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HYDROGEN PRODUCTION BY STEAM GASIFICATION OF HUNGARIAN BROWN COAL CHAR

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Abstract
Presently, the hydrogen is mainly generated from fossil fuels (natural gas and coal). The thermochemical conversion processes of carbon-based materials to hydrogen-rich gas has been widely described in the literatures. In which, the steam gasification is highly recommended for this purpose. The experimental steam gasification of Hungarian brown coal char was conducted in a single stage fixed-bed reactor with 80 mm of inner diameter and 1,200 mm of effective length. The experiments were carried out at 700, 800 and 900 °C of gasification temperature within 5 g/min of steam flow rate and 5, 7.5 and 10 g/min of steam flow rate within 900 °C of gasification temperature. The aim of this study is to determine the performance of Hungarian brown coal char during the gasification with the variation of gasification temperature and steam flow rate, as well as the hydrogen production during the gasification process. The gasification temperature plays an important role in the conversion process in the gasifier, as well as the change in the synthesis gas composition. The introduction of higher steam flow rate can also improve the reduction process. The highest total hydrogen yield of 1.8 Nm3 was gained at 900 °C of gasification temperature and 10 g/min of steam flow rate for 3 kg of initial coal.

Keywords: *thermochemical processes, gasification process, coal char gasification, synthesis gas*

1. Introduction

In 2020, more than 60% of the global electricity generation had been achieved by the fossil fuels (oil, natural gas, and coal) (BP, 2021). However, the combustion process of fossil fuels in the power generation brings a massive problem to both human well-being and environmental pollution (Mukhopadhyay and Forssell, 2005; Oliveira et al., 2011; Perera, 2017). Currently, hydrogen is considered as a promising candidate for a green future energy. Using hydrogen as an alternate fuel has received significant attention in recent years. Hydrogen is well-known as the most abundant element, the lightest element with highest heating value content, non-toxic, and no-hazardous product in the energy conversion process (Abe et al., 2019). In the term of application, hydrogen can be used in internal combustion engines (Deb et al., 2015; Mohammadi et al., 2007; Verhelst, 2014), gas turbine (Valera-Medina et al., 2019), fuel cells (Breen et al., 2002; Fierro et al., 2002; Galvagno et al., 2013; Soler et al., 2007), and as feeding material for chemical process (Bicer et al., 2016; Galindo Cifre and Badr, 2007).

Unlike fossil fuels, hydrogen is not a primary energy source (Acar and Dincer, 2020). Therefore, it is produced from hydrogen containing materials, such as water, biomass, natural gas, or fossil fuels. There are several approaches for hydrogen production. They can be generally grouped into electrolysis process, thermochemical process, photolytic and biological process (Holladay et al., 2009; Yukesh Kannah et al., 2021). In which, the thermochemical process can be listed as natural gas reforming, pyrolysis and gasification of fossil fuels and biomass, or biomass-based liquid reforming. Currently, the hydrogen production is particularly depended on the fossil fuels. In 2020, nearly 80% of total hydrogen demand was generated from natural gas and coal (IEA, 2021).

In the case of carbon-based solid fuel (coal, biomass, municipal solid waste), pyrolysis and gasification are used as major pathways to convert them into the synthesis gas. Pyrolysis is a thermal decomposition process in the non-oxidation reactant (as air or oxygen). The main products of pyrolysis process are char, condensate liquid, and syngas. Unlike the pyrolysis process, the gasification process is a thermochemical process, using heat and gasifying agents to convert carbonaceous containing material into synthesis gas. There are many types of gasifying agents used during gasification process, such as air, oxygen, carbon dioxide, steam, super critical steam, and their combination. The selectivity of gasifying agents affects to the synthesis gas composition and the heating value of produced gas (Yang and Chen, 2015). In which, the steam gasification process is highly recommend for the hydrogen-rich gas production (40 ÷ 62 %V) (Howaniec et al., 2011; Li et al., 2017; Xiao et al., 2018).

