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Lab-scale Simulation of Coal-Nuclear Synergy. Utilization of High Temperature Reactor Excess Heat in Synthesis Gas and Hydrogen-Rich Gas Production

ABSTRACT. The paper presents the results of the experimental study on simulated application of High Temperature Reactor (HTR) excess heat in the allothermal coal gasification to synthesis gas and hydrogen-rich gas. The effects of application of gasification agents pre-heating was tested in a laboratory scale fixed bed reactor installation. The installation was equipped with a specially designed auxiliary pre-heating system for gasification agents applied (air, oxygen or steam), simulating the utilization of the HTR excess heat. The results of the study proved the feasibility of the utilization of the external excess heat in air and steam coal gasification.

KEY WORDS: High Temperature Reactor; HTR; coal gasification; excess heat; synthesis gas; hydrogen

Introduction

Coal is the dominant fossil fuel in terms of global energy supply security. Over 90% power generation is based on coal in Poland and the figure is approximately 40% worldwide (Metz et al., eds. 2001, 2007). Coal is considered to be the most carbon-intensive fuel and its share of the global CO_2 emission is expected to grow from 44% in 2010 to 45% in 2040 (International… 2013).

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Nuclear energy, already providing approximately 7% of the total primary energy, could make an increasing contribution to carbon-free electricity and heat generation in the future (Sims et al. 2007). However, a wider implementation of nuclear energy faces several barriers including nuclear fuel waste management constraints, economic and safety aspects as well as adverse public perception. A mix of energy sources i.e. fossil fuels, renewable and nuclear, combined with improved end-use efficiency is required to meet the growing energy demand.

A High Temperature Reactor (HTR) consists of spherical fuel elements surrounded by a cylindrical graphite vessel applied as a neutron reflector (Lozovetskii 2001; Virgil et al. 2007). The fuel elements i.e. graphite balls with a diameter of 60 mm, contain small grains composed of uranium dioxide and thorium dioxide or uranium carbide and thorium carbide. Helium, applied as a reactor core cooling medium, is heated from 700 to 950°C flowing through a HTR (Becker, Laurien 2003; Belov et al. 2012; Filippov et al. 2004; Mitenkov et al. 2004). The excess heat from a HTR can be utilized in several chemical processes, e.g. in coal gasification (Mitenkov et al. 2004).

Gasification process, considered as an alternative to combustion, offers increased efficiency, lower negative environmental impact and wider application range of the main product, synthesis gas, in power generation and chemical synthesis (Chiesa et al. 2005; Collot 2006; Minchener 2005; Gasification… 2013). Coal gasification to hydrogen-rich gas, as an environment friendly energy carrier, is widely discussed in the literature in terms of system analysis process modeling and its optimization (Belghit 2011; Cormos 2009a,b, 2010, 2011; Cormos et al. 2008; Gordillo, Gungor 2011; Howaniec, Smoliński 2013; Zhao et al. 2011; Skoulou et al. 2008; Smoliński 2008, 2011; Su et al. 2010), as a particulary attractive option of coal utilization.

Nuclear energy may also be considered as one of the ways of mitigating global warming if it could be taken into account as a way to replace fossil fuel based energy systems with high greenhouse gas (GHG) emission levels. Highly efficient fossil fuel utilization in gasification systems of the energy sector may be further combined with utilization of the excess heat from nuclear reactors within a coal-nuclear synergy concept. In these terms the most advanced studies have been performed in Germany by companies Hochtemperatur-Reaktorbau GmbH, Gesellschaft für Hochtemperaturreaktortechnik mbH, Bergbauforschung GmbH, Essen, Rheinische Braunkohlenwerke, research center Kernforschungsanlage Jülich (Arndt et al. 1979). Based on the results of the research, the systems based on one-stage and two-stage gasifiers coupled with a High Temperature Gas Cooled Reactor (HTGR) reactor were developed (Tomeczek 1990; Kubiak et al. 1993).

In the paper the results of the experimental study on the effects of simulated HTR excess heat application on efficiency of the allothermal coal gasification to synthesis gas and hydrogen-rich gas are presented. Three variations of a pre-heating system were tested in coal gasification with various gasification agents, i.e., oxygen, air and steam. The results proved the feasibility of the utilization of the external excess heat in air and steam coal gasification.

1. Methods and materials

The experimental study on coal gasification to synthesis gas and hydrogen-rich gas was performed in a laboratory scale fixed bed gasifier equipped with an auxiliary gasification agents pre-heating system, simulating the utilization of an excess HTR heat.

1.1. Materials

Hard coal samples were provided by Piast coal mine in Bieruń, Poland. The proximate and ultimate analyses of the tested fuels were performed in the accredited laboratory of the Department of Solid Fuel Quality Assessment of the Central Mining Institute with the application of relevant standards, testing procedures and analyzers. These included: automatic thermogravimetric analyzers LECO: TGA 701 or MAC 500 (contents of moisture, ash, volatiles acc. to PN-G-04511:1980, PN-G-04560:1998 and PN-G-04516:1998, respectively), calorimeters LECO: AC-600 and AC-350 (heat of combustion acc. to PN-G-04513:1981), TruSpecCHN analyzer (contents of carbon, hydrogen, nitrogen acc. to PN-G-04571:1998) and TruSpecS analyzer (sulfur acc. to PN-G-04584:2001) and ash fusibility temperatures under oxidizing atmosphere (acc. to PN-G-04535:1982). The results are given in Table 1.

