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INVESTIGATION OF MULTI-STAGE COAL GASIFICATION FOR ENHANCED CONTROL OF SYNTHESIS GAS COMPOSITION

A Thesis Booket

By

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1. Introduction

In fact, the International Energy Agency's Global Energy and CO_2 status report showed that the global greenhouse gas (CO_2) emission reached a historic highest level in 2018, approximately 33.1 Gt CO_2 , by 1.7 % higher than that of 2017 [1]. The main reason is the ever-increasing energy demand in developed and developing countries. Among the emission sources, the utilisation of coal in energy generation contributed the highest amount, nearly 44.1% of the total CO_2 emission, especially in coal-fired power generation. On one hand, it cannot be denied that electricity generation has been highly dependent on fossil fuels, like coal, at least until the half of this century. But on the other hand, the use of traditional technologies in coal power generation will lead to even higher emission levels. Clean coal technology (CCT) could be one of the solutions to this conflict.

CCT could be grouped into two main categories, environmental protection and global warming [2]. Environmental protection includes NO_x combustion and flue gas treatment. Global warming is divided into high-efficiency cycle, CO_2 capture, and biomass co-combustion technology. Presently, the gasification of coal is considered the centre of CCTs.

Gasification is an incomplete combustion process of coal or another solid feedstock (biomass, municipal solid waste) [3]. The primary goals of gasification are, firstly, to convert the entire non-ash content of the feedstocks into gas, and to produce the gas with the highest heating value as much as possible. The main products of the gasification process are gas, ash, and tar.

The gasification produced gas could be used either as a base material in chemical synthesis processes or as fuel in the power generation [4]. The advantages of the gasification process can be listed as:

- The high flexibility of synthesis gas products (power and chemical) application
- The carbon dioxide emission rate is lower, even if the syngas is used as fuel in the further combustion process.
- The sulphur released from steam gasification is in form of H₂S rather than SO₂, which can be removed from the synthesis gas and can be used in Sulphur and Sulphur acid production, thus, producing a valuable product as a by-product.
- Nitrous oxides are primarily converted into ammonia, which can be removed from the synthesis gas at a lower cost.
- Emission rates of furan and dioxin are significantly lower compared to combustion.
- The total volume of gas that must be cleaned after gasification is lower than that of combustion.

2. Gasification process – Literature review

2.1. Principle of the gasification process

Generally, gasification is a thermochemical process, in which using heat and gasification agents carbon-based materials (coal, biomass, MSW, natural gas, natural oil...) can be converted into a combustible or synthesis gas. The gasification mechanism is illustrated in Figure 2-1.

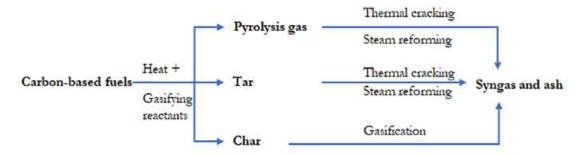


Figure 2-1. Gasification mechanism

During the gasification process, the principal chemical reactions may include species such as carbon, oxygen, carbon dioxide, carbon monoxide, hydrogen, steam, and methane. The main reaction of the gasification process can be listed as [5]:

Combustion reaction:

$$C + \frac{1}{2}O_2 \to CO \ (\Delta H_{298} = -111 \ kJ/mol)$$
 (2-1)

$$CO + \frac{1}{2}O_2 \to CO_2 \ (\Delta H_{298} = -283 \ kJ/mol) \tag{2-2}$$

$$H_2 + \frac{1}{2}O_2 \to H_2O \; (\Delta H_{298} = -242 \; kJ/mol) \tag{2-3}$$

Boudouard reaction:

$$C + CO_2 \leftrightarrow 2CO \ (\Delta H_{298} = 172 \ kJ/mol) \tag{2-4}$$

Water-gas reaction:

$$C + H_2 O \leftrightarrow CO + H_2 (\Delta H_{298} = 131 \, kJ/mol)$$
 (2-5)