In this study, the steam gasification characterization of Hungarian brown coal char was investigated in a single stage fixed-bed reactor. The purpose of this study is to determine the influence of gasification temperature and steam flow rate on products distribution, synthesis gas composition, energy, and chemical point of view, as well as the hydrogen production.

2. Material and methods

2.1. Materials

In this study, a Hungarian brown coal was adopted as starting material. The coal sample was directly used as input material without any pre-heat treatment processes. The particle size distribution was from 10 to 20 mm of diameter. *Table 1* presents the results of the ultimate analysis, proximate analysis, and heating value analysis of the coal sample. In which, the elemental composition of coal sample was examined under the standard ISO 29541:2010 Solid mineral fuels – Determination of total carbon, hydrogen, and nitrogen content – Instrumental method within a Carlo Erba EA 1108 analyser. The proximate analysis of coal sample was performed by thermal gravimetric analysis in a MOM Derivatograph-C type with the N_2 ambient and air ambient. The high heating value of the coal samples was 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.

***Table 1**. Elemental analysis, proximate analysis, and heating value analysis*

	C_{db} (wt.%)	H_{db} (wt.%)	N_{db} (wt.%)	S_{db} (wt.%)	O^{*}_{db} (wt.%)
Brown coal	44.68	2.45	0.75	5.14	19.24
	M (wt.%)	V (wt.%)	FC (wt.%)	A (wt.%)	HHV_{ad} (MJ/kg)
Brown coal	12.05	34.63	28.92	24.40	15.77
Db: dry basis; ad: air dry; *: by difference					

2.2. Methods

The lab-scale fixed bed gasification system is depicted in *Figure 1*. A tubular gasifier was used for the pyrolysis and gasification process. The gasifier is made from heat resistant steel with 80 mm of inner diameter and 1,200 mm of effective length. The heat requirement was supplied by a Carbolite 12/900 electrical furnace. Initially, there were 3 kg of coal loaded in the gasifier. The gasifier was then heated up to the desired temperature. After the pyrolysis process finished, the steam was introduced from the steam generator at the top of gasifier. In each experiment, the coal char was gasified for 6 hours. The produced synthesis gas exiting at the bottom of gasifier was cleaned using a venturi scrubber for the particle and tar removal and cooling of the synthesis gas. Gas samples were collected every 30 minutes during gasification process. The synthesis gas composition was analysed with a Dani Master GC, equipped with a flame ionization detector for CH_4 and hydrocarbons compositions, a thermal conductivity detector for H_2, CO and CO_2 compositions. After each experiment, the remaing char was removed and weighed for further analysis. The experiments stated above were performed at 700, 800, and 900 °C of gasification temperature with 5, 7.5 and 10 g/min steam flow rate at 900 °C of gasification temperature.

