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Gasification Characteristics of High Moisture Content Lignite under CO_2 and Auto-Generated Steam Atmosphere in a Moving Bed Tubular Reactor

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Abstract: An external thermal high-temperature continuous feed moving bed tubular reactor was used for the gasification of high moisture content lignite (30.41 wt.%) under CO_2 and an auto-generated steam atmosphere. The objectives of this study are to illustrate the synergistic gasification characteristics of high moisture content lignite and CO_2 in the tubular reactor; CO_2 and auto-generated steam (steam released from the lignite) were used as gasification agents for lignite gasification. The effects of temperature and CO_2 flow rate were also investigated. Experimental results showed that when the gasification temperature increased from 800 °C to 1000 °C, the H_2 yield also increased from 8.45 mol kg^{-1} to 17.86 mol kg^{-1}. This may indicate that the H_2O-CO_2 gasification of semi-coke was enhanced with the rise in temperature. At 900 °C, the gas yield increased with the increase in CO_2 flow rate, while the yield of char and liquid product showed an opposite trend. The lower heating value of the H_2-rich syngas varied from 11.73 MJ m^{-3} to 12.77 MJ Nm^{-3}. The experimental results proved that the high moisture content lignite in-situ CO_2 gasification process is an effective methodology for the clean and efficient utilization of lignite.

Keywords: lignite; CO_2 gasification; moisture content; temperature

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1. Introduction

The rapid development of the economy has led to drastic fossil energy consumption in China [1]. Among the assorted energy supplies, coal will continue to be a significant energy source [2]. There are about 212 billion tons of lignite in China, which is inexpensive and has high gasification reactivity [3]. Lignite contains more inherent moisture (approximately 25–50%) than traditional high-rank coals and is usually used for direct combustion. Approximately 20–25% of the energy in lignite is consumed to evaporate the internal moisture contained in lignite itself which results in lower power plant efficiency [4].

Gasification is a clean and effective technology for the utilization of low-rank lignite [5–7]. Gasification is a promising technology that offers a clean and efficient way to convert solid coal into syngas with an H_2/CO (molar ratio) of 0.5 to 3. Syngas from lignite-gasification, primarily consisting of H_2 and CO, has been employed as a versatile platform for generating various desired products, such as electric power, heat, hydrogen, methane, methanol, dimethyl ether, Fischer–Tropsch (FT) liquids, and ammonia [8]. Low-rank coal such as lignite is more likely to produce hydrogen-rich syngas by steam gasification because of its high reactivity [9]. Traditionally, O_2 and H_2O are used as the gasification agents of lignite [10]. The catalytic steam gasification of lignite was carried out in a decoupled triple bed reaction system by Xiao et al. [11] and fuel gas with a 51.7 vol.% H_2 concentration was obtained. The gasification of a high moisture biomass was also carried out in a supercritical water rector, which provided high cold gas efficiency and hydrogen yield [12]. However, compared with tubular furnace reactor gasification, supercritical water gasification is accepted as a sustainable sludge disposal technology but has many problems, such as requiring a high temperature and high pressure, high risk of manual operation, higher energy consumption, and less processing capacity [13,14].

The gasification of high moisture content lignite in an auto-generated steam agent (steam evaporated from the lignite) was also carried out in our previous research [15]. In the fixed-bed external thermal tubular furnace, the auto-generated steam generated by the evaporation of the moisture of the high wet lignite is used as the gasification agent. However, the reaction between the auto-generated steam and lignite char did not occur at an appreciable rate, thus the conversion ratio was relatively low.

Recently, the use of CO_2 as a supplement to traditional gasification agents is attracting more and more attention [16–18]. There are many advantages to using CO_2 as a gasification agent, such as the flexible adjustment of the H_2/CO ratio to meet the specific requirements for high-value fuels or chemical production [19]. Experimental studies on CO_2 gasification of Victorian lignite were performed at different temperatures and CO_2 concentrations in an entrained-flow gasifier [20]. The results revealed that the carbon conversion ratio rose from 58 to 80% with an increase in CO_2 concentration from 10 to 30%. Nevertheless, these results are not directly applicable to raw lignite which has higher moisture content.