1.2. Test stand

Simulated High Temperature Reactor (HTR) excess heat was applied in the allothermal coal gasification system of the Laboratory of Advanced Energy Technologies of the Department of Energy Saving and Air Protection, Central Mining Institute (see Fig.1). Synthesis gas and hydrogen-rich gas generation with pre-heated gasification agents, such as oxygen, air and steam, was tested. The main element of the test stand is a fixed bed reactor of a volume of approximately 0.8 L, heated with a resistance furnace. The installation is equipped with an auxiliary gasification agents pre-heating system, simulating the utilization of a HTR excess heat. The furnace system and gasification agents pre-heating system operation are controlled by a computer. The process temperature and pressure are monitored with a thermocouple and a manometer, respectively.

1.3. Operational conditions and test procedure

Coal sample of 3 g, grain size below 0.2 mm, in an analytical state, prepared according to the standard PN-G-04506:1996, was placed at the bottom of the reactor between two layers of a quartz wool for better temperature distribution and avoidance of the entrainment of fuel particles by the gaseous media. The experiments were performed in three series with application of steam, oxygen and air as gasification agents. In the first series (system I) the reactor was heated up with a resistance furnace with a heating rate of 1.33°C/s to the temperature of 700, 800

TABLE 1. Basic physical and chemical parameters of hard coal tested

TABELA 1. Właściwości fizykochemiczne badanego węgla

No	Parameter	Value	Unit
As received			
1.	Moisture W_t^r	10.76	%
2.	Ash A^r	6.94	%
3.	Total sulfur S_t^r	1.85	%
4.	Calorific value Q_i^r	25,583	kJ/kg
Analitical state			
5.	Moisture W^a	7.44	%
6.	Ash A^a	7.20	%
7.	Volatiles V^a	32.37	%
8.	Heat of combustion Q_s^a	27,815	kJ/kg
9.	Calorific value Q^i	26,626	kJ/kg
10.	Ash sintering point t_s	940	°C
11.	Ash softening point t_A	1,280	°C
12.	Ash melting point t_B	1,360	°C
13.	Ash flow temperaturę t_C	1,430	°C
14.	Total sulfur S^a	1.92	%
15.	Carbon C_t^a	67.36	%
16.	Hydrogen H_t^a	4.14	%
17.	Nitrogen N^a	0.92	%
Dry state			
18.	Ash A^d	7.78	%
19.	Total sulfur S_t^d	2.07	%
20.	Volatiles V^{daf}	37.92	%
21.	Heat of combustion Q_s^{daf}	32,586	kJ/kg

or 900°C in the inert gas (nitrogen) atmosphere. Next, the gasification agent of the temperature of approximately 100°C was introduced into the reactor. In the second series (system II) a sample was heated up in the reactor to one of the set temperatures mentioned above. Next, the heating of the reactor was switched off and the gasification agents were pre-heated to the temperature of 700, 800 or 900°C. The pre-heated gasification media were fed into the reactor. In the third series (system III) the reactor was heated to the set temperature with a resistance furnace and the set temperature was maintained by the resistance furnace. An auxiliary gasification agents pre-heating system was also applied to pre-heat the gasification media to the temperature of 700, 800 or 900°C. The pre-heated gasification agents were introduced into the reactor. The gasification process was assessed in terms of the gasification product volume and composition. Dry and clean samples of a product gas were analyzed automatically every 192s

Fig. 1. Fixed-bed reactor experimental stand

Rys. 1. Instalacja laboratoryjna z reaktorem ze złożem stałym

via the two-channel gas chromatograph Agilent 3000A. In the first channel, a column PLOT U (8 m x 0.32 mm) with a TCD detector was applied (for separation of CO_2 and C_2–C_5). In the second channel, a backflush injector module with a pre-column PLOT U (3 m x 0.32 mm) and an analytical column MS5A PLOT (10 m x 0.32 mm) with a TCD detector (for separation of H_2, O_2, N_2, CH_4 and CO) were applied. Helium and argon were the carrier gases applied in the PLOT U and MS5A PLOT columns, respectively. The temperature of the inlet, injector and the columns was 60°C. The injection time was $50 \cdot 10^{-3}$ for both columns, and the run time and post run time were 150 s and 10 s for PLOT U and MS5A PLOT, respectively. The backflush time for column PLOT U was 12 s.

2. Results and discussion

The study on the allothermal coal gasification with utilization of the simulated HTR excess heat and oxygen, air and steam as gasification agents was performed to determine the optimal conditions, within the parameters tested, for synthesis gas and hydrogen-rich gas production. The major findings are presented below.

