An ex Situ Underground Coal Gasification Experiment with a Siderite Interlayer. Course of the Process, Production Gas, Temperatures and Energy Efficiency

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An ex situ underground coal gasification experiment with a siderite interlayer. Course of the process, production gas, temperatures and energy efficiency

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Abstract

A 72-hour ex situ hard coal gasification test in one large block of coal was carried out. The gasifying agent was oxygen with a constant flow rate of 4.5 Nm³/h. The surroundings of coal were simulated with wet sand with 11% moisture content. A 2-cm interlayer of siderite was placed in the horizontal cut of the coal block. As a result of this process, gas with an average flow rate of 12.46 Nm³/h was produced. No direct influence of siderite on the gasification process was observed; however, measurements of CO₂ content in the siderite interlayer before and after the process allowed to determine the location of high-temperature zones in the reactor. The greatest influence on the efficiency of the gasification process was exerted by water contained in wet sand. At the high temperature that prevailed in the reactor, this water evaporated and reacted with the incandescent coal, producing hydrogen and carbon monoxide. This reaction contributed to the relatively high calorific value of the resulting process gas, averaging 9.41 MJ/kmol, and to the high energy efficiency of the whole gasification process, which amounted to approximately 70%.

Keywords

Underground coal gasification, UCG, siderite interlayer, water, ex situ, hydrogen

1 Introduction

Underground coal gasification (Bhutto et al. 2013; Cena and Thorness 1981; Perkins 2018) consists of the reaction of the oxidizing reagent (mainly oxygen) and water with the raw coal located underground, which results in the production of process gas and byproducts such as

postprocess water and tar substances. The main idea of this process is to obtain process gas with the highest possible amount and calorific value. The main components of this gas are H₂, CO, CH₄ and CO₂. After cleaning, this gas can be a valuable raw material for the chemical industry (Gregg and Edgar 1978; Maev et al. 2018). The course of this process is influenced by many factors, the most important of which are the oxygen concentration in the gasifying agent and its flow rate, process pressure and temperature, coal composition and geological conditions (Kreinin et al. 1982; Blinderman and Friedmann 2006). From the literature (Shu et al. 2020; Hamanaka et al. 2017), it is known that the higher the concentration of oxygen in the gasifying agent, the higher the temperature, and in turn the higher the concentration of CO and H₂ and the lower the concentration of CH₄ and CO₂ in the process gas. With increasing pressure (Wiatowski et al. 2016; Roberts and Harris 2000; Kapusta et al. 2020; Wall et al. 2002), the content of CH₄ and CO₂ increases, and the content of CO and H₂ decreases. As far as the role of water (Dvornikova 2018; Perkins and Prabu 2017; Surya and Prabu 2017) in underground gasification is concerned, it is essential because it is the raw material from which H₂ and CO are produced. Reactions in which water is essential are described by the following equations:

Reaction (1) is endothermic, while reaction (2) is slightly exothermic. The source of water is the moisture contained in coal and water inflowing to the gasified seam. If there is a lack of water, it has to be supplied from the outside; but on the other hand, when there is too much water, the temperature in the gasified seam decreases and the consumption of heat energy necessary to evaporate the excess water increases. Evaporated water in the form of steam increases the moisture content of the process gas, making it necessary to remove excess water in the gas purification process. In addition, the presence of a large amount of steam in the gas

increases its volume, which increases the cost of gas transport. At low process gas temperatures, steam may condense inside the pipeline, transporting the gas. This can, in extreme cases, result in water and tar jams, which are not beneficial for this process. All phenomena related to excess water cause an increase in energy consumption, which in the final effect reduces the efficiency of the process.

