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Performance analysis of an O_2/CO_2 power plant based on chemical looping air separation

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Abstract: The process of an O_2/CO_2 power plant based on chemical looping air separation (CLAS) is modeled using the Aspen Plus software. The operating parameters and power consumption of the CLAS unit are analyzed. The CLAS system, thermal power generation system and flue gas cooling and compression unit (CCU) are coupled and optimized, and the temperature and flow of the flue gas extraction are determined. The results indicate that the net plant efficiency of CLAS O_2/CO_2 power plant is 39.2%, which is only 3.54% lower than that of the conventional power plants without carbon capture. However, the O_2/CO_2 power plant based on cryogenic air separation technology brings 8% to 10% decrease in the net plant efficiency. By optimizations, the net plant efficiency increases by 1.65%. The energy consumption of the CCU accounts for 59.7% and the pump accounts for 27.1%. The oxygen concentration from the chemical looping air separation unit is 12.2%.

Key words: chemical looping air separation; O_2/CO_2 combustion; performance analysis

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Coal, as a kind of primary energy, has the largest reserves and the most widely distribution in the world, and it is also the cheapest source of energy. About 45% to 50% of electricity around world is provided by coal-fired power plants. With the advantages of small investment and a short construction period, coal-fired power plants will still play a major role for a long time in the future[1]. In recent years, CO_2 emissions have caused global warming and frequent extreme weather, which has attracted much attention. Coal-fired power plants contribute much more CO_2 emissions, and the reduction in CO_2 emissions of power plants is necessary. However, the CO_2 emissions in power plants are characterized by low CO_2 partial pressure and high quantity of flue gas. By conventional air combustion, the main component in the flue gas is N_2, while the concentration of CO_2 is only 10% to 15%, which means that the cost of CO_2 separation is extremely expensive and causes the net plant efficiency decline[2].

To increase the concentration of CO_2 in flue gas and decrease the cost of CO_2 separation, the oxy-fuel coal-fired technology (also called O_2/CO_2 recycle combustion technology) was proposed, which uses O_2/CO_2 mixed gas instead of air as the oxidant of fuel combustion to increase the CO_2 concentration in the flue gas. In the dry flue gas, the CO_2 concentration can be as high as 95% or more, which means that a high concentration of carbon dioxide can be gained easily after flue gas condensation and dehydration. It is a CO_2 capture technology of great potential. Nevertheless, the technology causes the net plant efficiency to decline deeply. The net plant efficiency decreases by 8% to 10% than that of the conventional pulverized coal power plant due to the large amount of oxygen for the combustion and much high power consumption of existing cryogenic air separation unit (ASU)[3].

Chemical looping air separation (CLAS) is a new oxygen generation technology, which is characterized by low investment and low power consumption. Applying this technology to generate oxygen instead of conventional ASU can promote the net plant efficiency of the oxy-fuel power plant. This work proposes a 1 000 MW ultra-supercritical coal-fired plant based on CLAS. Using Aspen Plus software, the process of the 1 000 MW CLAS O_2/CO_2 power plant is modeled.

1 Chemical Looping Air Separation

Conventional oxy-fuel plant combusts oxygen produced by the cryogenic ASU. The ASU is the only large-scale oxygen generation method under operation commercially. This technology has a long history and is rather mature, and the oxygen content of the product reaches above 99%. However, the technology is energy-intensive, which decreases the net plant efficiency of an oxy-fuel power plant by about 10%.

Chemical looping combustion (CLC) was first proposed by Richter et al[4]. CLC is composed of two independent reactors: an air reactor and a fuel reactor. A suitable kind of metal oxide is selected as an oxygen carrier