Figures 2 and 3 present the average total volumes of H_2, CO, CO_2 and CH_4 and the average composition of gas generated with steam, oxygen and air as gasification agents.

Based on the average gas volume and composition the calorific value of gas generated at 700, 800 and 900°C was determined (see Table 2).
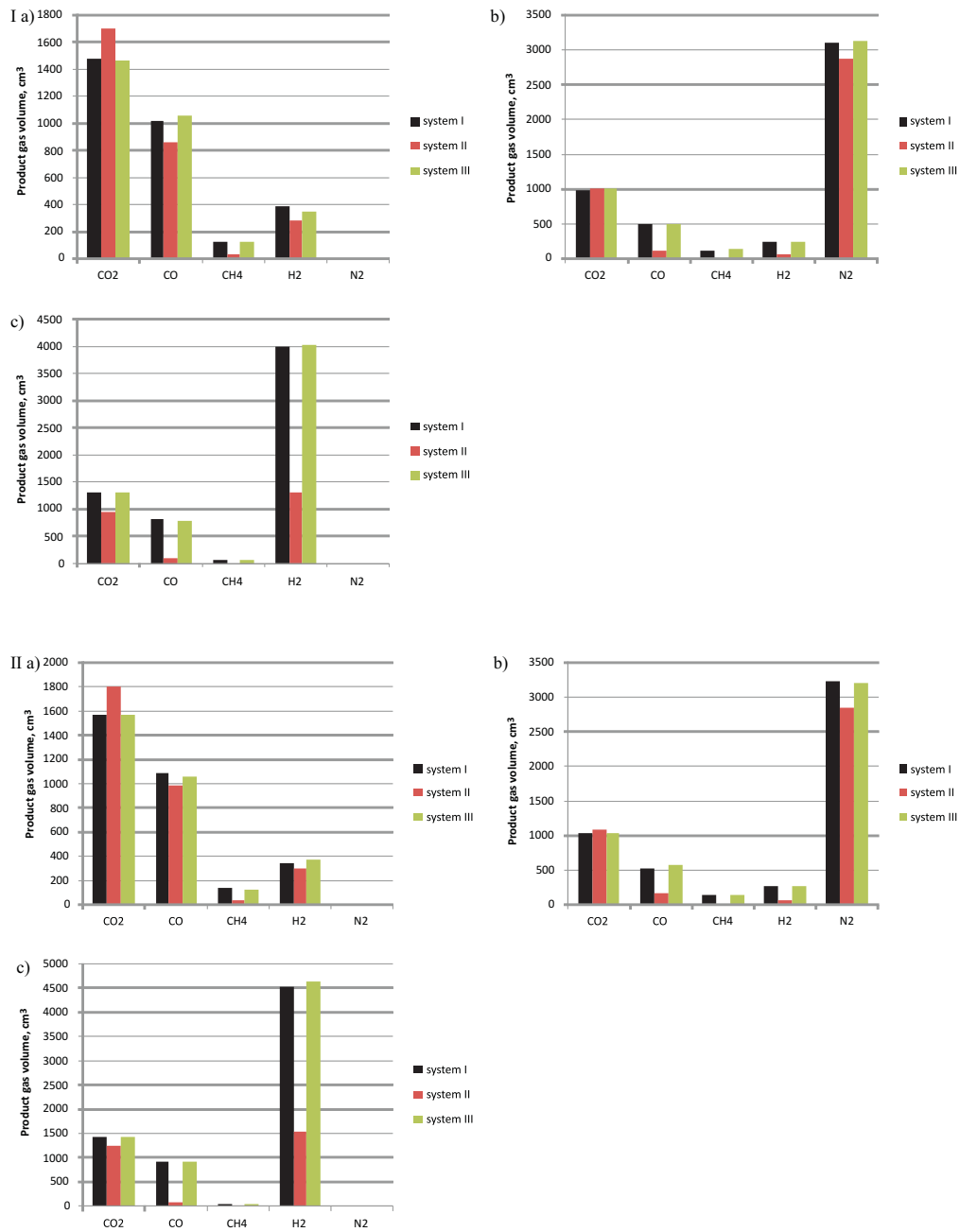
Fig. 2. Average total product gas volume in coal gasification with a) oxygen, b) air and c) steam at I) 700°C,
II) 800°C and III) 900°C

