

Article

Utilization of Carbon Dioxide in Coal Gasification—An Experimental Study

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Received: 20 November 2018; Accepted: 27 December 2018; Published: 1 January 2019



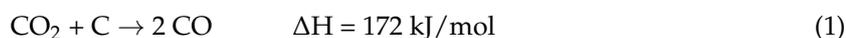
Abstract: Utilization of coal in the current energy sector requires implementation of highly-efficient technologies to meet the dual targets of increased energy-efficiency and reduced carbon footprint. Efforts are being made to develop gasification systems with lower unit emissions of carbon dioxide and other contaminants, capable of handling various feedstocks and flexible in terms of products generated (synthesis gas, hydrogen, heat and electricity). The utilization of captured carbon dioxide and waste heat in industrial processes are considered to further contribute to the advancements in energy-efficient and low-emission technological solutions. This paper presents the experimental results on the incorporation of carbon dioxide into the valorization cycle as a reactant in coal gasification. Tests were performed on a laboratory scale moving bed gasifier using three system configurations with various simulated waste heat utilization scenarios. The temperature range covered 700, 800 and 900 °C and the gasification agents used were carbon dioxide, oxygen and the mixture of 30 vol.% carbon dioxide in oxygen. The combined effect of the process parameters applied on the efficiency of coal processing in terms of the gas yields, composition and calorific value was studied and the experimental data were explored using Principal Component Analysis.

Keywords: carbon dioxide; utilization; carbon capture and utilization (CCU); carbon capture and storage (CCS); gasification

1. Introduction

The leading role of coal in the world energy resources balance stems from its high reserves to production ratio, which doubles the respective reported values for crude oil and natural gas, as well as world-wide availability [1]. Notwithstanding the strong pressure on EU's countries to make their economies more energy-efficient, competitive and zero-emission [2–4], the projected world coal production is still increasing by approximately 3%, and coal consumption is expected to remain at a level of approximately 190 quadrillion Btu during the 2015–2040 period, while the share of coal in the world electricity generation is expected to decline moderately, from 40% in 2015 to 31% in 2040, in the 25-years prognosis [5]. At the same time, the estimated world coal-related carbon dioxide emissions from the energy sector will increase 0.1%/year between 2015 and 2040, while liquid- and natural gas-related emissions are expected to be reduced by 0.7 and 1.4%/year, respectively [5]. The carbon dioxide emission reduction targets of the coal-based energy sectors are to be reached with the development and implementation of clean coal technologies, which include advanced gasification systems, as well as carbon capture storage and utilization techniques. The gasification technologies have been developed and implemented for several decades with entrained flow, fluidized bed and

moving bed reactors, and coal as the major feedstock [6]. The main challenges addressed today in terms of their advancement are highly efficient cogeneration systems (integrated gasification combined cycles) with carbon dioxide separation, as well as adaptation of gasifiers to alternative fuels, like biomass or industrial waste [7–10]. Co-gasification of coal with waste biomass is also considered to give the benefits of lowered carbon footprint, and potential synergy effects in terms of process efficiency and/or product quality [11,12]. The carbon capture and storage (CCS) technology chains still require advancements in terms of cost reduction, increased efficiency, environmental safety and social acceptance [13–16]. Efforts are also being made to develop and demonstrate technologies for the efficient utilization of the captured carbon dioxide (carbon capture and utilization, CCU) delaying the carbon emissions to the atmosphere, and making possible more sustainable management of natural resources, even though the market for captured carbon dioxide is quite limited compared to the anthropogenic emission potential [17]. The most viable CCU technological options considered today include the production of chemicals and fuels, biofuels from microalgae and mineral carbonation, with the latter one representing the actual carbon dioxide climate mitigation potential [18–20]. The main chemicals and fuels produced from carbon dioxide are urea, various polymers, synthetic natural gas, methanol, dimethyl-ether and oxymethylene ethers. Carbon dioxide may be also converted into the fuel gas, carbon monoxide, by the Boudouard reaction:



In this way the undesired product of thermochemical conversion of coal may be incorporated into the valorization cycle as a reactant in a highly-efficient and low-emission gasification technology. The gasification of chars of carbonaceous materials, including coal, biomass and waste with carbon dioxide has been tested in terms of the effects of various variables on char reactivity [21,22] as well as process kinetics and thermodynamics [23–26]. These include the properties of the feed material, the process temperature, pressure, char particle size and porous structure properties as well as the use of catalysts [27–30].