Methane reaction:

$$C + 2H_2 \leftrightarrow 2CH_4 \ (\Delta H_{298} = -75 \ kJ/mol) \tag{2-6}$$

Water-gas shift (WGS) reaction:

$$CO + H_2O \leftrightarrow CO_2 + H_2 (\Delta H_{298} = -41 \, kJ/mol)$$
 (2-7)

Steam methane reforming reaction:

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 (\Delta H_{298} = 206 \, kJ/mol)$$
 (2-8)

Reaction (2-1), (2-4), (2-5), (2-6) describe how char is gasified by oxygen, carbon dioxide, steam, and hydrogen. The reactions, where the enthalpy change is a negative value, are exothermic reactions. While the reactions with the positive enthalpy value are endothermic reactions. The heat requirement for the endothermic reaction is supplied from the partial combustion of feedstocks, known as the direct gasification (autothermal gasification), or the external heat source – the indirect gasification (allothermal gasification).

2.2. Review of multi-stage fixed bed gasification process

The most valuable benefit of the gasification process is the great flexibility of synthesis gas utilisation, especially in the chemical process. Regarding the control of H_2 /CO ratio during the gasification process, there are several effective ways in changing the gasification parameters (temperature, pressure, etc.), the selection of reactant types and their ratio, the utilisation of catalysts, and the gasifier setup modification. The multi-stage gasification is well-known as a promising approach in the enhancement of process efficiency and quality of product gas as well. In the gasification process, the sub-processes include drying, devolatilization, partial

oxidation, and reduction process. In multi-stage gasification, the separation or combination of these subprocesses is highly dependent on the fuel properties such as reactivity, ash content, sulphur content, and volatile content [6]. The multi-stage gasification can be commonly categorised as single-line and double-line process, are shown in Figure 2-2.

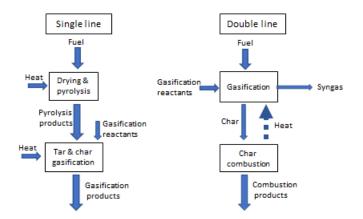


Figure 2-2. Single line and double line gasification process [6]

In the term of the single-line multi-stage gasification process, Peder Brandt et al. [7] studied the tar reduction of biomass air-gasification in the two-stage gasifier (Viking two-stage gasifier). Firstly, the biomass fuel was pyrolyzed in an allothermal auger reactor with the exhaust heat from the flue gas of a gas engine. After that, the pyrolysis products (including pyrolysis char and gas) were transported to an autothermal downdraft fixed-bed gasifier for the partial oxidation process of volatile matter and char reduction processes, as shown in Figure 2-3. The air was supplied from the top of the second stage reactor above the char bed of reduction zone for the partial oxidation process. The pyrolysis temperature was 420 and 600 °C at two different test series. While the temperature in the partial oxidation zone was 1050 and 1100 °C, respectively. Within the biomass air gasification in the two-stage gasifier, the tar content was less than 15 mg/Nm³ without heavy tar. Using the same Viking two-stage gasifier above, Jesper Ahrenfeldt et al. [8] experimented with wood chips, a range of 550÷600 °C of pyrolysis temperature and 1100-1300 °C at the oxidation zone. The experiment results showed that there were only minor amounts of naphthalene in the raw synthesis gas (0.1 mg/Nm³). The experiments were performed using the same Viking two-stage gasifier in another study [9] at 550-600 °C of pyrolysis temperature, 1150-1300 °C of oxidation temperature and O₂-CO₂ mixture as a reactant. It concluded that the synthesis gas during the O₂-CO₂-blown experiment gained a higher quality than the typical air-blown mode at <11 mg/Nm³ of tar content and <3 ppm of Sulphur content. From the chemical point of view, the H_2/CO ratio was around 1.76 in the air-blown mode. While there was a significant increase in CO concentration due to the high CO₂ concentration in the reactant. It led to a decreasing trend in H_2 /CO ratio, a range of 0.8÷0.89 in the case of 21 V/V% O2-CO2-blown experiments and 1.11÷1.14 in the case of 25 V/V% O2-CO2 blown experiments. Zhiqi Wang et al. [10] studied the biomass gasification process in a pilot-scale two-stage gasifier with oxygen-enriched air. During the stable operation time of the gasification process, the heat demand for the pyrolysis process was supplied from the flue gas of a syngas burner. It helped to retain the pyrolysis temperature around 350 and 450 °C. With an increase in O₂ concentration, the total content of H₂ and CO increased in all cases of experiments. The H_2/CO ratio was around 1 with the O₂ concentration at 50÷100 V/V%.