Rys. 2. Średnia objętość gazu produkowanego w procesie zgazowania węgla a) tlenem, b) powietrzem
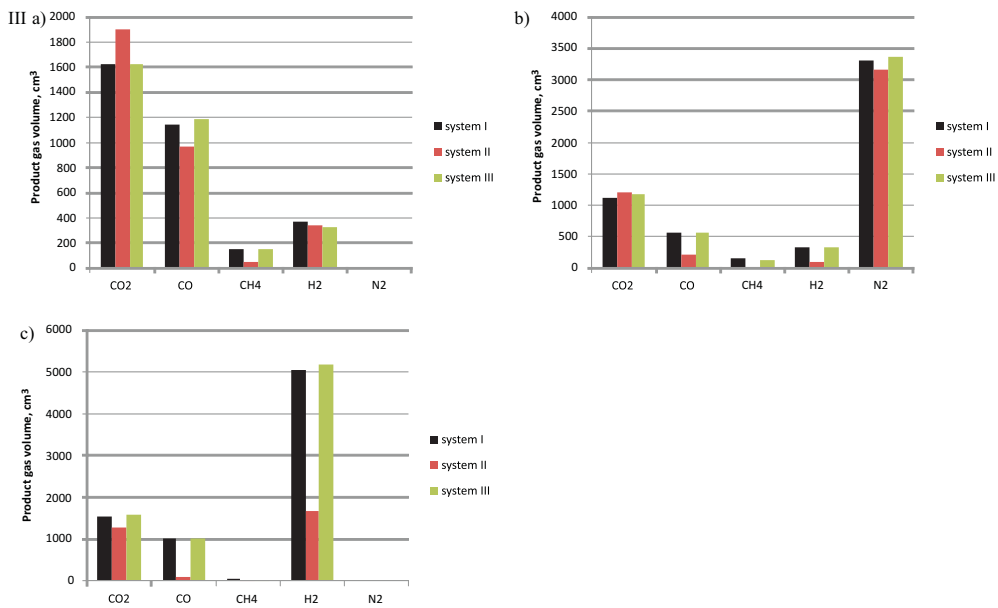i c) parą wodną w temperaturze I) 700°C, II) 800°C i III) 900°C

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Fig. 2 cont. Average total product gas volume in coal gasification with a) oxygen, b) air and c) steam at I) 700°C, II) 800°C and III) 900°C

Rys. 2 cd. Średnia objętość gazu produkowanego w procesie zgazowania węgla a) tlenem, b) powietrzem i c) parą wodną w temperaturze I) 700°C, II) 800°C i III) 900°C
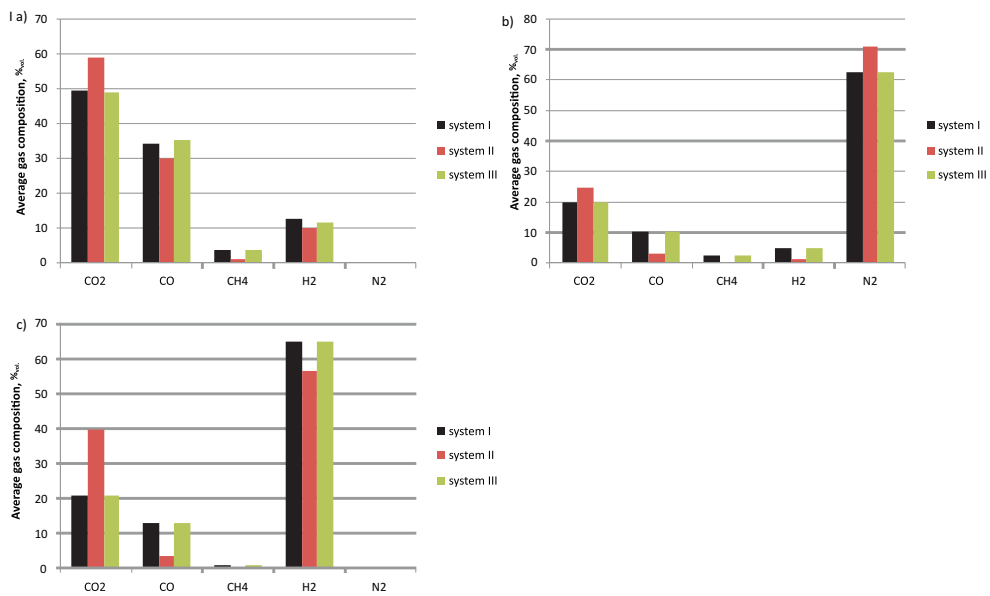
Fig. 3. Average gas composition in coal gasification with a) oxygen, b) air and c) steam at I) 700°C, II) 800°C and III) 900°C

