

Research on coal staged conversion poly-generation system based on fluidized bed

Mingjiang Ni · Chao Li · Mengxiang Fang · Qinhui Wang · Zhongyang Luo · Kefa Cen

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Abstract A new coal staged conversion poly-generation system combined coal combustion and pyrolysis has been developed for clean and high efficient utilization of coal. Coal is the first pyrolysed in a fluidized pyrolyzer. The pyrolysis gas is then purified and used for chemical product or liquid fuel production. Tar is collected during purification and can be processed to extract high value product and to make liquid fuels by hydro-refining. Semi-coke from the pyrolysis reactor is burned in a circulating fluidized bed (CFB) combustor for heat or power generation. The system can realize coal multi-product generation and has a great potential to increase coal utilization value. A 1 MW poly-generation system pilot plant and a 12 MW CFB gas, tar, heat and power poly-generation system was erected. The experimental study focused on the two fluidized bed operation and characterization of gas, tar and char yields and compositions. The results showed that the system could operate stable, and produce about $0.12 \text{ m}^3/\text{kg}$ gas with $22 \text{ MJ}/\text{m}^3$ heating value and about 10 wt% tar when using Huainan bituminous coal under pyrolysis temperature between 500 and 600 °C. The produced gases were mainly H_2 , CH_4 , CO , CO_2 , C_2H_4 , C_2H_6 , C_3H_6 and C_3H_8 . The CFB combustor can burn semi-coke steadily. The application prospect of the new system was discussed.

Keywords Poly-generation · Coal pyrolysis · Tar utilization · Combustion · Gas conversion

1 Introduction

Due to the abundant reserves and stable supply, coal would be one of the main energy sources in the next decades in China. The conventional coal conversion technologies, such as liquefaction, gasification, combustion and carbonization, had been developed, and so far, the coal was utilized widely not only as the power energy, but also as resources to produce chemical liquids and gas fuels, whereas the coal extensive utilization induced a large quantity of serious effects on environment. What's worse, the traditional coal utilization did not realize coal resource efficiency utilization. And so, it was significant to develop a new conversion technologies to adapt to demanding for

higher efficiency and less pollutant emissions. The poly-generation was an integrated utilization approach to realize the maximum adding-value to coal. A number of researchers devoted their focus on poly-generation based on the coal pyrolysis.

Paterson (1997) developed the air-blown gasification cycle (ABGC) and investigated the flexibility of the reaction process in the pressured pilot-scale reactor. Zhu et al. (2010) studied the performance behaviors of coal-to-fuel poly-generation systems of coal-to-FT oil and coal-to-SNG, and the results demonstrated the coal-to-SNG process owned higher energy and efficiencies due to its low power consumption and less heat loss. Yi et al. (2012) proposed a new optimized method for integrated poly-generation system and investigated the effect of key operating parameters on the chemical conversion and energy utilization process of the whole system. Chiesa et al. (2005) and Kreutz et al. (2005) investigated performances, costs and prospects of poly-generation system

M. Ni · C. Li · M. Fang (✉) · Q. Wang · Z. Luo · K. Cen
State Key Laboratory on Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China
e-mail: mxfang@zju.edu.cn

which converted coal to hydrogen and electricity with carbon dioxide capture and storage. Through this system the effects of changing the ratio of electricity/H₂, gasifier pressure and purity of hydrogen were explored.

Solomon and King, Squire et al. (1984, 1986) made researches on coal pyrolysis property and indicated that the temperature of tar formation for bituminous coal and sub-bituminous coal was higher than for the lignite coal. Xu and Tomita (1987) revealed higher hydrocarbon yields were obtained from bituminous coals due to the different structure from other coal types. The researchers (1987) from CSIRO in Australia built and operated bench-scale experimental fluidized bed reactors of 1 g/h and 20 kg/h to get liquid fuels and investigated pyrolysis performances of two pyrolysers for three different Australian coals: Loy Yang brown coal, Liddell bituminous coal and Millmerran sub-bituminous coal, they confirmed the maximum tar yields from 3 coals were achieved at the temperature of 550–600 °C, and the amount of tar yields was directly related to the atomic ratio of H/C in parent coal (Cen et al. 2004; Wang et al. 2010).

By integrating a circulation fluidized bed (CFB) boiler and a fluidized bed pyrolyzer into one system, a new coal staged poly-generation system was developed in this paper. Coal will be pyrolyzed in the pyrolyzer first. Hydrogen-rich components were converted into gas and tar, and the remaining semi-coke for heating and power generation. Thus, poly-generation of gas, tar, heat and power can be realized in one system.

The main objective of the present research work was to analyze the new polygeneration process and evaluate both the process performance and characteristics of the products. Furthermore, the application prospect of the new system was discussed here.

