

Article

Effect of Lignite Properties on Its Suitability for the Implementation of Underground Coal Gasification (UCG) in Selected Deposits

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Abstract: Two experimental simulations of underground coal gasification (UCG) processes, using large bulk samples of lignites, were conducted in a surface laboratory setup. Two different lignite samples were used for the oxygen-blown experiments, i.e., “Velenje” meta-lignite (Slovenia) and “Oltenia” ortho-lignite (Romania). The average moisture content of the samples was 31.6wt.% and 45.6wt.% for the Velenje and Oltenia samples, respectively. The main aim of the study was to assess the suitability of the tested lignites for the underground coal gasification process. The gas composition and its production rates, as well as the temperatures in the artificial seams, were continuously monitored during the experiments. The average calorific value of gas produced during the Velenje lignite experiment (6.4 MJ/Nm^3) was much higher compared to the result obtained for the experiment with Oltenia lignite (4.8 MJ/Nm^3). The Velenje lignite test was also characterized by significantly higher energy efficiency, i.e., 44.6%, compared to the gasification of Oltenia lignite (33.4%). The gasification experiments carried out showed that the physicochemical properties of the lignite used considerably affect the in situ gasification process. Research also indicates that UCG can be considered as a viable option for the extraction of lignite deposits; however, lignites with a lower moisture content and higher energy density are preferred, due to their much higher process efficiency.



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Keywords: lignite; underground coal gasification; UCG; ex situ tests

1. Introduction

Despite the current shift to renewable sources of energy, fossil fuels, and, in particular, coal, will remain a meaningful fuel in some parts of the world, for a long time into the future [1]. Increased energy demand will ultimately lead to the mining of deep coal deposits. As a consequence of the increasing depths, conventional underground mining is more difficult, more dangerous, and more expensive nowadays. Underground coal gasification is considered to be a technology that will enable the safe and economical exploitation of coal resources that could not otherwise be mined [2–4]. In recent decades, advances in directional drilling and monitoring technologies have changed the way that UCG can be conducted, at great depths. This, coupled with the current energy security issues and the need to reduce the environmental footprint, has sparked a global revival of interest in UCG. During UCG, coal deposits are transformed into gaseous combustible products (syngas) and extracted to the surface. The quality of the product gas depends, to a large extent, on the gasification medium used, operating conditions, coal rank, and local hydrogeology [5–10]. UCG can be a viable extraction technology for coal seams, for which conventional coal mining technologies are technically, economically, or environmentally not feasible [11]. Moreover, with the high-profile mine disasters, UCG is deemed safer than conventional mining, since it does not require staff below the ground.

Low-value coals, predominantly lignites, constitute a considerable proportion of global coal reserves [12]. The specific physicochemical properties of lignites result in many limitations in their extraction and further utilization. Lignites are characterized

by high humidity, up to 60 wt.%, and thus low calorific values [13]. The suitability of lignites for UCG is therefore questionable [14]. The high moisture content of typical lignite results in low thermal efficiency and poor quality of gas from UCG, because a significant amount of thermal energy is used to evaporate the water. Another challenge would be performing UCG at relatively shallow depths, which may negatively affect the groundwater quality [15–17]. Although UCG has been tested in many parts of the world, many technological issues still need to be solved before it is commercially applied.

The former UCG tests using lignite samples showed that the moisture content is one of the key parameters influencing the gasification performance, the quality of the gaseous products, and, finally, the suitability of lignite for UCG [18–21]. Lignite gasification using oxygen-enriched air (OEA) resulted in a low gas calorific value, 4.18 MJ/Nm³, for the experimentally optimized oxygen/air ratio [18]. It was experimentally proven that the consumption of heat for water evaporation leads to a very poor thermal efficiency (20%). Another study showed that lignite gasification under elevated pressure resulted in increased methane formation, with a lower H₂ and CO content in the UCG gas produced [20]. During a multiday UCG experimental simulation on lignite, conducted in a large-scale ex situ test facility, gas with an average calorific value of about 7.2 MJ/Nm³ was produced, with a process energy efficiency 59%. The results suggested that the crucial issue for the improvement of process efficiency in lignite seams may be an appropriate geometry of the underground reactor. Nevertheless, it was revealed that extraction of the high-moisture lignites using oxygen-blown UCG may be a feasible option [14]. This paper presents the results of laboratory experiments on the suitability of two European lignites for UCG. Two multiday UCG experimental simulations were conducted in a laboratory ex situ test stand, dedicated for experiments with large coal samples. Most of the experimental studies presented in the previous works were carried out for relatively small seam geometries. In the presented research, the volumes of the tested lignite samples significantly exceeded those used in the earlier studies. Such geometries allow for a more accurate estimation of the feasibility of the gasification process of the examined lignite deposits.

2. Materials and Methods

2.1. UCG Experimental Installation

The test facility used for the ex situ UCG tests is presented in Figures 1 and 2. The installation enables the simulation of the UCG process in laboratory conditions. The main section of the test facility is a gasification chamber, where in situ geological conditions are simulated both in respect to coal seam and surrounding strata. The maximum length of the artificial coal seam is about 7 m.

