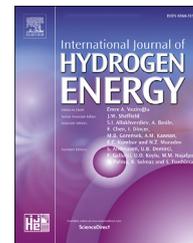




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Experimental simulations of methane-oriented underground coal gasification using hydrogen - The effect of coal rank and gasification pressure on the hydrogasification process

Krzysztof Kapusta ^{a,*}, Marian Wiatowski ^a, Hywel R. Thomas ^b, Renato Zagorščak ^b, Sivachidambaram Sadasivam ^b, Shakil Masum ^b, Thomas Kempka ^{c,d}, Christopher Otto ^c, Wioleta Basa ^a, Marcin Szyja ^a, Krzysztof Stańczyk ^a

^a Główny Instytut Górnictwa (Central Mining Institute), Plac Gwarków 1, 40-166 Katowice, Poland

^b Geoenvironmental Research Centre (GRC), School of Engineering, Cardiff University, Cardiff CF24 3AA, UK

^c GFZ German Research Centre for Geosciences, Telegrafenberg, Potsdam, 14473, Germany

^d University of Potsdam, Institute of Geosciences, Karl-Liebknecht-Str. 24-25, 14476 Potsdam, Germany

HIGHLIGHTS

- Large scale gasification experiments of methane-rich gas production using hydrogen (hydrogasification) were conducted.
- The effect of pressure and coal rank on the methane concentrations and gas production rates was confirmed in the study.
- The technical feasibility of the underground coal hydrogasification (UCHG) concept was confirmed.

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ABSTRACT

This paper presents a series of surface experimental simulations of methane-oriented underground coal gasification using hydrogen as gasification medium. The main aim of the experiments conducted was to evaluate the feasibility of methane-rich gas production through the in situ coal hydrogasification process. Two multi-day trials were carried out using large scale gasification facilities designed for ex situ experimental simulations of the underground coal gasification (UCG) process. Two different coals were investigated: the “Six Feet” semi-anthracite (Wales) and the “Wesoła” hard coal (Poland). The coal samples were extracted directly from the respective coal seams in the form of large blocks. The gasification tests were conducted in the artificial coal seams (0.41 × 0.41 × 3.05 m) under two distinct pressure regimes - 20 and 40 bar. The series of experiments conducted demonstrated that the physicochemical properties of coal (coal rank) considerably affect the hydrogasification process. For both gasification pressures applied, gas from “Six Feet” semi-anthracite was characterized by a higher content of methane. The average CH₄ concentration for “Six Feet” experiment during the H₂ stage was 24.12% at 20 bar and 27.03% at 40 bar. During the hydrogasification of “Wesoła” coal, CH₄ concentration was 19.28% and 21.71% at 20 and 40 bar, respectively. The process was characterized by high stability and reproducibility of conditions favorable for methane formation in the whole sequence of gasification cycles. Although the feasibility of methane-rich gas production by

* Corresponding author.

E-mail address: kkapusta@gig.eu (K. Kapusta).

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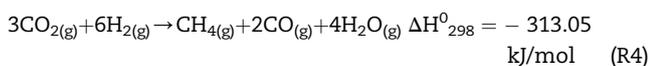
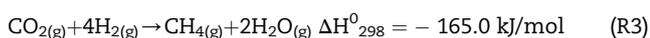
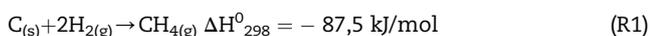
underground hydrogasification was initially demonstrated, further techno-economic studies are necessary to assess the economic feasibility of methane production using this process.

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Introduction

The process of underground coal gasification (UCG) is based on the direct, in-situ conversion of the coal seam into process gas that can be further used for the production of heat, electricity or as a raw material for the chemical industry [1–3]. The gasifying reagent is usually air, oxygen or mixtures thereof, and water. The most valuable components of the process gas are its combustible components, therefore the UCG process is carried out in such a way that the calorific value of the obtained gas is as high as possible. The most important products of the UCG process are methane, hydrogen and carbon monoxide. The factors that determine this are the process conditions such as temperature, pressure, amounts of water involved in the gasification, oxygen concentration in the gasifying reagent and its flow rate [4–8]. A typical UCG gas has the following composition: 10–30% H₂, 15–25% CO, 5–8% CH₄ and 15–60% CO₂. The content of ethane and hydrogen sulfide usually does not exceed 1% [9–11]. The component of the UCG gas that has the highest calorific value is methane. Its calorific value is 35.81 MJ/m³ and is approximately three times higher than the calorific values of carbon monoxide and hydrogen of 12.68 MJ/m³ and 10.79 MJ/m³, respectively.

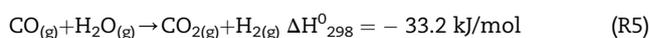
During the thermochemical conversion of carbonaceous feedstocks, methane is formed in the following reactions:



From the equations presented, it can be seen that all reactions are highly exothermic which reduces the amount of heat energy required for coal gasification. Therefore, all these reactions are favored by lower temperature. Furthermore, equations (R1-R4) show that with an increase in the partial pressure of hydrogen, the concentration of methane increases. This is due to the hydrogenation and methanation reactions that convert C, CO or CO₂ and H₂ into CH₄ at high pressures, leading to a reduction in the number of molecules in the reacting system. Reaction (R1) - hydrogasification is heterogenic while the other reactions (R2-R4) occur only in the gas phase. The hydrogasification reaction is favored by high pressure with the effect of increased process pressure on methane yield [15,16]. This reaction is often accelerated by a catalyst (coal ash and metal oxides Ni, Rh, Ru). Typically the

temperature used is not lower than about 650 °C. At lower temperature (400–450 °C) the process is performed with low efficiency, while after exceeding 1200 °C, the resulting methane is pyrolyzed to carbon and hydrogen.

Methane can also be formed [17,18] in three reversible methanation reactions (R2-R4) in which the ratio of hydrogen to carbon monoxide/dioxide should be at least 3:1 or 4:1. These reactions are also favored by high pressure. Reaction (R3) is known as Sabatier reaction [19,20] and it proceeds catalytically at relatively low temperature (300–400 °C) on catalysts [20] such as Ru, Co, Ni, Fe. Reactions (R2-R4) are linked with the water gas shift (WGS) reaction (R5) that always occurs when catalysts are used [21,22].