Another factor influencing the UCG process is the composition and properties of the gasified coal. Hard coal (Elliott 1981; Chodyniewska and Walanus 1985) is a very complicated mixture of organic carbon substances, mineral substances and moisture. The mineral substances contained in coal may affect the course of gasification. One of the minerals usually found in coal is siderite (Retallack 2007). Its main component is iron (up to 48%) in the form of iron carbonate FeCO_3 . Most often, such siderite is partially oxidized, which is manifested by its brown colour. Apart from iron, siderite often contains elements such as Si, Al, Mg and Ca. Siderite in coal may occur in the form of discontinuous inserts, thin layers, sideritized limestones and mudstones or nodules made of compacted oolite-type siderites (Maes 2000). Hard coal may have a siderite content between zero and a few percent. In a study (Riley 2012) concerning research on Australian bituminous coals, the siderite content was 0.11-2.72%. Siderite is also found in rocks accompanying coal seams in the form of clusters of variable thickness between 2-40 cm. Its content in these rocks may be higher than the content in coal and can reach several dozen percent (Chodyniewska and Gabzdyl 1986).

Under high-temperature conditions that occur during UCG, the siderite undergoes thermal decomposition (Dhupe and Gokarn 1990) to wustite. Siderite decomposition is an endothermic reaction that occurs at temperatures of 350-530 °C according to a simplified equation (variant I):

The resulting iron oxide FeO may react with carbon dioxide to magnetite:

In variant II (Zhu et al. 2016), siderite may decompose according to the following equation:

The resulting magnetite may undergo the opposite reaction described in equation (4) to wustite. Siderite can also react with the hydrogen contained in the process gas according to the following equation:

If the decomposition of siderite takes place in an atmosphere containing oxygen, at 600-750 °C, iron is oxidized to magnetite (Lee et al. 2009). This reaction is exothermic and can be described by the following equation:

The iron oxides formed from siderite have a catalytic effect (Smirnov et al. 2019; Wang et al. 2016; Yang et al. 2009) in the reaction of water gas conversion (2) and the endothermic Boudouard reaction:

The catalytic properties are dependent on the degree of surface development and contact with substrates. If iron oxides are small in size and dispersed in coal, they can have a catalytic effect. If they are concentrated in one place in the form of larger grains, then the catalytic effect will be marginal (Smirnov et al. 2019).

The Boudouard reaction (8) is reversible and only occurs at low pressures above 800 °C. From the equations presented, it follows that the decomposition of siderite causes the release of CO₂. If the siderite is located close to the high-temperature zone, the sample taken after the gasification process should contain a lower amount of CO₂ than before the process. It is therefore possible, if only in a very approximate way, to determine the location of high-

temperature zones in the reactor. Under real conditions, this is not feasible, but during the ex situ gasification process, such an experiment is possible.

The main purpose of this study was to investigate the effect of siderite addition on the course of the underground coal gasification process in ex situ conditions and to attempt to determine the temperature distribution in the UCG reactor. The impact of moisture contained in the surroundings of the gasified coal seam on the composition and yield of the process gas was also determined. Research on the influence of siderite and the mapping of temperatures in the UCG process described in this work has not been conducted thus far. The results obtained can therefore contribute to the expansion of knowledge in this field of science.

2 Experimental

2.1 Installation for ex situ coal gasification

The coal gasification test was conducted in an experimental reactor designed to simulate the UCG process under surface conditions. The installation enables leading of the underground coal gasification process on the surface (ex situ) in a simulated coal seam (max. seam length 7.0 m, cross section 1.0×1.0 m) under atmospheric pressure. The maximum design process temperature can be up to 1600 °C. The gasification reactor was equipped with appropriate technical infrastructure to carry out the gasification process (gasification agent dosing system, takeoff, cleaning and disposal of the resulting process gas). Gasification tests can be carried out using oxygen, air and steam, either individually or in mixtures. Nitrogen is used to inertise the installation and in the final phase to cool the reactor after the process. A schematic view of the installation is presented in Fig. 1, and details of the reactor construction are shown in Fig. 2a-c.

The process gas was treated in a dedicated separation and purification module, the first element of which was a water scrubber (quick gas cooling and condensation of process tars).

Next, the gas was directed to an air cooler, moisture separators, oily substances and solid particles.