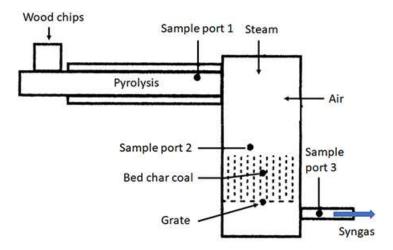


Figure 2-3. Two-stage gasification process

2.3. Scientific gaps and objects of research

The previous studies on the gasification process of Hungarian low rank coals have identified a scientific gap in investigating the synergy effects of gasification temperature and S/C ratio on both the gasification products and syngas composition.

Although several concepts of single line multi-stage gasification processes have been developed since the 1990's, these studies primarily focused on reducing tar formation during biomass gasification in multistage reactors. The produced gas generated was predominantly used for combined heat and power systems. Consequently, the existing knowledge gap is the performance of multi-stage gasification process for low rank coals, specifically in achieving the desired H₂/CO ratio suitable for chemical applications.

The general objective of this study is to increase the knowledge on a new advanced process of multistage fixed bed gasification using steam as the reactant.

Specific objectives are:

- Fully demonstration of multi-stage fixed bed gasification with the continuous fuel line.
- To study the effects of gasification temperature on the multi-stage gasification process, such as gasification products, syngas quality, carbon conversion, as well as gasification efficiency.
- To study the effects of steam flow rate on the multi-stage gasification process, such as gas quality, tar content, carbon conversion, as well as gasification efficiency.
- To generate the optimisation conditions of the operation parameters for H₂/CO ratio.
- The gasification performance of coal samples from gravity separation process.

3. Materials and experimental apparatuses

3.1. Materials

The results of proximate, elemental, and heating value analysis of samples from separation process are presented in Table 3-1, in which Corg-original coal sample, C1 and C2- coal samples from separation process.

	-	Fuel name		
		Corg	C1	C2
Proximate analysis-wt%	Moisture	10.37	6.97	8.59
	Volatile	32.33	43.93	40.96
	Fixed carbon	25.22	35.80	32.25
	Ash	32.08	13.30	18.20
Elemental analysis-wt%	Ν	0.72	0.83	0.84
	С	35.60	51.95	48.17
	Н	3.39	4.68	4.39
	S	3.07	5.14	5.38
	O (by diff.)	25.14	37.40	41.22
Lower heating value (MJ/kg)		14.02	20.62	18.62

Table 3-1. Proximate, elemental, and heating value analysis of samples (as-received basis)

3.2. Multi-stage gasification apparatuses

The design and construction of the single line multi-stage gasifier started in 2020. The first trial operation was in September 2021. Both the construction and the operation of the multi-stage gasifier have required a large contribution from the people of the Institute of Energy and Quality, University of Miskolc. The schematic diagram of the multi-stage gasification system is depicted in Figure 3-1.