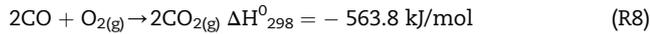
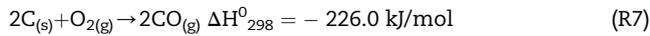


The hydrogen formed in the reaction (R5) can then react as described by reactions (R1-R4). Increasing the methane content of the process gas in the UCG process by adding water to the reacting system is a well-known method [23], because the participation of water in the UCG process is essential. During coal methanation at 650 °C and above, the dominant reaction is heterogeneous exothermic hydrogasification (R1) while the other reactions (R2-R5) occur to a lesser extent. The methanation process has been known and used for many years [12,13]. It is used for the production of substitute natural gas (SNG) from syngas (H₂ and CO), most often obtained via surface gasification of carbon-containing feedstocks (coal, lignite and biomass). A practical example of the application of the methanation process is the Great Plains Synfuel Plant [14], which opened in 1984 in Beulah, North Dakota. It produces 4.1 million m³ of SNG per day by gasifying 18,000 tons of lignite.

In order to carry out the underground coal hydrogasification (UCHG), it is necessary to conduct the process under increased pressure and to provide thermal energy to sustain the reaction. The pressure of the UCG process is related to the depth of the coal seam underground. In-situ coal gasification can be carried out at a pressure that does not exceed the hydrostatic pressure of the water that prevails at a given depth. For example, at a depth of 500 m the maximum hydrostatic pressure of the water is 50 bar and this pressure must not be exceeded by the UCG process. Otherwise, the pressure of the generated gases would exceed the hydrostatic pressure, and as a consequence, contaminants could spread in the rock mass, which should be prevented [24].

The main goal of the experiments presented in this paper was to assess the feasibility of producing high-methane gas by hydrogasification of coal in the simulated underground conditions (via reaction R1). For this purpose, it was assumed that the gasification process would be two-stage. In the first stage, oxygen was used as the reagent, the main purpose of which

was to accumulate thermal energy for the subsequent hydrogasification reaction (in the next stage). This stage involved the highly exothermic combustion reactions:



In the next stage, only hydrogen was injected into the incandescent coal seam in order to carry out the hydrogasification process.

The experimental studies were conducted in a high-pressure ex situ laboratory installation. To determine the effect of pressure and coal rank on the hydrogasification process, the UCHG experiments were conducted at two pressure regimes - 20 and 40 bar with two coals of different ranks. To the best of our knowledge, there is no information on underground coal gasification with hydrogen in the available

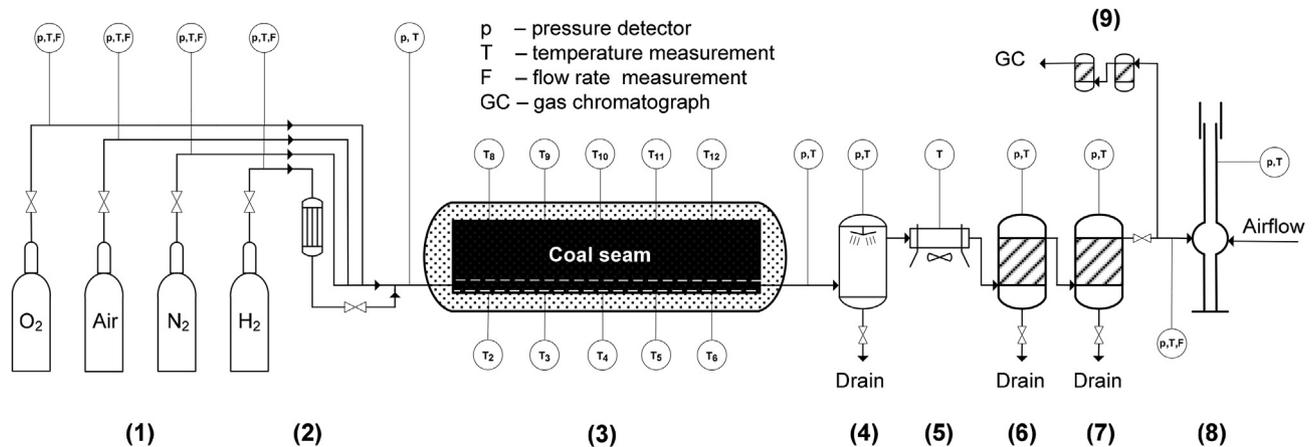


Fig. 1 – Scheme of the ex situ high pressure UCG installation: (1) compressed reagents, (2) hydrogen preheater, (3) gasification chamber, (4) wet scrubber for gas cleaning, (5) air cooler, (6,7) gas separators, (8) thermal combustor, (9) gas treatment module prior to GC analysis [23].

Table 1 – Proximate and ultimate characteristics of coals used for the gasification tests [23].

No.	Parameter	Coal sample		Standards
		“Six Feet” semi-anthracite	“Wesota” hard coal	
As received				
1	Total moisture, %	1.15 ± 0.40	3.60 ± 0.40	PN-G-04511:1980
2	Ash, %	4.61 ± 0.30	8.74 ± 40	PN-G-04560:1998
3	Volatiles, %	9.92 ± 0.12	27.67 ± 0.50	PN-ISO 1171:2002 PN-G-04516:1998
4	Total sulfur, %	1.55 ± 0.04	0.31 ± 0.02	PN ISO-562:2000 PN-G-04584:2001
5	Calorific value, kJ/kg	33,416 ± 220	28,798 ± 200	PN-ISO 334:1997
6	Specific gravity, g/cm ³	1.35 ± 0.028	1.40 ± 0.018	PN-G-04513:1981
Analytical				
7	Moisture, %	0.84 ± 0.30	2.18 ± 0.27	PN-G-04513:1981
8	Ash, %	4.62 ± 0.30	8.87 ± 0.63	PN-G-04560:1998
9	Volatiles, %	9.95 ± 0.13	28.08 ± 0.92	PN-ISO 1171:2002 PN-G-04516:1998
10	Heat of combustion, kJ/kg	34,414 ± 228	30,317 ± 161	PN ISO-562:2000
11	Calorific value, kJ/kg	33,527 ± 221	29,258 ± 201	PN-G-04513:1981
12	Roga index RI	1 ± 1	34 ± 4	PN-G-04518:1981
13	Total sulfur, %	1.55 ± 0.04	0.31 ± 0.08	PN-G-04584:2001
14	Carbon, %	87.31 ± 0.66	75.35 ± 1.13	PN-ISO 334:1997
15	Hydrogen, %	3.97 ± 0.28	4.61 ± 0.40	PN-G-04571:1998
16	Nitrogen, %	1.29 ± 0.12	1.20 ± 0.22	PN-G-04571:1998
17	Oxygen, %	0.50 ± 0.05	7.65 ± 0.1	from difference to 100%

literature. The research on the methane-oriented UCHG process can therefore be considered as innovative and unique.

Materials and methods

Description of the UCG installation used for the hydrogasification tests

The schematic view of the surface installation (ex situ) used for the hydrogasification experiments is presented in Fig. 1.

