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COMPARATIVE ANALYSIS OF EXTERNAL COMBUSTION SCHEMES IN THE THREE-STEP COAL GASIFICATION TECHNOLOGY WITH CO_2 RECYCLING

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ABSTRACT

Chemical energy conversion has a great influence on the cold gas efficiency of coal gasification technology. In this paper, a three-step gasification technology with CO_2 recycling is introduced and two external combustion schemes (CO-fueled chemical looping combustion and unconverted coke combustion) are compared. Results showed that the CO-fueled chemical looping combustion scheme has a higher cold gas efficiency of 90.1%, while cold gas efficiency of the unconverted coke combustion scheme is 88.4%. Before the water gas shift subprocess, the chemical energy conversion efficiency in the unconverted coke combustion scheme is 93.2%, which is 1.6 percentage points higher than that in the CO-fueled chemical looping combustion scheme. However, more chemical energy is consumed for CO_2 regeneration in the unconverted coke combustion scheme, which results in chemical energy conversion efficiency decreases from 93.2% to 88.4%. Therefore, better energy matching between reactions can effectively improve the cold gas efficiency of the coal gasification technology. Besides, chemical energy consumption for CO_2 regeneration should be reduced for gasification technology adopting CO2 as a gasifying agent.

Keywords: coal gasification; CO_2 recycling; Chemical looping combustion; Unconverted coke combustion;

NONMENCLATURE

Abbreviations	
AR	Air reactor
CFCLC	CO-fueled Chemical looping combustion
CGE	Cold gas efficiency
CLC	Chemical looping combustion
COG	Coke Oven Gas
FR	Fuel reactor
LHV	Lower heating value
ST	Steam turbine
UCC	Unconverted coke combustion
WHB	Waste heat boiler
WGS	Water gas shift

1. INTRODUCTION

Coal, which accounts for approximately 30% of the global primary energy consumption, will play a significant role in the future global energy system [1]. However, there are three main challenges in clean coal conversion technologies: enhancement of energy conversion efficiency, effective control of hazardous pollutants emission and CO_2 capture [2]. Compared with coal directly combustion, coal gasification technology has been proved to be a preferred scheme to realize high efficiency utilization, clean coal conversion and carbon management.

Coal gasification process is the thermochemistry conversion of coal with gasifying agents including oxygen, steam, carbon dioxide, air, hydrogen and a combination of these [3]. In the coal gasification process, coal is partially oxidized by the gasifying agent at high temperature, accompanying with chemical energy of coal converted to chemical energy (LHV, lower heating value) of syngas. Consequently, cold gas efficiency (CGE) is an important criterion to evaluate coal gasification performance, which is defined as the ratio of chemical energy between syngas and coal. Though after a long-term development, CGE of current coal gasification technologies is still limited to 70-83%, which restrains the efficiency enhancement of coal-based energy systems [4].

To further enhance the CGE of the coal gasification technology, a three-step coal gasification technology with CO_2 recycling technology is introduced and two external schemes are compared. The three-step gasification technology is composed of pyrolysis subprocess, CO_2-coke gasification subprocess, water gas shift (WGS) subprocess. Since the pyrolysis subprocess and CO_2-coke gasification subprocess are endothermic, two external combustion schemes are proposed to

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supply heat for the pyrolysis subprocess and CO_2-coke gasification subprocess. The two external combustion schemes are unreacted char combustion (UCC) scheme and CO-fueled chemical looping combustion (CFCLC) scheme, respectively. In the UCC scheme, coke is partially converted in the CO_2-coke gasification subprocess and unconverted coke is combusted directly in the external combustion chamber. While in the CFCLC scheme, CO generated from the CO_2-Coke gasification subprocess is combusted through chemical looping combustion method and production CO_2 from the fuel reactor is recycled back as the gasifying agent.

In this paper, a three-step coal gasification technology with CO_2 recycling technology is proposed and two external schemes are introduced. Besides, a comparative analysis is conducted to reveal the CGE difference of the two external combustion schemes.