Another aspect of a more sustainable and energy-efficient system is the waste heat recovery from various industrial processes found in metallurgy, ceramic, food industry [31,32] or from high-temperature nuclear reactors [33–35]. The application of a high temperature waste heat in the highly endothermic gasification of coal with carbon dioxide as a gasification agent would make the system even more advantageous in terms of mitigating the greenhouse gas emissions and increasing the energy efficiency [36].

Therefore, within the experimental study presented in this paper, gasification of a bituminous coal with the use of carbon dioxide as a gasification agent, and the simulated process waste heat as an external, thermal-driven heat source for the endothermal reactions was performed in a moving bed gasifier. The process temperature applied was 700, 800 or 900 °C. The gasification agent used was pure carbon dioxide, or 30 vol.% carbon dioxide in oxygen or pure oxygen, for comparison of the effects of their various oxidizing potentials on the process performance under the experimental conditions adopted. The combined effects of gasification agent composition, process temperature and configuration of the waste heat utilization system on the process efficiency in terms of product gas composition, yield and calorific value were assessed with the application of Principal Component Analysis.

2. Materials and Methods

2.1. Experimental Procedure

The study on gasification of bituminous coal chars with carbon dioxide, 30 vol.% carbon dioxide in oxygen or oxygen was performed under the atmospheric pressure and at the temperature of 700, 800 or 900 °C. A laboratory scale installation with a moving bed reactor and an auxiliary gasification agent pre-heating system, simulating the waste heat recovery, was employed (see Figure 1). The working volume of the batch gasifier is 0.8 L. The gasifier and the gasification agents pre-heating unit are

heated with computer-controlled electric resistance furnaces. The process temperature is monitored with thermocouples and controlled with temperature controllers. Further details on the experimental stand may be found in [37]. Coal samples of 3 g (grain size below 0.2 mm) were heated in the nitrogen atmosphere to the set process temperature. Next, the gasification agent was injected into the reactor with a flow rate of 1.17 cm³/s, in the following three system configurations. In system I, the gasification zone was heated with a resistance furnace through the entire trial, and no preheating of gasification agents was applied. In system II, the gasification agents were heated to the process temperature with the simulated waste process heat, and the gasifier with the use of the resistance furnace; heating of the reactor was stopped once the set process temperature was reached. In system III, both the gasification agent and gasifier were preheated to the process temperature and the temperature was maintained during the process with the use of the resistance furnace as the source of the external heat. The product gas was treated in a water trap and filtered before its yield and composition were analyzed on-line with the application of a mass flowmeter and an Agilent 3000A gas chromatograph (Agilent Technologies Inc., Santa Clara, CA, USA), respectively.

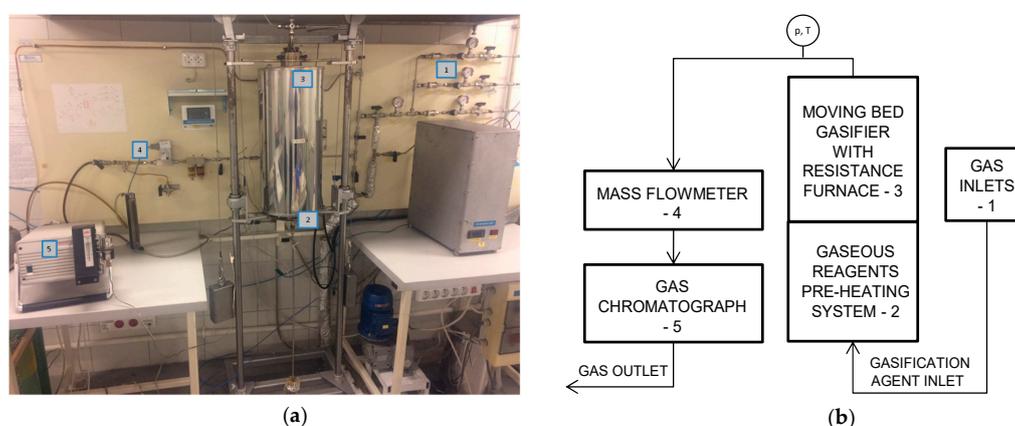


Figure 1. Laboratory scale installation with a moving bed reactor coupled with a gasification agent pre-heating system: (a) view and (b) schematic diagram.

2.2. Materials

Bituminous coal was provided by a coal mine located in the Upper Silesia Coal Basin (Poland). Coal was sampled and pre-treated according to the relevant standard [38] and characterized in terms of moisture, ash and volatiles contents [39], heat of combustion and calorific value [40], ash fusion temperatures [41], sulfur content [42], as well as carbon, hydrogen and nitrogen contents [43] (see Table 1).