However, to the best of our knowledge, an experimental study on the gasification of high moisture content lignite using CO_2 in the presence of auto-generated steam has not yet been carried out, and an accurate measurement of the gas yield, gas composition, and proportion of each species has not been found. The present paper is devoted to exploring the optimum process for the efficient conversion of high moisture content lignite under a CO_2 gasification agent. A continuous feed external thermal moving bed pyrolysis experimental system was constructed, and the screened high moisture lignite was sent to the experimental system at a certain feed rate to investigate the feed rate, residence time, and reactor temperature. The product yield, gas composition, and carbon conversion ratio were accurately measured and analyzed. The reaction mechanism of in-situ water and CO_2, as well as the in-situ gasification reaction mechanism and the water migration law in the reaction process, will be discussed.

2. Experimental Procedure

2.1. Preparation of Lignite Samples

The low-rank lignite samples studied in this paper were freshly mined from Hailer, inner Mongolia, China. The proximate and ultimate analysis data are listed in Table 1. The ultimate analysis of lignite was carried out in CHNS/O (Vario micro cube Elementar). On an 'as received' basis (ar) the lignite had a high moisture content of 30.41 wt.%. Therefore, the raw samples were shredded and sieved into particles 0.1–3.2 mm in size and kept in airtight bags to prevent moisture dissipation.

Table 1. The proximate and ultimate analysis of Hailer lignite (%).

Proximate Analysis (wt.%, ar)				Ultimate Analysis (wt.%, daf)				
M	A	V	FC	C	H	O	S	N
30.41	15.26	30.97	23.36	79.81	4.17	14.33	0.56	1.13

Note: ar: as received basis; daf: dried and ash-free basis; M: moisture; A: ash; V: volatile species; FC: fixed carbon; C: Carbon; H: hydrogen; O: oxygen; S: sulfur; N: nitrogen.

2.2. Experimental Instruments and Procedure

Experimental tests were carried out in a self-designed moving bed tubular reactor. As shown in Figure 1, the moving bed tubular reactor consists of a spiral-propelled reactor, an electric heating furnace, a temperature controller, and a gas cleaning and analysis line. Among them, the spiral-propelled reactor consists of a lignite hopper screw driver and a stainless tube. During the test, lignite was loaded into the sealed hopper which can be flushed with nitrogen or CO_2. The residence time and feeding rate of the lignite in the reactor were controlled via the speed of revolution which was 20 min and 16 g min^{-1}, respectively.

The temperature of the reactor was monitored by K-type thermocouples. The maximum operating temperature of the laboratory moving bed tubular furnace was 1000 °C.

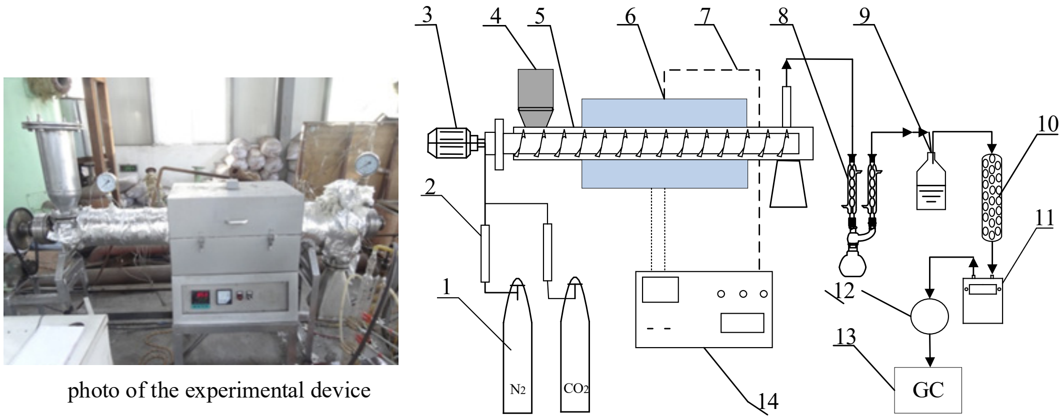

Figure 1. Schematic diagram of experimental apparatus. 1—gas cylinder; 2—mass flow meter; 3—motor; 4—feed-bunker; 5—spiral propelled tubular reactor; 6—Electric heating furnace; 7—Thermalcouple; 8—Spherical condenser; 9—water filter; 10—galss fiber filter; 11—gas meter; 12—sampling bag; 13—gas chromatograph; 14—temperature control.