Oxygen, steam and air, supplied individually or in mixtures may be used as gasification reagents. N₂ is used as a safety and inertizing agent. The gas produced is water scrubbed to lower the temperature to remove particulates and gasification tar. Subsequent step involves the separation of aerosols. Gas is combusted in a thermal combustion. Concentrations of the main components are determined using chromatographic (GC) technique. The temperatures during the gasification tests are measured by 14 sensors (Pt10Rh-Pt) installed directly in various zones of the gasification chamber.

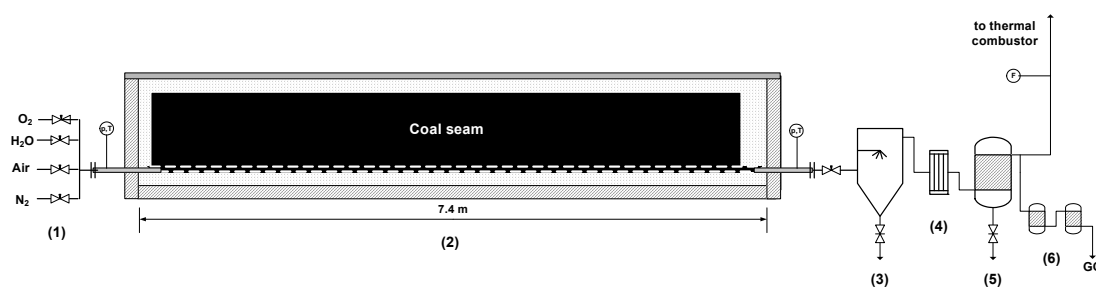


Figure 1. Scheme of the experimental installation: (1) supply of reagents, (2) gasification reactor, (3) scrubber, (4) cooler, (5) dust separator, (6) gas filters.



Figure 2. Ex situ UCG installation used in the study.

2.2. Lignite Samples and Creation the Coal Seams

The lignite samples for the gasification experiments were obtained from two different locations. The first selection of coal samples was gathered from an underground mine, Premogovnik Velenje, Slovenia. The sampling location was in the underground workings at a depth of approximately 350 m below the ground level (sample labelled as Velenje). The second selection of blocks was obtained from the Peșteana open cast mine (Complexul Energetic Oltenia Company), Romania. The sampling location was approximately 100 m below the ground level from a coal seam no. 8. This sample was labelled in the study as Oltenia. Physicochemical characteristics of the lignites used are showed in Table 1. Both the lignites are characterized by high moisture content. Oltenia sample is characterized by significantly higher moisture, ash and sulfur contents and lower content of volatile matter compared to lignite from Velenje. This resulted in considerably lower calorific value (as received basis). All analyses were performed by a certified laboratory (accreditation certificate according to ISO/IEC 17025).

The sulfur speciation studies revealed that in both lignites, pyritic sulfur is the main chemical form of sulfur (Figure 3). The Oltenia lignite is characterized by considerably higher content to total sulfur compared to the Velenje sample, i.e., 2.43% and 0.66%, respectively (analytical conditions). With respect to the UCG process, the ratio of ash sulfur-to-combustible sulfur is a crucial parameter governing partitioning of the total sulfur between post-gasification ash/slag left underground and product gas recovered on the surface. For the Oltenia lignite more than 90% of total sulfur occurs in combustible form. About 55% of total sulfur of Velenje lignite is supposed to remain as the post-gasification ash/slag.

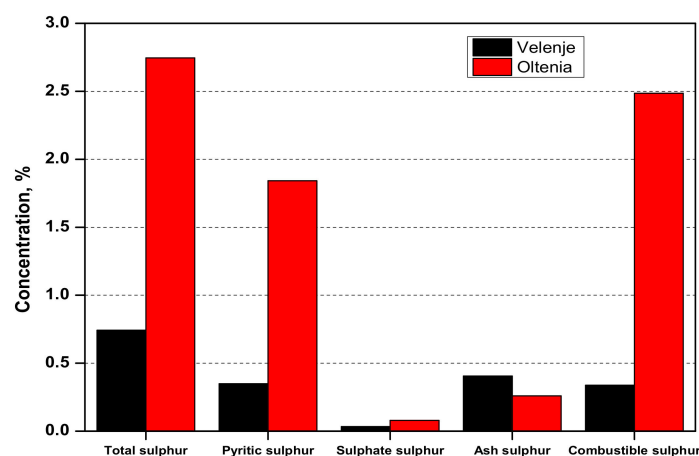


Figure 3. Sulfur forms in lignites used for ex situ gasification tests (dry basis).

Table 1. Physicochemical characteristics of lignites used for the ICG tests.