Table 1. Analytical properties of the tested coal.

| No | Parameter, unit | Value |
|----|---------------------------|--------|
| 1 | Moisture, %w/w | 7.4 |
| 2 | Ash, %w/w | 7.2 |
| 3 | Volatiles, %w/w | 32.4 |
| 4 | Heat of combustion, kJ/kg | 27,815 |
| 5 | Calorific value, kJ/kg | 26,626 |
| 6 | Sintering point, °C | 940 |
| 7 | Softening point, °C | 1280 |
| 8 | Melting point, °C | 1360 |
| 9 | Flow temperature, °C | 1430 |
| 10 | Sulfur, %w/w | 1.9 |
| 11 | Carbon, %w/w | 67.4 |
| 12 | Hydrogen, %w/w | 4.1 |
| 13 | Nitrogen, %w/w | 0.9 |

2.3. Data Analysis

The complex effects of the application of the gasification agents of various carbon dioxide content, various process temperatures and waste process heat recovery configurations were analyzed with the use of Principal Component Analysis (PCA) [12,44–46]. This method enables effective reduction of data dimensionality, its visualization and interpretation. In PCA the original experimental data matrix $\mathbf{X}(m \times n)$ is decomposed into two matrices, called score matrix $\mathbf{S}(m \times f)$ and loading matrix $\mathbf{D}(f \times n)$, with m , and n denoting number of objects and variables, respectively and f denoting number of significant factors (principal components—PCs). Columns of matrix \mathbf{S} , and rows of matrix \mathbf{D} (PCs) are built as a linear combination of original variables with the weights maximizing the description of the data variance.

3. Results and Discussion

The combined effect of the simulated waste heat utilization, process temperature and gasification agent composition on the efficiency of coal processing in terms of the total gas yields, gas composition and calorific value of produced gas was studied. The average total gas yields reported for coal gasification with various gasification agents and in different heating system configurations at 700, 800 and 900 °C are presented in Figures 2–4.

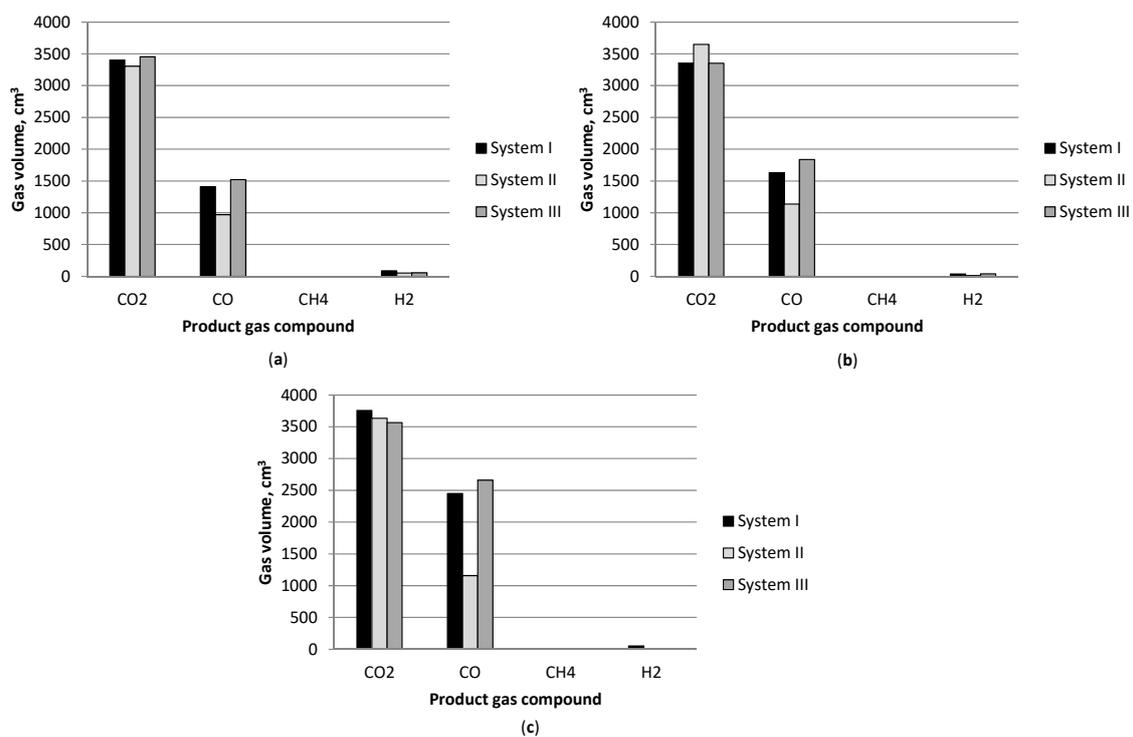


Figure 2. Average total gas yield in coal gasification with carbon dioxide at: (a) 700 °C, (b) 800 °C and (c) 900 °C in system I–III.

