

# The single-stage steam gasification of magnetite heavy suspension separated coal samples from hungarian brown coal

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## Abstract

Currently, gasification of coal is considered the centre of the clean coal technology. In this study, there were two coal samples from the magnetite heavy suspension separated process used in the single stage fixed bed gasification within the non-moving of material. These two types of samples were marked as A1 and A2 sample, with the specific densities of  $\leq 1.8 \text{ g/cm}^3$  and  $\leq 1.6 \text{ g/cm}^3$ , respectively and diameters 1-20 mm. The gasification experiments were conducted at steam flow rates of 5 and 10 g/min and gasification temperatures of 700, 800, and 900 °C. The main purpose of the experiments was to consider the effects of temperature and steam flow rate during the coal gasification from both, the energetic and chemical utilisation point of views. With higher temperature and steam flow rate, the experiments within A1 sample performed a better char yield and higher volume of produced syngas. In the case of A1 sample, the lowest char yield was 23.78 wt% at 900 °C gasification temperature and 10 g/min steam flow rate. At 900 °C of gasification temperature, the range of low heating value of synthesis gas were from 9.13 to 9.49 MJ/Nm<sup>3</sup> within 5 and 10 g/min of steam flow rate, indicating that the produced syngas is sufficient for energetic application. The range of H<sub>2</sub>/CO ratio were from 1.96 to 2.52 at 900 °C of gasification temperature, which has a great potential for a further chemical application process.

Keywords: low rank coal, thermochemical process, coal gasification, synthesis gas

Kulcsszavak: gyenge minőségű szén, termokémiai folyamat, szénelgázosítás, szintézisgáz

## 1. Introduction

Coal is known to be the most abundant and widespread among fossil fuel resources. Today, coal is mainly used in the cement industry, iron and steel manufacturing, and electric power generation [1]. In 2018, the utilisation of coal accounted for the highest proportion in the electrical generation, approximately 37.93% [2]. Still now, the electricity generation from coal has been the most important resource and couldn't be directly replaced by the other energy primary sources.

The range of coal can be classified from peat to anthracite. In which, low-rank coals include lignite and sub-bituminous coals. These types of coal characterised by a lower heating value than high-rank coals (bituminous to anthracite) as a result of their lower carbon and higher moisture content. Lignite (brown coal) is a coal formed in the early stages of coalification, with the properties intermediate to those of bituminous coal and peat [3]. The major benefit of lignite combustion is the abundance and low cost of mining. On the other hand, the use of lignite has a long list of disadvantages. Low calorific value, high moisture content that reduces the boiler efficiency, and ash properties that require specific combustion processes. Nevertheless, lignite has been successfully used as a raw material for gasification, liquefaction, and pyrolysis processes. The advantages of using lignite over higher rank coals in

gasification and liquefaction relies on their high reactivity, non-caking properties, and catalytic property of alkali and alkaline-earth elements [4], [5].

Gasification is a thermochemical process, in which, using heat and gasifying agents to convert carbon-based materials (coal, biomass, municipal solid waste) into synthesis gas. In comparison with the conventional coal combustion in power generation, the gasification process exhibits several advantages [6]–[8]: (i) high ability to use a wide range of starting materials (coal, biomass, municipal solid waste), (ii) multi-production capacity (power generation, hydrogen fuel, or chemical products), (iii) lower emission of hazardous products (for example nitrogen, sulphur, mercury-based products, ash), (iv) reduced power and total budget for CO<sub>2</sub> capture. The major advantage of gasification is the high flexibility of synthesis gas utilisation. The quantity and quality of synthesis gas depend on different factors, such as the property of starting material, type of gasifiers, operational parameters, gasifying reactants, as well as catalyst [9]–[13].