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which cycles between the two reactors. Chemical looping with oxygen uncoupling (CLOU) based on CLC was proposed by Shulman et al[5]. Gaseous oxygen is released in the fuel reactor rather than lattice oxygen, which is mainly separated from CLC and avoids solid-solid reactions. CLAS technology was proposed based on CLOU. Moghtaderi[6] studied the performance of CLAS under a vapor atmosphere and the results show that a mixed oxygen carrier made from Mn_3O_4/Mn_2O_3 and CoO/Co_3O_4 at a mass ratio of 1∶1 achieves a better performance. The preliminary cost analysis shows that the specific power consumption of the CLAS process is about 0.045 (kW·h)/m^3 of air produced, which is approximately 11% of the specific power consumption of conventional cryogenic air separation systems. Mei et al.[7] studied the thermodynamics and reaction mechanism of the deoxygenation reaction of Cu/Co/Mn based oxygen carriers. They found that the deoxygenation reactivity of the Mn-based oxygen carrier is higher when the Cu-based oxygen carrier is lower and the Co-based oxygen carrier is moderate. Zhao et al.[8] modeled CLAS systems using Mn-oxide and Co-oxide as oxygen carriers, and CO_2 as the medium gas, and analyzed their performances. The results show that when the oxidation temperature is 890 ℃ and the reduction temperature is 880 ℃, the performance of the Mn-based oxygen carrier is better with specific power consumption of 0.151 (kW·h)/kg and 13.2% oxygen concentration. Mn_3O_4/Mn_2O_3 is used as the oxygen carrier of the CLAS system in this work.

The CLAS concept using CO_2 as the medium gas is shown in Fig. 1. Due to the higher oxygen partial pressure in the oxidation reactor, Mn_3O_4 is oxidized to Mn_2O_3 at a high temperature. In the reduction reactor, Mn_2O_3 is reduced back to Mn_3O_4 and releases oxygen because of the lower partial oxygen pressure in CO_2.

$$4Mn_3O_4 + O_2 \longrightarrow 6Mn_2O_3 \tag{1}$$

$$6Mn_2O_3 \longrightarrow 4Mn_3O_4 + O_2 \tag{2}$$

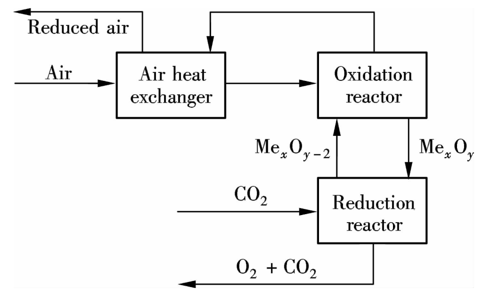

Fig. 1 Chemical looping air separation concept

2 O_2/CO_2 Recycle Combustion Plant

As shown in Fig. 2, an O_2/CO_2 recycle combustion plant integrating CLAS is proposed and the system mainly consists of three sections: 1) CLAS unit; 2) Conventional power plant; and 3) Flue gas cooling and compression unit.