2 Coal staged conversion poly-generation system

To realize comprehensive utilization of coal, a new staged conversion poly-generation system combined coal combustion and pyrolysis has been put forward which combines a circulating fluidized bed boiler and a fluidized bed pyrolyzer to realize coal pyrolysis and combustion in one system (Fig. 1). Here are the processes. Firstly coal is fed into a pyrolyzer to be heated and pyrolysed. Pyrolysis gas is then cooled and purified and used for chemical product or liquid fuel production. Tar is collected during purification and can be processed to extract monocyclic aromatic hydrocarbons and polycyclic aromatic hydrocarbons (PAHs) etc. and to make liquid fuels by hydro-refining. The heat absorbed in the pyrolyzer is supplied by high temperature circulating solids from a CFB boiler furnace. Semi-coke produced in the pyrolyzer is transported into the

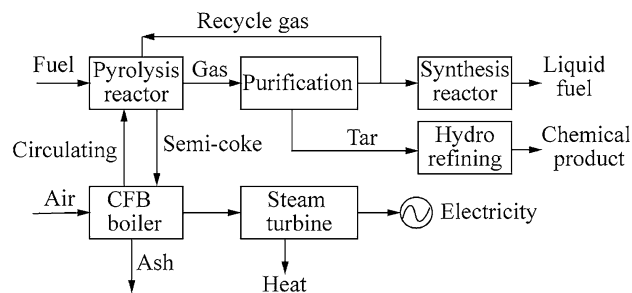


Fig. 1 Coal staged conversion poly-generation system combined combustion and pyrolysis

CFB boiler furnace with circulating solids to burn out, and to produce steam. Steam can be used to generate electricity, supply heat, etc.

The system has following advantages:

- (1) It produces not only steam and electricity, but also tar and gas, which can be converted as chemicals, liquid fuels, city gas or syngas, etc. So it provides a promising alternative energy.
- (2) Good fuel adaptability. Most bituminous coal and lignite coal can be used for the system.
- (3) Using staged conversion technology, valuable fractions of coal, such as the volatile fraction, are extracted and semi-coke is burned at boiler. Coal composition is fully utilized.
- (4) Most of sulfur compounds, nitrogen bases, and undesirable pollutant are removed during the pyrolysis. Therefore, pollutant emission from the boiler is very low.

On basis of many verification tests of poly-generation technology at 1 MW pilot plant, a 12 MW demonstration project, designed by Zhejiang University in cooperation with the Huainan Mining (Group) Co., Ltd. had been erected and operated successfully from August 2007. Many tests had been conducted at this system. Results show that it could attain stable and continuous operation and produced not only high yield tar and high quality gas, but also heat and power.