Relative works regarding the gasification of Hungarian brown coal are quite rare. A. Pettinau et al. [14] studied in both bench-scale and pilot-scale fixed bed gasifier to investigate the potential industrial applications. The bench-scale gasification experiments carried out at 800 °C with steam and mixture of

steam and oxygen. While the experiments took place at a range of 900 to 950 °C with steam/air mixture in an up-draft pilot-scale gasifier. L. Bokányi and Á. Pintér-Móricz [15] investigated the plasma gasification of sub-bituminous Hungarian coal for the potential methanol-ethanol synthesis process. The experiments carried out within 30 kVA of plasma reactor with the temperature range of 1240-1588 °C. Oxygen was used as the gasifying agent in these experiments. There is a clear gap in the description of synergy effects of temperature and steam flow rate during the gasification of different Hungarian coal samples. In this study, there were two coal samples from the magnetite heavy suspension separated process used in the single stage fixed bed gasification. The main propose was to consider the effects of temperature and steam flow rate in both product distribution and gasification efficiency for these two coal samples and asses the significance of the separation method.

## 2. Materials and experiments

The main goal of this study is the investigation of gasification behaviour of two coal samples from the magnetite heavy suspension separated process. These two samples were marked as A1 and A2 sample, with the specific densities of  $\leq 1.8 \text{ g/cm}^3$  and  $\leq 1.6 \text{ g/cm}^3$ , respectively and diameters 1-20 mm. The elemental analysis and heating values are presented in *Table 1*. In this scope, the elemental composition of coal samples was examined under the standard ISO 29541:2010 Solid mineral fuels - Determination of total carbon, hydrogen and nitrogen content - Instrumental method [16] within a Carlo Erba EA 1108 equipment analyser. The high heating value of coal samples were determined by a bomb calorimeter – Parr 6200 Isoperibol Calorimeter type analyser, using the ISO 1928:2009 - Solid mineral fuels - Determination of gross calorific value by the bomb calorimetric method and calculation of net calorific value standard [17]. The proximate analysis of coal samples was performed by thermal gravimetric analysis in a MOM Derivatograph-C type with the nitrogen and air ambient. The TG and DTG curves of coal samples are presented in *Fig. 1*.

Elemental analysis in air-dry samples, wt%						
	N-nitrogen	C-carbon	H-hydrogen	S-sulphur	O-oxygen (by difference)	
<b>A1</b>	0.83	51.62	3.99	4.87	19.64	
<b>A2</b>	0.82	48.38	3.92	5.18	16.65	
Proximate analysis in air-dry samples, wt%						Heating value [MJkg <sup>-1</sup> ]
	M-moisture	V-volatile	FC-fixed carbon	A-ash	HHV	LHV
<b>A1</b>	9.37	36.75	34.84	19.05	20.029	18.966
<b>A2</b>	8.56	34.44	31.95	25.05	16.889	15.699

Table 1 Elemental analysis, proximate analysis, and heating value analysis of two different coal samples

1. táblázat Két különböző szénminta elemvizsgálata, nedvességtartalma és fűtőértéke

The experimental apparatus used in this study mainly consisted of heat resistant steel reactor (1200 mm long and 80 mm inner diameter), electrical heater (Carbolite 12/900), data logger, steam generator, manometer, venturi scrubber, cotton filter and gas meter, as illustrated in *Fig. 2*. The coal pyrolysis and steam gasification experiments were carried out at room pressure. The procedure of experiments is described as below:

In each experiment, there were 3 kg of brown coal loaded in the reactor. The electrical heater was then turned on to heat up the reactor. The starting point of the gasification with the introduction of steam into the reactor started when the pyrolysis process finished. The outlet of produced synthesis gas was in the bottom of the reactor, from where the gas was introduced into the venturi scrubber. In the venturi scrubber, the particles and tar removal process and cooling of the synthesis gas has taken place. The cooled synthesis gas was then filtered through a cotton wool filter before entering the gas meter and then combusted in a gas torch. After each experiment, the char was collected and weighted as the gasification char yield. During the gasification period, the temperature along reactor was displayed and recorded by a data logger in every minute. The gas volume was measured by a gas meter and gas flow rate calculated and also recorded by a data logger at every minute. The synthesis gas samples were analysed with an Agilent 490 micro-GC in every ~4 minutes using micro thermal conductivity detector ( $\mu$ -TCD).