An essential part of the installation is a gasification chamber (3) where underground geological conditions of the coal seam are reproduced. The installation enables simulations of the underground coal gasification process on surface (ex situ), in an artificial coal seam with a maximum seam length of 3.05 m and a cross-sectional area of $0.41 \times 0.41 \text{ m}^2$. Gasification reagents can be supplied individually or in mixtures. To avoid contact with oxygen, hydrogen is supplied to the reactor through a separate line. This line is equipped with a preheater that heats the hydrogen up to $400 \text{ }^\circ\text{C}$ before its injection into the reactor, which reduces the rate of the temperature drop in the reactor due to the high specific heat of hydrogen ($14.3 \text{ kJ/kg}\cdot\text{K}$). Nitrogen is used as a safety agent for

inertizing and cooling down the reactor after gasification. The raw UCG-derived gas is subject to scrubbing with water to reduce its temperature, remove particulate matter and condense high boiling tar components. The subsequent gas treatment steps involve separation of aerosols. Produced gas is finally burned in the thermal combustor.

Coal samples and preparation of the artificial seam

Properties of coals used for the experiments

The coal block samples for the UCHG tests were sourced from two different mines. The first samples - "Six Feet" were obtained from the South Wales Coalfield, UK (semi-anthracite coal). The average thickness of the coal seam was 1.2 m and the sampling location was 88 m below the ground level. The second samples of coal were derived from the "Wesola" mine in the Upper Silesia Basin, Poland (bituminous coal). The sampling location was 950 m below the ground level from a coal seam of an average thickness of 5 m. The results of proximate and ultimate analyses of the tested coals are presented in Table 1. All analyses were performed in a certified laboratory at the Solid Fuels Quality Assessment Department of the Central Mining Institute (accreditation certificate ISO/IEC 17025).



a)



b)



c)



d)

Fig. 2 – Preparation of artificial seam for the high-pressure hydrogasification tests: a) sample cutting, b) prepared coal block, c) inlet cross-section, d) reactor closing.

The preparation of coal samples for the hydrogasification tests. The raw coal samples after initial processing were used to create a continuous artificial coal seam of a total length of 3.05 m, a width of 0.41 m and a thickness of 0.41 m. The procedure for the preparation of the coal seams for the tests is presented in Fig. 2. The cross-sections of the reactor for the UCHG tests are presented in Fig. 3.

The distribution of temperatures during the UCHG process was controlled by 10 high-temperature thermocouples (Pt10Rh–Pt). Fig. 3b shows that the thermocouples are located in the insulating layer of the reactor and do not reach the coal seam. The distance of the thermocouples from the bottom and roof of the artificial seam was about 2 cm. This placement of the thermocouples is necessary to protect them from direct contact with oxidizers.

The coal seams were ignited using a pyrotechnic charge located inside the gasification channel on the bottom of the coal seam at a distance of approx. 1 m from the face of the coal seam.

Experimental procedure

In order to simulate the coal hydrogasification process under underground conditions, a two-stage process for each coal type was used. In the first stage, oxygen was used as reagent with the main aim to accumulate thermal energy for the subsequent hydrogasification reaction in the second stage. In the next stage, only hydrogen was injected into the incandescent coal seam to carry out the hydrogasification process.

Between the two alternate stages, nitrogen was put into the experimental installation in order to remove the remaining oxygen and hydrogen from the system. The main aim of this inertization stage was to prevent formation of explosive gas mixtures. The experimental procedure used is shown in Fig. 4.

For each type of coal, the initial hydrogasification pressure was 20 bar. The first 24 h of the test were used for the heat-up process of the coal seam. The next stage of oxygen injection lasted 12 h. After reaching the required temperatures, the reaction zone was flushed with nitrogen (inertization) for 15 min. Subsequently, the hydrogen stage was started, which lasted for 12 h. After the hydrogen step, the reactor was re-flushed with nitrogen for 15 min. Then, after 4 oxygen and 4 hydrogen cycles, the reactor pressure was increased to 40 bar and the hydrogasification process was repeated. The full UCHG experiment consisted of 16 successive stages: 8 oxygen-blown and 8 hydrogen-blown, separated by inertization with nitrogen. The flow rate of oxygen and hydrogen in each cycle was 4 Nm³/h, and nitrogen during the inertization of the reactor was 3 Nm³/h (see Fig. 4).

Results and discussion

Gas production rates

The evolution of the UCG gas product over the course of the experiments with “Six Feet” and “Wesöta” samples is

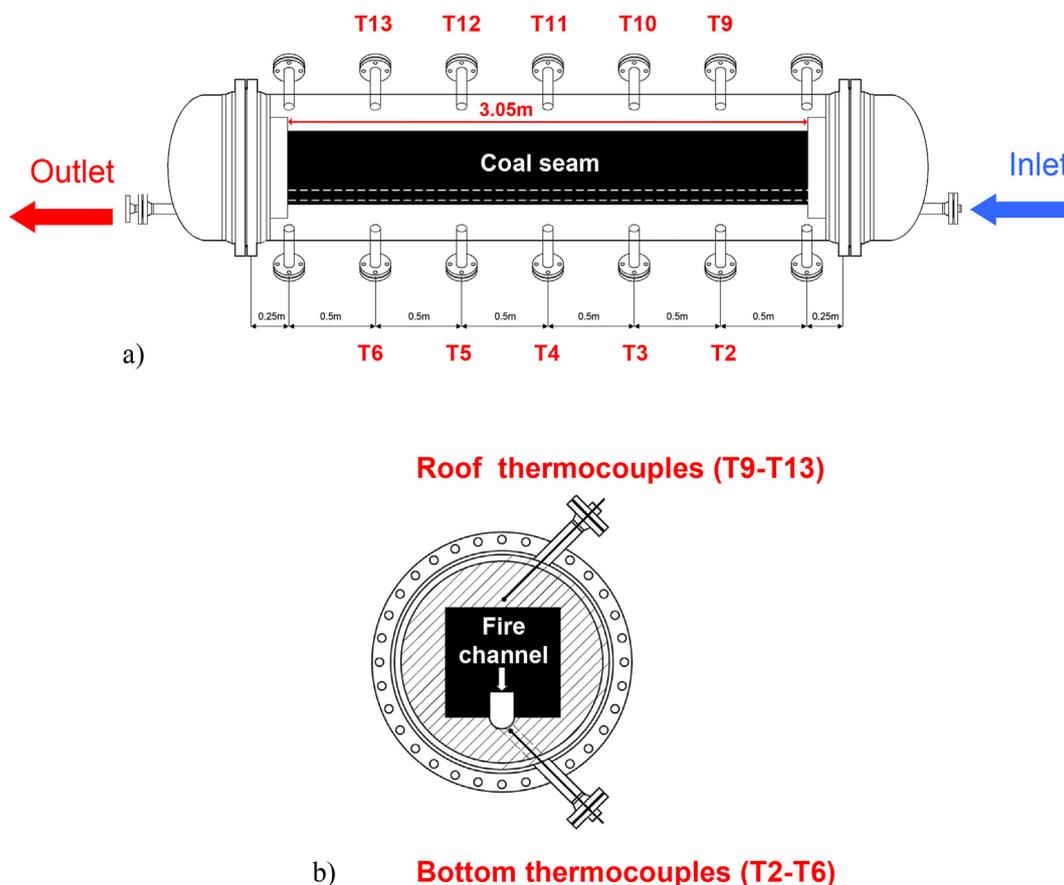


Fig. 3 – Cross-sections of the ex situ reactor prepared for the UCHG tests: a) side cross-section, b) vertical cross-section [23].