Fig. 1 Scheme of ex situ UCG installation: (1) reagent supply system, (2) gasification reactor, (3) connection for tar sampling, (4) water scrubber, (5) air cooler for process gas, (6,7) gas separators, (8) centrifugal suction fan, (9) thermal combustor, (10) gas purification module for GC analysis

Fig. 2 Details of reactor construction: a) longitudinal cross section: arrangement of coal seam and thermocouple positions, b,c) cross-section: dimensions of coal seam (b) and thermocouple distances (c)

Part of the gas stream was directed through a separate gas path for chemical analysis, where concentrations of basic gas components such as hydrogen, carbon monoxide and dioxide, methane, ethane and hydrogen sulfide were determined by chromatographic methods. The temperature profiles inside the reactor were measured using a set of up to 18 thermocouples placed at different heights of the simulated coal seam and overburden layer. The length of each thermocouple (0.45 m) was selected so that it is possible to measure the temperature as close as possible to the fire channel both inside and outside the gasified bed. The control equipment recorded data every 10 seconds. All results obtained for gas products were converted to normal conditions ($T=273.15\text{K}$, $p=1013.25\text{ hPa}$).

2.2 Materials

As a material for the gasification test, hard coal “Piast” was used, and the mineral interlayer was a layer of siderite. The insulation of the gasified coal from the reactor walls was wet sand, which filled the empty spaces of the reactor and simulated the moisture of the surroundings of the gasified coal. The gasifying agent was technical oxygen of 99.95% purity taken from the bundle of oxygen cylinders. The basic properties of the “Piast” hard coal used for gasification are presented in Table 1. Analysis of coal was carried out in a certified laboratory at Silesian Technical University according to Polish Standards.

Table 1 Proximate and ultimate characteristics of coal from Piast mine and standard analytical methods

Parameter		Value	Standard
As received			
Total moisture	W_t^r (%)	4.70	PN-G-04511:1980
Ash	A_t^r (%)	16.30	PN-G-04560:1998
Volatiles matter	V_r^a (%)	30.10	PN-G-04516:1998
S total	S_t^r (%)	0.83	PN-G-04584:2001
Lower heating value	Q^r (kJ/kg)	22 719	PN-G-04513:1981
Analytical			
Moisture	W^a (%)	2.95	PN-G-04560:1998
Ash	A^a (%)	16.60	PN-G-04560:1998
Volatiles matter	V^a (%)	30.65	PN-G-04516:1998
Lower heating value	Q^a (kJ/kg)	23 137	PN-G-04513:1981
S_t^a total	(%)	0.85	PN-G-04584:2001
C^a	(%)	62.50	PN-G-04571:1998

H ^a	(%)	4.39	PN-G-04571:1998
N ^a	(%)	1.04	PN-G-04571:1998
O ^a *	(%)	11.67	

*Oxygen calculated as $(O^a) = 100 - (W^a) - (A^a) - (C^a) - (H^a) - (S_t^a) - (N^a) (\%)$

The raw coal used was characterized by low moisture (as received) at 4.7%, a relatively high content of ash at 16.30% and volatile parts at 30.10%. The lower heating value of coal was 22 719 kJ/kg. Because the raw coal did not contain siderite, it was necessary to deliver it from outside. The siderite used to make the interlayer was a mixture of several siderites from the area of the Chwałowice Basin, from the Rudzkie layers. It was obtained from a heap located on the border of the Marklowice and Świerklany communes using mining waste from deposits 408 and 409 of the Chwałowice mine. Prior to the gasification process, the elemental chemical composition in the siderite interlayer was determined using X-ray fluorescence (XRF) from the ZSX Primus II Rigaku spectrometer, and the results were converted into oxides, while the CO₂ content was determined by the volumetric method in the Scheibler-Dietrich apparatus. The method consists of measuring the volume of CO₂ released during the reaction of the tested sample with hydrochloric acid. To obtain accurate results of the CO₂ content in the siderite interlayer, samples for analysis were taken every 0.2 m (measuring grid 0.2 × 0.2 m). In total, approximately 80 samples were analysed. The obtained results showed that the tested siderites were characterized by similar CO₂ content in the range of 33.1-35.7% by mass and 34.6% on average. The detailed mineral composition of the siderites is presented in Table 2.