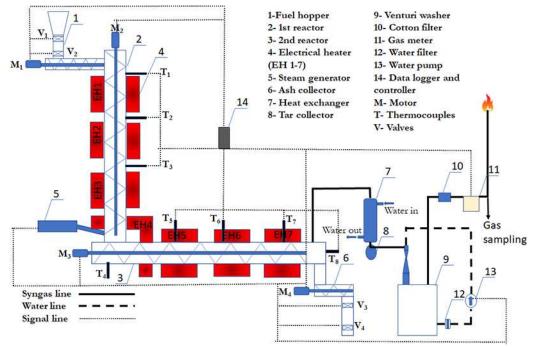


Figure 3-1. Schematic diagram of the multi-stage gasification system

In each experiment, the reactors were heated up to the desired temperature. When the reactor reached the set temperature, the starting material was fed from the fuel hopper to the first stage of the gasifier through a screw conveyor. In the first stage of the gasifier, the pyrolysis process was carried out. Following, the pyrolysis char and gas flowed through to the second stage of the gasifier. In the second stage, the steam was introduced into the gasifier for the reduction reactions. The ash produced from the gasification then was collected in the ash collector. After the gasification process took place, the synthesis gas entered the downstream section. In this section, the removal of tar and particle content took place within the heat exchanger and the venturi scrubber. In the heat exchanger, the synthesis gas was cooled down to collect the condensable components from the produced synthesis gas. The venturi scrubber was mainly used to control the flow rate of

synthesis gas and to separate the remaining particles and volatiles from synthesis gas. In the venturi scrubber, the scrubbing water from the bottom of the water tank was pumped through a water filter to remove large size particles before entering the spaying nozzle. The high-pressure ensures the atomizing of the washing liquid, which then is turbulently mixed at the throat section of the scrubber with the synthesis gas. In addition, the high-pressure spray of scrubbing liquid creates a vacuum at the gas inlet of the scrubber. Therefore, through the regulation of the liquid pressure in the venturi scrubber we can set the pressure within the reactor. Finally, the dry synthesis gas passes through the gas meter and combusted in a burner. During the experiments, the synthesis gas composition was in-situ analysed with a GASBOARD- 3100P gas analyser. The synthesis gas composition of CO, CO2, CH4 and CnHm was measured within a non-dispersive infrared sensor (NDIR sensor). The H2 concentration was determined by a thermal conductivity detector (TCD sensor), while the O2 concentration was evaluated by an electron capture detector (ECD sensor).

3.3. Experimental parameters

The experimental conditions are summarized in Table 3-2.

Mass flow rate coal	of Gasific temper		S/C ratio	Steam flow rate	Time of experiment
g/h	°C	1 -	mol/mol	g/min	days
			0.75	4.5	3
1084	700)	1.00	6.7	3
			1.25	8.8	3
			0.75	4.5	3
1084	800	1.00	6.7	3	
			1.25	8.8	3
	900		0.75	4.5	3
1084			1.00	6.7	3
			1.25	8.8	3
Sample name	Mass flow rate of coal	Gasification temperature	S/C ı	ratio Steam flow rate	Time of experiment
	g/h	°C	mol/	mol g/min	days
C1	991.70	900	1.0	00 10.06	5 3
CI	991.70	900	1.2	25 12.87	7 3
\mathbf{C}^{2}	1043.00	900	1.0	9.91	1 3
C2			1.2	25 12.67	7 3

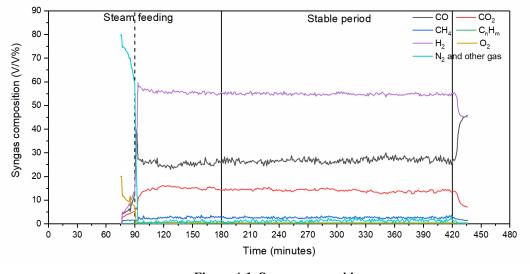
Table 3-2. Experimental conditions

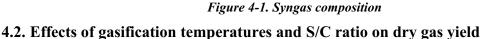
4. The effects of gasification temperature and steam-to-carbon (S/C) ratio on the multi-
stage gasification process of Hungarian brown coal

4.1. Data monitoring of a typical experimental day

Figure 4-1 illustrates the volume fraction of syngas components, including carbon monoxide (CO), carbon dioxide (CO₂), methane (CH₄), long-chain hydrocarbons (C_nH_m), hydrogen (H₂), oxygen (O₂), and nitrogen (N₂) and other gas.