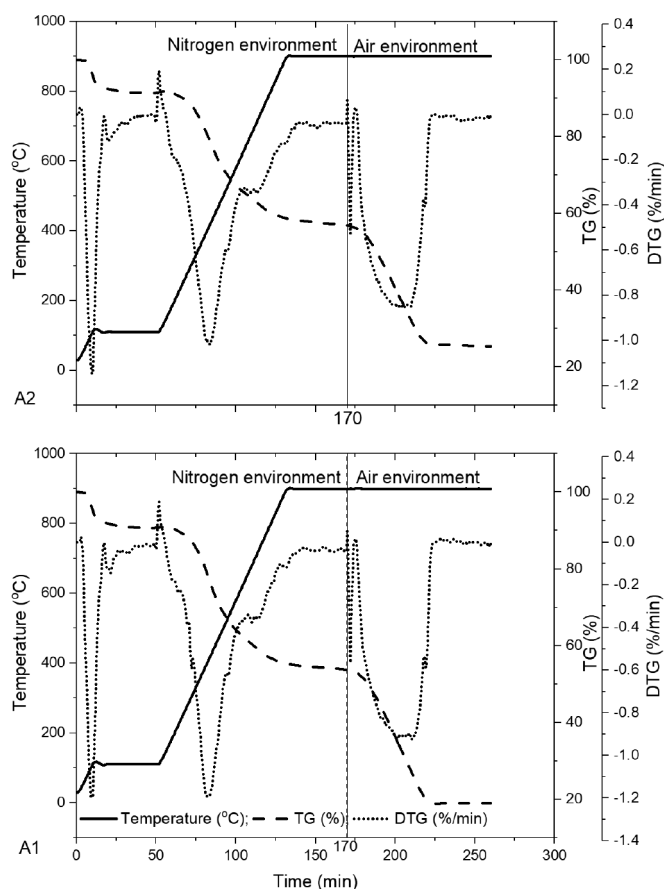


Fig. 1. Thermal analysis of coal samples (A2-top, A1-bottom)  
1. ábra Szénminták termokémiai vizsgálata

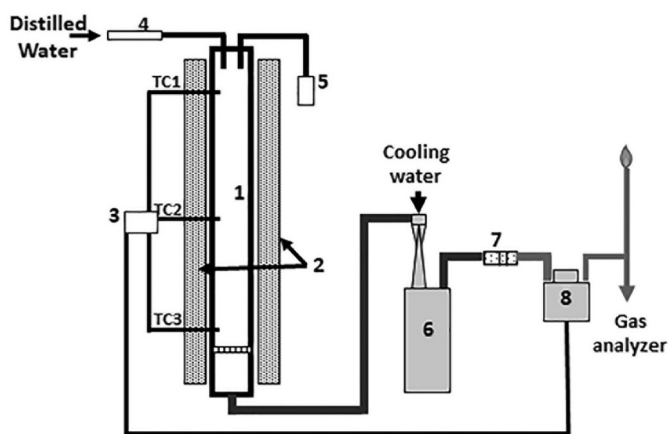
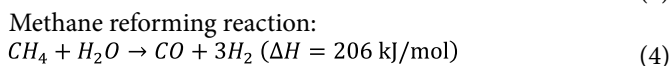
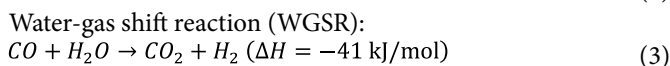
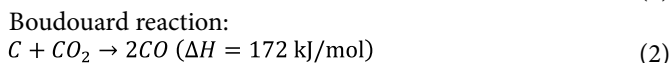
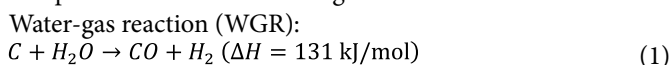


Fig. 2. The single stage lab-scale gasification system (1. Reactor, 2. Electrical heater, 3. Data logger, 4. Steam generator, 5. Manometer, 6. Venturi scrubber, 7. Cotton wool filter, 8. Gas meter, TC. thermocouple)

2. ábra Az egyfokozatú laboratóriumi elgázosításó rendszer (1. Reaktor, 2. Elektromos fűtőtest, 3. Adatgyűjtő, 4. Gőzfejlesztő, 5. Manométer, 6. Venturi mosó, 7. Vattaszűrő, 8. Gázmérő, TC. termoelem).