Table 2 Average composition of used mixture of siderites

Component	Composition (wt %)
FeO	41.08
SiO ₂	10.72
Al ₂ O ₃	5.03
MgO	3.33
CaO	2.37
Na ₂ O	0.79
MnO	0.63
K ₂ O	0.47
P ₂ O ₅	0.43

Cl	0.24
TiO ₂	0.19
SO ₃	0.05
BaO	0.03
SrO	0.02
ZnO	0.02
CO ₂	34.60
Total	100.00

The data show that the dominant component is iron oxide - 41.08% FeO. The content of other minerals was much lower and amounted (up to 1% by mass): silicon dioxide - 10.72%, aluminium trioxide - 5.03%, magnesium oxide - 3.33% and calcium oxide - 2.37%.

2.3 Preparation of coal seam with siderite interlayer and reactor for gasification

The block of coal used for gasification was 3.8 m long, 0.7 m wide and 0.7 m high. It was cut horizontally at a height of 0.5 m counting from underneath. The 2-cm-thick siderite interlayer was placed in the cutting of the block. The weights of the blocks of coal and siderite were 2300 kg and 150 kg, respectively. To fill the empty space in the reactor, 7900 kg of wet sand of 11.0% humidity was used. The moisture content in the sand was measured according to the PN-G-04511:1980 standard. The arrangement of thermocouples in the reactor is shown in Fig. 2a. Thermocouples no. 2-4, 8-11 and 15-18 measured temperatures in the gasified coal seam and others in the sand stratum.

2.4 Apparatus for measuring physicochemical properties of gaseous media

- The gas temperature inside the reactor was measured with thermocouple-type Pt10Rh-Pt
- At the reactor inlet, outlet and scrubber thermocouples Pt100 were used
- The installation pressure was measured by a WIKA digital transmitter type IS-20-S
- The process gas flow was measured using an ELSTER bellows gasometer type BK-G10M
- The composition of gases was measured using an Agilent 3000A gas chromatograph
- The oxygen flow was regulated by the Bronkhorst EL-FLOW type mass flow controller, model F-202AV-M20-RAD

2.5 Experimental procedure

The experiment began with switching on the suction fan and then igniting the coal seam with a pyrotechnic charge. The oxygen flow rate was set at 4.5 Nm³/h, and this value was kept constant throughout the experiment. To prevent clogging of the process gas pipes by the liquefied tar, a pump feeding water to the scrubber was switched on simultaneously (water injection 14 kg/h). Every 2 hours, the scrubber was emptied, and the amount of wastewater received was weighed. The gas produced was analysed every hour or more often as required. After three days of the gasification process (72 hours), the oxygen supply was stopped, and nitrogen in the amount of 2 Nm³/h was started until the gasified coal seam was cooled down (to internal temperatures below 100 °C). After the gasification test was completed, the reactor was disassembled, the sand overburden was removed, its moisture content was examined, and samples of thermally transformed siderites were taken to analyse the CO₂ content.

3 Results and discussion

3.1 Process gas flow and gas calorific value

Fig. 3 and Fig. 4 show the flow of the process gas obtained and its calorific value.

Fig. 3 Process gas flow rate

Fig. 4 Process gas calorific value

The data received (Fig. 3) show that from the beginning of the process until approximately 46 hours, the flow of gas received was relatively stable and amounted to 10-14 Nm³/h. After 46

hours of the process, the amount of gas produced started to increase to the final value of approximately 18 Nm³/h. The total amount of gas produced was 896.94 Nm³ with an average flow of 12.46 Nm³/hour. The calorific value of the gas (Fig. 4) showed a downward trend from 5 and 46 hours. Between the next 46-64 hours of the process, there was an upward trend in the calorific value of the gas; then, after 64 hours, the calorific value decreased slightly until the end of the experiment. During the entire gasification period, quite rapid increases and decreases in caloric value were visible. For example, between 25-28 hours and 56-58 hours of the process, the calorific value of the gas increased from approximately 9-10.5 MJ/Nm³ and from 9-11 MJ/Nm³, respectively. The calorific value of the gas obtained was very high, ranging from 8-11 MJ/Nm³, while the average value over the entire gasification period was 9.41 MJ/Nm³.

3.2 Process gas composition and mass flow of its components

Measurements of the composition of the obtained gas are presented in Fig. 5, while the mass stream of its individual components is presented in Fig. 6.