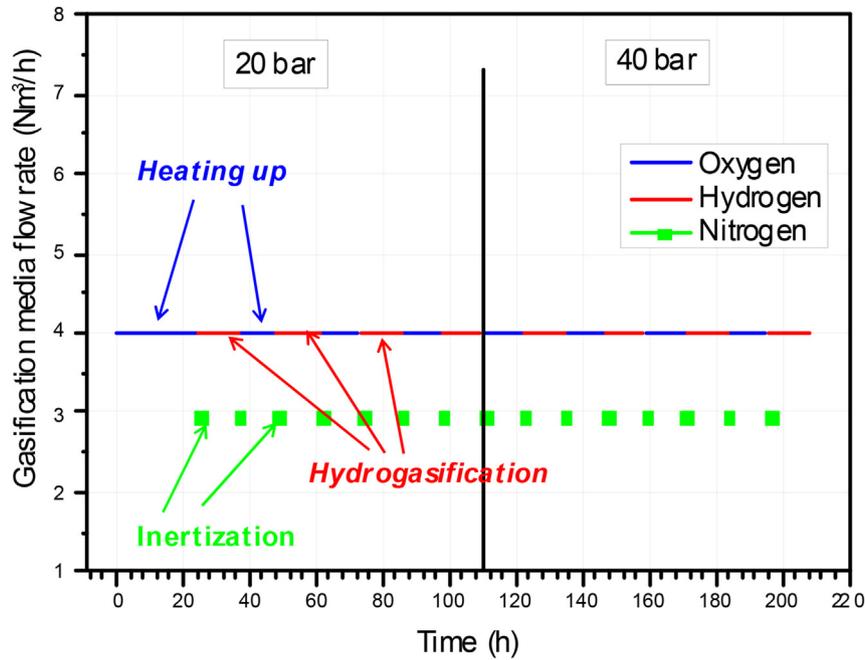


Fig. 4 – Experimental procedure for the hydrogasification tests.

presented in Fig. 5a and b, respectively. The relatively intensive gas production rate in the initial, oxygen-blown gasification stage during both experiments was mainly due to an intensive devolatilization (pyrolysis) of coal feed at the beginning of the process. As can be seen from the gas production graphs and from the data presented in Table 2, for

each of the hydrogasification experiments conducted, the maximum gas yields were obtained in the oxygen (heating-up) stages. The reasons for this are twofold. The first is the significant contribution of pyrolytic gas and CO_2 during the oxygen stage owing to relatively high temperature in the reactor. On the contrary, in the hydrogen stage, the observed decrease

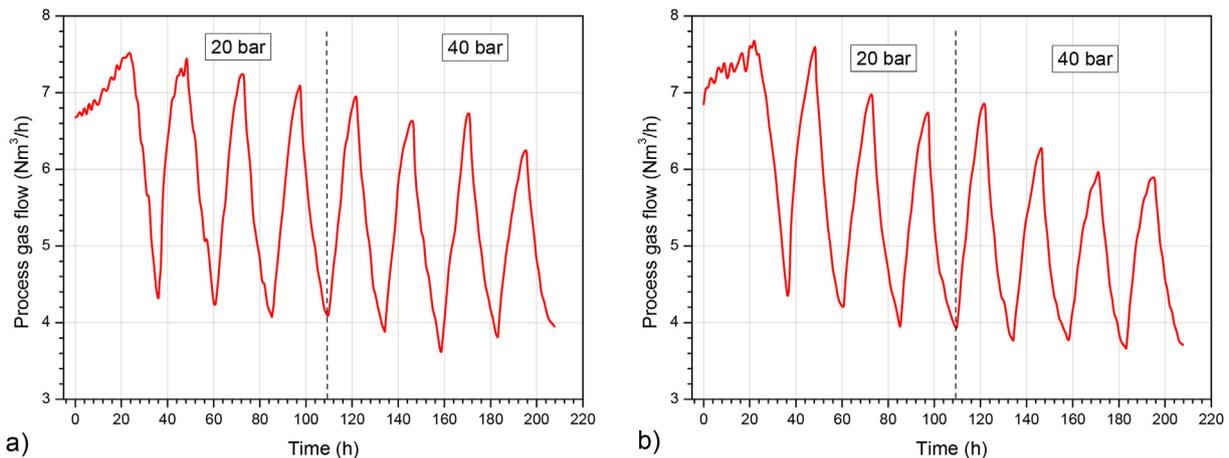


Fig. 5 – Gas production rates over the course of hydrogasification experiments: a) “Six Feet” semi-anthracite, b) “Wesota” hard coal.

Table 2 – Average gas production parameters for the UCHG experiments conducted.

Gas production parameter	“Six Feet” semi-anthracite				“Wesota” hard coal			
	20 bar		40 bar		20 bar		40 bar	
	O ₂ stage	H ₂ stage	O ₂ stage	H ₂ stage	O ₂ stage	H ₂ stage	O ₂ stage	H ₂ stage
Average gas production rate, Nm ³ /h	6.49	5.43	5.57	4.75	6.45	5.24	5.38	4.52
Gas yield, Nm ³ /kg of coal consumed	1.92	2.63	1.85	2.51	1.65	2.43	1.57	2.30

in the total gas volume was due to the reduction in the number of gas moles in the reaction of carbon with hydrogen. Additionally, during the hydrogen stage, pyrolysis was limited due to a significant temperature drop. The temperature drop during the hydrogen stage was most noticeable in the upper (roof strata) parts of the coal seam (Figs. 8 and 9).