In the heating-up period, there was only a small amount of pyrolysis gas, hence the N_2 and O_2 were dominant components. Following the introduction of coal and steam, the concentrations of H_2 , CO, and CO₂ increased significantly, while those of O_2 and N_2 decreased dramatically, almost to 0 V/V% in the case of O_2 and around 1.3 V/V% in the case of N_2 and other gas. After 90 minutes, the volume fractions of syngas reached a stable state. During the stable period, the concentrations of the long-chain hydrocarbon (C_nH_m) and O_2 in the produced gas were nearly unnoticed, which indicated the high reaction rate of the steam reforming reaction of the long-chain hydrocarbons in the steam gasification process. The N_2 and other gas concentrations were under 2% during the stable period. This N_2 content was mainly due to the air entering during the coal feeding process. H_2 was the most abundant gas compound, accounting for around 55.5 V/V% during the stable period. Other main gas components were CO and CO₂, around 27.55 and 13.7 V/V%, respectively. The produced gas also included a small amount of CH₄, lower than 4 V/V% in this case.





The effects of gasification temperature and S/C ratio on dry gas yield are illustrated in Figure 4-2. The positive impact of increasing gasification temperature on dry gas yield was more remarkable at a higher S/C ratio. The dry gas yield generated at 900 °C was 2.3 and 2.6 times higher than that at 700 °C for the S/C ratios of 1.00 and 1.25, respectively. At constant gasification temperature, the dry gas yield increased remarkably if the S/C ratio increased from 0.75 to 1.00 and they increased slowly if the S/C ratio increased further from 1.00 to 1.25. This means that the positive effects of increasing S/C ratio on char gasification were limited at this temperature below the 1.25 S/C ratio. The highest dry gas yield was 1.14 Nm³/kg_{coal} at 900 °C of gasification temperature and 1.25 of S/C ratio.

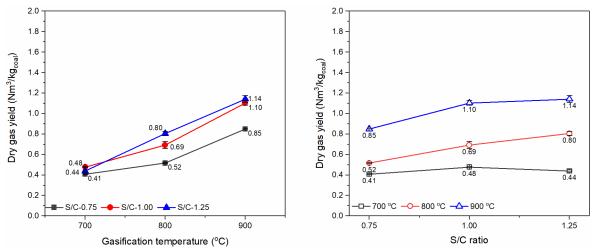


Figure 4-2. The effects of gasification temperature and S/C ratio on dry gas yield

4.3. Effects of gasification temperature and S/C ratio on syngas composition

Regarding the syngas composition, the main components of produced gas under all experiments were H_2 , CO, CO₂, and CH₄, accounting for at least 94 V/V% of the total volume. The effects of gasification temperature and S/C ratio on synthesis gas composition are illustrated in Figure 4-3.

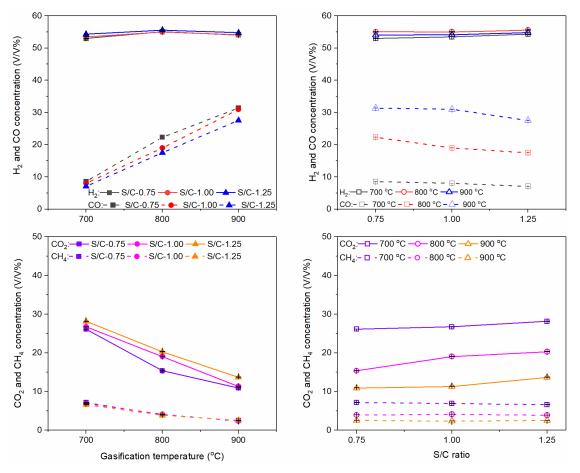


Figure 4-3. The effects of gasification temperature and S/C ratio on syngas composition

Increasing the gasification temperature had a significant effect on the syngas composition. Meanwhile, the volume fraction of syngas varied slightly when the S/C ratio increased from 0.75 to 1.25. The mean volume fraction of CO, CO₂, and CH₄ showed a significant variation when the gasification temperature increased from 700 to 900 °C for all S/C ratios. On the other hand, the changes in H₂ concentration did not follow a monotonic trend under the same conditions. From a chemical utilisation point of view, the gasification temperature at 900 °C and S/C ratio of 1.25 resulted in the most promising H₂/CO ratio of 1.99 (~2). From an energetic point of view, the highest LHV_{syngas} was observed at 900 °C and 0.75 of S/C ratio, approximately 10.69 MJ/Nm³.