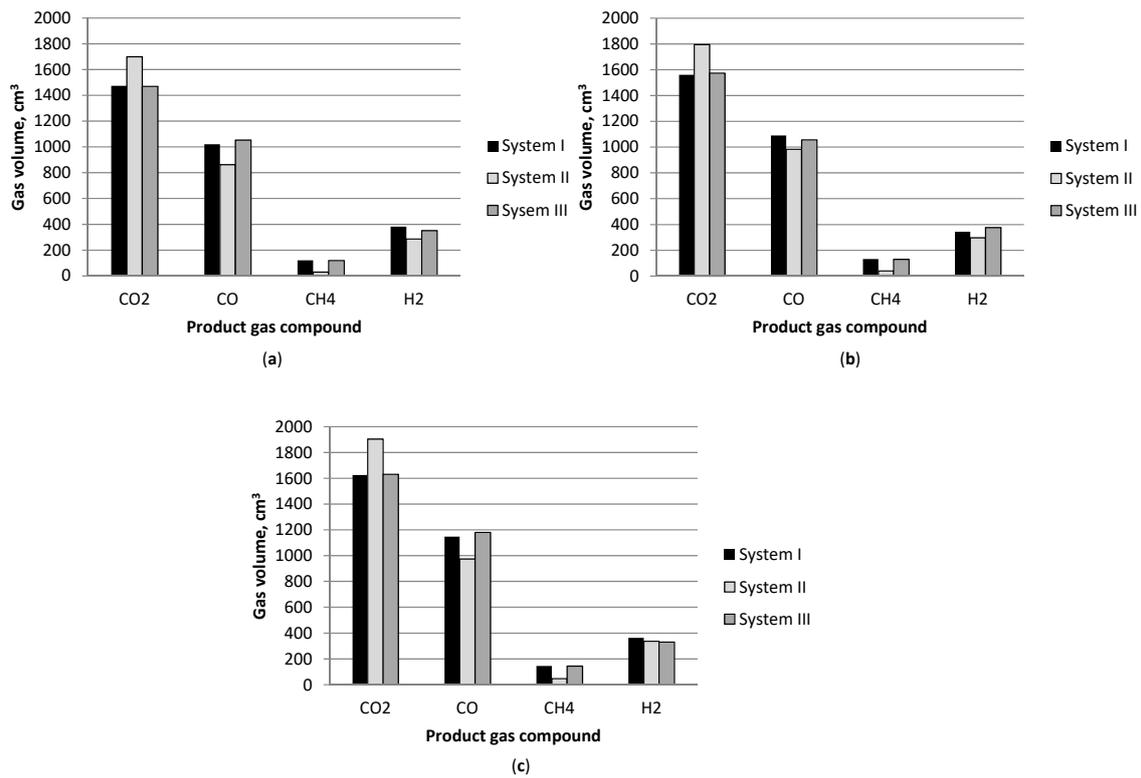


Figure 3. Average total gas yields in coal gasification with oxygen at: (a) 700 °C, (b) 800 °C and (c) 900 °C in system I–III.