As can be seen from Table 2, for both coals used, the gas production rates and gas yields per mass of gasified coal are significantly affected by the gasification pressure and the correlation is negative for both the oxygen and hydrogen stages. Higher gas yields from gasified coal mass during gasification of “Six Feet” anthracite resulted from more

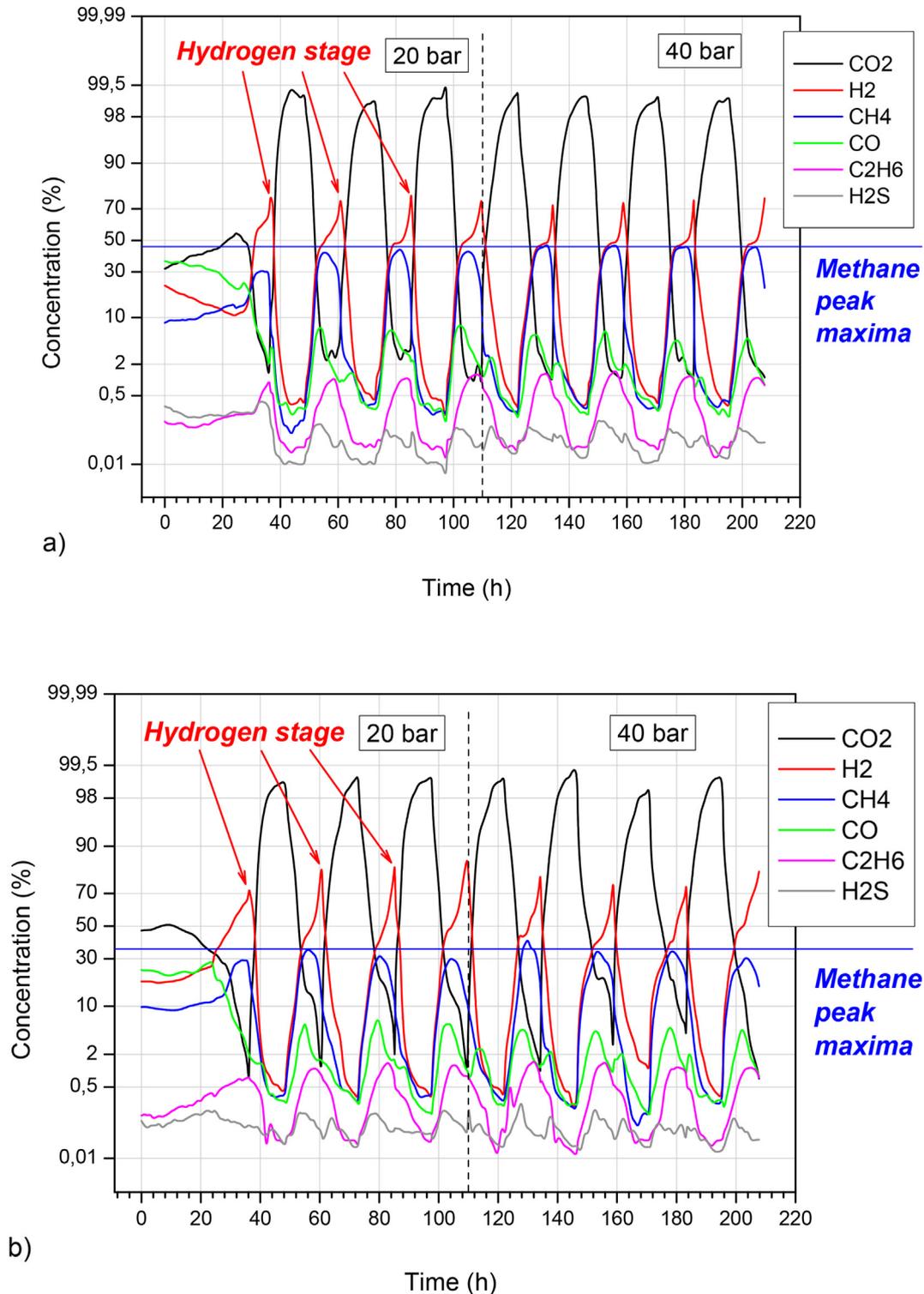


Fig. 6 – Changes in gas composition over the course of hydrogasification gasification experiments a) “Six Feet” semi-anthracite, b) “Wesoła” hard coal.

favorable gasification conditions due to higher calorific value and lower ash content in the gasified sample.

Product gas composition and gas calorific value

Changes in the product gas composition and gas calorific value for the experiments carried out are presented in Figs. 6 and 7. Average gas compositions obtained in the particular gasification experiments are presented in Table 3 and Table 4.

The calorific value of the produced gas was calculated by summing the molar fractions of the particular gas components multiplied by their calorific values according to the following equation:

$$Q = \sum x_i \cdot Q_i$$

where:

x_i - the mole fractions of particular components,

Q_i - heating values of the particular components (MJ/Nm³).

As can be seen from the data presented, the gas composition was significantly dependent on both coal properties and gasification pressure. For both 20 and 40 bar experiments, gas from “Six Feet” semi-anthracite was characterized by higher contents of methane. The average CH₄ concentrations during the “Six Feet” semi-anthracite experiment in the hydrogen stages were: 24.12% and 27.03% at 20 and 40 bar, respectively.

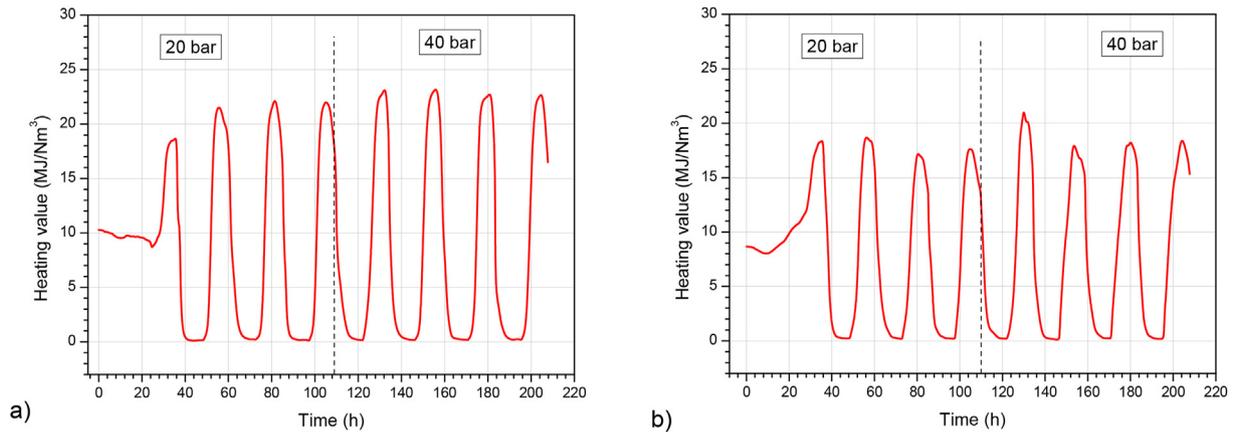


Fig. 7 – Changes in gas calorific value over the course of hydrogasification experiments: a) “Six Feet” semi-anthracite, b) “Wesola” hard coal.