5. Multi-stage gasification of Hungarian brown coal samples from gravity separation process with magnetite suspension media

5.1. Dry syngas yield

Figure 5-1 illustrates the dry gas yield obtained from the gasification experiments conducted on C_{org} , C1, and C2 samples at a temperature of 900 °C and S/C ratio of 1.00 and 1.25. Obviously, there was a higher

dry gas yield generated in the case of C1 and C2 samples at all S/C ratios, with increasing dry gas yield observed in order of C_{org} , C1, and C2, respectively. At 1.25 of the S/C ratio, the gasification process of the C2 sample produced the highest produced gas yield (1.70 Nm³/kg_{coal}) and followed by the gasification process of the C1 sample, with 1.52 Nm³/kg_{coal}.

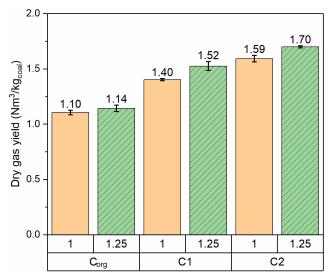


Figure 5-1. Dry gas yield in the gasification process of C1 and C2 samples at 1.00 and 1.25 of S/C ratio **5.2. Syngas composition**

Figure 5-2 presents the syngas concentration obtained from the gasification experiments of C1 and C2 samples with the gasification temperature at 900 °C and 1.00 and 1.25 S/C ratios. The H₂ concentration remained relatively constant among the gasification experiments of C_{org}, C1, and C2 samples, ranging from 53.66 to 54.76 V/V%. It can be observed that an increase in the S/C ratio resulted in a slight increase in H₂ concentration for all samples. When the S/C ratio increased from 1.00 to 1.25, the H₂ concentration increased from 53.66 to 54 V/V% in the case of the experiment of the C1 sample, while from 53.89 to 54.38 V/V% for the C2 sample.

At the same S/C ratios, the CO concentration was lower for the gasification experiments of the C1 and C2 samples compared to that of the C_{org} sample. Increasing of S/C ratio led to a decrease in the CO concentration for both gasification experiments of C1 and C2 samples but to different extents. When the S/C ratio increased from 1.00 to 1.25, the CO concentration decreased by 0.69 V/V% for the experiment of the C1 sample. In the case of the C2 sample, that number was by 1.75 V/V%.

In contrast with the CO volume fraction, the CO_2 volume fraction increased in the order of C, C2, and C1 at all S/C ratios. Furthermore, the volume fraction of CO_2 expanded as the S/C ratio increased. In the case of the gasification experiment of the C1 sample, the volume fraction of CO_2 increased from 15.54 to 16.90 V/V% at 1.00 and 1.25 S/C ratios, respectively. These numbers were 12.79 and 14.14 V/V% in the case of the C2 sample.

At all S/C ratios, the syngas produced from the gasification experiments of C1 and C2 samples had a higher volume fraction of CH_4 than that of C_{org} sample.

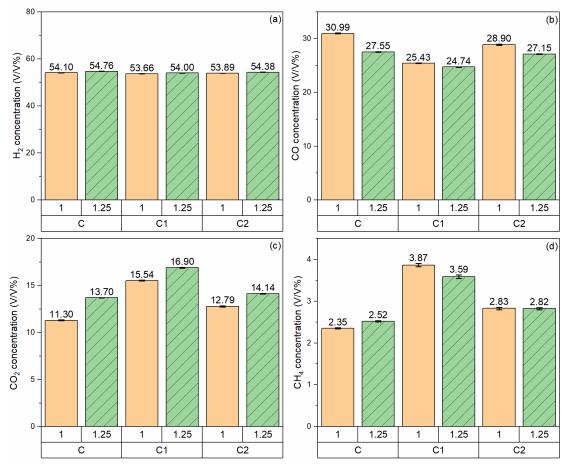


Figure 5-2. Syngas concentration from the gasification process of C1 and C2 samples