No.	Parameter	Lignite Sample	
		Velenje	Oltenia
	As received		
1	Total moisture W_t^r , %	31.62	45.64
2	Ash A_t^r , %	4.29	8.86
3	Volatiles V^r , %	43.67	25.78
4	Total sulfur S_t^r , %	0.51	1.49
5	Calorific value Q_i^r , kJ/kg	13,615	10,642
	Analytical		
6	Moisture W^a , %	11.13	11.49
7	Ash A^a , %	5.57	14.42
8	Volatiles V^a , %	56.76	41.98
9	Heat of combustion Q_s^a , kJ/kg	19,719	20,001
10	Calorific value Q_i^a , kJ/kg	18,427	18,860
11	Total sulfur S^a , %	0.66	2.43
12	Carbon C_t^a , %	49.86	49.49
13	Hydrogen H_t^a , %	4.67	3.94
14	Nitrogen N^a , %	0.64	1.34
15	Oxygen O_d^a , %	27.83	17.12

The ash fusion test describes the behavior of the ash residues at high temperature and it is a crucial factor in selecting solid fuels for energy use. The following ash parameters were determined for the two lignites under study:

- Sintering temperature;
- Softening temperature;
- Melting temperature;
- Flow (fluid) temperature.

The ash softening temperature is the temperature at which the ash softens beyond some arbitrary softness and the melting temperature is a measure of when the ash will melt and transform from solid to liquid. Since the UCG process has a zonal character (oxidation and reduction zones), the ash fusion temperatures are determined for both the oxidizing and reducing atmosphere. With regard to the UCG process, the key parameter is the ash flow temperature, at which slag begins to flow in the underground cavity/channels, which can cause clogging of the gas paths. Higher ash flow temperatures were observed for Velenje lignite, i.e., 1270 and 1300 °C, under oxidative and reducing conditions, respectively (Figure 4). Contrary to the Velenje sample, for Oltenia lignite, lower values of flow temperature were reported under reducing environment. This suggests that during UCG of Oltenia lignite, there may be a greater risk of channel clogging in the reduction (gasification) zone of the UCG reactor than in the oxidation zone. The observed significant differences in the sintering temperatures of both coal samples result from the different compositions of the mineral matter.

The raw samples provided by the industrial partners were used to prepare the continuous artificial coal seams of the total length of 6.0 m, width of 0.7 m and thickness of 0.7 m for experiments (Figure 5).

The gasification channel was prepared along the bottom part of the seams and its dimensions were 0.1 × 0.1 m. Sand was used to fill the voids between coal blocks and the reactor's walls as well as for the creation of the roof stratum. Fourteen temperature sensors were installed inside the gasification chamber to measure distributions of temperature during the UCG tests (Figure 5). The Nos. 1–4 were installed in the gasification channel, Nos. 5–14—inside the coal artificial seam.

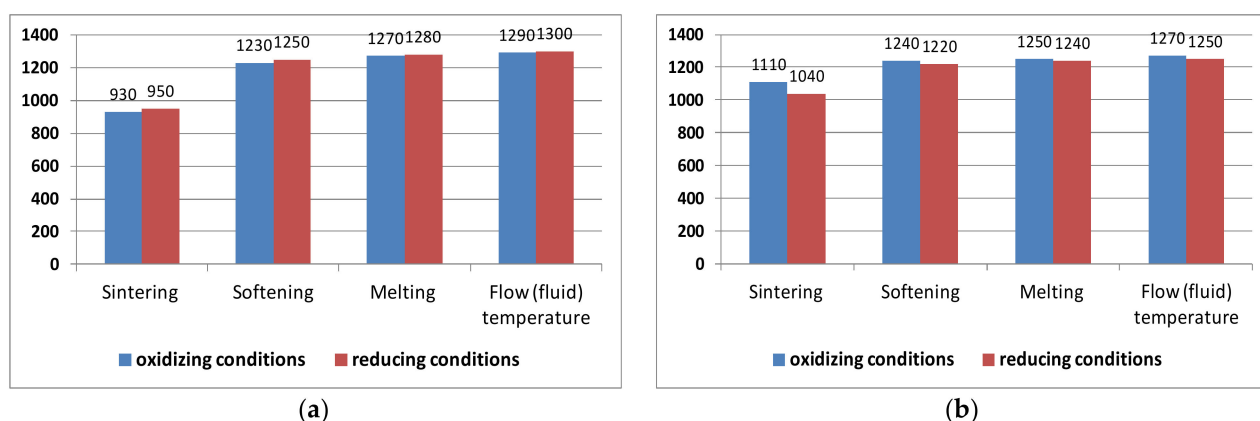


Figure 4. Ash fusion temperatures in oxidizing and reducing environments for lignites used for the UCG tests: (a) Velenje, (b) Oltenia.