Rys. 3. Średni skład gazu w procesie zgazowania węgla a) tlenem, b) powietrzem i c) parą wodną w temperaturze I) 700°C, II) 800°C i III) 900°C
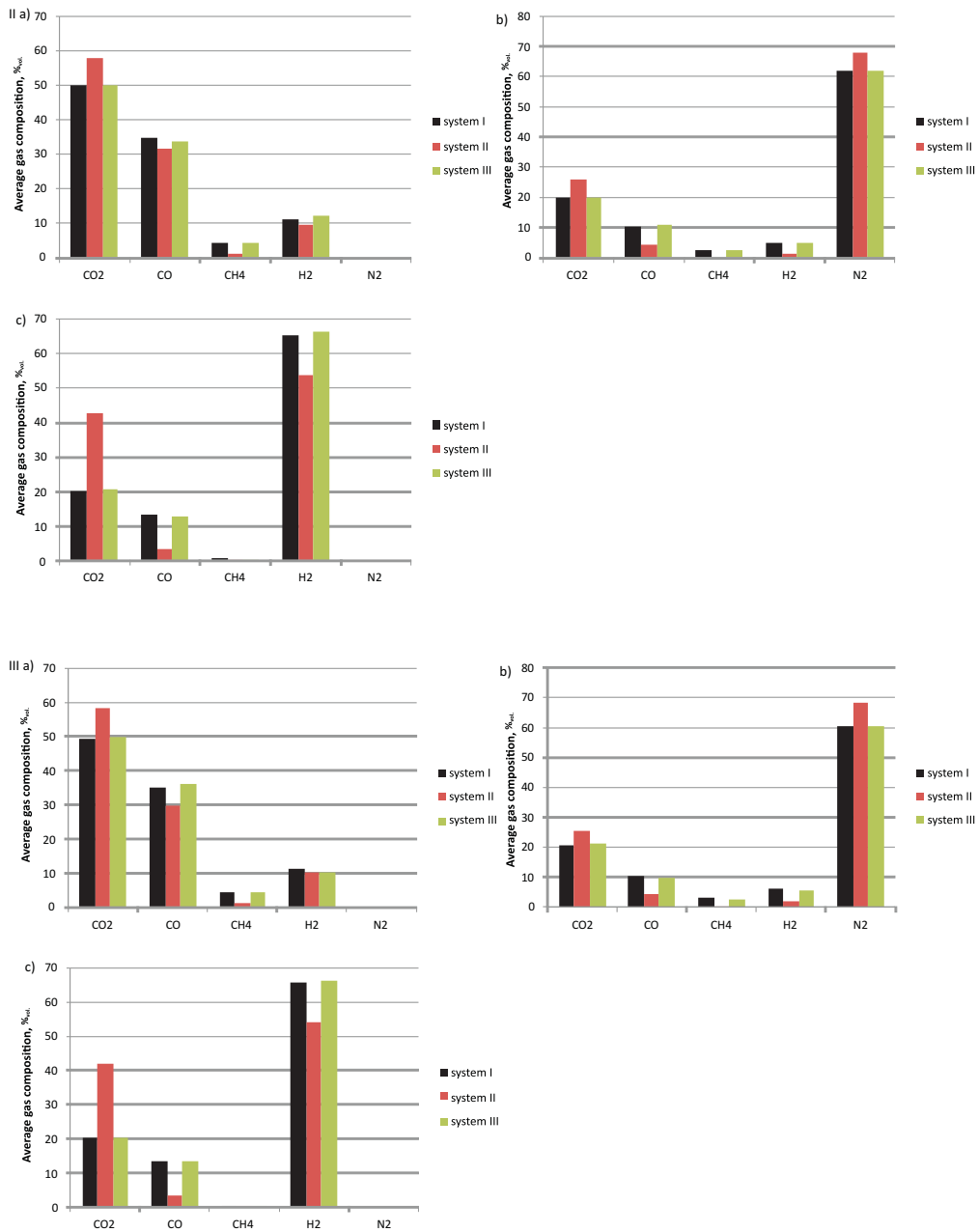
Fig. 3 cont. Average gas composition in coal gasification with a) oxygen, b) air and c) steam at I) 700°C, II) 800°C and III) 900°C

Rys. 3 cd. Średni skład gazu w procesie zgazowania węgla a) tlenem, b) powietrzem i c) parą wodną w temperaturze I) 700°C, II) 800°C i III) 900°C

TABLE 2. Calorific value, Q_g, of gas generated in coal gasification with oxygen, air and steam at 700–900°C in three system options tested

TABELA 2. Wartość opałowa, Q_g, gazu z procesu zgazowania węgla tlenem, powietrzem i parą wodną w temperaturze 700–900°C w trzech badanych opcjach sytemu

System option	Q_g [MJ/m³]								
	System I			System II			System III		
Gasification agent	oxygen	air	steam	oxygen	air	steam	oxygen	air	steam
700°C	6.884	2.692	8.926	4.914	0.507	6.509	6.890	2.688	8.904
800°C	6.888	2.755	8.885	5.167	0.673	6.234	6.809	2.815	8.789
900°C	6.976	2.942	8.852	5.120	0.734	6.312	6.953	2.755	8.713

The calorific value of the product gas was similar in the system with the heating of the reactor (system I) and with the pre-heating of gasification agents and heating of the reactor during the experiment (system II), irrespective of the gasification agent applied at 700, 800 and 900°C. Howver, the variations were observed in terms of the gas volume. The total gas volume generated in the system I at 900°C was 1.09, 1.10 and 1.24 times higher than at 700°C, with gasification agents applied: oxygen, air and steam, respectively. In the system III the total gas volume at 900°C was 1.10, 1.11 and 1.26 times higher than at 700°C, when oxygen, air and steam were applied as gasification agents, respectively (see Fig. 4). The total hydrogen volume is a key parameter of steam gasification process efficiency assessment. It was from 3,997 and 5,048 cm³ at 700°C to 5,048 and 5,197 cm³ at 900°C in the systems I and III, respectively.