Table 3 – Average gas yields and composition obtained during the “Six Feet” hydrogasification experiment.

Stage	Gas yield Nm ³	Gas production rate Nm ³ /h	Gas composition (vol. %)						Gas calorific value MJ/Nm ³
			CO ₂	H ₂	CH ₄	CO	C ₂ H ₆	H ₂ S	
20 bar gasification pressure									
Stage 1 O ₂	176.16	7.34	41.26	15.88	10.8	31.73	0.13	0.20	9.73
Stage 3 H ₂	70.92	5.91	26.28	37.98	22.47	12.71	0.40	0.16	14.06
Stage 5 O ₂	79.68	6.64	80.31	17.11	1.38	1.02	0.15	0.03	2.57
Stage 7 H ₂	66.96	5.58	33.41	38.72	24.54	2.78	0.49	0.06	13.66
Stage 9 O ₂	73.20	6.10	83.57	14.29	1.31	0.73	0.08	0.02	2.16
Stage 11 H ₂	61.68	5.14	32.17	39.01	24.59	3.62	0.56	0.05	13.86
Stage 13 O ₂	70.32	5.86	85.61	12.46	1.11	0.69	0.11	0.02	1.90
Stage 15 H ₂	61.20	5.10	31.36	39.34	24.87	3.70	0.68	0.05	14.08
Average for O₂ stages	99.84	6.49	72.69	14.94	3.65	8.54	0.12	0.07	4.09
Average for H₂ stages	65.19	5.43	30.81	38.76	24.12	5.70	0.53	0.08	13.92
40 bar gasification pressure									
Stage 17 O ₂	69.96	5.83	84.01	13.47	1.24	1.03	0.20	0.05	2.17
Stage 19 H ₂	59.16	4.93	34.27	35.02	27.29	2.74	0.62	0.06	14.32
Stage 21 O ₂	68.16	5.68	84.38	12.57	1.96	0.89	0.17	0.04	2.29
Stage 23 H ₂	56.52	4.71	34.49	34.86	27.38	2.51	0.68	0.08	14.35
Stage 25 O ₂	64.92	5.41	85.39	11.86	1.79	0.78	0.15	0.03	2.12
Stage 27 H ₂	56.40	4.70	31.45	37.93	27.64	2.31	0.62	0.05	14.70
Stage 29 O ₂	64.44	5.37	88.35	10.34	0.65	0.51	0.12	0.03	1.50
Stage 31 H ₂	55.92	4.66	35.63	35.79	25.79	2.17	0.56	0.06	13.76
Average for O₂ stages	66.87	5.57	85.53	12.06	1.41	0.80	0.16	0.04	2.02
Average for H₂ stages	57.00	4.75	33.96	35.90	27.03	2.43	0.62	0.06	14.28

Table 4 – Average gas yields and composition obtained during the “Wesola” hydrogasification experiment.

Stage	Gas yield Nm ³	Gas production rate Nm ³ /h	Gas composition (vol. %)						Gas calorific value MJ/Nm ³
			CO ₂	H ₂	CH ₄	CO	C ₂ H ₆	H ₂ S	
20 bar gasification pressure									
Stage 1 O ₂	183.12	7.63	45.50	20.68	9.81	23.72	0.19	0.10	8.89
Stage 3 H ₂	72.60	6.05	16.39	51.58	21.26	10.10	0.58	0.09	14.86
Stage 5 O ₂	77.88	6.49	83.23	12.47	3.22	0.81	0.22	0.05	2.75
Stage 7 H ₂	63.12	5.26	37.82	38.44	20.70	2.35	0.61	0.08	12.29
Stage 9 O ₂	71.40	5.95	84.86	12.26	1.97	0.66	0.20	0.05	2.25
Stage 11 H ₂	59.64	4.97	39.98	38.78	17.85	2.66	0.65	0.08	11.35
Stage 13 O ₂	68.88	5.74	85.59	11.94	1.62	0.62	0.18	0.05	2.07
Stage 15 H ₂	56.04	4.67	35.89	43.32	17.32	2.73	0.66	0.08	11.67
Average for O₂ stages	100.32	6.45	74.80	14.34	4.16	6.45	0.20	0.06	3.99
Average for H₂ stages	62.85	5.24	32.52	43.03	19.28	4.46	0.63	0.08	12.54
40 bar gasification pressure									
Stage 17 O ₂	68.64	5.72	84.00	12.63	1.94	1.14	0.23	0.06	2.36
Stage 19 H ₂	56.88	4.74	35.86	40.31	20.65	2.35	0.75	0.08	12.55
Stage 21 O ₂	63.84	5.32	85.31	11.78	1.74	0.93	0.19	0.05	2.14
Stage 23 H ₂	53.28	4.44	35.17	38.87	22.97	2.21	0.71	0.07	13.18
Stage 25 O ₂	62.88	5.24	85.79	11.54	1.58	0.87	0.17	0.05	2.04
Stage 27 H ₂	53.64	4.47	34.69	39.89	22.53	2.19	0.65	0.05	13.09
Stage 29 O ₂	62.76	5.23	87.33	10.61	1.21	0.66	0.15	0.04	1.77
Stage 31 H ₂	53.04	4.42	34.37	42.35	20.67	1.95	0.61	0.05	12.63
Average for O₂ stages	64.53	5.38	85.61	11.64	1.62	0.90	0.19	0.05	2.08
Average for H₂ stages	54.21	4.52	35.02	40.36	21.71	2.18	0.68	0.06	12.86

The average CH₄ concentrations during the “Wesola” coal tests in the hydrogen stages were: 19.28% and 21.71% at 20 and 40 bar, respectively. The maximum (peak) methane concentration in the product gas was 27.64% and it was achieved at 40 bar hydrogasification pressure for the “Six Feet” sample. The positive effect of gasification pressure on the methane yields was therefore evident.

The higher concentrations of CH₄ in gas produced during hydrogasification of “Six Feet” sample resulted in relatively higher gas calorific values, i.e. 13.9 and 14.3 MJ/Nm³ at 20 and 40 bar, respectively compared to 12.5 MJ/Nm³ at 20 bar and 12.9 MJ/Nm³ at 40 bar during gasification of “Wesola” hard coal.

The both processes were stable and conditions conducive for the methane formation were reproducible in each hydrogen stage.