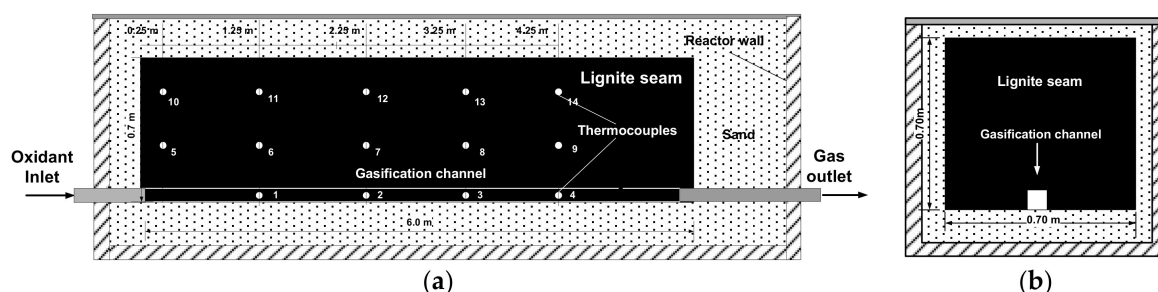


Figure 5. Sections of the artificial coal seam (proportions are not respected): (a) longitudinal (b) cross-section.

2.3. Gasification Test Procedure

The lignite samples were ignited using a pyrotechnic charge. The charge consisted of 800 g granulated pyrotechnic mass typically used in the mining industry. Appropriate modification of the composition allowed the achievement of an appropriately long combustion time (3–5 min) at high temperature (800–900 °C). The charge was placed 0.7 m from the front face of the seam. The pyrotechnic material was ignited by two fuses actuated by a capacitor electric igniter. The ignition was considered complete when the O₂ concentration in the outlet gas dropped below 1% vol. The gasification process was started by putting oxygen (99.5% purity) into the ignited coal seam. The initial oxygen supply rate was 2–3 m³/h and it was gradually increased over the course of the experiment, up to a maximum value of 5 m³/h in the final phases of the UCG tests. No additional water was supplied during the first test with Velenje lignite. In the second UCG trial with Oltenia lignite, a short test to investigate the influence of steam was carried out. The decision to add steam was made during the gasification experiment, taking into account the deteriorating quality of the gas produced and a rapid drop in the gas production rate (see Figure 6). Both UCG experiments were carried out under near-atmospheric pressure conditions. Concentrations of the gas components (H₂, CO, CH₄, CO₂, N₂, O₂, C₂H₆, H₂S) were analyzed in one hour time intervals. The UCG experiments lasted for 120 and 96 h for Velenje and Oltenia lignites, respectively.

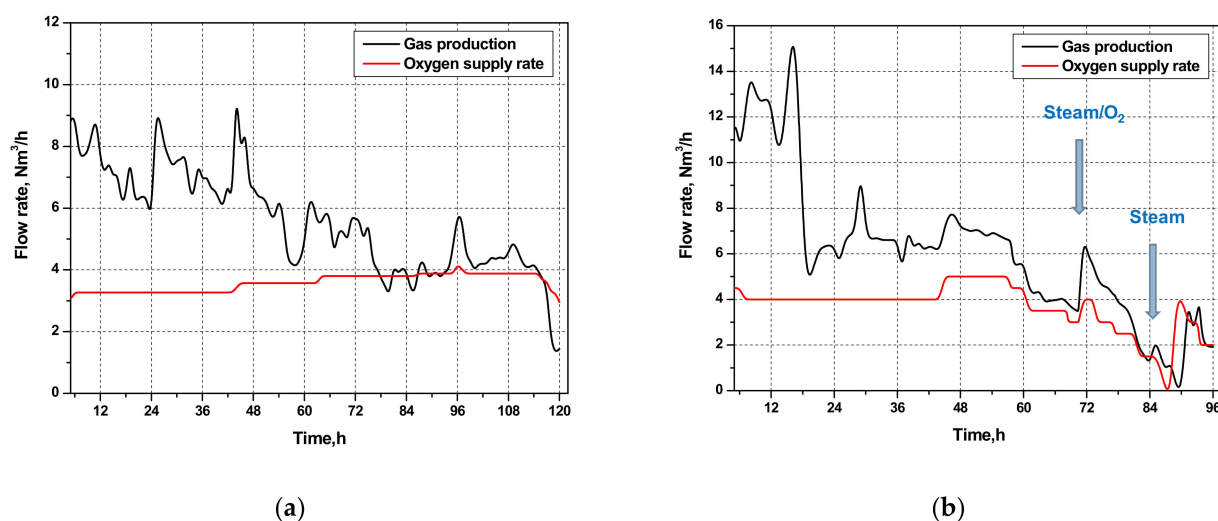


Figure 6. Gas production rates and oxygen supply rates during the UCG experiments: (a) Velenje lignite, (b) Oltenia lignite.