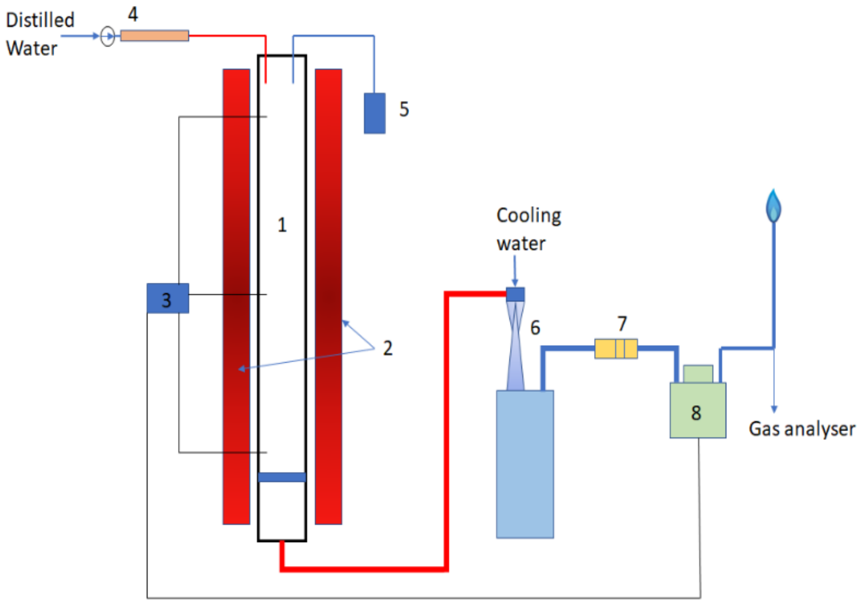

Figure 1. *The single-stage lab-scale gasification system; (1. Reactor, 2. Electrical heater, 3. Data logger, 4. Steam generator, 5. Manometer, 6. Venturi scrubber, 7. Cotton wool filter, 8. Gas meter)*

3. Results and discussion

In this research, the gasification process was conducted after the pyrolysis process. Therefore, the synthesis gas anaylsis during the pyrolysis process was excluded from the dataset. The synthesis gas composition of the gasification process at ambient pressure is the result of the main reactions as follows:

Boudoard reaction:

$$C + CO_2 \rightarrow 2CO \quad (1)$$

Steam reforming reaction:

$$C + H_2O \rightarrow H_2 + CO \quad (2)$$

Water-gas shift reaction:

$$CO + H_2O \rightarrow H_2 + CO_2 \quad (3)$$

Methane reforming reaction:

$$CH_4 + H_2O \rightarrow CO + 3H_2 \quad (4)$$

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

$$LHV_{syngas} = \frac{H_2\% * 10.783 + CO\% * 12.633 + CH_4\% * 35.883}{100} \left(\frac{MJ}{Nm^3}\right) \quad (5)$$

3.1. Effects of temperature

The effects of gasification temperature on gasification products are shown in *Table 2*. The effects of gasification temperature were examined at 700, 800, and 900 °C within 5 g/min of steam flow rate. The char yields were calculated by the weight percentage of the residual ash divided by the weight of initial fuel. The synthesis gas yields were determined by the total gasification syngas divided by the weight of initial fuel.

Table 2. *Effects of gasification temperature on gasification products*

No.	Temperature (°C)	Steam flow rate (g/min)	Total char yield (wt.%)	Total syngas yield (Nm^3/kg of coal)
1	700	5	49.37	0.41
2	800	5	38.32	0.65
3	900	5	36.39	0.98

As is observed, the higher temperature of gasification improved the gasification process by significantly decreasing the char yield from 49.37 wt.% at 700 °C to 36.39 wt.% at 900 °C within 5 g/min of steam flow rate. While the highest synthesis gas yield was 0.98 Nm^3/kg of coal reached at 900 °C of gasification temperature.

Figure 2 illustrates the composition, H_2/CO ratio, and low heating value of synthesis gas during the coal char steam gasification at 5 g/min of steam flow rate. Typically, the H_2 concentration remained roughly 45–55 V% during the gasification process. A study published by Pettinau et al. (Pettinau et al., 2014) presents the results of gasification carried out in a lab-sclae updraft reactor at 800 °C with Hungarian brown coal. The H_2 concentration was around 60 ÷ 65 V% with the 5.2 g/min of pure steam introduction. In the case of using the O_2/steam mixture, the CO concentration was dominant with 35 ÷ 40 V% and the H_2 concentration was only around 35 V%. The highest H_2 concentration was produced at 700 °C of gasification temperature. There were reverse trends in the CO and CO_2 concentration. With the increase in gasification temperature, the concentration of CO drastically increased, while that of CO_2 declined. During the gasification process, there was a small amount of CH_4, and the present of longer chain hydrocarbons were under detectable level. The CH_4 concentration decreased below 2 % in the gasification process. The obtained trends could be explained by the higher temperature favouring the endothermic reactions *[equation (1), (2), (4)]* in the gasification process (Chang et al., 2020; Chen et al., 2017; Yuan et al., 2015), which led to higher CO rate production.

