the material balance. The obtained dependences of calorific value, H_2/CO ratio in the synthesis gas, and gasification efficiency as a function of the steam/fuel ratio are shown in fig. 6. The dependence of the conversion degree of coal, carbon, and hydrogen on the steam/fuel ratio is shown in fig. 7.

Table 6. The results of material balance calculation

No. of regime	Air excess (by output)	Output of wet synthesis gas [m^{3}h^{-1}]	Synthesis gas humidity [% vol.]	Unbalance by O [% mol.]	Degree of carbon conversion X_C	Degree of hydrogen conversion X_H	Degree of coal conversion
1	1.08	231.4	7.5	–7.50	0.49	0.29	0.76
2	0.74	237.3	8.9	4.81	0.65	0.47	0.83
3	0.67	239.6	11.1	4.38	0.72	0.49	0.87
4	0.70	240.3	17.0	2.64	0.70	0.35	0.86
5	0.73	236.6	22.9	3.43	0.66	0.25	0.84

Table 7. The results of heat balance calculation

No. of regime	Chemical energy of coal [kW]	Physical heat of steam [kW]	Physical heat of synthesis gas [kW]	Chemical heat of synthesis gas [kW]	Chemical heat of coke-ash residue [kW]	Physical heat of coke-ash residue [kW]	Unbalance (heat losses) [% of input]	Chemical efficiency [%]
1	91.9	0.00	23.8	8.9	37.2	2.2	23.98	10.5
2	85.1	0.46	24.2	24.7	23.9	1.7	15.02	34.1
3	79.1	1.13	24.7	27.3	17.4	1.4	13.65	43.7
4	74.1	1.02	25.2	22.5	17.8	1.4	12.96	41.1
5	69.5	1.43	25.0	19.1	18.8	1.4	11.65	39.5

According to fig. 5, the addition of steam to air blowing increases significantly the hydrogen content in the synthesis gas. The degree of carbon conversion with the steam addition increases from 30% to 45% depending on the steam/fuel ratio. Dependences of the calorific value of synthesis gas, chemical efficiency, and degree of coal conversion on the steam/air ratio are non-monotonous with a maximum at a steam/air ratio of 0.4 kg/kg. The physical heat of the synthesis gas increases with increasing steam/fuel ratio. The degree of fixed hydrogen conversion does not exceed 50% and decreases sharply with increasing steam/fuel ratio of more than 0.5 kg/kg.

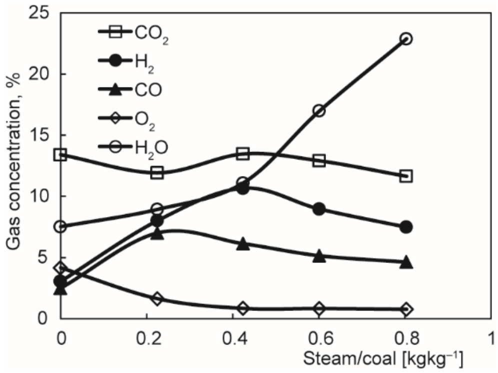

Figure 5. The dependence of the of wet synthesis gas composition at the reactor outlet on the steam/fuel ratio

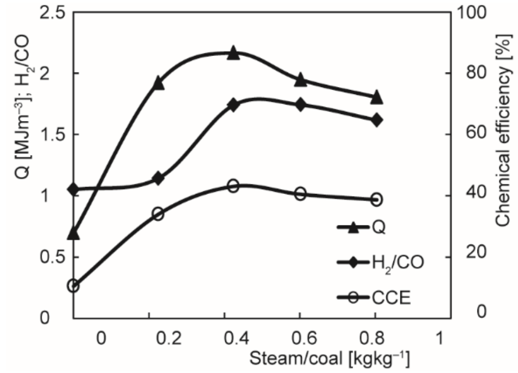

Figure 6. The dependence of calorific value, H_2/CO ratio in synthesis gas and gasification efficiency on the steam/fuel ratio

The discrepancy in the material balance for the regimes with steam supply is less than 5%. Heat losses in the regime with steam supply are relatively constant and make up about 11% of the incoming heat to the reactor, which confirms the stationary nature of the obtained regimes. The somewhat larger values of discrepancies of the material and heat balance for the regime without steam supply are related to the fact that this regime was worked out first, and the reactor heating to the operating temperature was not completed.

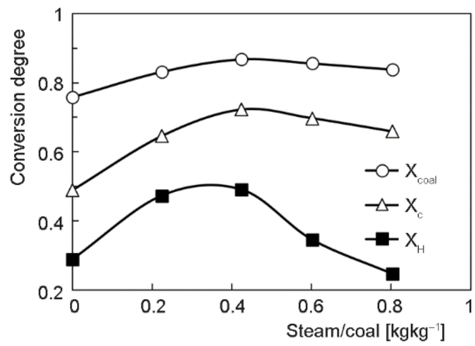

Figure 7. The conversion degree of coal, carbon and hydrogen *vs.* steam/fuel ratio

Conclusion

The stationary regimes of steam-air gasification of mechanically activated coal were obtained due to experimental studies on a modernized flow coal reactor. The positive effect of steam addition to air blowing was revealed. In the obtained regimes, the steam addition increases the hydrogen content in the synthesis gas by 2-2.5 times and increases the degree of carbon conversion from 0.49 to 0.72. For the studied coal, the optimal steam/fuel ratio of 0.4 kg/kg was determined. At that, the chemical efficiency of gasification is about 44%. The relatively low values of gasification efficiency and high values of H_2/CO (1.2-1.7) are explained by incomplete conversion of fuel carbon due to the short time of fuel particle stay in the reactor because of the limited size of the reaction chamber. The reliability of results is confirmed by the convergence of material and heat balances.

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