Temperature distribution

Distributions of temperatures over the course of the experiments in the gasification channel and in the roof strata are presented in Figs. 8 and 9. The highest temperatures during the “Six Feet” semi-anthracite hydrogasification test were approximately 900 °C and were recorded in the roof of the seam during the oxygen-blown stage of the process (heating-up). The relatively high calorific value of the feed coal was the main reason of the higher gasification temperatures. The high temperatures consequently resulted in relatively high methane yields. As presented in Fig. 9, during the hard coal “Wesola” hydrogasification experiment, the recorded temperatures were lower, with the maximum value of approximately 800 °C. Another observation that may be drawn from the graphs is that temperatures in the bottom strata

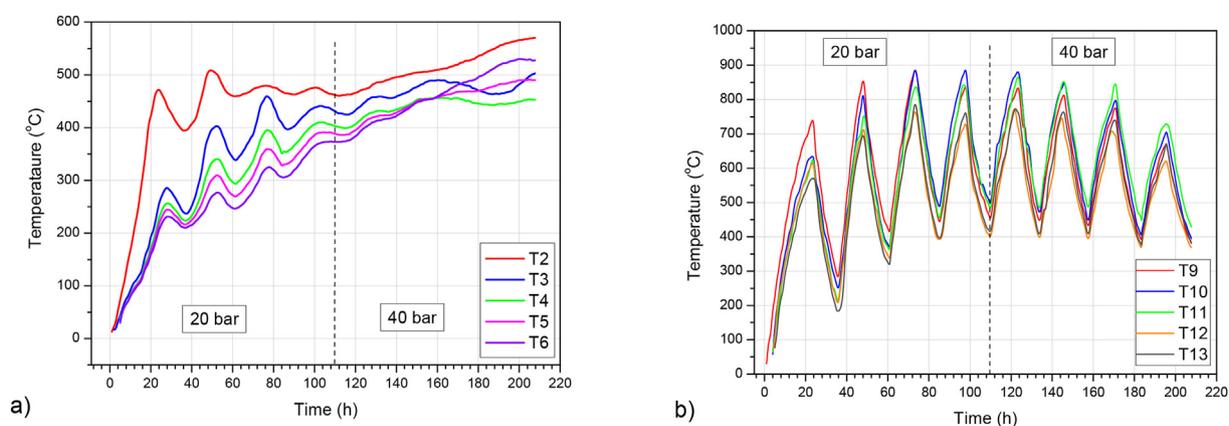


Fig. 8 – Distributions of temperatures during UCHG test with “Six-Feet” semi-anthracite: a) seam bottom, b) roof strata.

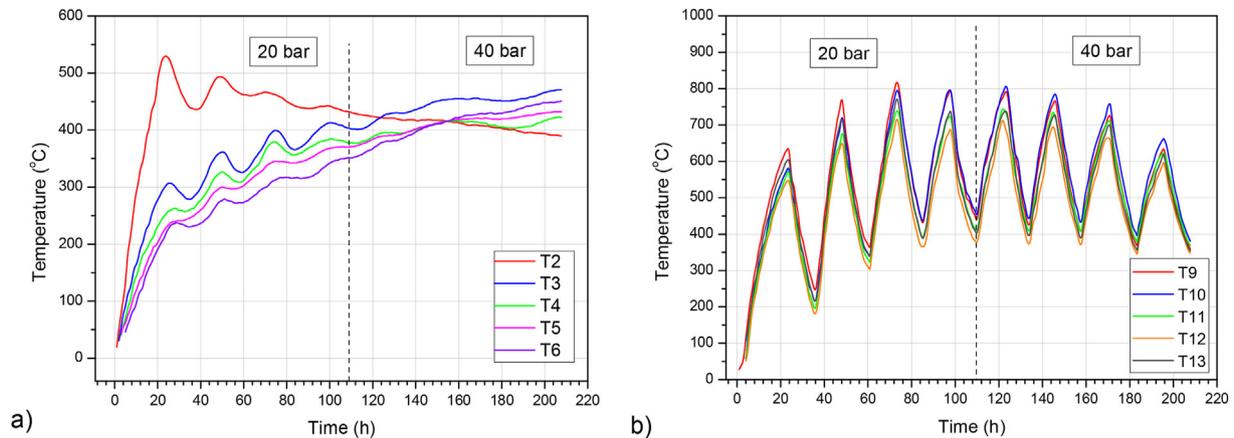


Fig. 9 – Distributions of temperatures during UCHG test with “Wesola” coal: a) seam bottom, b) roof strata.

Table 5 – Mass and energy balance calculations for the “Six Feet” semi-anthracite UCHG experiment.

Stage/reagent	Duration, h	Gas yield, Nm ³	Average gas production rate, Nm ³ /h	Energy in gas, MJ	Mass of coal gasified, kg	Gasification rate, kg/h	Energy in coal consumed, MJ
20 bar gasification pressure							
Total for O ₂ stages	60	399.4	6.5	2210.5	208.4	3.4	7050.9
Total for H ₂ stages	48	260.8	5.4	3628.4	99.1	2.1	2569.7
40 bar gasification pressure							
Total for O ₂ stages	48	267.5	5.6	542.2	144.9	3.0	4903.0
Total for H ₂ stages	48	228.0	4.8	3256.8	90.7	1.9	3070.2

Table 6 – Mass and energy balance calculations for the “Wesola” coal UCHG experiment.

Stage/reagent	Duration, h	Gas yield, Nm ³	Average gas production rate, Nm ³ /h	Energy in gas, MJ	Mass of coal gasified, kg	Gasification rate, kg/h	Energy in coal consumed, MJ
20 bar gasification pressure							
Total for O ₂ stages	60	401.3	6.5	2145.3	243.2	4.0	7003.8
Total for H ₂ stages	48	251.4	5.2	3185.5	103.6	2.2	2983.3
40 bar gasification pressure							
Total for O ₂ stages	48	258.1	5.4	538.0	164.7	3.4	4743.0
Total for H ₂ stages	48	216.8	4.5	2788.1	94.3	2.0	2714.2

(gasification channel) for each experiment were about 200–400 °C lower compared to the roof strata. This phenomenon confirms that the post gasification ash/slag and molten roof may effectively insulate against heat conduction to the bottom strata during the UCG operations. It should be emphasized, however, that the actual process temperatures were much higher, but due to the insulating phenomena (refractory materials and ceramic thermocouples casings used), the records had lower values.

Process balance data

The energy and mass balance calculations for the “Six Feet” and “Wesola” UCHG experiments are presented in Table 5 and Table 6, respectively. The energy in the process gas at each stage of the gasification was calculated by multiplying the gas yield by the corresponding average gas calorific value. The amount of carbon contained in the process gas was calculated

based on the gas composition and its yield. Then, the mass of coal gasified during the particular stages was calculated based on technical and elemental analysis of the raw coal.

The study revealed that at the same experimental conditions, hydrogasification of “Wesola” coal took place at much higher coal consumption rates, i.e. 4.0 kg/h compared to 3.4 kg/h at 20 bar and 3.4 kg/h compared to 3.0 kg/h at 40 bar for “Wesola” and “Six feet” coal, respectively. These differences can be explained by the higher reactivity of “Wesola” sample (lower rank coal).