From an energetic point of view, the lower heating value (LHV) is one of the most important properties of synthesis gas and the H_2/CO ratio is an important factor from chemical point of view. The H_2/CO ratio was in a range of 6 ÷ 8 in case of 700 °C of gasification temperature. At the starting period of gasification process, the H_2/CO ratio was 2.6 and 1.3 in cases of 800 and 900 °C, respectively. After that, they increased over time reaching the ratio of 4.5 and 3.6 at the end of experiments. The LHV of syngas remained roughly around 8 MJ/Nm^3 in the case of 700 and 800 °C. While the highest low heating value of synthesis gas was observed at 900 °C of gasification temperature. But it was slightly decreased during the second haft of gasification time.

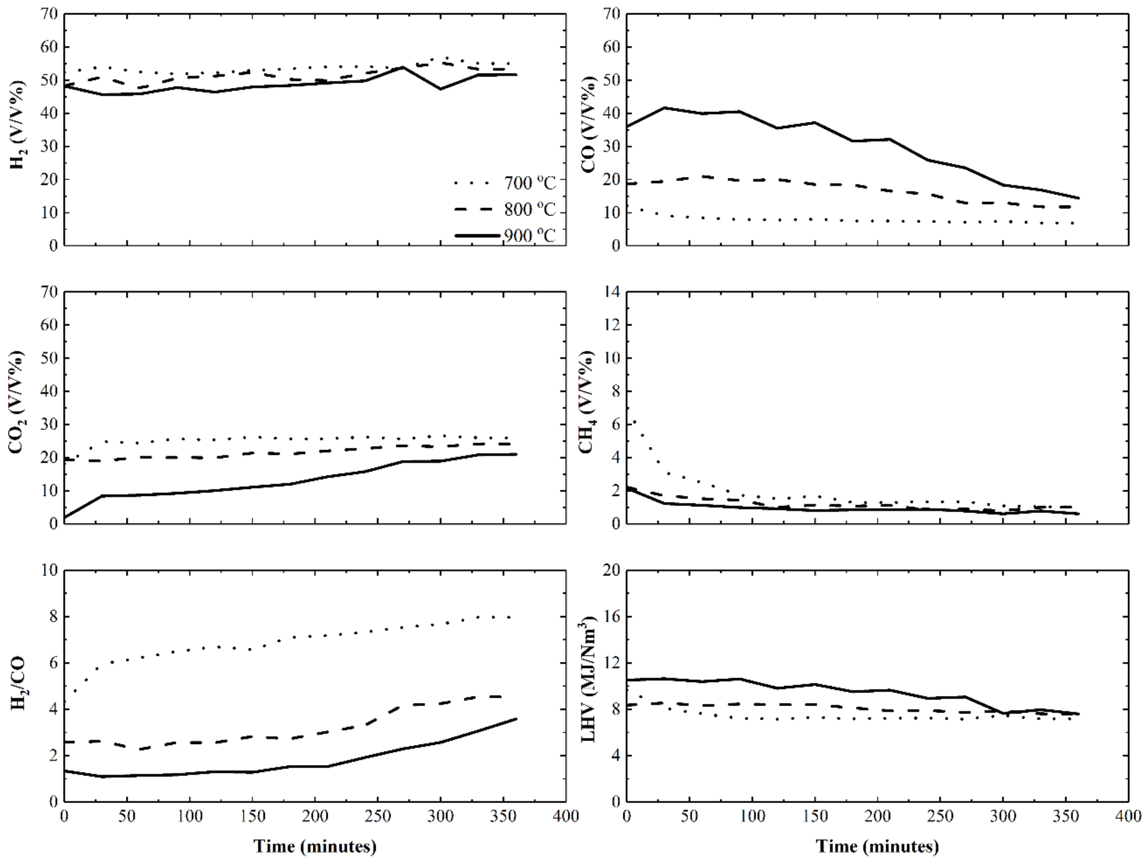

Figure 2. *The effects of gasification temperature on syngas composition, H_2/CO ratio and LHV*

The effect of temperature on the total hydrogen yields is depicted in *Figure 3*. The total hydrogen yield was only 0.66 Nm^3 at 700 °C of gasification temperature for 3 kg of initial coal. That value was doubled at 900 °C. This result indicates that the gasification temperature has a significant effect on the hydrogen yield at similar steam flow rate. The significant increase of H_2 yield with the increase of gasification temperature (600 ÷ 900 °C) was measured by Yang et al. (Yang et al., 2018).