For each experimental run, the reactor was preheated to the desired temperature with a multi-stage precise temperature control tubular electric heating furnace and kept isothermal for 30 min. The screw driver of the spiral-propelled reactor was then turned on to start feeding. In order to ensure in situ steam (evaporated from the lignite) and the added CO_2 could participate in the gasification reaction, the pressure in the furnace was maintained at about 50 kPa by adjusting the opening of the gas valve. The lignite was then propelled through the stainless tube with the help of the screw driver. The condensable products (liquid products) were captured by tar collectors and activated carbon filters, while the condensable gas products were collected by gas sampling bags. Then, the components of the gas products were analyzed offline by a SHIMADZU GC-14C gas chromatograph. Finally, the coal char was collected in a char hopper and weighted by a Sartorius electronic balance. The design parameters of the experimental apparatus are given in Table 2.

Table 2. The design parameters of the experimental apparatus.

System Parameter	Range/Set Point
Lignite feed rate	10–30 g min^{-1}
Lignite particle size	0.5–3.0 mm^{-1}
Residence time	20 min
Isothermal temperature of the spiral propelled reactor	800–1100 °C
Reactor hot zone length	1200 mm
Reactor diameter	50 mm
Volume of the spiral propelled reactor	2.3 L
System pressure	50 kPa

2.3. Analysis of the Gasification Product

The carbon conversion ratio refers to the proportion of carbon atoms transferred from the lignite to combustible gas, which is one of the most important parameters to evaluate the effect of the lignite thermo-chemical conversion. The carbon conversion ratio is calculated by Equation (1):

$$\eta_c = \frac{12(V_{CO} + V_{CH_4} + V_{CO_2} + 2V_{C_2H_n})}{22.4 \times (298/273) \times C\%} \times G_V \quad (1)$$

The lower heating value of the gas species is calculated according to [21]

$$Q_{LHV} = (107.98 \times V_{H_2} + 126.36 \times V_{CO} + 358.18 \times V_{CH_4} + 629.09 \times V_{C_2H_n}) \times 10^{-3} \quad (2)$$

where G_V represents the dry basis gas yield, $C\%$ is the carbon content of the lignite in the ultimate analysis, and V_i is the volume content of H_2, CO, CH_4, CO_2, and C_2H_n.

3. Results and Discussion

3.1. Brief Description of the Gasification Process

Because of relatively high moisture content (generally 20–50%), lignite is usually first subjected to a drying pretreatment [22]. The theoretical minimum energy consumption for water evaporation in lignite is 2341 kJ kg^{-1}, and some water must be removed at a higher temperature which brings out high drying costs [23]. The flue gas produced contains a large amount of low-pressure steam and is difficult to reuse, while the steam gasification of dry coal needs to re-prepare a large amount of steam [24].

In this paper, the gasification process of high moisture content lignite using CO_2 and in situ steam (evaporated from the lignite) as gasification agents was proposed, as seen in Figure 2. As shown, the drying, pyrolysis, steam, and CO_2 gasification of lignite were concentrated in the continuous feeding spiral-propelled tubular reactor. The external heating spiral propelled tubular reactor was used to make the lignite react synergistically in a steam and CO_2 atmosphere while avoiding the dilution of the product gas. Finally, a high purity and H2-rich fuel gas were produced. Xiong et al. [25] carried out similar work in a tubular reactor, which showed that the inherent water in the raw material enhanced the production of H2-rich fuel gas.

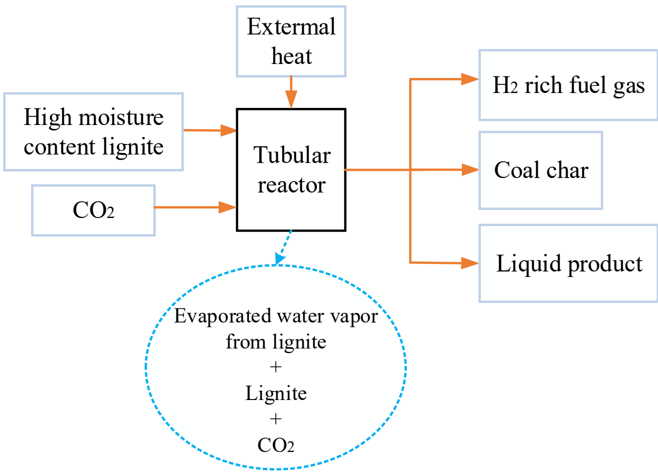

Figure 2. Schematic of the high moisture content lignite gasification under a CO_2 atmosphere.