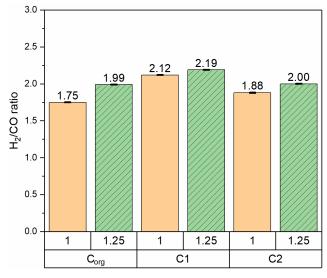


Figure 5-3. H₂/CO ratio from the gasification process of C1 and C2 samples

The H₂/CO ratio obtained from the gasification experiments of C1 and C2 samples is illustrated in Figure 5-3. It is observed that, with a stable H₂ concentration, the H₂/CO ratio was primarily driven by the CO concentration. Consequently, the H₂/CO ratio was higher in the case of the C1 sample experiment. When the S/C ratio increased from 1.00 to 1.25, the H₂/CO ratio also increased for all samples, albeit to varying degrees. In the case of the C_{org} sample, the H₂/CO ratio increased from 1.75 at 1.00 of the S/C ratio to 1.99 at 1.25 of

the S/C ratio. These numbers were from 2.12 to 2.19 and 1.88 to 2.00 for C1 and C2 samples, respectively, over the same range of S/C ratios.

6. Conclusion

Experiments were carried out in a single line multi-stage fixed bed gasifier arranged in an L shape, with a vertical reactor for the pyrolysis process and a horizontal reactor for the reduction process.

In the first part of this study, the original Hungarian brown coal sample- C_{org} was examined at gasification temperatures of 700, 800 and 900 °C with S/C ratios of 0.75, 1.00, and 1.25, while maintaining the pyrolysis temperature at 600 °C.

Both increasing gasification temperature and S/C ratio resulted in a higher conversion rate of the gasification process, leading to a decrease in residual char and condensate liquid yields, and an increase in produced gas yield. However, the positive effects of increasing S/C ratio on char gasification were limited when the S/C ratio exceeded 1.00. The highest conversion rate was achieved at a gasification temperature of 900 °C and an S/C ratio of 1.25, with an average syngas yield of 1.14 Nm³/kg_{coal}.

From a chemical utilisation point of view, the gasification temperature at 900 °C and S/C ratio of 1.25 resulted in the most promising H_2 /CO ratio of 1.99 (~2). From an energetic point of view, the highest LHV_{syngas} was observed at 900 °C and 0.75 of S/C ratio, approximately 10.69 MJ/Nm³.

In the second part of this study, the original Hungarian brown coal samples were subjected to a gravity separation process using magnetite suspension fluid. The separation process resulted in two types of coal samples, namely C1 (≤ 1.6 g/cm³) and C2 (between 1.6- 1.8 g/cm³).

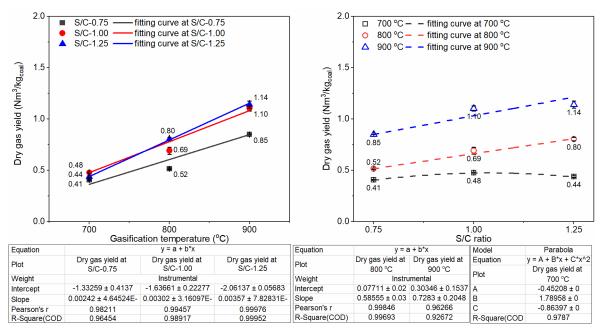
The results indicated that the gasification process of the separated coal samples produced a higher specific volume of syngas compared to the original coal sample.

Regarding the H_2/CO ratio, for the experiments of the C2 sample, the H_2/CO ratio reached 1.88 and 2.00 at S/C ratios of 1.00 and 1.25, respectively. Therefore, the optimal conditions for the C2 sample were identified as a gasification temperature of 900 °C and an S/C ratio of 1.25.