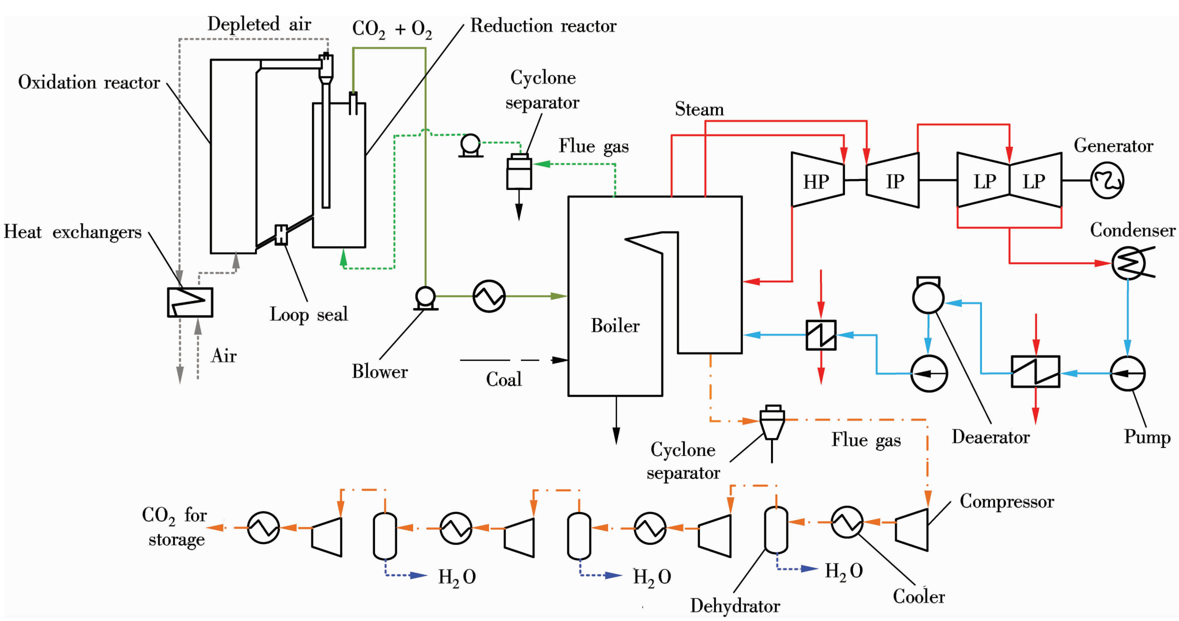

Fig. 2 O_2/CO_2 recycle combustion plant based on CLAS

High temperature flue gas extracted from the boiler furnace exit flows into the reduction reactor after undergoing cyclone separation and the booster fan. The flue gas is rich in carbon dioxide and water vapor, and flows into the bottom of reduction reactor of the CLAS unit. The CLAS unit is composed of an oxidation reactor, a reduction reactor, and air heat exchangers. After being preheated in heat exchangers, fresh air is induced from the bottom to the oxidation reactor where the reduced oxygen carrier from the reduction reactor is regenerated. After solid oxygen carrier separation, the oxygen depleted air is discharged into the atmosphere after heat recovery in heat exchangers. The oxygen carrier goes into the reduction reactor and releases oxygen into high temperature flue gas. The oxygen concentration in the flue gas increases and is ready for the combustion of coal after the removal of the solid oxygen carrier. The reduced oxygen carrier recycles into the oxidation reactor through the loop seal. The air preheater is no longer needed, which differs from the conventional coal-fired boiler. Liquid carbon dioxide is obtained after multi-stage water vapor condensations and compressions of exhaust flue gas, and the non-condensate gases, mainly comprised of oxygen, can be used as combustion air.

3 O_2/CO_2 Combustion System Modeling and Analysis

3.1 CLAS unit

The CLAS unit is modeled using Aspen Plus software, as shown in Fig. 3. Fresh air preheated in the heat exchanger (EX block) flows into the oxidation reactor (OXD block), and oxidizes Mn_3O_4. The solid product in the oxidation reactor is mainly Mn_2O_3 and the fresh air is turned into oxygen depleted air. After undergoing the cyclone separator (OX-SP block), the oxygen depleted air flows into the heat exchanger (EX block) for heat recovery to heat the fresh air. Then, the separated solid Mn_2O_3 enters the reduction reactor (RED block). It is reduced to Mn_3O_4, and it releases oxygen to the flue gas. The oxygen-containing flue gas is induced to the boiler and burns with coal. Solid Mn_3O_4 recycles back to the oxidation reactor (OXD block) for regeneration.

The simplified air components are N_2 (78.12%), O_2 (20.95%) and Ar (0.93%) with the ambient pressure of 101.325 kPa and the temperature of 25℃. Reaction (1) is exothermic while reaction (2) is endothermic. Therefore, heat of reaction (2) is mainly supplied by the sensible heat of high temperature flue gas or the electric oven.

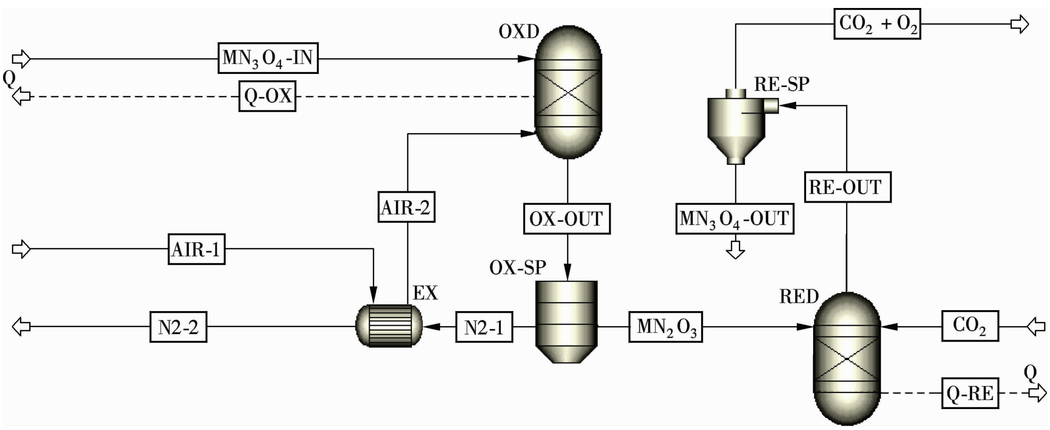

Fig. 3 The model of CLAS unit

Tab. 1 shows the results of the simulation of CLAS unit under different flue gas temperatures. Coal burnout is similar in air and under the oxy-fuel (O_2/CO_2) conditions with 11% to 14% O_2[9]. Under higher flue gas temperatures as in Case 2, less flue gas is needed to produce 1 kg O_2, which means that there is a high heat exchange surface between flue gas and water/steam in the boiler and a higher power generation. Moreover, less air means lower power consumption from the booster fan, lower oxygen depleted air heat losses, and a higher O_2 concentration in the oxygen-containing flue gas. Case 2 has an obvious advantage compared with Case 1.