### 3. Results and discussions

The experiments focused on the gasification performance of coal char. Therefore, the analysis of pyrolysis gas was not included in this report. The gasification temperature was set to 700, 800, and 900 °C, with steam flow rate of 5 and 10 g/min for each temperature. The main chemical reactions within the steam gasification process that determines the syngas composition were the following:

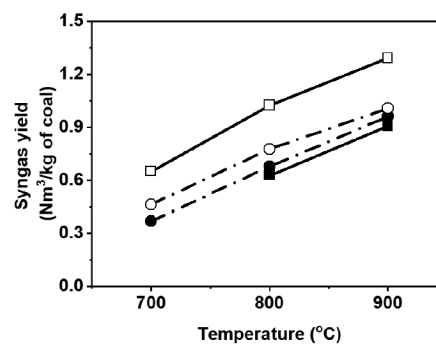
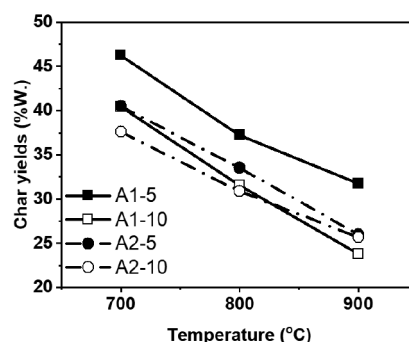


#### 3.1 Effect of temperature and steam flow rate on products distribution of gasification process

The effects of temperature and steam flow rate on gasification products are illustrated in Fig. 3. During the gasification experiments of A1 sample, the analysis of synthesis gas was absent in the experiment at 700 °C of gasification temperature and 5 g/min of steam flow rate due to technical problem. As consequently, further analysis for syngas composition and gasification performance (heating value, carbon conversion, and cold gas efficiency) are not include in the report. The char yields were calculated from the mass percentage of residual ash.

As the temperature increased, the char yields decreased, and the gas yields increased at all examined steam flow rate. In the case of A1 sample, the char yields declined sharply from 46.21 wt% and 40.39 wt% at 700 °C to 31.75 wt% and 23.78 wt% at 900 °C of gasification temperature, within the steam flow rate at 5 and 10 g/min, respectively. In the case of the A2 sample, from 40.44 wt% and 37.58 wt% at 700 °C decreased to 25.99 wt% and 25.68 wt% at 900 °C at the same steam flow

rate conditions. Regarding to the synthesis gas yields, the increase in gasification temperature resulted in the increasing of synthesis gas production per kg of coal. With the gasification experiment of A1 sample at 10 g/min of steam flow rate, the synthesis gas yields rose drastically from 0.65 Nm<sup>3</sup>/kg<sub>coal</sub> at 700 °C to 1.29 Nm<sup>3</sup>/kg<sub>coal</sub> at 900 °C of gasification temperature. In the gasification process of A2 sample, the gas yields increased significantly from 0.37 Nm<sup>3</sup>/kg<sub>coal</sub> and 0.46 Nm<sup>3</sup>/kg<sub>coal</sub> at 700 °C to 0.96 Nm<sup>3</sup>/kg<sub>coal</sub> and 1.01 Nm<sup>3</sup>/kg<sub>coal</sub> at 900 °C, within the steam flow rate at 5 and 10 g/min. It can clearly be seen that the lowest char yield was at 900 °C and 10 g/min of steam flow rate. The trends shows that the endothermic reactions (Eq. (1), (2), (4)) were strongly promoted with the higher gasification temperature [18]–[20].



3. ábra Az elgázosítási folyamat termékeinek eloszlása a hőmérséklet és a gőz áramlási sebessége szerint (5 g/perc - szilárd szimbólum és 10 g/perc - nyitott szimbólum; A1 - szilárd vonal és négyzet szimbólum és A2- szaggatott pontvonal és kör szimbólum).

Fig. 3 Products distribution of gasification process by temperature and steam flow rate (5 g/min-solid symbol and 10 g/min - open symbol; A1-solid line and square symbol and A2- dash-dot line and circle symbol)

At each gasification temperature, the higher steam flow rate resulted in the lower char yields and the higher gas yields. It was illustrated that the more addition of steam increased the rate of reactions of Eq. (1), Eq. (3), and Eq. (4) [21]. However, the effects of steam flow rate were different in the gasification process of sample A1 and A2. At 900 °C of gasification temperature, the effect of steam flow rate on char yields showed clearly for A1 sample, 31.75 wt% at 5 g/min and 23.78 wt% at 10 g/min. While that for A2 sample was relatively similar, 25.99 wt% at 5 g/min and 25.68 wt% at 10 g/min of steam flow rate and gasification temperature at 900 °C. The comparison trends were also presented in the change of synthesis gas flow rate. With higher temperature and steam flow rate the gasification process of A1 sample had a better char yield and higher volume of syngas production.