Fig. 5 Composition of process gas

Fig. 6 Mass flow of process gas components

The data presented in Fig. 5 show that the composition of the gas produced was quite stable throughout the gasification process. Between 25-28 hours and from approximately 46 hours of the process, a significant decrease in carbon dioxide content was observed with a simultaneous increase in hydrogen and carbon monoxide content. From approximately 46 hours of gasification, the methane content started to gradually increase, and after 60 hours, it stabilized at

a value of approximately 5%. Changes in the concentrations of process gas components caused an increase in its calorific value (Fig. 4) in the given time intervals. Throughout the whole experiment, high concentrations of hydrogen (23-38%) and carbon monoxide (27-46%) were observed. The methane concentration during the whole process was in the range of 2-5.6%, while the content of ethane and hydrogen sulfide was practically below 0.5% and 0.2%, respectively.

Fig. 6 shows that the hydrogen and carbon monoxide flow increased twice between 25-28 hours and twice from 46 hours to the end of the gasification process. As far as methane is concerned, in the second half of the gasification process, a clear increase in its flow was observed. From approximately 46 hours to the end of gasification, the flow of methane increased by up to three times in the same period of time as the increase in flow of hydrogen and carbon monoxide. The relationships presented in Fig. 6 describe the changes in process gas during the gasification process in more detail than in Fig. 5 because they take into account the concentrations of individual components of the process gas and their flow.

The average composition of the process gas is shown in Table 3. Notably, the high average content of hydrogen and carbon monoxide and low CO₂ content are similar to those in the UCG process.

Table 3 Composition of process gas and its calorific value

Average composition of process gas (% vol.)							Lower heating value (Q)
CO ₂	N ₂	H ₂	CH ₄	CO	C ₂ H ₆	H ₂ S	(MJ/Nm ³)
27.27	0.80	30.93	3.53	37.11	0.09	0.27	9.41

3.3 Amount of water obtained and humidity of process gas

As a result of coal gasification, in addition to gas, process water was obtained. The amount of this water produced and the gas moisture content calculated on this basis are shown in Fig. 7.

The data presented only relate to the amount of water that entered the scrubber from the

reactor due to evaporation and subsequent condensation, which was created as a result of chemical reactions occurring during gasification. The amount of water that was injected into the scrubber to cool hot gases was subtracted. At the beginning of gasification, the mass of condensate liquefied grows quickly and then drops rapidly. It reaches a minimum of 4.5 kg/h in 10 hours. This stage is related to the evaporation of moisture contained in the gasified coal close to the ignition place. As time passed, the gasification zone expanded, the temperature increased, and as a result, the mass of the condensate also increased.

Fig. 7 Weight of water condensate and humidity of process gas

Between 26-32 hours, a rapid increase in the weight of the condensate was observed with a maximum of 10 kg/h in 30 hours. This stage is most likely related to the increase in temperature of the gasified coal seam and the evaporation of part of the moisture from the wet sand surrounding the coal seam. As the gasification zone expanded, the wet sand heated more and more, resulting in approximately 7 kg/h of condensate being produced between 40-52 hours. The moisture content of the sand decreased, the mass of condensate decreased, and the gasification zone moved to further regions of the coal seam. At the end of the process, starting from approximately 58 hours, the amount of water released increased again. During the whole process, 521 kg of condensate was obtained, and its average rate of release was ~7.2 kg/h. On the basis of the mass of the separated condensate, the moisture content in the

process gas was calculated. The shape of this dependence is similar to the curve of the condensate release. The course of the curve has a declining tendency that results from the increasing amount of the process gas. The average calculated moisture content in the process gas converted into dry gas was 0.62 kg H₂O/Nm³.

3.4 Determination of CO₂ content in siderite interlayer after gasification process

After the process, samples of thermally transformed siderite were taken again to re-determine the CO₂ content. The samples taken came from the same locations as for the determination of CO₂ content before gasification. This allowed us to compare the CO₂ content in the siderite layer before and after the process. The composition of the separated CO₂ was additionally controlled by a gas chromatograph. The results are presented graphically in Fig. 8 in the form of isolines of CO₂ concentrations in the siderite layer (top view).