Tab. 1 Simulation under different flue gas temperatures

Parameter	Case 1	Case 2
Flue gas temperature/℃	1 000	1 100
O_2 concentration/%	11	12.2
Oxidation reactor temperature/℃	783.3	787
Reduction reactor temperature/℃	757.5	762.4
Flue gas flow/(kg · kg^{-1})	11.14	9.9
Air flow/(kg · kg^{-1})	352.4	234

3.2 CLAS-based O_2/CO_2 combustion power plant

The thermal power generation section of the O_2/CO_2 combustion power plant is the same as that of the conventional power plant.

Assumptions on the simulations of boiler combustion are made as follows: 1) Coal combustion is divided into two processes: pyrolysis and coal char burning; 2) Burnout of coal is 99.5%; 3) Combustion is under barometric pressure. The proximate analysis and ultimate analysis of coal are shown in Tab. 2, and the lower heating value (LHV) is 22.55 MW/kg[10]. Tab. 3 and Tab. 4 show the results of simulations.

Tab. 2 Proximate analysis and ultimate analysis of coal %

Proximate analysis					Ultimate analysis			
w_C	w_H	w_O	w_N	w_S	w_M	w_A	w_{FC}	w_V
60.17	2.68	4.92	0.67	0.26	8.32	22.98	57.7	11

As shown in Tab. 3, the simulation results of the conventional power plant are similar to those of the CLAS-based power plant, which indicates that the simulations are reasonable. To maintain the thermal equilibrium of the boiler, circulating flue gas is composed of two parts:

one from the furnace exit (1 099 ℃) and the other from exhaust flue gas (378.1 ℃), which results in the decrease in circulating flue gas temperature. Therefore, the power consumption of CLAS increases.

Tab. 3 Summary of simulation results

Items	Conventional power plant	CLAS-based power plant
Coal feed/($t\cdot h^{-1}$)	362 116	362 116
Air/flue gas volume/($m^3\cdot h^{-1}$)	2.451×10^6	3.715×10^6
Live steam flow/($t\cdot h^{-1}$)	2 981	3 000
Live steam temperature/℃	605	607
Live steam pressure/MPa	27.46	27.76
Reheat temperature/℃	603	603
Reheat pressure/MPa	5.94	6.3
Power output/MW	1 028.69	1 043.37

As seen from Tab. 4, the CO_2 concentration of exhaust flue gas of the CLAS-based O_2/CO_2 plant is apparently higher than that of the convention power plant, over 97% in dry flue gas, and the CO_2-rich flue gas is ready for CO_2 capture.

Tab. 4 Mole fraction of gas components in furnace exit %

Mole fraction	Conventional power plant	CLAS-based power plant
x_{N_2}	74.94	0.24
x_{O_2}	2.54	1.89
x_{H_2O}	5.68	17.68
x_{NO}	5.72×10^{-2}	1.31×10^{-3}
x_{NO_2}	8.62×10^{-5}	2.76×10^{-6}
x_{SO_2}	2.53×10^{-2}	7.84×10^{-2}
x_{SO_3}	1.14×10^{-4}	6.75×10^{-4}
x_{CO_2}	15.89	80.1
x_{Ar}	0.89	0

3.3 Cooling and compression unit (CCU)

The components of the flue gas of the CLAS O_2/CO_2 combustion power plant, as shown in Tab. 4, are mainly CO_2, H_2O, and slight N_2, O_2, etc. After multi-stage cooling, water vapor condensation and compression, CO_2 in the flue gas is liquefied and is available for large-scale transport and reserve[11-12], while the non-condensate, mainly oxygen, is recycled as combustion air.