Based on the results it may be concluded that the application of the gasification agents pre-heating system (system III) is reasonable only in the case of air and steam gasification, irrespective of process temperature. The total gas volume generated in air gasification in option III was from 0.4% (at 800°C) to 1.7% (at 900°C) higher than that reported under respective process conditions in system I. The increase in the total gas volume in steam gasification from system I to system III was from 0.6% at 700°C to 2.4% at 900°C.

The variations were also observed in the concentrations of the main gas components.

Slightly higher methane and hydrogen contents were observed in the process of gasification with oxygen in system I, irrespective of the process temperature, than in system III. These amounted to 4.01 and 12.74% vol. (system I) and 3.97 and 11.74% vol. (system III) of methane and hydrogen, respectively, at 700°C. The respective values at 900°C were 4.43 and 11.06% vol. (system I) and 4.37 and 10.06% vol. (system III) at 900°C. Relatively higher carbon dioxide and monoxide contents were reported for system III. The similar trend was observed in case of air gasification. In steam gasification in system I, relatively higher content of carbon dioxide, carbon monoxide and methane and lower content of hydrogen (64.91% vol. at 700°C and 65.82% vol. at 900°C) were observed than in system III (65.23% vol. at 700°C and 66.34% vol. at 900°C), irrespective of process temperature. Increase in carbon monoxide content and decrease in carbon dioxide concentration with the temperature increase from 800 to 900°C were observed in steam gasification in systems I and III, resulting from the reverse Boudouard
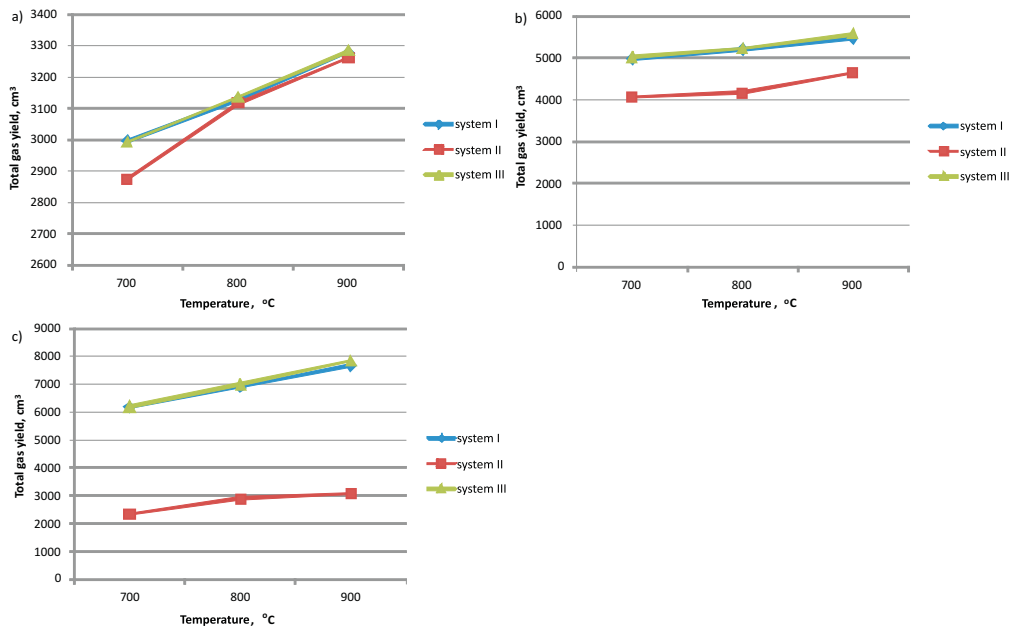
Fig. 4. Total gas volume generated in coal gasification with a) oxygen, b) air and c) steam at 700, 800 and 900°C, with simulated utilization of a HTR excess heat

Fig. 4. Całkowita objętość gazu wyprodukowanego w procesie zgazowania węgla a) tlenem, b) powietrzem i c) parą wodną w temperaturze 700, 800 i 900°C, z symulacją wykorzystania ciepła nadmiarowego z wysokotemperaturowego reaktora jądrowego

reaction and weak water gas shift reaction at temperatures over 800°C. Relatively low methane content in steam gasification: 1.06 and 0.96% vol. at 700°C and 0.42 and 0.11% vol. at 900°C in systems I and III, respectively, resulted from steam and dry reforming of methane.