3.2. Effects of steam flow rate

The gasification experiments were conducted at 5, 7.5 and 10 g/min of steam flow rate. The influence of steam flow rate on the char yields and synthesis gas yields is showed in *Table 3*. The results indicated that the reduction process was promoted with the increase of steam flow rate. The char yields decreased from 36.39 wt.% at 5 g/min of steam flow rate to 32.15 wt.% at 10 g/min of steam flow rate. While the total syngas yields increased from 0.98 to 1.24 Nm^3/kg of coal, respectively.

The effects of steam flow rate on the synthesis gas composition, H_2/CO ratio and LHV are presented in *Figure 4*. There is no significant variation observed in H_2 concentration as the steam flow rate increased from 5 to 10 g/min. The H_2 concentration remained around 50% during the first haft of gasification time at all steam flow rates. Then it increased slightly for the remaining time of the gasification. It is reasonable with the experiment results in Ref. (Yang et al., 2018), the variation of H_2 concentration at 50 ÷ 60% with the increase of steam volume percentage. Regarding the CO and CO_2 concentrations, they were rather stable in the first 60 minutes at nearly 40 V% of CO and 10 V% of CO_2 in all cases of steam flow rate. After that there was a decreasing trend in the CO concentration, which resulted in the increase of CO_2 concentration during the gasification process. The CH_4 concentration during the gasification process showed a slight decrease, under 1 V% at all steam flow rate conditions. However, the differences in the CH_4 concentrations amongst steam flow rates were not obvious. These variations in the synthesis gas composition could be explained by the higher steam quantities, that led to the higher partial steam pressure in the gasifier, which in turn resulted in the product formation described in *equation (2), (3),* and *(4)* (Li et al., 2010; Lin and Weng, 2017; Mondal et al., 2005).

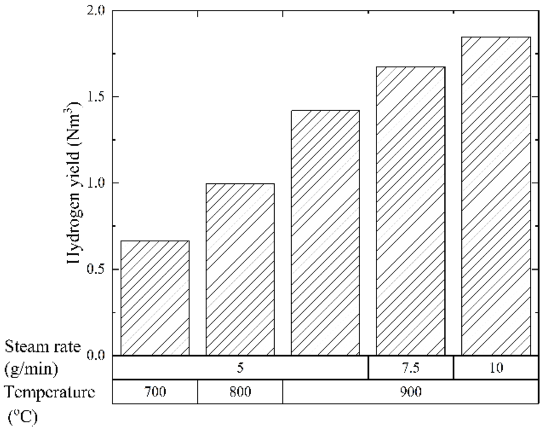

Figure 3. *Hydrogen yields*

Table 3. *Effects of steam flow rate on gasification products*

No.	Temperature (°C)	Steam flow rate (g/min)	Total char yield (wt.%)	Total syngas yield (Nm^3/kg of coal)
1	900	5	36.39	0.98
2	900	7.5	35.04	1.10
3	900	10	32.15	1.24

As mentioned above, the CO concentration was nearly constant in the first 60 minutes at every steam flow rate. This resulted in a stable H_2/CO ratio as well, around 1.2. The H_2/CO ratio reached at 3.6, 7, and 9 by the end of experiments when 5, 7.5, and 10 g/min of steam flow rate were applied, respectively. As decreased in the CO concentration, the LHVs of synthesis gas declined as well in all experiments. At the beginning of experiments, the LHVs were at the highest values, around 10.5 MJ/Nm^3. And then, they tended to decrease over time.