Four stages in compression, cooling and water vapor separation are selected for CO_2 liquidation. Exhaust flue gas from the boiler exit is cooled to 25 ℃. After the removal of the condensate, it is compressed to a higher pressure, and then flows to the next stage for further cooling, water vapor condensation, and compression in four stages. The flue gas is compressed to 10 MPa in the fourth stage and cooled to 25 ℃, and CO_2 is liquefied as fluid. Centrifugal-flow compressors with 85% isentropic efficiency and 98% mechanical efficiency are chosen in the simulations with the compression ratio of 3.5, 3.5, 3.5 and 2.5 in four stages. The simulation results show that CO_2 in the flue gas is as high as 97.14%, as shown in Tab. 5. The non-condensate components are N_2, O_2, water vapor, NO_x and SO_x. Also, the power consumption of the whole CCU process is 101.96 MW.

Tab. 5 Simulation results of CCU

Component	Import		Export	
	Flow/($kg\cdot h^{-1}$)	Concentration/%	Flow/($kg\cdot h^{-1}$)	Concentration/%
N_2	1 602.19	0.24	1 602.19	0.28
O_2	14 704.52	1.89	14 704.5	2.29
H_2O	77 406.175	17.68	661.079	0.18
NO	9.57	0.001 3	9.57	0.001 6
NO_2	0.03	0.00	0.03	0.00
SO_2	1 220.71	0.078	1 220	0.095
SO_3	13.13	0.000 67	13	0.000 8
CO_2	857 249.93	80.1	857 222.24	97.14

4 Performance Analysis and Optimization

4.1 Performance analysis of CLAS

Reaction (2) in the reduction reactor of the CLAS unit is endothermic. To keep the reactor temperature, a quantity of heat is needed and may come from: 1) The heat carried by solid materials from the oxidation reactor, 2) The heat brought by higher temperature flue gas; 3) The electric oven. The reduction temperature affects the performance of the CLAS unit. Tab. 6 shows the results when the reduction temperature is increased by 5 ℃.

Tab. 6 Effects of the reduction temperature on the performance of the CLAS unit

Items	Before	After
Reduction temperature/℃	762.4	767.4
O_2/CO_2 gas temperature/℃	762.4	767.4
O_2 concentration/%	12.2	13.0
Furnace exit gas temperature/℃	1 099	1 100.3
Power of electric oven/kW	672.55	761.27

As seen from Tab. 6, with the increase in the reduction temperature by 5 ℃, the O_2/CO_2 gas temperature increases by 5 ℃ as well, and the O_2 concentration is increased by 0.8%. The flue gas temperature in the furnace exit is increased by 1.3 ℃. It has little effect on the thermal load of the boiler and the power output, but the power of the electric oven is increased by 88.72 kW, and more power is consumed and so the net plant efficiency decreases.

However, decreasing the reduction temperature means a decrease in the O_2 concentration in O_2/CO_2 and the decrease in the burnout of coal. Hence, it is reasonable to maintain the reduction temperature at about 762 ℃ and an O_2 concentration of 12.2%.

4.2 Optimization of CCU

In the simulation, the boiler exhaust flue gas is about 378.1 ℃. Particulate removal, cooling, water vapor condensation and separation are needed before flue gas compression. To remove water vapor in the flue gas, an appropriate cooling temperature is needed. The volume fraction of CO_2 in flue gas is calculated according to the flue gas condensing temperature. The volume fraction of CO_2 increases when the temperature decreases, especially when the temperature is lower than 50 ℃. Circulating water (CW) is usually adopted as the cooling medium. Assuming that the flue gas is cooled to 25 ℃, about 140 MW heat may be recovered for further use.

After being cooled to 25 ℃, the flue gas is compressed for cooling and water vapor separation. Higher outlet pressure means higher power consumption. Water vapor volume fraction and power consumption are also simulated according to the outlet pressure. Water vapor volume fraction almost stays unchanged when the outlet pressure is higher than 3 MPa. In order to decrease the power consumption, the optimum option is to compress the flue gas to 3 MPa in the first three stages and then to 10 MPa in the 4th stage, and it has the minimum power consumption. The first three stages are of the same compression ratio of 3.1. The power of 23.56 MW can be saved.

In order to make full use of exhaust flue gas heat energy and improve the net plant efficiency, the heat from the flue gas is used to heat the low-pressure feed water in stage 5 and stage 6, and after each stage of compression, the compressed flue gas is used to heat the low-pressure feed water in stage 7 and stage 8 before the CW coolers, as shown in Tab. 7. Steam extractions from steam turbine for the regenerative system decrease and turbine power increases by 17.5 MW.