Fig. 8 Isolines of carbon dioxide content in siderite layer after gasification process (vt %)

The largest decreases in CO₂ content were observed mainly in the first half of the gasified coal seam close to the fire channel located underneath. The farther away from the fire channel, the lower the thermal transformation of siderite. Fig. 8 also shows local and lateral deviations of the gasification directions. This direction usually follows the flow direction of the gasifying agent because the oxygen concentration in this stream is the highest. The visible deviations are most likely due to local, higher oxygen concentrations in these areas, which intensified the gasification reaction in these places. The higher the gasification intensity, the

higher the temperatures and the higher the level of thermal degradation of the siderite. As a result, the CO₂ content in the siderite layer was located in areas where temperatures were high. This suggests that in real conditions, such a phenomenon may also occur.

The most visible changes in CO₂ concentration occurred from 0-0.5 m and between 1-1.7 m long of the siderite interlayer. In these areas, the CO₂ content fell to the lowest value of 0-2% by mass. Starting from the length of 1.7 m, the CO₂ content in the siderite layer gradually increased, which means that the farther away from the beginning of the deposit, the smaller the amount of siderite that decomposed to CO₂. This indicates a gradual decrease in the gasification process temperature in these areas. Using data on the CO₂ content in siderite before the process (34.6% by mass) and the data from Fig. 8, it was calculated that only approximately 34.1% of the used siderite was decomposed to CO₂, producing 8.99 Nm³ ± 2 Nm³ of CO₂. This represents approximately 2.9-4.5% of the total amount of CO₂ produced in gasification. This is a small amount that has little effect on the CO₂ balance and the course of the process.

3.5 Pressure and temperature profiles

The following pictures (Fig. 9-12) show the pressure and temperature diagrams during the gasification process.

Fig. 9 Pressures and temperatures at reactor inlet, outlet and inside scrubber
a) pressures, b) temperatures

To prevent the air from being sucked in by the suction fan (Fig. 1), the experiment was carried out in such a way that a minimum overpressure of 5-10 mbar was maintained in the reactor. Pressure sensors were located at the inlet and outlet of the reactor and in the wet scrubber. The pressure diagram (Fig. 9a) shows that between hours 10-15 and 56 of the process, quite rapid pressure peaks were observed.

Fig. 10 Temperature graph in fire channel, first row of thermocouples

Fig. 11 Temperature diagram in second row of thermocouples

Fig. 10 shows the temperatures obtained in the lowest layer of the gasified coal seam. The highest temperatures in this layer were recorded by thermocouple T2 at over 1100 °C. The next diagram (Fig. 11) shows the temperature profiles in the second row of the gasified coal seam. These thermocouples were located at a height of approximately 0.4 m from the bottom of the gasified coal seam. In contrast to the first row of thermocouples (Fig. 10), the highest temperatures in the second row were recorded by the second thermocouple (T9). The maximum temperatures indicated by these thermocouples were over 1200 °C. Further thermocouples recorded lower values. The third row of thermocouples (T15-21) was located above the layer of siderite in the coal seam at a height of approximately 0.65 m from the bottom. The temperature measurement results recorded by these thermocouples are shown in Fig. 12.

Fig. 12 Temperatures in third layer of thermocouples over siderite interlayer

The maximum measured temperatures in the third layer were up to approximately 300 °C. The exception is the second T16 thermocouple, which measured a temperature of approximately 1000 °C. Fig. 2a,b and Fig. 10-13 show that the T2, T8, T9 and T16 thermocouples were located in the zones where the most carbon dioxide was released from siderite. These thermocouples indicated values exceeding 500 °C. Because siderite decomposes above this temperature, it is clear that there is a certain correlation between the temperature in these places and the amount of CO₂ released from the siderite.

3.6 Balance calculation

3.6.1 Energy and power

The amount of thermal energy contained in the process gas (after it burned) was calculated on the basis of data on the process gas stream and its calorific value. Then, by dividing the result obtained by the whole gasification time (72 hours), the thermal power of the gasification experiment was calculated. The obtained results are shown in Fig. 13. Until approximately 46 hours had elapsed, the process gas had a relatively high thermal energy at 100-140 MJ. Starting from the 46th hour of the process, the energy contained in the gas began to increase

to a high value of approximately 200 MJ. The calculated average heating power of the process gas obtained during the whole experiment was 32.56 kW.