3. Results and Discussion

3.1. Gas Production Rate and Gas Composition

The evolution of UCG gas product during the experiments, and the oxidant supply rates (Nm^3/h) are presented in Figure 6. As can be observed from the graphs, the values of the production rates changed over the course of the experiments, with maximum values of about 9 Nm^3/h and 15 Nm^3/h for the Velenje and Oltenia experiments, respectively. The relatively intensive gas production rates in the initial gasification stages were mostly because of an intensive coal devolatilization (pyrolysis) at the beginning of the process. This intense gas evolution was especially evident in the Oltenia experiment. This can be explained by the specific physicochemical properties of the lignite sample. The Oltenia sample is classified as soft a ortho-lignite, which, compared to the Velenje meta-lignite, is geologically younger, contains more moisture, and has lower mechanical strength. Consequently, as a result of the intensive heating at the early stage of the experiment, the gasification and pyrolysis reactions were more intense compared to the Velenje experiment, mainly due to the mechanical disintegration of the seam and the rapid release of volatiles. This resulted in the relatively high gas production rates.

Changes in the UCG gas composition and in the gas calorific value for the experiments carried out are presented in Figures 7 and 8, respectively. As can be observed from the graphs, the initial gasification periods for both the experiments were characterized by a good-quality product gas, with a relatively high calorific value. In both the experiments, from about the 48th hour, a gradual decrease in the gas production rate was observed, with a significant deterioration in the gas quality, expressed in the content of combustible components (H_2 , CH_4 , CO) and its calorific value. The very high concentrations of CO_2 observed in the gas suggest that the combustion was the main chemical reaction when the gasification conditions deteriorated. This may be due to a limited availability of water for the gasification process. Consequently, during the Oltenia test, in the 70th hour of the experiment, steam was additionally supplied to the reactor, at a constant rate of 2 $\text{kg H}_2\text{O}/\text{h}$. As can be observed from Figure 7b, this resulted in a slight increase in the gas quality, which then gradually decreased. Although the gas quality improved again after increasing the oxygen supply rate at the 84th hour of the experiment, the effect was short-lived. Since the addition of water did not significantly improve the calorific value of the gas, the deterioration in the gasification conditions could be associated with a change in cavern geometry and disturbances in the gas flow, resulting in the combustion of the gasification products.

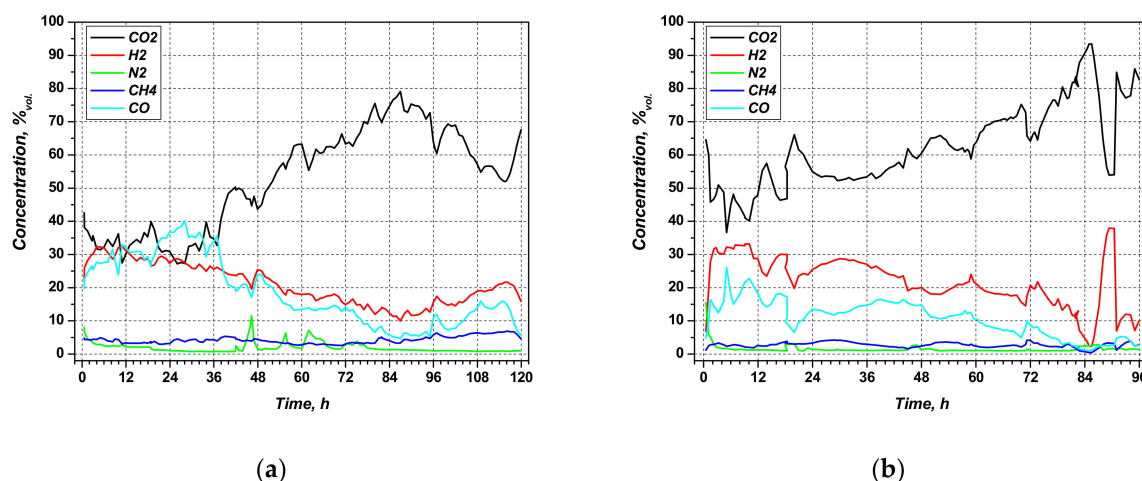


Figure 7. Changes in gas composition during the experiments: (a) Velenje lignite, (b) Oltenia lignite.