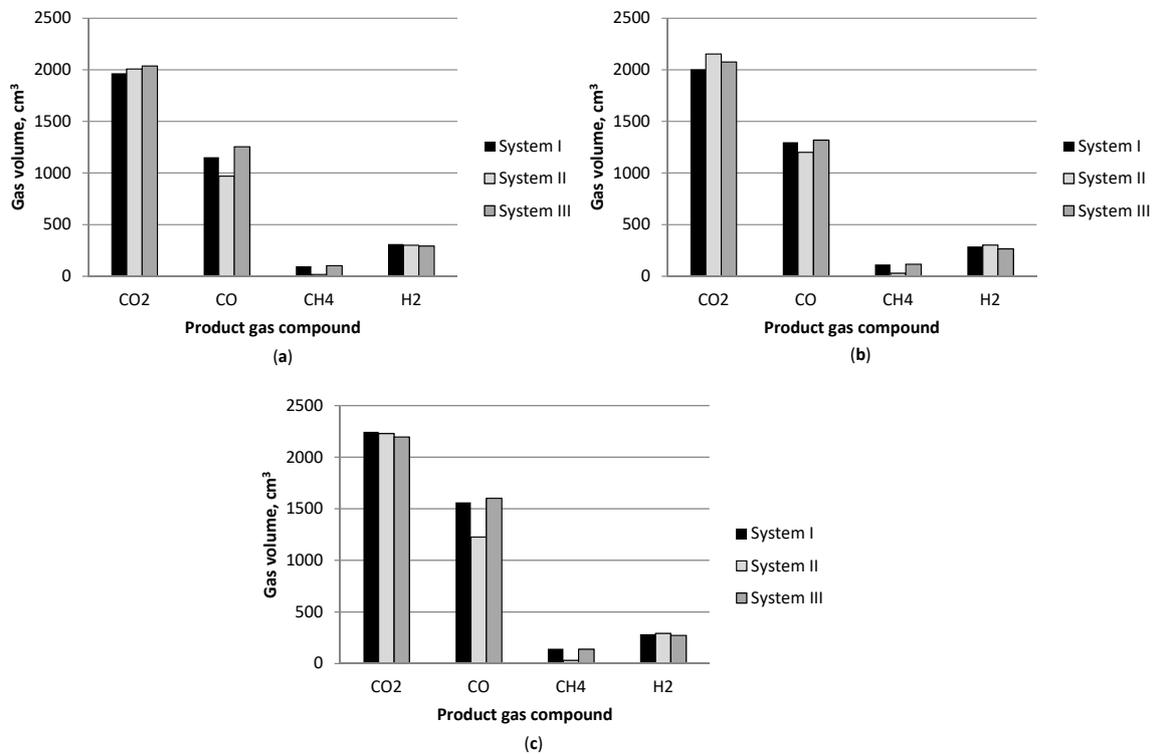


Figure 4. Average total gas yields in coal gasification with 30%vol. carbon dioxide in oxygen at: (a) 700 °C, (b) 800 °C and (c) 900 °C in system I–III.

3.1. Effect of Temperature

The conversion rate of carbonaceous material in gasification is affected by a combination of physical and chemical processes covering diffusion of the gasification agent to the char surface and next, to its porous structure, chemical reaction of the oxidant with carbon, and transport of the gaseous product to the char surface and next to the gas phase [27]. The carbon conversion rate reported in this study increased with increasing temperature which resulted in the highest gas yield at 900 °C, at each of the system configuration applied. These results clearly indicate the chemical reaction rate control within the operating parameters range applied in this study. They are in line with the observations made by Ye et al. [22] and Everson et al. [24] who determined the reaction rates of coal chars in a fluidized bed reactor experiments to be increasing with temperature from 765 to 891 °C and from 850 to 900 °C, respectively. The carbon conversion rates of coal chars observed by Wang and Bell in a drop tube reactor also increased with the temperature within the tested range of 833–975 °C [25]. Such effects were also observed for other carbonaceous materials chars, e.g., Guizani et al. reported over 3.5-fold reduction in time required for a 90% conversion of biomass chars with the temperature increase from 850 to 950 °C in a macro thermogravimetric device [21]. The lowest total gas volume and the lowest product gas calorific value in coal gasification with various gasification agents tested were reported for system II, where no external heat was provided during the process, at each of the process temperatures tested (see Table 2). This is because the temperature is the controlling parameter in the endothermic gasification reactions, in particularly with carbon dioxide as a gasification agent [28].

Table 2. Calorific value, Q_g , of gas generated in coal chars gasification with carbon dioxide, oxygen or 30%vol. carbon dioxide in oxygen at 700, 800 and 900 °C in various system configurations.

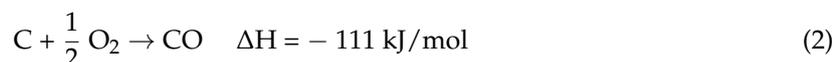
| No | Gasification Agent | Temperature, °C | Q_g , MJ/m ³ | | |
|----|-----------------------------------|-----------------|---------------------------|-----------|------------|
| | | | System I | System II | System III |
| 1 | carbon dioxide | 700 | 3.50 | 2.70 | 3.59 |
| 2 | carbon dioxide | 800 | 3.81 | 2.75 | 4.12 |
| 3 | carbon dioxide | 900 | 4.59 | 2.80 | 4.92 |
| 4 | oxygen | 700 | 6.73 | 4.88 | 6.73 |
| 5 | oxygen | 800 | 6.72 | 5.12 | 6.65 |
| 6 | oxygen | 900 | 6.80 | 5.06 | 6.78 |
| 7 | 30 vol.% carbon dioxide in oxygen | 700 | 5.70 | 4.56 | 5.77 |
| 8 | 30 vol.% carbon dioxide in oxygen | 800 | 5.97 | 4.91 | 5.87 |
| 9 | 30 vol.% carbon dioxide in oxygen | 900 | 6.17 | 4.86 | 6.26 |