The hydrogen yields as a function of steam flow rate are illustrated in *Figure 3*. As is observed, the total hydrogen yields increased when the steam flow rate varying from 5 to 10 g/min. The highest total hydrogen yield was 1.8 Nm^3 at 10 g/min of steam flow rate for 3 kg of initial coal.

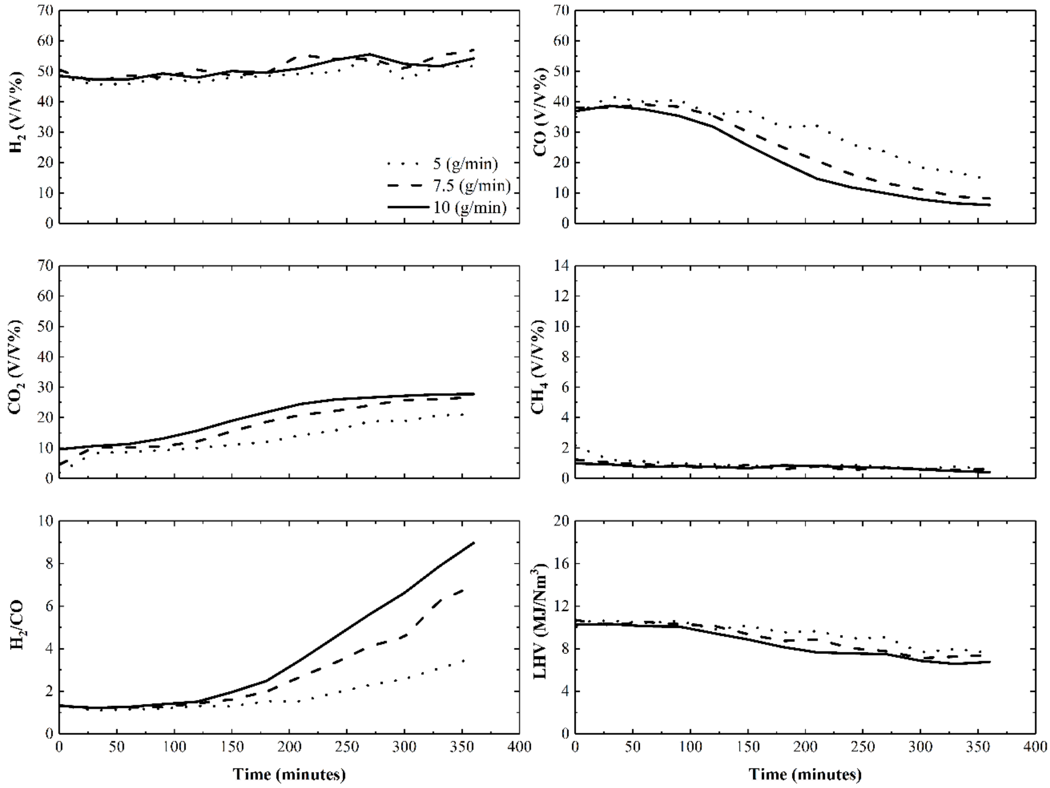

Figure 4. *The effects of steam flow rate on syngas composition, H_2/CO ratio and LHV*

4. Summary

The Hungarian brown coal char was gasified in a single stage fixed bed reactor at 700, 800 and 900 °C of gasification temperature and 5, 7.5 and 10 g/min of steam flow rate. The gasification temperature plays an important role in the conversion process in the gasifier, as well as the change in the synthesis gas composition. The higher gasification temperature contributes to a significant increase of the hydrogen yield. The introduction of higher quantity of steam can also improve the reduction process and the hydrogen yield as well. However, the excess steam can reduce the gasification temperature and then decrease the gasification efficiency. The highest total hydrogen yield of 1.8 Nm^3 was gained at 900 °C of gasification temperature and 10 g/min of steam flow rate for 3 kg of initial coal.

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