Calorific values of gas generated in coal gasification with oxygen and steam in system II were slightly lower than in systems I and III. A significant decrease in gas calorific value was observed in air gasification in system II (507 MJ/m^3 at 700°C and 734 MJ/m^3 at 900°C) when compared to options I (2.69 MJ/m^3 at 700°C and 2.94 MJ/m^3 at 900°C) and III (2.69 MJ/m^3 at 700°C and 2.76 MJ/m^3 at 900°C). Steam gasification resulted in the highest hydrogen content (from 56.48% vol. at 700°C to 53.68% vol. at 800°C) generated in a highly endothermic reaction. Furthermore, a decrease in carbon monoxide and increase in carbon dioxide concentrations were reported, resulting from carbon monoxide conversion to carbon dioxide in water gas shift and Boudouard reactions. The total gas volume in steam gasification in system II was three times lower than in systems I and III, irrespective of process temperature (see Fig. 3). Decrease in the total gas volume in steam gasification in system II was related to the significant decrease in process temperature. Pre-heating of gasification agents to the required temperature of 700, 800 or 900°C with the simulated HTR excess heat was not sufficient to maintain the highly endothermic reactions. The temperature profiles recorded in steam gasification experiments in system II are presented in Fig. 5. It can be seen that a significant decrease in temperature was reported for this option (see Fig. 5-IIIb). Such effects were not
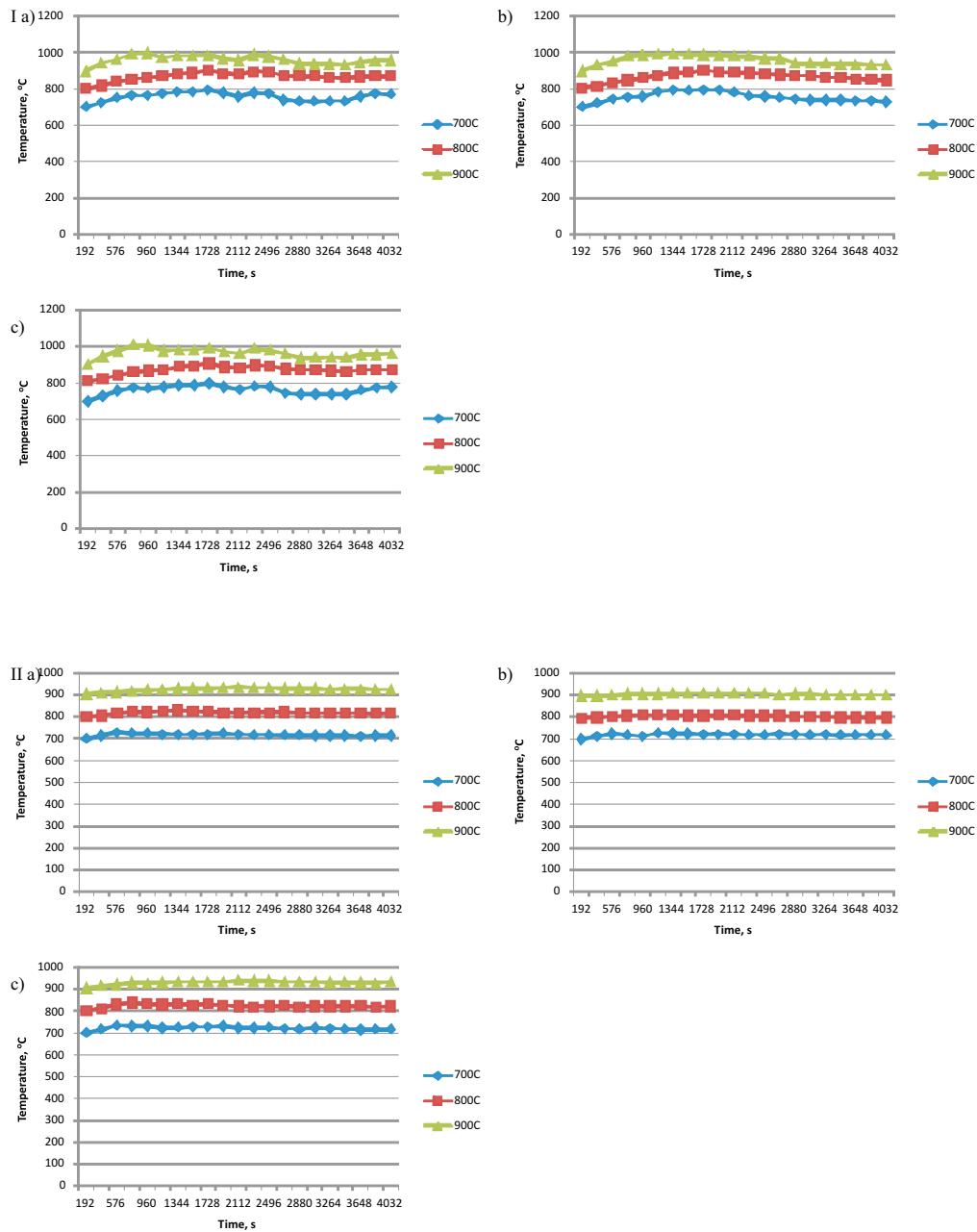
Fig. 5. Temperature profiles in coal gasification with I) oxygen, II) air and III) steam in system a) I, b) II and c) III at 700, 800 and 900°C