Conclusions

The hydrogasification experiments conducted demonstrated a significant influence of coal properties and operational pressure on the main process parameters, including gas composition and methane formation, in particular:

- The UCHG gas composition was significantly dependent on both coal properties (coal rank) and gasification pressure. For both 20 and 40 bar gasification pressure, gas from "Six Feet" semi-anthracite was characterized by a higher content of methane. The average CH₄ concentration for "Six Feet" anthracite during the H₂ stage was 24.12% at 20 bar and 27.03% at 40 bar. During the hydrogasification of "Wesola" coal, the CH₄ concentration was 19.28% and 21.71% at 20 and 40 bar, respectively. The study revealed that the hydrogasification process was characterized by high stability and reproducibility of conditions favorable for methane formation in the whole sequence of the gasification cycles.
 - For both coals under study, the gas production rates and gas yields (volume of gas per mass of gasified coal) were significantly dependent on gasification pressure and the correlation was negative both for the O₂ and H₂ stage. The higher gas yields obtained during gasification of "Six Feet" coal, resulted from more favorable gasification conditions due to higher calorific value and lower ash content in the gasified sample.
 - The study revealed that at the same experimental conditions, hydrogasification of "Wesola" coal took place at much higher coal consumption rates, i.e. 4.0 kg/h compared to 3.4 kg/h at 20 bar and 3.4 kg/h compared to 3.0 kg/h at 40 bar for "Wesola" and "Six Feet" coal, respectively. These differences can be explained by the higher reactivity of "Wesola" sample (lower rank coal).
 - Because during UCG operational pressure increases with the coal seam depth (increase of the hydrostatic pressure available) the results obtained imply that the in-situ hydrogasification of coal (UCHG) is more effective at higher depths.
 - Although the feasibility of methane-rich gas production by underground hydrogasification was initially demonstrated, further techno-economic studies are necessary to assess the economic feasibility of producing methane through UCG using hydrogen as gasification reagent.
- Burton E, Friedmann J, Upadhye R. Best practices in underground gasification. Draft. US DOE contract no W-7405-Enfg-48. Livermore, CA, USA: Lawrence Livermore Laboratory; 2005.
 - Thorsness CB, Cena RJ. Underground coal gasification, data base. Lawrence Livermore National Laboratory Livermore, CA 94550.
 - Prog Energy Combust Sci 2018;67:158–87.
 - Perkins G. Underground coal gasification - Part II: fundamental phenomena and modeling. Prog Energy Combust Sci 2018;67:234–74.
 - Dvornikova EV. The role of groundwater as an important component in underground coal gasification. Underground Coal Gasification and Combustion 2018:253–81.
 - Shu X, Li J, Hao J, Liu Z, Wang Q, Lu X. Effect of atmosphere and temperature on syngas production during gasification of Zhundong lignite and water-washed Zhundong lignite in a fixed-bed reactor. Chem Paper 2020;74:555–69.
 - Surya Kanta D, Prabu. Experimental studies on humidified/water influx O₂ gasification for enhanced hydrogen production in the context of underground coal gasification. Int J Hydrogen Energy 2017;42:14089–102.
 - Su F, Hamanaka A, Itakura K, Degouchi G, Sato K, Kodama J. Evaluation of coal combustion zone and gas energy recovery for underground coal gasification (UCG) process. Energy Fuels 2017;31(1):154–69. 2017.
 - Wang ZT, Ding XP, Huo LW, Wang GX, Rudolph V. A remining technology of underground coal gasification at Zhongliangshan Coal Mine. J Coal Sci Eng 2008;14(3):469–73. 2008.
 - Wiatowski M, Stańczyk K, Świądrowski J, Kapusta K, Cybulski K, Krause E, et al. Semi-technical underground coal gasification (UCG) using the shaft method in, Experimental Mine "Barbara". Fuel 2012;99:170–9.
 - Fredersdorf CG, Elliot MA. Coal gasification. In: Lowry HH, editor. Chemistry of coal utilization. New York: Wiley J and Sons; 1963. p. 892–1022.
 - An analysis of coal hydrogasification processes. San Francisco, CA: Bechtel Corporation; 1978. Final Report by Dr. Michael Epstein, Dr. Tang-Ping Chen, Dr. Mohamed A. Ghaly. Prepared For The United States Department Of Energy Under Contract No. EF-77-A-01-2565.
 - Miller WR, Honea FI. Great plains coal gasification plant start-up and modification report. Fluor Technology Inc.; 1986. Technical report.
 - Roddy Dermot J. A syngas network for reducing industrial carbon footprint and energy use. Appl Therm Eng 2013;53:299–304.
 - Byron SRJ, Muruganandam L, Shekhar SM. A review of the water gas shift reaction Kinetics. Int J Chem React Eng 2010;8:1–34.
 - Takenaka S, Shimizu T, Otsuka K. Complete removal of carbon monoxide in hydrogen-rich gas stream through methanation over supported metal catalyst. Int J Hydrogen Energy 2004;29(10):1065–73.
 - Schaaf T, Grünig J, Schuster MR, Rothenfluh T, Orth A. Methanation of CO₂ – storage of renewable energy in a gas distribution system. Energy Sustain Soc 2014;4:2. <https://doi.org/10.1186/s13705-014-0029-1>.
 - Verostko C, Forsythe R. A study of the sebatier-methanation reaction. 1974. SAE Technical Paper 740933.
 - Wai SH, Ota Y, Nishioka K. Performance analysis of sabatier reaction on direct hydrogen inlet rates based on solar-to-gas conversion system. Int J Hydrogen Energy 46(2021);2681-26808
 - Minutillo M, Perna A. Renewable energy storage system via coal hydrogasification with co-production of electricity and synthetic natural gas. Int J Hydrogen Energy 2014;39:5793–803.

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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REFERENCES

- Gregg DW, Edgar TF. Underground coal gasification. AIChE J 1978;24:753–81.

- [22] Perkins G, Sahajwalla V. Steady-state model for estimating gas production from underground coal gasification. *Energy Fuel* 2008;22:3902–14.
- [23] Kapusta K, Wiatowski M, Stańczyk K, Zagorščak R, Thomas HR. Large-scale experimental investigations to evaluate the feasibility of producing methane-rich gas (SNG) through underground coal gasification process. Effect of coal rank and gasification pressure. *Energies* 2020;13:1334.
- [24] Burton E, Friedmann J, Upadhye R. Best practices in underground coal gasification. Livermore (CA): Lawrence Livermore National Laboratory; 2006. Contract W-7405-Eng-48 Contract No.: Contract W-7405-Eng-48].