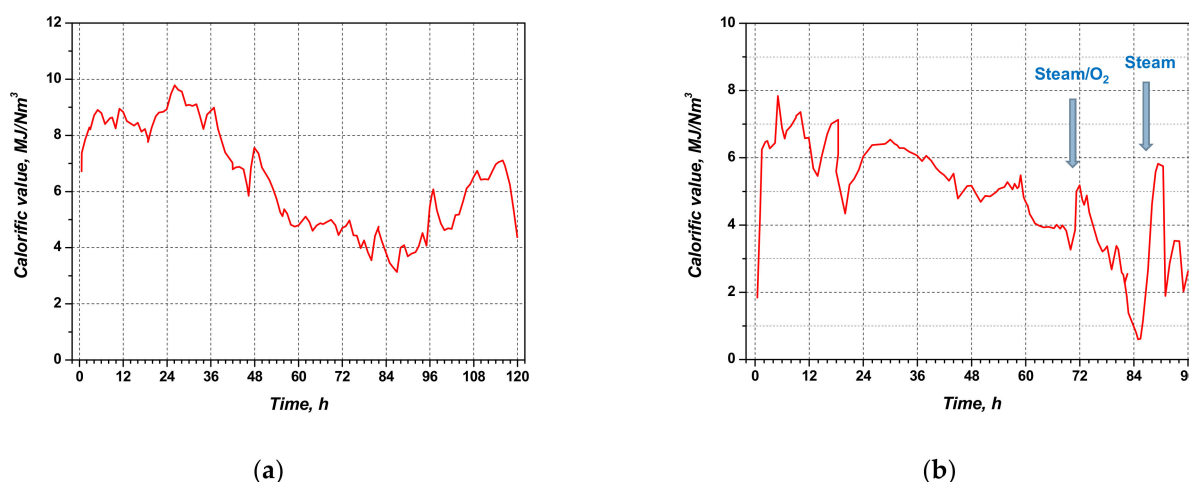


Figure 8. Changes in gas calorific value during the experiments: (a) Velenje lignite, (b) Oltenia lignite.

The average gas compositions for the experiments are presented in Table 2. The final gaseous products from both the experiments are characterized by relatively high hydrogen contents, i.e., 21.0% vol. and 21.3 % vol., for the experiments with Velenje and Oltenia lignite, respectively.

It was revealed that the Velenje gas product contained relatively high amounts of carbon monoxide compared to the product obtained during the gasification of Oltenia lignite. This may be due to the creation of more-favorable conditions for the Boudouard reaction, during the Velenje experiment, which resulted in a lower CO₂ concentration and higher calorific value of the gas. The average calorific value of gas, which was obtained for Velenje lignite - 6.4 MJ/Nm³, was comparable with the results of a previous UCG experiment with Polish lignite, which resulted in gas with an average calorific value of 7.2 MJ/Nm³ [14]. The UCG test was carried out in the same experimental installation and for a similar geometry of the artificial lignite seam. The differences in gas composition can be attributed to the coal properties, e.g., elemental composition, petrography, and ash mineralogy. The average calorific value of gas produced during the Oltenia lignite experiment (4.8 MJ/Nm³) was much lower compared to the result obtained for the experiment with Velenje lignite. This is mainly due to the higher moisture and ash content in the Oltenia lignite sample used for the test (Table 1). Therefore, the conducted study showed that the physicochemical properties of lignite strongly affect the chemical composition of the UCG gas produced.

Table 2. Average compositions of gas obtained in the UCG tests.

Lignite Sample	Gas Composition, %vol.								Q, MJ/Nm ³
	CO ₂	C ₂ H ₆	H ₂	O ₂	N ₂	CH ₄	CO	H ₂ S	
Velenje	52.5	0.2	21.0	1.0	2.0	4.3	18.6	0.5	6.4
Oltenia	63.3	0.2	21.3	0.2	1.5	2.7	10.2	0.6	4.8

3.2. Balance Calculations Results

The results of the balance calculations for the two gasification experiments conducted are showed in Table 3. Approximately 730 kg and 790 kg of raw lignite feed were consumed during the gasification tests with the Velenje and Oltenia samples, respectively. The remaining part the coal feed was left in the reactor as a gasification char, or unreacted, dried coal. This was confirmed by the post-gasification examination of the UCG cavity.

Table 3. Balance calculations for the Velenje and Oltenia atmospheric pressure experiments.

Parameter	Velenje	Oltenia
Total coal consumption (kg)	730	790
Average coal consumption rate (kg/h)	6.1	8.2
Average gas production rate (Nm ³ /h)	5.7	6.1
Average reactor power (kW)	10.3	8.1
Gross energy efficiency (%)	44.6	33.4

The study showed that at similar conditions, the gasification of Oltenia lignite took place with a significantly higher coal consumption rate, i.e., 8.2 kg/h compared to 6.1 kg/h for Velenje lignite. These differences can be explained by the different reactivities of the samples used. According to the energy balance calculations, the Velenje process was characterized by a much higher energy efficiency, i.e., 44.6% (calculated as a ratio of the energy in coal-to-the energy output in gas), compared to the gasification of Oltenia lignite (33.4%). The lower energy efficiency obtained for the Oltenia experiment was due to the higher moisture content of the raw lignite sample. The over-stoichiometric moisture content in the lignite sample eliminates the need to add additional reagent water, at least in the early stage of the gasification. However, excess water may result in a poor gasification efficiency, due to significant heat losses for the evaporation of coal moisture. The obtained results suggest that the appropriate length of gasification channels may be a key issue in improving the efficiency of the process in lignite seams. Recovering the heat of the gas at the surface is an option that can be considered to improve the energy efficiency of the process. This issue may be particularly important in the case of Oltenia lignite, the gasification of which resulted in a significantly lower energy efficiency.

The maximum energy efficiency of approximately 45%, obtained during the gasification of the Velenje lignite sample, was still much less than the values that are typical for UCG of hard coals, i.e., 60–70%.