Tab. 7 Steam extraction flow for regeneration before and after optimization t/h

Stage	Before	After
5	86.20	45.90
6	95.79	50
7	80.54	59.70
8	148.63	109.80

4.3 Power consumption analysis

We can see from Tab. 5 that SO_2 in the flue gas is also liquefied when CO_2 is liquefied. The critical point of CO_2 is 7.4 MPa/31 ℃ and that of SO_2 is 7.9 MPa/157.8 ℃. Therefore, the desulfurization of flue gas is unnecessary. The power consumption of desulfurization in the conventional power plant accounts for 1% of total capacity, and about 10 MW is saved from the O_2/CO_2 plant.

From Tab. 8 we can see that, in the CLAS O_2/CO_2 plant, the power consumption of CCU is the largest, accounting for 66.2% of the total, and that of the pump accounts for 22.6%. After optimization, the power consumption of CCU declines to 59.7% of the total while that of the pump is 27.1%. Due to the recycling of flue gas, the exhaust flue gas flow of the O_2/CO_2 system is much less than that of the conventional power plant and the heat losses are much lower than those of the conventional plants. More steam and more power are produced as shown in Tab. 3. The net plant efficiency of the optimized CLAS O_2/CO_2 plant is 1.65% higher than that of the un-optimized, and only 1.89% lower than that of the conventional plant without CO_2 capture.

Tab. 8 Power consumption comparison between the conventional plant and the O_2/CO_2 power plant

Items	Conventional power plant	O_2/CO_2 power plant	O_2/CO_2 power plant (optimized)
CLAS/MW		1.8	1.8
Desulfurization/MW	10.28		
Pumps/MW	34.61	34.83	35.55
Air blowers/MW	14.36	15.52	15.52
CCU/MW		101.96	78.4
Output power/MW	1 028.69	1 043.37	1 060.91
Efficiency/%	45.35	46	46.77
Net power/MW	969.44	889.26	926.64
Net efficiency/%	42.74	39.2	40.85

5 Conclusion

A 1 000 MW O_2/CO_2 power plant based on chemical looping air separation is proposed. The system performances are simulated using the Aspen Plus software, and are compared with the conventional power plant. The aim is to determine the possibility of using the CLAS O_2/CO_2 power plant. High temperature flue gas extracted from the furnace exit is used to absorb oxygen in the CLAS unit. The concentration of oxygen from the CLAS unit is 12.2% under a 787 ℃ oxidation temperature and 762.4 ℃ reduction temperature. The oxygen containing flue gas, together with the flue gas extracted from the exhaust, is used as the circulating gas. The net plant efficiency of the CLAS O_2/CO_2 system is 39.2% when the CO_2 is compressed to 10 MPa, 3.54% lower than the conventional power plant without carbon dioxide capture. When the heat of the exhaust flue gas in CCU is used in the regenerative system, the net plant efficiency of the optimized CLAS O_2/CO_2 plant is 40.85%, 1.89% higher. The CO_2 concentration of 97.14% is achieved after CCU.

Nonetheless, further work is needed to verify the possibility of CLAS, for example, to find a proper oxygen carrier to design the CLAS unit, etc. Due to the overwhelming performance in the carbon capture, the CLAS O_2/CO_2 plant is worthwhile investigating in the later work.

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基于化学链制氧的 O_2/CO_2 燃烧电站性能分析

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摘要：采用 Aspen Plus 软件对基于化学链高温空分制氧技术(CLAS)的 O_2/CO_2 燃烧电厂全过程进行建模，对化学链高温空分单元进行运行参数及功耗分析，并对化学链高温空分单元、锅炉热力发电系统和烟气冷却压缩单元(CCU)进行耦合并优化，确定高温烟气抽取温度及抽取流量. 结果表明，O_2/CO_2 燃烧系统的净效率为 39.2%，仅比不能进行碳捕集的常规电厂低 3.54%. 然而，基于深冷空分技术的 O_2/CO_2 燃烧系统会使得全厂净效率下降 8% ~10%. 当采取优化措施后，O_2/CO_2 燃烧系统效率能够提高 1.65%. 烟气冷却压缩单元能耗占总能耗的 59.7%，泵能耗占 27.1%. 化学链制氧单元的供氧浓度为 12.2%.

关键词：化学链空分制氧；O_2/CO_2 燃烧；性能分析

中图分类号：TK114