2. PROCESS DESCRIPTION

2.1 The three-step coal gasification technology with CFCLC scheme

The simplified flowsheet of three-step coal gasification technology with the CFCLC scheme is shown in Fig. 1. In the three-step coal gasification process with the CFCLC scheme, coal is firstly sent to the pyrolyzer where coal is thermally decomposed into raw coke oven gas (COG) and coke. The hot coke from the pyrolyzer is sent to the gasifier to react with gasifying agent CO_2. In the gasifier, the coke gasification reaction occurs and coke is converted to CO. Considering that the pyrolysis subprocess and CO_2-coke gasification subprocess are endothermic, the CO generated from the gasifier is divided into two streams. one stream of CO is combusted in the external CLC process and CO_2 from the fuel reactor is recycled back as the gasifying agent. Another stream of CO is sent to the waste heat boiler (WHB) to generate high-pressure steam for steam turbines (ST). After heat recovery by the WHB, a bypass configuration is adopted to satisfy the requirement of gasifying agent CO_2. In the bypass configuration, part of CO is converted to CO_2 and H2 through the WGS reaction. CO_2 is separated from shifted gas and recycled back as the gasifying agent. As a result, three streams of gases are exported as gasification productions including H_2 from CO_2 separation subprocess, CO from gasifier and COG from the pyrolyzer.

2.2 The three-step coal gasification with UCC scheme

In the three-step coal gasification with the UCC scheme (shown in Fig. 2), the hot coke from the pyrolyzer is partially gasified with CO_2, then the unconverted coke is sent to the external combustion chamber to supply heat for the pyrolyzer subprocess and CO_2-coke gasification subprocess. After partially CO_2-coke gasification, the WHB is employed to recover the sensible heat of CO from the gasifier. Similarly, a bypass