		Syngas composition (V/V%)		H <sub>2</sub>		CO		CH <sub>4</sub>		CO <sub>2</sub>					
		Steam flow rate (g/min)		5	10	5	10	5	10	5	10				
Samples	A1	Gasification temperature (°C)	700		60.88		5.34		2.25		23.12				
			800	57.6	58.96	14.29	11.83	2.11	1.36	18.42	19.84				
			900	53.91	55.83	23.78	22.17	1.43	0.85	13.14	14.42				
	A2	700		60.22		61.09		5.84		5.19		2.75	2.14	22.63	23.4
		800	58.02	58.49	13.28	12.61	1.81	1.49	18.71	19.48					
		900	53.06	55.46	27.05	22.74	0.97	0.93	11.45	14.25					

Table 2 Total synthesis gas composition by temperature and steam flow rate  
2. táblázat A teljes szintézisgáz összetétele hőmérséklet és gőzáramlás szerint.

### 3.2. Effect of temperature and steam flow rate on synthesis gas composition

Table 2 illustrates the effects of gasification temperature and steam flow rate on the syngas composition of total syngas volume. The main components of synthesis gas were H<sub>2</sub>, CO, CO<sub>2</sub>, and CH<sub>4</sub> with over 92 V/V% under all experiments.

As the gasification temperature increased from 700 to 900 °C, the total composition of H<sub>2</sub>, CO<sub>2</sub>, and CH<sub>4</sub> showed a decreasing trend in the experiments for both sample type. While CO concentration increased with the increase of gasification temperature. The trends could be results of the improved rate of endothermic reactions (water gas reaction- Eq. (1), Boudouard reaction-Eq. (2), and methane reforming reaction-Eq. (4)) [18], [19] therefore the more CO produced and the more CO<sub>2</sub> and CH<sub>4</sub> were consumed. The water-gas shift reaction- Eq. (3) is a light exothermic reaction consequently, it was reversed when the temperature was higher than the equilibrium temperature [20]. Therefore, it led to a slight drop in H<sub>2</sub> concentration.

The synthesis gas composition changed at different rates with the increase of temperature. In case of the gasification experiment of A1 sample at a steam flow rate of 10 g/min, the H<sub>2</sub> concentration decreased by 1.92 V/V% when temperature increased from 700 to 800 °C. While that was 3.13 V/V% when temperature increased from 800 to 900 °C. In case of the A2 sample at the same gasification conditions, these numbers were 2.6% and 3.03%, respectively. The trend was even more emphasised in the changing of CO and CO<sub>2</sub> concentrations.

As steam flow rate increased from 5 to 10 g/min in the gasification process, the H<sub>2</sub> and CO<sub>2</sub> concentration increased in both samples. While that of CO and CH<sub>4</sub> showed a decreasing trend at all temperature conditions. The higher steam flow rate increased the reaction rate of water gas reaction- Eq. (1), water gas shift reaction- Eq. (3) and methane reforming reaction- Eq. (4) [22]–[24].

The composition of the synthesis gas at higher gasification temperature presented a higher differentiation with the increased steam flow rate. At lower gasification temperatures (700 and 800 °C) the difference in the changing of H<sub>2</sub>, CO and CO<sub>2</sub> composition were not as significant when the steam flow rate was changed from 5 to 10 g/min. However, the difference was significant at a gasification temperature of 900 °C. In the case of the A2 sample, the H<sub>2</sub> concentration increased only by 0.87 V/V% at 700 °C and 0.47 V/V% at 800 °C when steam flow rate from was increased from 5 to 10 g/min. While that change was 2.4 V/V% at 900 °C. Similar trends were observed in the variation of CO and CO<sub>2</sub> concentrations. This could be the

consequence of the change in the reaction rate of water gas shift reaction- Eq. (3). In the one hand, the higher concentration of steam enhanced the water gas shift reaction to produce a higher concentration of CO<sub>2</sub> and H<sub>2</sub>. In the other hand, the higher gasification temperature led to the reverse direction of the water gas shift reaction. Therefore, the temperature had a significant effect on water-gas shift reaction when the gasification temperature was over the equilibrium temperature.