3.2. Effect of Temperature

Under the condition of a 1.1 L min^{-1} CO_2 flow rate, experimental research on the gasification characteristics of lignite (moisture content 30.41%) was carried out in the tubular reactor. In this study, the gasification temperature varied from 800 to 1000 °C, with an interval of 50 °C. Figure 3 shows the effect of temperature on product distribution (dry base). It can be seen that when the gasification temperature rose from 800 to 1000 °C, the yield of gas increased from 41.17 to 71.89%, while the liquid and semi-coke yield decreased from 33.37 and 69.16% to 21.26 and 50.55%, respectively. Along with the increase in temperature, the scission of carbon bonds was promoted, which contributes to the better chemical reactivity of lignite [26].

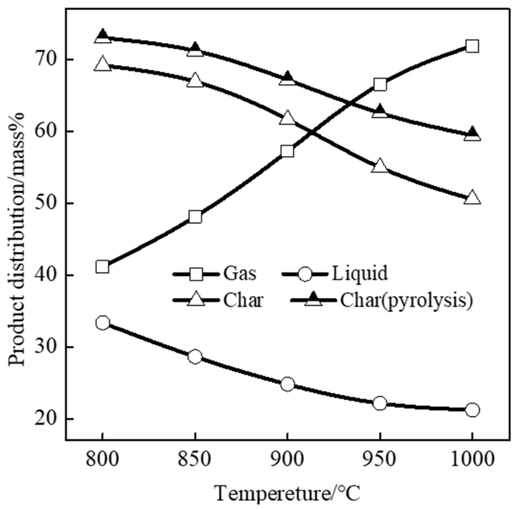

Figure 3. The effect of temperature on the product yield.

Furthermore, the semi-coke yield of lignite CO_2 gasification was lower than that of lignite in situ pyrolysis (0 L min^{-1} CO_2 flow rate), and the difference between them increased with the rise in temperature. A similar conclusion can be found in Roberts' research [27]. This indicates that the gasification process is endothermic; a higher temperature promoted the CO_2 and steam gasification of lignite, which led to the secondary decomposition of tar and macromolecule structure in coal, resulting in an increase in gas yield and a corresponding decrease in liquid and semi-coke yields. Similar conclusions can be found in Tanner's work [20].

The effect of temperature on the in-situ gasification characteristics is given in Table 3. As shown, when the gasification temperature increased from 800 to 1000 °C, the total gas yield raised from 21.17 mol kg^{-1} to 39.62 mol kg^{-1}. The volume fractions of H_2 and CO were observed to increase with the rise in temperature. The yields of H_2, CO, CH_4, and C_2H_n at different temperatures are given in Figure 4. Apparently, when the gasification temperature increased from 800 to 1000 °C, the H_2 yield raised from 8.45 to 17.86 mol kg^{-1}, which indicates that the steam gasification of coal char was enhanced. The CO yield rose from 5.31 to 13.72 mol kg^{-1}, indicating that the gasification reaction between semi-coke and CO_2 accelerated with the increase in temperature. Note this phenomenon was also demonstrated in Tong's work [28]. The yield of CH_4 increased at the initial stage and then decreased with the increase in temperature. However, the yield of C_2H_n decreased with the increase in temperature. This is mainly because (1) the increase in H_2 and CO volume fraction intensified the methanation reaction [29]; (2) with the rise of temperature, the cracking reaction of C_2H_n and CH_4 is accelerated by the newly formed active char.

Table 3. Effect of temperature on the gas characteristics *.

Temperature /°C	Volume Fraction/%					H_2/CO	Q_{LHV} /(MJ Nm^{-3})	Gas Yield/mol kg^{-1}	Carbon Conversion Ratio/%
	H_2	CO	CH_4	CO_2	C_2H_n				
800	39.92	25.08	9.17	24.47	1.36	1.59	11.62	21.17	25.75
850	41.23	26.14	8.92	22.36	1.35	1.58	11.80	24.69	29.39
900	43.72	29.81	8.12	17.04	1.31	1.47	12.22	29.88	34.07
950	43.98	31.62	6.67	16.81	0.92	1.39	11.71	34.95	39.39
1000	45.08	34.64	5.04	14.63	0.61	1.30	11.43	39.62	43.55

Note: *—Dry basis.