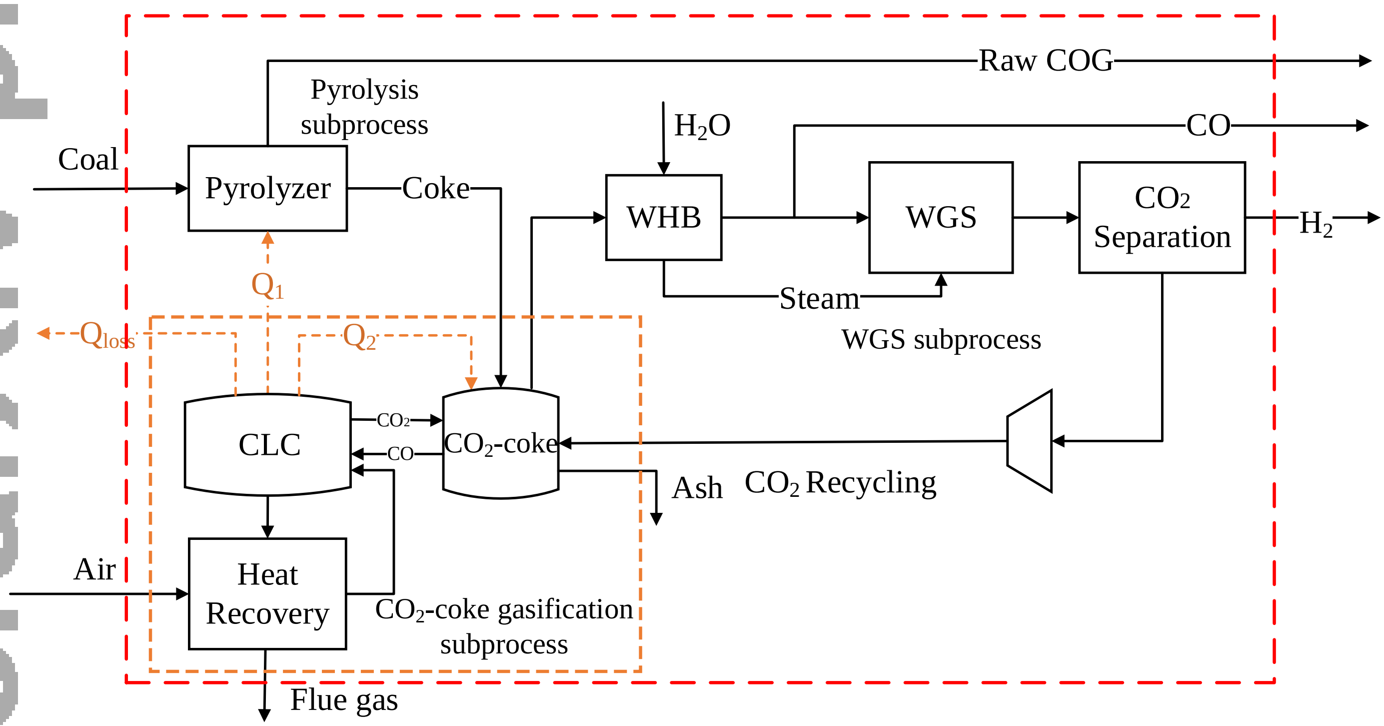

Fig. 1 The simplified flowsheet of the three-step coal gasification with CFCLC scheme