### 3.3. Effect of temperature and steam flow rate on gasification performance

The total of H<sub>2</sub>/CO ratio, the low heating value of synthesis gas, carbon conversion and cold gas efficiency by gasification temperature and steam flow rate are shown in Fig. 4.

The low heating value of synthesis gas is defined as follow:

$$LHV_{\text{synthesis gas}} = \left( \frac{H_2\% * 10.783 + CO\% * 12.633}{+CH_4\% * 35.883} \right) / 100 \quad (5)$$

The carbon conversion efficiency of the gasification process is calculated by:

$$\eta_{\text{carbon conversion}} = \left( \frac{12 * Y * (CO\% + CO_2\% + CH_4\%)}{22.4 * C\%} \right) * 100 \quad (6)$$

The cold gas efficiency is calculated by:

$$\eta_{\text{cold gas}} = \left( \frac{LHV_{\text{synthesis gas}} * Y}{LHV} \right) * 100 \quad (7)$$

in which, H<sub>2</sub>%, CO%, CO<sub>2</sub>% and CH<sub>4</sub>% are volume concentration of H<sub>2</sub>, CO, CO<sub>2</sub> and CH<sub>4</sub> in synthesis gas V/V%. Y is the yield of synthesis gas Nm<sup>3</sup>kg<sub>coal</sub><sup>-1</sup>. C% is carbon content of raw material in weight percentage wt%.

The H<sub>2</sub>/CO ratio, heating value of synthesis gas, carbon conversion and cold gas efficiency were obviously driven by the components in synthesis gas (H<sub>2</sub>, CO, CO<sub>2</sub> and CH<sub>4</sub>). The results showed that the CO concentration increased drastically with the increased gasification temperature. It led to a decline in the H<sub>2</sub>/CO ratio and an increasing trend in the low heating value of synthesis gas, carbon conversion and cold gas efficiency. Furthermore, the increase in gasification temperature improved the synthesis gas yield in the gasification process. It likewise enhanced carbon conversion and cold gas efficiency.

As an increase in gasification temperature, the H<sub>2</sub>/CO ratio showed a converging trend in both samples. In the case of A1 sample at 900 °C of gasification temperature, the H<sub>2</sub>/CO ratio was 2.27 at 5 g/min and 2.52 at 10 g/min of steam flow rate. In the case of A2 sample that was 1.96 and 2.44, respectively. However, this trend was only in the case of the A2 sample for the carbon conversion and cold gas efficiency.