Fig. 13 Heat energy contained in process gas and its heating power

3.6.2 Coal balance

The process gas contained 327.17 kg of carbon. The coal balance showed that if such a quantity of coal was contained in the process gas, then 532.80 kg of raw coal had to be gasified. Taking into account that the calorific value of coal subjected to gasification was 22.719 MJ/kg, it can be calculated that if such a quantity of coal was burned, then 12 104.82 MJ of thermal energy would be released. It was calculated that the gasification efficiency was 69.73%. The mass and energy balance results are presented in Table 4.

Table. 4 Mass and energy balance results

Time [h]	Process gas yield (Nm ³)	Average process gas flow rate (Nm ³ /h)	Average process gas calorific value (MJ/Nm ³)	Total heat energy in the process gas (MJ)	Average reactor operating power (kW)	Average gasification rate (kg/h)	Energy contained in coal gasified (MJ)	Gasification energy efficiency (%)
72	896.94	12.46	9.41	8 440.21	32.56	7.40	12 104.82	69.73

This is a very good result because it means that only approximately 30% of the energy contained in the calculated amount of gasified coal was used to heat the coal seam to a sufficiently high temperature, evaporate the water and lose to the surrounding strata. On the

basis of data on the amount of coal contained in the gasified coal seam before gasification and in the gases obtained, it was calculated that only 23.17% of the total mass of raw coal contained in the reactor was gasified.

3.6.3 Hydrogen balance

The 896.94 Nm³ of process gas contained 30.86 kg of hydrogen. 532.80 kg of raw coal containing 4.39% hydrogen was gasified. This amount can theoretically produce 22.98 kg of hydrogen. The missing amount of hydrogen (7.88 kg) could have come from the evaporation of part of the water contained in the coal seam and its reaction with glowing coal. The raw coal used contained 4.70% moisture, and 532.80 kg of coal contained 25.04 kg of water. This amount of water contained 2.78 kg of hydrogen (H₂). The total mass of hydrogen was 22.98+2.78= 25.76 kg. In the balance sheet, 5.10 kg of hydrogen was still missing. However, it is difficult to assume that this calculated amount of water reacted with coal. A large part of it could evaporate and liquefy in the scrubber. It is more likely that the missing amount of hydrogen came from the reaction with coal of part of the water contained in wet sand. Fig. 8 shows that the greatest changes in the CO₂ content in the siderite layer occurred in the length of the coal deposit of 1.7 m. These changes are mainly due to the highest temperatures prevailing in this area during gasification. To estimate the amount of water evaporated from the sand, the initial 1.7 m of coal deposit length was taken into account.

In the immediate surroundings of this part of the coal seam, there was approximately 1600 kg of wet sand containing 176 kg of water. This amount of water can theoretically produce 19.6 kg of hydrogen. Since only 5.10 kg of hydrogen was missing in the balance, it can be estimated that only approximately 26% of this amount of water reacted to hydrogen, and the rest condensed in the scrubber. It should be taken into account that the calculations made are only approximate, as they do not take into account all factors that may have influenced the results.

3.6.4 Water balance

During the whole experiment, 521 kg of water was condensed in the scrubber. Raw coal in the reactor (2300 kg) contained approximately 108 kg of water. Even if all of this water was evaporated and condensed in the scrubber, 413 kg of water was still missing. The only source of such water is the moisture contained in wet sand. Assuming the simplifying assumption that all of the water contained in 7900 kg of wet sand was evaporated, it was calculated that approximately 869 kg of water could be released from this amount of wet sand. In addition, after taking into account that 46 kg of this water can supplement the hydrogen balance (producing the missing 5.10 kg of hydrogen), $869 - 46 = 823$ kg of water was still available. This amount twice exceeds the water deficiency (413 kg) in the balance. This result proves that the main source of water obtained from gasification is water from wet sand and confirms previous assumptions that part of this water is in the form of steam reacted with coal to water gas, thus increasing the amount of hydrogen and carbon monoxide produced.