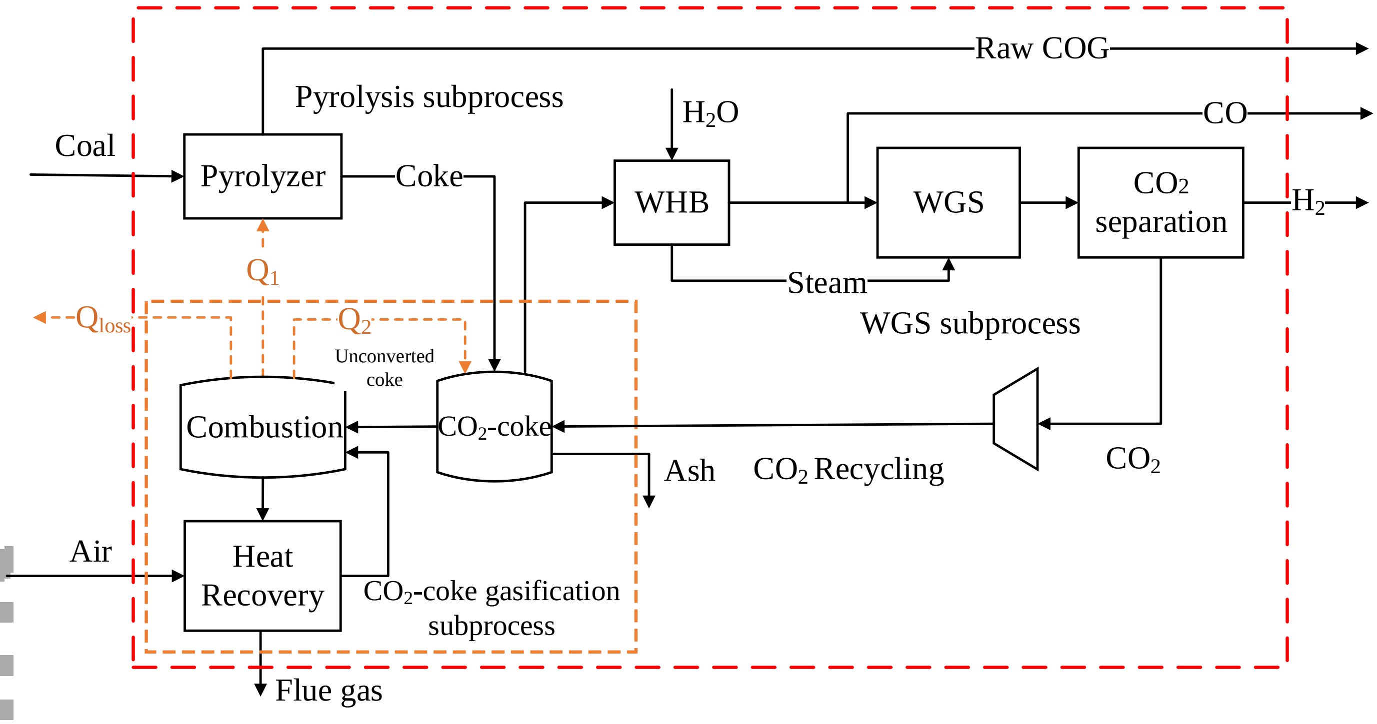

Fig. 2 The simplified flowsheet of the three-step coal gasification with UCC scheme

configuration with the WGS subprocess and CO_2 separation subprocess is adopted to regenerate gasifying agent CO_2.

There are two main differences between the two schemes: (1) Reactions matching between the CO_2-coke gasification subprocess and the external combustion subprocess. In the CFCLC scheme, the CO-fueled chemical looping combustion method is adopted to supply heat, while unconverted coke is directly combusted in the UCC scheme. The different configurations between the coke-CO_2 gasification subprocess, external combustion subprocess have a significant influence on the chemical energy output of CO. (2) CO_2 regeneration method. In the UCC scheme, the required gasifying agent CO_2 is regenerated from WGS and CO2 separation subprocess. While in the CFCLC scheme, gasifying agent CO_2 is composed of two streams: combustion productions from fuel reactor and separated CO_2 from the WGS subprocess.

3. KEY DESIGN PARAMETERS

In this section, details of the main facilities in the three-step gasification model are mentioned. Three-step gasification technology with two external combustion schemes is both simulated by Aspen Plus v8.4. In the simulation, PR-BM is selected as the global method and the bituminous coal is selected in this work. The mass flow of feeding coal is 28.64 kg/s.

In the pyrolysis subprocess, coal is decomposed into raw coke oven gas and coke at 900 ℃ and atmospheric pressure, the hot coke from the pyrolyzer is directly sent to the gasifier. The pyrolysis subprocess is simulated by RYield reactor model in Aspen plus and product yield distribution is based on experimental data, which are presented in Table. 1.

To simulate the coke-CO_2 gasification subprocess, the RGibbs and RYield reactor blocks are employed. Firstly, coke is designated as the non-conventional component and RYield reactor block is used to convert coke into a series of stable simple substances including C(s), H_2, N_2, S(s), O_2 and ash according to coke ultimate analysis [5]. Then RGibbs reactor block is adopted to calculate the gasifier production distribution, which operates at 1100 ℃ and 20 bar.

In the UCC scheme, the hot coke is partially gasified and unconverted coke is sent to the combustion chamber, which is modeled by RStoic reactor block. To supply heat for the pyrolyzer and gasifier, about 48.6% of hot coke is combusted with air in the combustion chamber with an operating temperature of 1200 ℃. The excess air ratio is set 30% to ensure the coke combusted completely.

In the CFCLC scheme, part CO from the gasifier is combusted by the chemical looping combustion method. In this model, the RGibbs reactors are used to simulate the two reactors and oxygen carriers Fe_2O_3/Fe_3O_4 are selected to transfer oxygen from the air reactor (AR) to fuel reactor (FR). In the FR, CO is oxidized and Fe_2O_3 is reduced to Fe_3O_4. The FR operates at 1200 ℃ and the excess molar ratio of oxygen carrier and CO is set as 1.2 to ensure CO completely converted to CO_2. The high-temperature CO_2 generated from FR is recycled back to the gasifier and the reduced oxygen carrier Fe_3O_4 is transported to AR. In the air reactor, the reduced oxygen carrier Fe_3O_4 reacts with preheated air at atmospheric pressure. The temperature of AR reactor is 1200 ℃ and the excess air coefficient is 1.3 to ensure complete oxidation of oxygen carrier Fe_3O_4. The oxygen-depleted air discharged from AR is sent to preheat the air and the Fe_3O_4 carriers are transported back to FR. After preheating air, the oxygen-depleted air is emitted at 135 ℃.