3.2. Gasification Products

The yields of carbon monoxide increased with process temperature applied in gasification within the temperature range tested and were the highest in gasification with carbon dioxide as a gasification agent (see Figures 2–4). The yields of hydrogen were considerably lower than those of carbon monoxide and resulted mainly from the devolatilization step. Similar trends of generation of carbon monoxide as the main gaseous compound, low yield of hydrogen and temperature-related increase in values of product gas components yields in gasification of coal chars with carbon dioxide were also observed by Porada et al. within the temperature range 850–950 °C [26]. Methane formation was negligible under the process conditions applied. Billaud et al. [29] also reported the increase in carbon monoxide and hydrogen yield in gasification of sawdust with carbon dioxide in a drop tube reactor with the process temperature rise from 800 to 1500 °C [29]. In terms of the effect of the waste heat utilization system configuration on the yields of carbon monoxide, the lowest values were reported for system II, as previously noted, and the amounts generated in system I and III were comparable with a slightly higher amounts for system III, where the pre-heating of gasification agent was applied along with the external heating of a gasification zone during the experiment (see Figures 2–4). The concentration of carbon monoxide varied with temperature from 29 to 39%vol. in system I, from 22 to 24%vol.

in system II and from 30 to 43%vol. in system III with the temperature increase from 700 to 900 °C in coal chars gasification with carbon dioxide. The tendency of carbon monoxide content increase with process temperature rise from 850 to 950 °C was also observed by Chen et al. [30] in CO₂ gasification of steam-activated carbon. Such effects are caused by thermodynamics of the reaction (1), the main reaction of CO₂ gasification, responsible for the production of carbon monoxide, which is thermodynamically favored at higher temperatures, starting from 700 °C.

The exothermic reaction between carbon and oxygen:



is thermodynamically feasible within the entire temperature range covered in the study presented. There were no significant differences observed between the effects of coal chars gasification with oxygen in terms of the volumes of carbon monoxide and hydrogen generated in system I, without gasification agent pre-heating, and system III, where the temperature of both the gasification agents and gasification reactor was maintained with the use of the external heat source (see Figure 3). The concentration of carbon monoxide was on a comparable level of 34–35 vol.% and 35–36 vol.% for systems I and III, respectively, and 30–32 vol.% in system II. The amount of carbon dioxide increased slightly with process temperature but this had no considerable effect on product gas calorific value of 6.7–6.8 MJ/m³ for systems I and II, and 4.9–5.1 MJ/m³ for system II, respectively, in the temperature range 700–900 °C (see Table 2).

Application of 30 vol.% of carbon dioxide in oxygen as a gasification agent resulted in the increase in the volume and content of carbon monoxide in the product gas, when compared to oxygen gasification (see Figures 3 and 4). The volumes and concentrations of carbon monoxide increased with temperature. The maximum concentrations of carbon monoxide were reported for 900 °C and amounted to 36 vol.%, 33 vol.% and 38 vol.%, for systems I–III, respectively, and slightly exceeded the maximum values reached in oxygen gasification as presented above. This was accompanied by the decrease in methane and hydrogen yields and increase in carbon dioxide yields when compared to oxygen gasification (see Figures 3 and 4). Interestingly, the yield of carbon monoxide in system II, with no external heat supply during the process, was higher in gasification with carbon dioxide/oxygen mixture than in gasification with pure carbon dioxide which proves the positive thermal effect of application of oxygen in a gasification agent on the process performance in this system option. The results show also a positive effect of the application of oxygen in the gasification agent mixture on the product gas calorific value, which increased of approximately 40%, when compared to carbon dioxide gasification (see Table 2).

3.3. Principal Components Analysis in Exploration of the Combined Effects of Temperature, Gasification Agent Composition and Waste Heat Utilization on Gasification Process Performance

The analysis of the complex effects of the temperature, gasification agent composition and waste heat utilization on the results of coal chars gasification was performed with the application of Principal Component Analysis (PCA) [12,44–46]. The experimental data were organized in a matrix $X(27 \times 6)$, with rows representing samples processed in gasification experiments performed at the temperatures of 700, 800, 900 °C in system I, II and III with the application of carbon dioxide (objects nos 1–9); oxygen (objects nos 10–18); and carbon dioxide/oxygen mixture (objects nos 19–27) as a gasification agent, respectively. The columns of the matrix X represent measured parameters, i.e., the amounts of the main gas components (carbon monoxide, carbon dioxide, methane and hydrogen), total gas yield and gas calorific value (parameters nos 1–6).