4 Discussion of results obtained

On the basis of the results obtained, the influence of the siderite interlayer on the amount and composition of the process gas was not found. This is because an insufficient amount of CO_2 separated from the siderite layer. The puzzling sudden decreases in the concentration of carbon dioxide in the process gas are shown in Fig. 5. There may be several reasons for this. For example, increasing the temperature of the gasified coal seam above 800 °C produces a Boudouard reaction that leads to an increase in carbon monoxide content. However, Fig. 5 shows that with an increase in CO concentration, the H_2 concentration increases at the same time. Thus, it is more probable that under the described conditions, with a large supply of steam, the production of water gas ($\text{H}_2 + \text{CO}$) was preferred over CO production alone. This was also confirmed during the water balance calculation in this process. The required temperature for water gas formation is 700-900 °C. Such temperatures were recorded during

this gasification process, and therefore the production of combustible components of this gas was possible. The results obtained indicate the important role of water in the underground gasification process. In the case of this experiment, its source was the moisture contained in the sand surrounding the gasified coal seam; however, under real conditions, in the absence of this moisture, it must be supplied from outside. The results obtained were also influenced by the correct operation of the reactor's oxygen supply system and the removal of produced gases from the reactor. Properly selected conditions of cooperation of both these systems also contributed to the high energy efficiency of the whole gasification process.

5 Conclusions

Based on the measurements carried out, it was concluded that

- 1) During 72 hours of coal gasification, 897 Nm³ of process gas was obtained at an average high calorific value, on average 9.41 MJ/Nm³.
- 2) The average gasification rate was 7.4 kg coal/h. The energy efficiency of the gasification process was approximately 70%. A total of 23.17% of the coal charge was gasified.
- 3) Due to the small amount of CO₂ produced from the thermal decomposition of the siderite interlayer, its influence on the coal gasification process was not observed. However, based on the differences in the CO₂ content in the siderite interlayer before and after the process, it was found that the gasification direction may differ from the gasification agent flow direction. This phenomenon may occur in the case of a real underground gasification process.
- 4) The use of wet sand to surround the gasified coal had an important impact on the gasification process. The water contained in the wet sand reacted with the coal, producing large amounts of hydrogen and carbon monoxide, which improved the

amount and composition of the gas as well as the energy balance of the process. This result shows the important role of water, which is necessary in the UCG process.

- 5) One of the important factors influencing the course of the process was the suitably fast take of the process gas produced. Efficient operation of this installation also contributed to the high efficiency of the gasification process.

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CRedit authorship contribution statement

Marian Wiatowski: Conceptualization, Validation, Investigation, Writing - original draft.

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Jacek Nowak: Conceptualization, Validation, Investigation, Formal analysis.

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All authors have read and agreed to the published version of the manuscript.

Ethical standards

The experiments comply with the current laws of Poland.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Figures

Figure 1

Scheme of ex situ UCG installation: (1) reagent supply system, (2) gasification reactor, (3) connection for tar sampling, (4) water scrubber, (5) air cooler for process gas, (6,7) gas separators, (8) centrifugal suction fan, (9) thermal combustor, (10) gas purification module for GC analysis

Figure 2

Details of reactor construction: a) longitudinal cross section: arrangement of coal seam and thermocouple positions, b,c) cross-section: dimensions of coal seam (b) and thermocouple distances (c)

Figure 3

Process gas flow rate

Figure 4

Process gas calorific value

Figure 5

Composition of process gas

Figure 6

Mass flow of process gas components

Figure 7

Weight of water condensate and humidity of process gas

Figure 8

Isolines of carbon dioxide content in siderite layer after gasification process (wt %)

a)

b)

Figure 9

Pressures and temperatures at reactor inlet, outlet and inside scrubber a) pressures, b) temperatures

Figure 10

Temperature graph in fire channel, first row of thermocouples

Figure 11

Temperature diagram in second row of thermocouples

Figure 12

Temperatures in third layer of thermocouples over siderite interlayer

Figure 13

Heat energy contained in process gas and its heating power