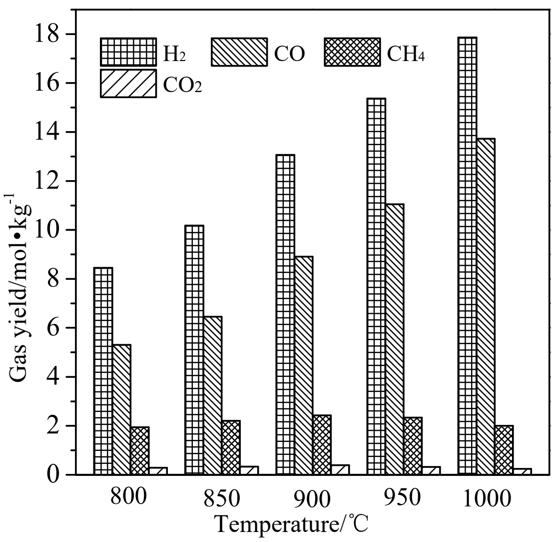

Figure 4. Effect of temperature on the gas yield of lignite gasification.

It can also be seen from Figure 4 and Table 3 that with the increase in gasification temperature, the carbon conversion rate increases from 25.75 to 43.55%, which indicates that the increase in gasification temperature can effectively increase the carbon conversion rate. Compared with the independent pyrolysis of high moisture lignite, the addition of CO_2 promoted the lignite gasification reaction, increasing the dry-base gas yield and a decreasing of the semi-coke yield. In addition, the H_2/CO decreased with the rise in temperature, ranging from 1.59 to 1.30, which is suitable for the Fischer–Tropsch process or chemical products such as ammonia, methanol, and dimethyl ether [30].

3.3. Effect of CO_2 Flow Rate

The influence of CO_2 addition on the gasification of high moisture content lignite characteristics is complex and requires a comprehensive consideration of temperature and CO_2 flow rate. The experimental results in the previous section have shown that increasing the gasification temperature led to a higher gas yield and carbon conversion rate, which agrees with Saucedo's research [31]. When the gasification temperature reached 900 °C, the low calorific value of the gas attained the maximum. According to a comprehensive analysis of the experimental data above, the greatest influence of the CO_2 flow rate on the direct gasification characteristics of high humidity lignite was selected at 900 °C.

Figure 5 shows the effect of CO_2 flow rate on the yield of the gas, liquid, and solid products on a dry base at 900 °C. In this work, the CO_2 flow rate varied from 0 L min^{-1} to 2 L min^{-1} with an interval of 0.3–0.5 L min^{-1}. As can be seen, with the rise in CO_2, the flow rate increased from 0 to 2.0 L min^{-1}, the gas yield of brown coal gasification raised from 35.50 to 78.13%, and the liquid yield and semi-coke yield decreased from 41.05 and 67.15% to 14.83 and 50.74%, respectively. This could be explained by the following: (1) The synergistic reaction between CO_2, steam, and lignite changed the internal structure of lignite while increasing the pore volume and specific surface area of semi-coke. This promoted the gasification of lignite and the precipitation of volatile matter, resulting in the reduction in semi-coke yield and the increase in volatile matter yield [32]; (2) Along with the increase in CO_2 flow rate, the Boudouard reaction between fixed carbon and CO_2 was strengthened and more semi-coke was converted to gaseous products.

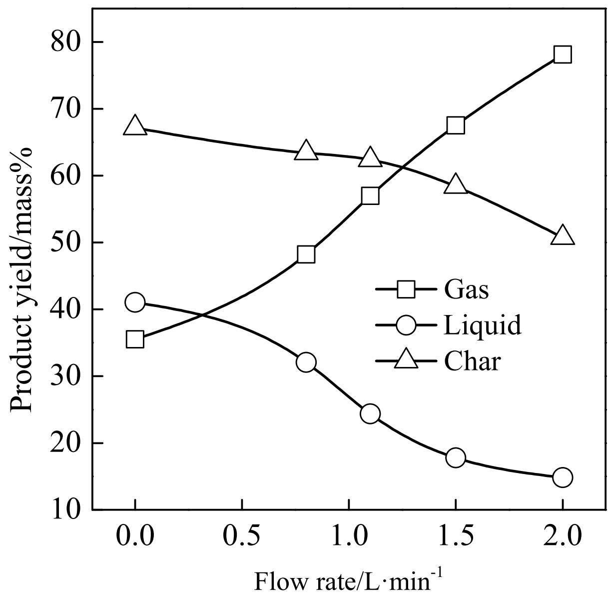

Figure 5. The effect of the CO_2 flow rate on the product yield at 900 °C.