PCA constructed for the studied data $X(27 \times 6)$ enabled their effective compression. The PCA model constructed with three PCs described 98.73% of the total data variance. The respective score plots and loading plots are presented in Figure 5.

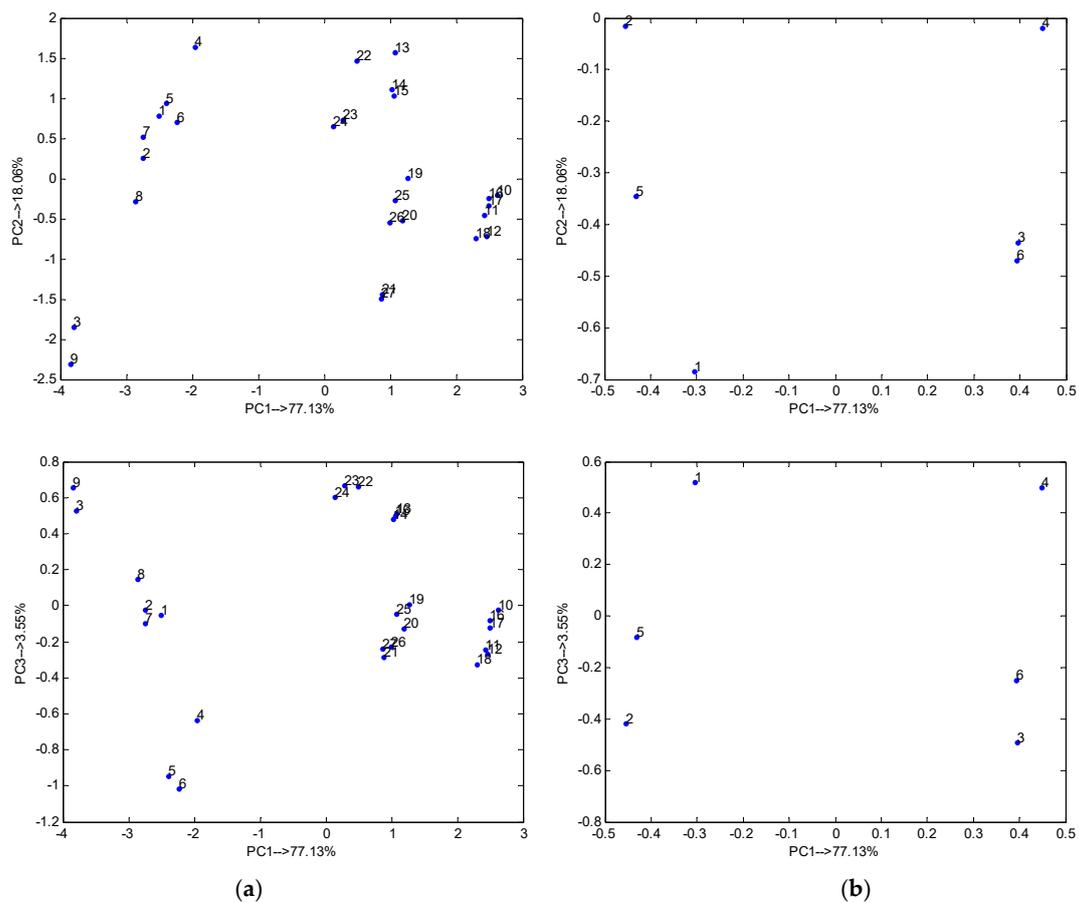


Figure 5. PCA score plots (a) and loading plots (b) for the studied data set $X(27 \times 6)$.

Four groups of objects defined as coal chars processed in gasification with various gasification agents, at various process temperatures, and with the application of various waste heat utilization configurations were distinguished along the PC1, describing 77.13% of the total data variance. The first group was composed of samples processed with the use of carbon dioxide as a gasification agent at 900 °C in systems I and III (objects nos 3 and 9), and the second group consisted of the remaining samples gasified with carbon dioxide (objects nos 1, 2, 4, 5, 6, 7 and 8). Within the third group samples processed in oxygen gasification at 700, 800 and 900 °C in system II, and all samples gasified with the mixture of carbon dioxide/oxygen at all studied temperatures in systems I, II and III (objects nos 13–15 and 19–27) were collected. The fourth group included samples gasified with oxygen at 700, 800 and 900 °C in systems I and III (objects nos 10–12 and 16–18), respectively.