Rys. 5. Wykres zmian temperatury w procesie zgazowania węgla I) tlenem, II) powietrzem i III) parą wodną w systemie a) I, b) II i c) III w temperaturze 700, 800 i 900°C
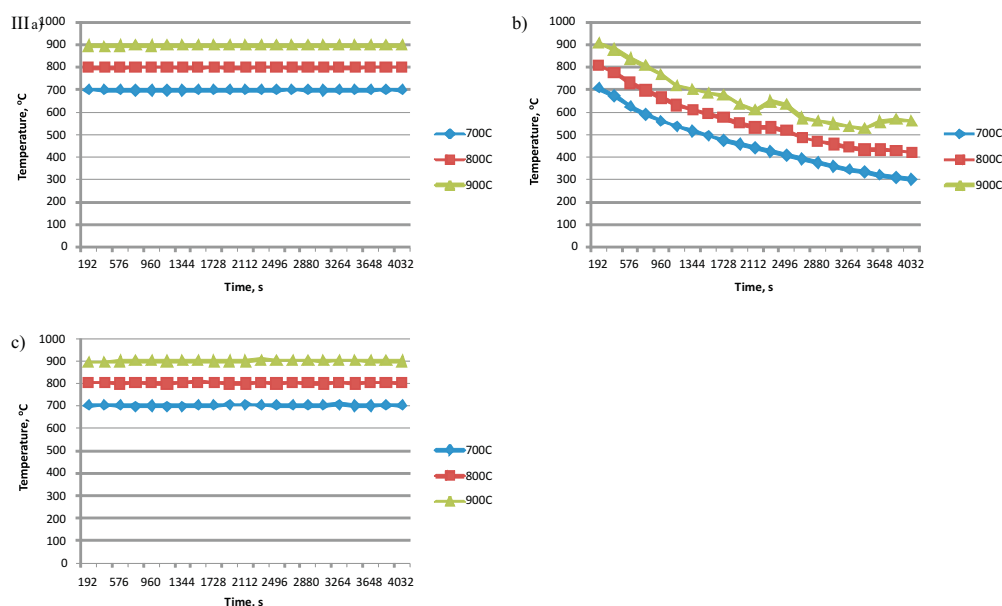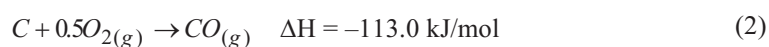
Fig. 5 cont. Temperature profiles in coal gasification with I) oxygen, II) air and III) steam in system a) I, b) II and c) III at 700, 800 and 900°C

Rys. 5 cd. Wykres zmian temperatury w procesie zgazowania węgla I) tlenem, II) powietrzem i III) parą wodną w systemie a) I, b) II i c) III w temperaturze 700, 800 i 900°C

observed in exothermic oxygen and air gasification in system II (Fig. 5-IIIa and c). The dominant role in all systems considered play two exothermic reactions (see temperature profiles in Fig. 5-IIIa and c for systems I and III):

$$C + O_{2(g)} \rightarrow CO_{2(g)} \quad \Delta H = -394.9 \text{ kJ/mol} \tag{1}$$

$$C + 0.5O_{2(g)} \rightarrow CO_{(g)} \quad \Delta H = -113.0 \text{ kJ/mol} \tag{2}$$

Low calorific value of gas generated in air gasification in option II irrespective of process temperature resulted from relatively low contents of combustible gas components. Hydrogen content in product gas was from 1.49% vol. at 700 to 1.89% vol. and 900°C, while in options I and III, the respective values were from 5.00 and 4.89 to 5.87 and 5.69% vol. at 700 and 900°C.

3. Conclusions

1. Application of the gasification agents pre-heating system with simulated HTR excess heat was reasonable only in the case of air and steam gasification, irrespective of process temperature.

2. Steam gasification resulted in the highest hydrogen content (from 56.48% vol. at 700°C to 53.68% vol. at 800°C) generated in a highly endothermic reaction.

3. Decrease in carbon monoxide and increase in carbon dioxide concentrations were reported in steam gasification resulting from the conversion of carbon monoxide to carbon dioxide in water gas shift and Boudouard reactions.

4. The total gas volume in steam gasification in system II was three times lower than in systems I and III, irrespective of the process temperature.

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Badania symulacyjne synergii węglowo-jądrowej. Zastosowanie ciepła nadmiarowego z reaktora HTR w procesie zgazowania węgla do gazu syntezowego i gazu bogatego w wodór

Streszczenie

W artykule przedstawiono wyniki badań symulacyjnych zastosowania ciepła nadmiarowego z wysokotemperaturowego reaktora jądrowego (HTR) w procesie allotermicznego zgazowania węgla do gazu syntezowego i gazu bogatego w wodór. Określono wpływ wstępnego przegrzania czynnika zgazowującego na wyniki procesu zgazowania w laboratoryjnej instalacji z reaktorem ze złożem stałym. Instalacja została wyposażona w specjalnie do tego celu zaprojektowany układ wstępnego przegrzania czynnika zgazowującego (powietrze, tlen lub para wodna), symulujący wykorzystanie ciepła nadmiarowego z reaktora HTR. Wyniki badań potwierdziły możliwość wykorzystania zewnętrznego źródła ciepła nadmiarowego w procesie zgazowania węgla powietrzem i para wodną.