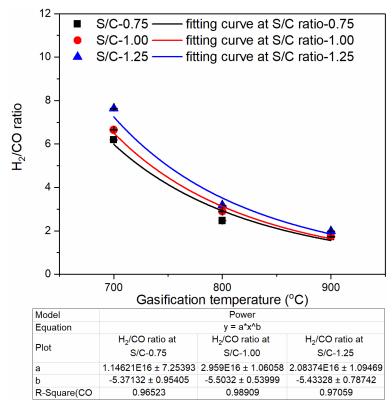
7. New scientific results

Thesis 1. The original low-rank coal sample from Hungary with a particle size of 10-30 mm was studied in the steam gasification process. The gasification experiments were carried out in a single-line multi-stage fixed bed configuration, with a mass flow rate of coal of 1084 g/h at the gasification temperatures of 700, 800 and 900 °C, while with S/C ratios were set at 0.75, 1.00, and 1.25.

To establish the relationship between the produced gas yield and the gasification parameters, a linear function and a parabolic were formulated considering the gasification temperatures and S/C ratios. These equations can be expressed as follows:



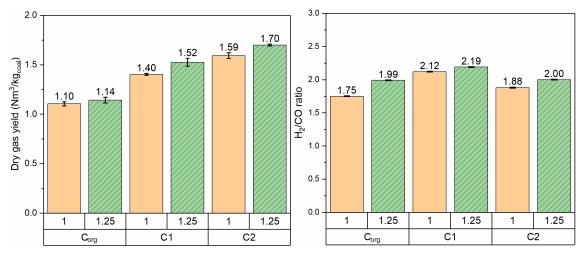
All linear functions describe well the relations between the dry gas yield, gasification temperature and S/C ratio, with the exception of dry gas yield and S/C ratio at 700 °C, that is best described with a parabolic function. **Thesis 2.** Under the same experimental conditions as shown in thesis 1, to investigate the relationship between the H_2 /CO ratio and the gasification parameters, an equation was formulated based on the experimental parameters. At a constant S/C ratio, the H_2 /CO ratio can be expressed as power functions of the gasification temperatures. The equations describing these relationships are as follows:



Thesis 3. The desired H_2/CO ratio for a specific chemical production process is typically 2.00. The experiments conducted in a single-line multi-stage gasifier demonstrated that an H_2/CO ratio of approximately 2.00 can be achieved at a gasification temperature of 900 °C using an original low-rank coal sample with a particle size of

10-30 mm and a mass flow rate of 1084 g/h. The optimal conditions for achieving an H_2/CO ratio of 2.00 were identified as a gasification temperature of 900 °C and an S/C ratio of 1.25.

Thesis 4. The original low-rank coal sample was subjected to a gravity separation process using a heavy magnetite suspension fluid to obtain two separated coal samples 1) with a specific density of up to 1.6 g/cm³ (C1) and 2) a specific density between 1.6 - 1.8 g/cm³ (C2). The heavy suspension-separated coal samples exhibited higher syngas yields compared to the original coal sample, under the same gasification temperature of 900 °C and S/C ratios of 1.00 and 1.25. The produced syngas from the coal sample with a specific density between 1.6 - 1.8 g/cm³ (C2) can reach an optimal H₂/CO ratio of 2.00 for chemical production processes, while the gas yield can be increased by 50 % compared to the original coal sample, under the same gasification conditions, with an S/C ratio of 1.25 at 900 °C.



8. Publications

- Mai, T.D.; Koós, T.; Sebe, E.; Siménfalvi, Z.; Kállay, A.A. *Efficiency Enhancement of the Single Line Multi-Stage Gasification of Hungarian Low-Rank Coal: Effects of Gasification Temperature and Steam/Carbon (S/C) Ratio.* Energies 2023, 16, 4427. https://doi.org/10.3390/en16114427
- Thuan Duc, Mai; Sebe, Emese; Kallay, Andras Arnold. The single-stage steam gasification of magnetite heavy suspension separated coal samples from hungarian brown coal, Építőanyag: Journal of Silicate Based and Composite Materials 74: 4 pp. 150-155., 6 p. (2022). https://doi.org/ 10.14382_epitoanyag-jsbcm.2022.23.
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