The objects of the first two groups differed from the remaining ones in terms of relatively high average amounts of carbon monoxide, carbon dioxide and the total amount of gas produced in gasification (parameters nos 1, 2 and 5), as well as low average amount of hydrogen generated (parameter no 4). Samples gasified with carbon dioxide at 900 °C in systems I and III (objects nos 3 and 9) were characterized by the highest average amount of carbon monoxide and the total amount of gas produced in gasification (parameters nos 1 and 5), and the lowest average volume of hydrogen (parameter no 4). Furthermore, the uniqueness of samples gasified with the use of oxygen at 700, 800 and 900 °C in systems I and III (objects nos 10–12 and 16–18) was observed resulting from relatively high average yields of methane and hydrogen (parameter nos 3 and 4), the highest calorific value of gas (parameter no 6) and the lowest average amount of carbon dioxide generated in gasification (parameter no 2).

The PC2 describing 18.06% of the total variance, was constructed mostly because of the differences between the sample processed with the application of carbon dioxide as a gasification agent at 900 °C

in system III (object no 9) and the sample gasified with oxygen at 700 °C in system II (object no 13). The PC3, describing 3.55% of the total variance, was developed on the basis of the differences between the samples processed with the use of carbon dioxide at 700, 800 and 900 °C in system II (objects nos 4, 5 and 6), and all the remaining samples. On the basis of the loading plots, the difference between the sample gasified with the application of carbon dioxide at 900 °C in system III (object no 9) and the sample processed in oxygen gasification at 700 °C in system II (object no 13) was observed and it was attributed to relatively low average yield of hydrogen (parameter no 4) for object no 9. Object no 13 was unique due to relatively high average volume of hydrogen produced in gasification (parameter no 4) and the lowest average yield of carbon monoxide (parameter no 1) among all the studied samples. The samples gasified with the application of carbon dioxide as a gasification agent at 700, 800 and 900 °C in system II (objects nos 4, 5 and 6) were characterized by low average yield of hydrogen (parameter no 4).

The loading plots revealed a positive correlation between the average yield of methane and gas calorific value (parameters nos 3 and 6). The negative correlation was reported between the average yield of carbon dioxide and hydrogen (parameters nos 2 and 4).

4. Conclusions

The idea of utilization of captured carbon dioxide in coal gasification with the use of waste process heat was experimentally tested as a method potentially contributing to the development of low-emission and highly-efficient coal-based energy technologies. The lowest total gas yield and the lowest gas calorific value in coal gasification with carbon dioxide were reported for systems where no external heat source was applied once the process temperature was achieved. This implies that the thermal energy provided in this case was insufficient for an effective gasification dependent on the highly endothermic Boudouard reaction. The highest average yield of carbon monoxide, and the total gas yield, as well as the lowest average amount of hydrogen were characteristic for gasification with carbon dioxide at 900 °C in systems with the supply of the external source heat to gasification zone, with pre-heating of gasification agent only slightly enhancing the process productivity. Gasification with 30%vol. carbon dioxide in oxygen improved the thermal conditions of gasification in system with no temperature maintenance during the gasification process when compared to gasification with pure carbon dioxide. This was reflected in higher yields of carbon monoxide at 800 and 900 °C than in carbon dioxide gasification. However, in systems with the external heat source applied throughout the gasification test, higher yields of carbon monoxide were achieved for the gasification agent of higher carbon dioxide content. The experiments performed proved the feasibility of production of gas of calorific value 4–6 MJ/m³ in gasification with carbon dioxide—containing gasification agent under the laboratory conditions adopted. The idea of carbon dioxide valorization and waste heat utilization in gasification of coal, although promising in the context of the development of energy-efficient and low carbon footprint systems, needs further advancements in terms of process integration as well as measures of improving its cost-competitiveness before it may be considered for wider implementation.

Author Contributions: Conceptualization, A.S. and J.Z.; Methodology, A.S., and J.Z.; Investigation, J.Z. and N.H.; Data Analysis, J.Z. and A.S.; Writing—Original Draft Preparation, J.Z., and N.H.; Writing—Review and Editing, J.Z. and N.H.; Supervision, A.S.

Funding: This research received no external funding.

Conflicts of Interest: The authors declare no conflict of interest.

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