3.3. Temperature Distribution

The distributions of temperatures over the course of the experiment in the gasification channel, and at two heights from the bottom of the seam are presented in Figures 9 and 10.

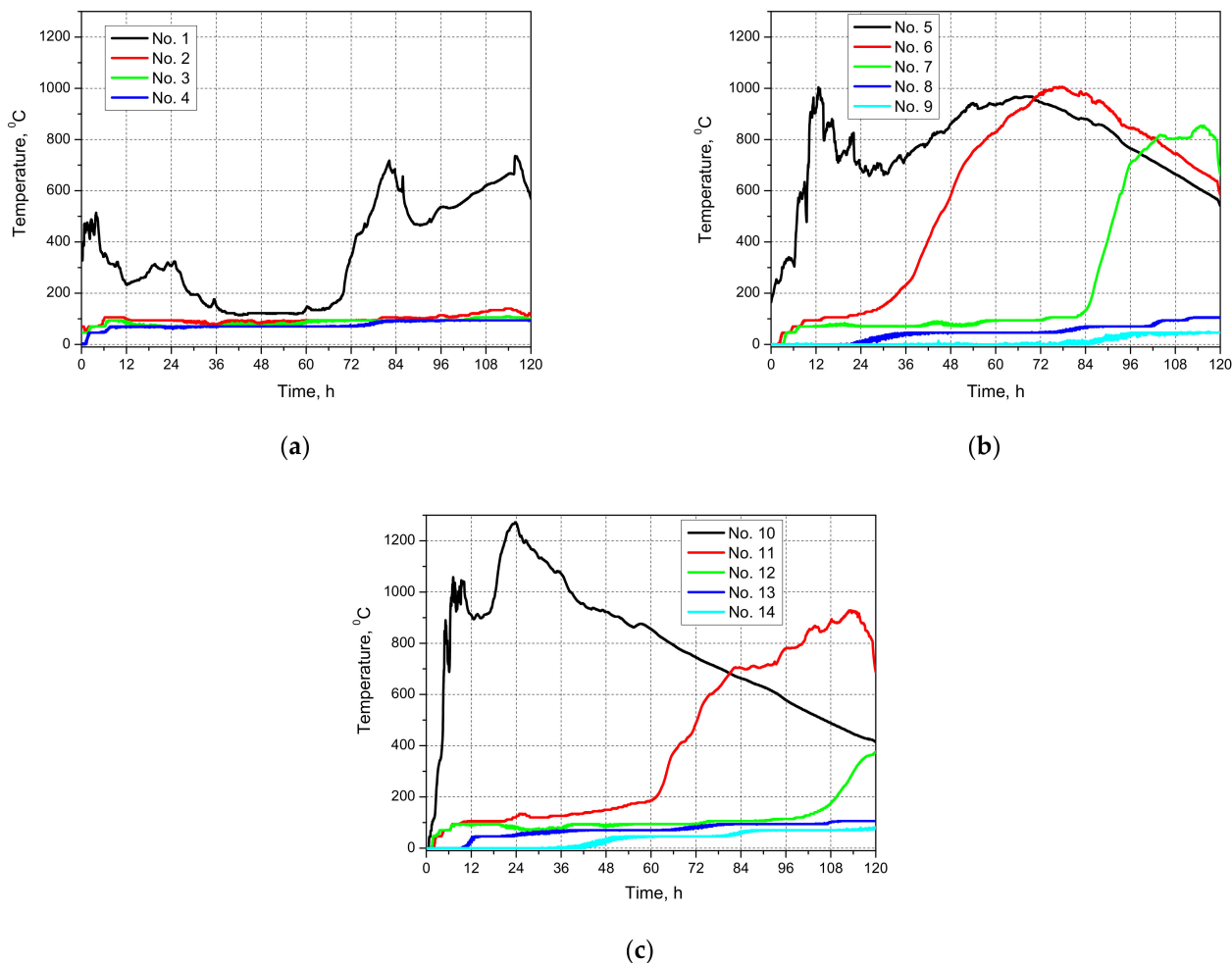


Figure 9. Temperature distribution during the Velenje gasification experiment: (a) gasification channel, (b) 0.3 m above bottom, (c) 0.6 m above bottom.

The maximum temperature during the Velenje test was approximately 1300 °C, and it was recorded at 0.3 m above the gasification channel, after approx. 24 h of the gasification run. The analysis of the temperatures indicates that during the whole course of the experiment, thermal conditions promoting the water gas reaction, as well as the Boudouard reaction (>750 °C), were achieved. Another observation that may be drawn from Figure 9 is that the temperatures in the bottom strata (gasification channel) were about 200 to 400 °C lower than the temperatures in the upper levels of the seam. This observation confirms that the gasification residues (ash and slag) may effectively insulate against heat conduction to the bottom strata, during the UCG process. For the Oltenia experiment, the maximum gasification temperature was about 1380 °C, and it was detected at 0.3 m above the gasification channel, at the early stage of the process (Figure 10). Similarly to the Velenje UCG experiment, during the whole course of the experiment, thermodynamic conditions promoting the water gas and Boudouard reactions were achieved.