Table. 1 Experimental results of pyrolysis

Products yields (Mass, fraction %)			
COG	31.9	Coke	59.3
Tar	1.5	H_2O	7.3
Coal ultimate analysis (Mass, fraction %)			
C	71.63	H	4.53
O	10.28	N	0.84
S	0.33	W	7.30
Ash	8.45	LHV (MJ/kg)	26.6
COG component (Volume, %)			
H_2	51.7	O_2	1.7
N_2	9.0	CH_4	11.6
CO_2	3.5	CO	21.9
C_2H_4	0.5	H_2S	0.1
Char ultimate analysis (Mass fraction, %)			
C	91.86	H	1.38
O	0.14	N	0.87
S	0.44	Ash	5.31

The next units of the two schemes are consistent. CO will be cooled to 230 ℃ and high-pressure steam of 535 ℃/120 bar is produced for electricity generation in the waste heat boiler (WHB) unit. After heat recovery in the WHB unit, two stages with the intercooling shifted approach are adopted to model the water gas shift (WGS) unit. The first stage is modeled by an adiabatic reactor, and the second is modeled by an isothermal reactor. In the adiabatic reactor which allows higher reaction temperature, CO and H_2O reacts rapidly. However, the conversion of CO is limited. Therefore, the isothermal reactor is adopted to achieve a higher conversion of CO. Both the adiabatic reactor and isothermal reactor are simulated by the REQUIL reactors.

In the CO_2 separation process, shifted syngas is cooled to 40 ℃ and the cooled syngas are sent to the Selexol process to separate CO_2. The separated CO_2 is compressed and recycled back to the gasifier as the gasifying agent. The key design parameters aforementioned are presented in the Table. 2.

Table. 2 Key design parameters

Item	Description
Pyrolyzer	T=900 ℃; P=1.013 bar
CO_2-coke gasifier	T=1100 ℃; P=20 bar
CO-fueled chemical looping combustion	AR: T=1200 ℃, P=1.013 bar; FR: T=1200 ℃, P=20 bar; Excess ratio of Fe_2O_3: 1.2; Excess ratio of air: 1.3; Heat loss: 9.0% of fuel input LHV [6]
Unconverted coke combustion	T=1200 ℃; Air excess ratio: 1.3; Heat loss: 9.0% of fuel input LHV [6]
WGS reaction	Two stages with inter-bed cooling; first stage adiabatic and second stage isothermal with 225 ℃ [7]
WHB & Steam turbine	Triple-pressure reheat steam: 126/25/5.5 bar, Steam temperature: 566 °C, Isentropic efficiency of ST: 0.88/0.89/0.87
CO_2 separation	Selexol technology; CO_2 recovery ratio: 98%; Sulfur recovery ratio: 100%
CO_2 compression	Isentropic efficiency of compressor: 0.85

4. RESULTS AND DISCUSSION

The composition and mass flow of gasification productions in the two schemes are presented in the Table. 5. The results indicate that the CGE of the two schemes is 88.4% and 90.1%, respectively. Compared with the chemical energy output of gasification productions, CO and H_2 chemical energy output difference contributes to the different CGE of the two schemes. In the UCC scheme, the chemical energy output of CO and H_2 is 221.0 MW and 230.1 MW, respectively. Compared with the UCC scheme, the chemical energy output of CO in the CFCLC scheme is 166.5 MW more and the chemical energy output of H_2 is 154.3 MW less.