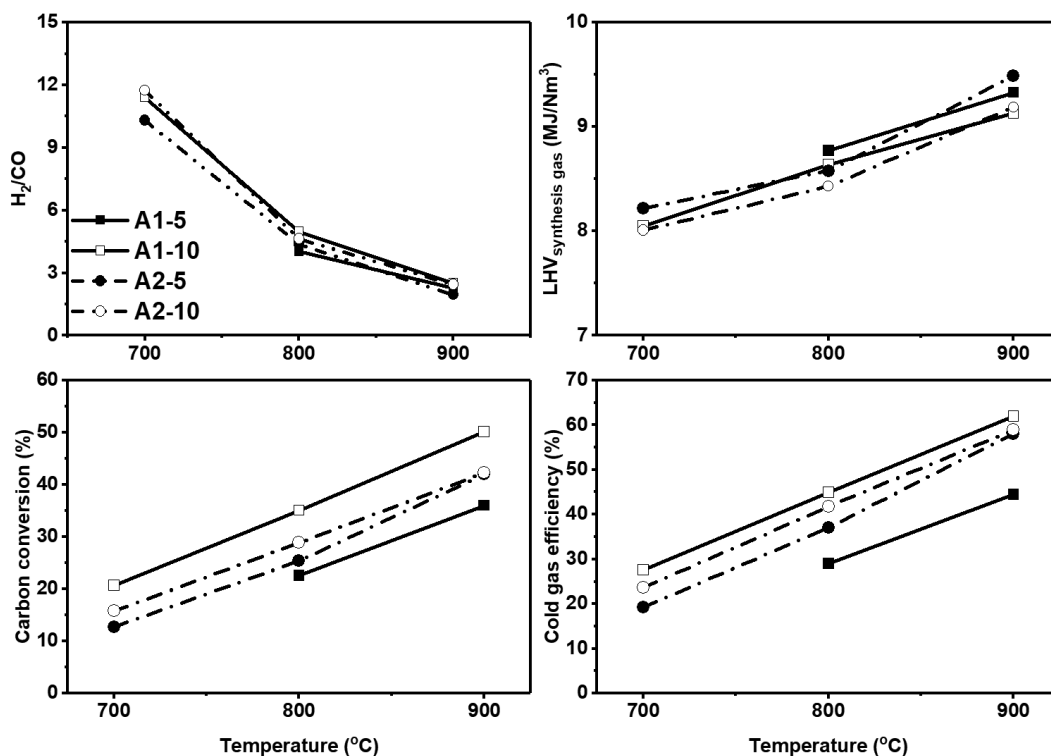


Fig. 4 Gasification performance by temperature and steam flow rate (5 g/min-solid symbol and 10 g/min – open symbol; A1-solid line + square symbol and A2- dash-dot line + circle symbol)

4. ábra Elgázosítás hőmérséklet és gőzarámlás szerint (5 g/min - szilárd szimbólum és 10 g/min - nyitott szimbólum; A1 - szilárd vonal + négyzet szimbólum és A2- szaggatott pontvonal + kör szimbólum).

As an increase in steam flow rate from 5 g/min to 10 g/min, the CO concentration decreased at all temperature conditions. As a result, the H<sub>2</sub>/CO ratio at 5 g/min was lower than that at 10 g/min of steam flow rate. While the low heating value of synthesis gas at 5 g/min was higher than that at 10 g/min of steam flow rate. The highest low heating value of synthesis gas reached at 5 g/min of steam flow rate and 900 °C of gasification temperature for both samples, 9.33 MJ/Nm<sup>3</sup> for A1 sample and 9.49 MJ/Nm<sup>3</sup> for A2 sample. In the other hand, the higher steam flow rate led to an increase in CO<sub>2</sub> and synthesis gas yield. Therefore, the carbon conversion and cold gas efficiency were increased with the increase of steam flow rate at all temperature conditions.

Regarding to the effect of samples in gasification performance, the A1 sample was shown a more promising starting material for gasification than the A2 sample in term of carbon conversion and cold gas efficiency, especially at 900 °C of gasification temperature. In case of A2 sample, the carbon conversion efficiency was 41.98% at 5 g/min and 42.28% at 10 g/min of steam flow rate. While that was 36.01% and 50.12%, respectively, for A1 sample. The highest cold gas efficiency was 61.97% in the case of A1 sample at 900 °C of gasification temperature and 10 g/min of steam flow rate.

#### 4. Conclusions

The two different coal samples were gasified at steam flow rates of 5 and 10 g/min and gasification temperatures of 700, 800, and 900 °C in a single stage fixed bed gasifier within the non-moving of material. The highest synthesis gas yields of

samples were achieved at 900 °C of gasification temperature and 10 g/min of steam flow rate. The synthesis gas yields from the gasification process of A1 sample were higher than that of A2 sample at all experiment temperature conditions within 10 g/min of steam flow rate. The gasification temperature had a significant effect on the concentration of CO and CO<sub>2</sub>. The higher gasification temperature led to a decrease trend in the H<sub>2</sub>/CO ratio due to the increasing CO concentration. The range of H<sub>2</sub>/CO ratio were from 1.96 to 2.52 at 900 °C of gasification temperature, which has a great potential for chemical application process. Furthermore, the A1 sample showed the better results in the carbon conversion and cold gas efficiency within 10 g/min of steam flow rate at all gasification temperature case. Based on the experiment results, the gasification process is a viable method to utilization the Hungarian low rank coals for the higher value application and the magnetite heavy suspension separation could further increase the carbon conversion rate and the cold gas efficiency.

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