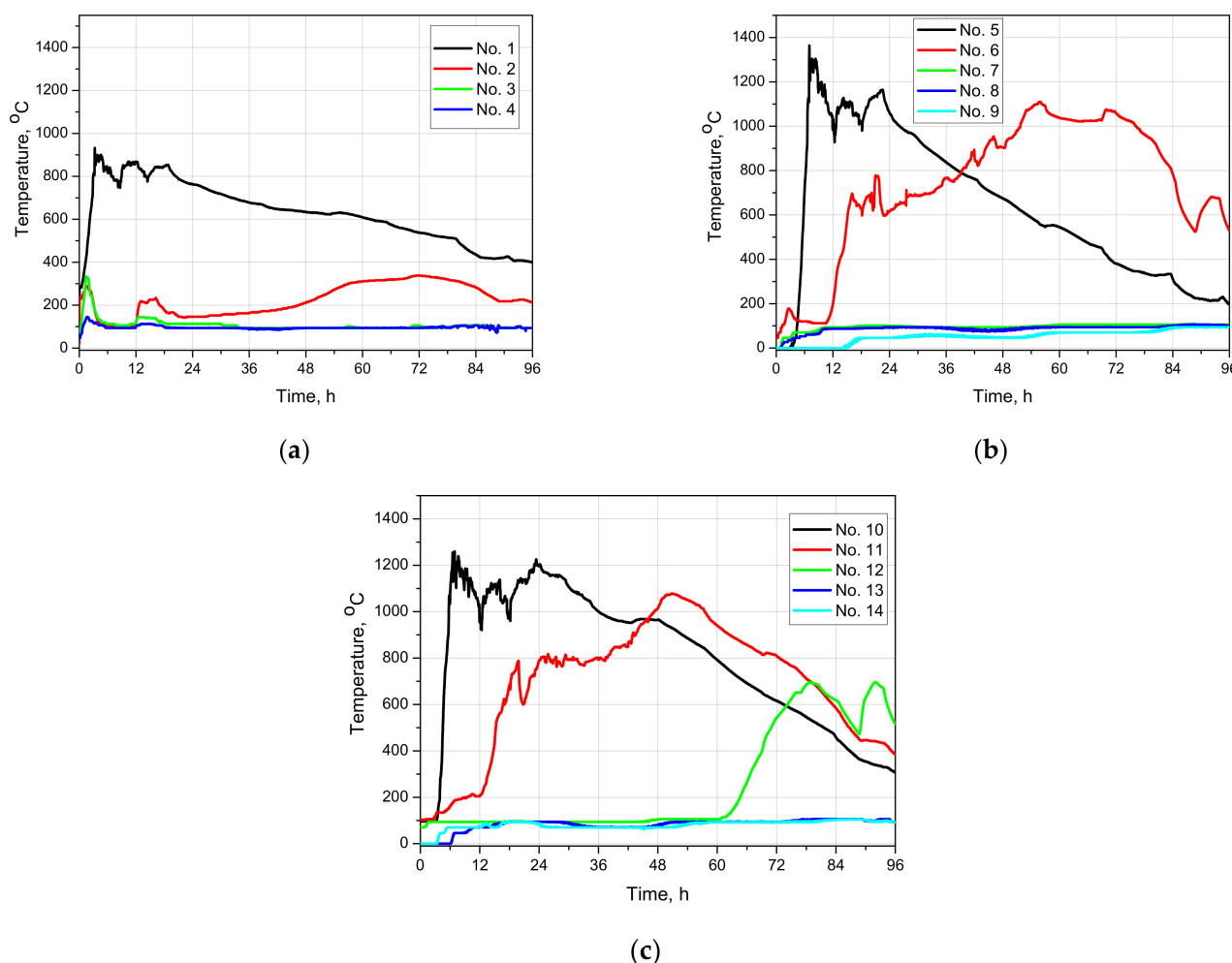


Figure 10. Temperature distribution during the Oltenia gasification experiment: (a) gasification channel, (b) 0.3 m above bottom, (c) 0.6 m above bottom.

4. Conclusions

The gasification tests that were conducted demonstrated that the physicochemical properties of the lignite used considerably affect the in situ gasification process. For similar process conditions (coal seam dimensions, oxygen supply rates), the gasification of Velenje lignite resulted in considerably better gas quality and process efficiency. The overall energy efficiencies, expressed as a ratio of energy in the obtained gas-to-the energy in the coal feed consumed, in the Velenje experiment, were significantly higher compared to the Oltenia test. The main reason behind this is the over-stoichiometric moisture content in the Oltenia lignite sample. The results of the study indicate that underground gasification may be a feasible option for the extraction of lignite deposits, especially in the case of Velenje lignite, which was characterized by a relatively higher calorific value, and lower moisture and ash content.

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Conflicts of Interest: The author declares no conflict of interest.

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