Table. 3 Gasification products comparison between the two external combustion schemes of three-step gasification technology

Item	UCC scheme			CFCLC scheme		
Feedstock coal, MW	761.8			761.8		
Syngas output	COG	CO	H_2	COG	CO	H_2
T, ℃	900	230	40	900	230	40
P, bar	1.013	20	20	1.013	20	20
Molar fraction, %						
CO_2	3.5	4.4	1.9	3.5	6.6	1.9
CO	21.9	88.5	1.7	21.9	86.3	1.7
H_2	51.7	6.0	95.1	51.7	5.7	95.0
H_2O	0	0.6	0.8	0	0.9	0.9
CH_4	11.6	0	0	11.6	0	0
N_2	9.0	0.3	0.3	9.0	0.3	0.3
O_2	1.7	0	0	1.7	0	0
C_2H_4	0.5	0	0	0.5	0	0
Others	0.1	0.2	0.2	0.1	0.2	0.2
LHV, kJ/kg	25341.9	9816.0	67318.1	25341.9	9421.1	66676.1
Mass flow, kg/h	31528.7	81102.6	12353.2	31528.7	148131.0	4142.3
Chemical energy output, MW	221.9	221.1	230.1	221.9	387.7	76.7
CGE	88.4%			90.1%		

Therefore, the CGE of the CFCLC scheme is 1.7 percentage higher than that in the UCC scheme.

In the three-step gasification technology, the chemical energy of CO generated from gasifier are decided by the thermally coupling between endothermic subprocesses and exothermic combustion subprocess. In the UCC scheme, coke is partially converted in the coke-CO_2 gasification subprocess and heat required is supplied by unconverted coke combustion. Differently in the CFCLC scheme, coke is nearly completely converted in the coke-CO_2 gasification subprocess and heat required is provided by CO-fueled chemical looping combustion subprocess. In the two schemes, the chemical energy of converted CO in the CFCLC scheme is 344.9 MW and the chemical energy of unconverted coke in the UCC scheme is 241.2 MW. which implies that more thermal energy discharged in the external combustion process is recovered and converted to chemical energy in the pyrolysis and CO_2-coke gasification subprocess. Due to better configuration between endothermic subprocesses and exothermic combustion subprocess, the chemical energy efficiency before WGS in the UCC scheme is 93.2%, which is 1.6 percentage points higher than that in the CFCLC scheme.

Furthermore, part of CO generated from are shifted to CO_2 and H_2 through the WGS reaction in the two schemes, so the amount of CO sent to the WGS unit also affects the chemical energy output of CO and H_2. There is part of the chemical energy of CO consumed for CO_2 regeneration in the WGS subprocess because the WGS reaction is exothermic. Therefore, chemical energy consumption for CO_2 regeneration also has a significant influence on the CGE of the three-step gasification technology. Chemical energy consumption for CO_2 regeneration is decided by CO_2 required in the coke-CO_2 gasification subprocess. Compared with the CO_2 recycling amount distribution, in the CFCLC scheme 1102.5 kmol/h CO_2 are regenerated from the WGS unit and the other CO_2 are recycled from the fuel reactor, which can be separated without energy consumption. While in the UCC scheme, 3231.0 kmol/h CO_2 are regenerated and recycled from the WGS unit. Consequently, more CO are shifted to CO_2 in the UCC scheme. After the WGS subprocess, the chemical energy efficiency in the UCC decreases from 93.2% to 88.4%, while in the CFCLC scheme, the chemical energy efficiency decreases from 91.7% to 90.1%. Therefore, less chemical energy is consumed for CO_2 regeneration in the CFCLC scheme, which brings about higher CGE than that in the UCC scheme.

5. CONCLUSION

In this paper, a three-step coal gasification technology with CO_2 recycling technology is introduced and two external combustion schemes are compared. Chemical energy consumption for CO_2 regeneration and reactions matching between endothermic subprocesses and exothermic combustion subprocesses have significant influence on the cold gas efficiency of the three-step gasification technology. The three-step gasification technology with the CFCLC scheme has a cold gas efficiency of 90.1%, which is 1.7 percentage points higher than that in the UCC scheme. Better reactions matching between endothermic subprocesses and exothermic combustion subprocesses can effectively improve the chemical energy conversion efficiency, but more CO converted in the WGS subprocess results in the lower cold gas efficiency in the UCC scheme.

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