The effect of the CO_2 flow rate on the direct gasification characteristics of lignite at 900 °C is given in Table 4. As can be seen, when the CO_2 flow rate increased from 0 L min^{-1} to 2.0 L min^{-1}, the total gas yield raised from 18.86 to 40.09 mol kg^{-1}. The combustible gas yields of H_2, CO, CH_4, and C_2H_n under different CO_2 flow rates at 900 °C are presented in Figure 6. Obviously, with the increase in CO_2 flow rate, the volume fraction and yield of CO raised greatly. Table 4 and Figure 6 indicate that although the volume fraction of H_2 decreased gradually, the yield of H_2 first increased and then decreased with the rise in the CO_2 flow rate. It also can be seen that the maximum H_2 yield reached 13.06 mol kg^{-1} when the CO_2 flow rate was 1.1 L min^{-1}. This variation was mainly attributed to three main reasons: Firstly, with the increase in CO_2 flow rate, the gasification reaction between semi-coke and CO_2 accelerated, which led to more CO output; Secondly, the porosity of char increases with the increase in CO_2 flow rate, which promoted the steam gasification reaction of char to a certain extent [33]. Thirdly, the steam was gradually diluted by the rising CO_2 flow in the reactor, which limited the steam gasification reaction of semi-coke. The syngas lower heating value varied from 11.73 to 12.77 MJ Nm^{-3}, much higher than Serhat's data [34].

Table 4. The effect of the CO_2 flow rate on the gas characteristics of lignite gasification at 900 °C.

Flow Rate/ (L min^{-1})	Volume Fraction/%					H_2/CO	Q_{LHV}/(MJ Nm^{-3})	Gas Yield /mol kg^{-1}	Carbon Conversion Ratio/%
	H_2	CO	CH_4	CO_2	C_2H_n				
0	51.72	24.13	7.90	14.41	1.84	2.14	12.62	18.86	18.71
0.8	47.45	27.04	8.06	15.32	2.13	1.75	12.77	25.33	27.41
1.1	43.72	29.81	8.12	17.04	1.31	1.47	12.22	29.88	34.07
1.5	35.68	34.92	8.87	19.61	0.92	1.02	12.02	34.45	44.49
2.0	31.15	37.56	7.74	22.86	0.69	0.81	11.73	40.09	55.19

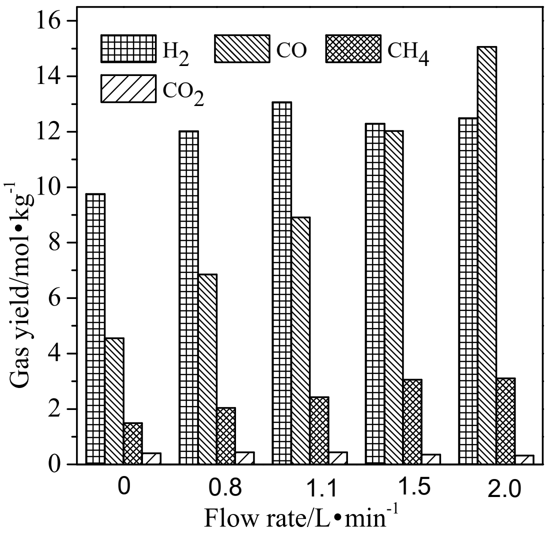

Figure 6. The effect of CO_2 flow rate on the gas yield of lignite gasification at 900 °C.

Figure 6 also illustrates that the yield of CH_4 increased gradually with the rising CO_2 flow rate, while that of C_2H_n decreased. This is mainly because, firstly, adding the appropriate amount of CO_2 promoted the fracture of methyl and methylene; secondly, more CO_2 promoted the decomposition reaction of tar and macromolecule C_2H_n., which was conducive to the formation of CH_4.

4. Conclusions

The gasification of high moisture content lignite under CO_2 and a steam agent was undertaken in an external thermal high-temperature continuous feed moving bed tubular reactor. Compared with the independent pyrolysis of high moisture lignite, the addition of CO_2 promoted the lignite gasification reaction. At 900 °C, the gas yield rose with the increase in CO_2 flow rate, while the yield of char and liquid product showed an opposite trend. When the CO_2 flow rate increased from 0 to 2.0 L min^{-1}, the total gas yield rose from 18.86 to 40.09 mol kg^{-1}. The lower heating value of the hydrogen-rich syngas varied from 11.73 to 12.77 MJ Nm^{-3}.

Under the condition of 1.1 L min^{-1} CO_2 flow rate, when the gasification temperature increased from 800 to 1000 °C, the H_2 yield raised from 8.45 to 17.86 mol kg^{-1}, indicating that the steam gasification of coal char was enhanced. The yield of CO also indicated growth, showing that the gasification reaction between semi-coke and CO_2 was accelerated.

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