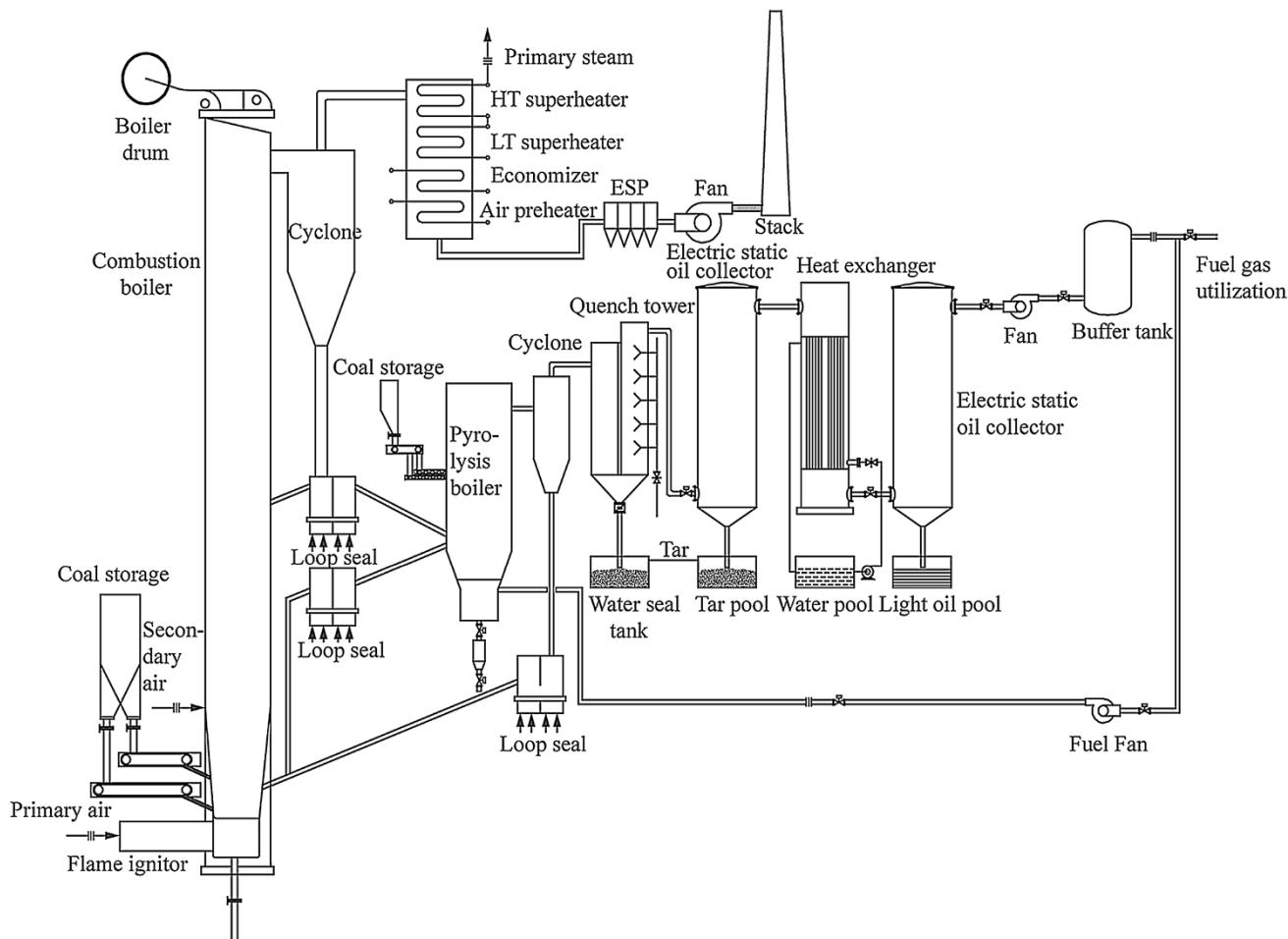
3 Experimental

3.1 Coal samples

The proximate and ultimate analysis of bituminous coal used in this experiment was given in Table 1. It was a low sulfur, high ash content bituminous coal. The particle size ranged 0–8 mm. According to the Gray-King assay, tar yield was 9.70 % wt/wt of coal. The bed materials used in this study were bottom ash of CFB boiler.

Table 1 Proximate analysis and ultimate analysis of sample bituminous coal

Proximate analysis (%)					$Q_{\text{net, ar}}$ (MJ/kg)	Ultimate analysis (%)					Gray-King assay tar yield Tar, ar (%)
M_{ar}	M_{ad}	A_{ad}	V_{ad}	FC_{ad}		C_{ad}	H_{ad}	N_{ad}	$S_{\text{t,ad}}$	O_{ad}	
3.87	2.07	30.41	28.78	38.74	21.4	54.72	4.65	0.98	0.25	6.92	9.7

**Fig. 2** The 12 MW gas, tar, and power poly-generation pilot plant

3.2 MW pilot-plant description

As shown in Fig. 2, the 12 MW pilot-plant mainly consisted of a 75t/h CFB boiler, a fluidized bed pyrolyzer, gas cool and clean system. Coal was first fed to the pyrolyzer, and pyrolyzed by high-temperature ash from the CFB boiler. Volatiles and easily gasified components were converted into gas and tar, while semi-coke was sent to the CFB boiler to burnout for heat or power generation. The high temperature ash from the boiler furnace is separated by high temperature cyclone and part of ash was sent to the pyrolyzer by a recycle device to supply the heat for pyrolyzer. Gas from pyrolyzer first was sent into the dust collector to remove the dust and then cooled and purified in a quench tower and electrical static oil

collector, and tar was collected, simultaneously. The gas was then cooled in a heat exchanger and purified by 2nd electrical static oil collector. Tar could be made as valuable chemicals or liquid fuels by hydro-finishing. After purification, the produced gas, H_2 , CO , CH_4 , CO_2 , C_2H_4 , etc., could be used as town gas or syngas for chemical product.

3.3 Sample collection and analysis

During experiments, coal, gas, tar, semi-coke and ash samples were taken for analysis. Tar sampling was carried out according to literature (Simell et al. 2000). Gas was sampled from the pyrolyzer via sampling line to gas bottles in the cold bath. The tar compounds in the gas were absorbed by solvent in

the bottles. The temperature, and flow rate of the gas flow during sampling were measured. The tar yield was obtained after the solvent and water was removed by distillation. Tar and gas compositions and concentrations were measured using GC-MS and gas chromatograph, respectively.

4 Results and discussion

4.1 Typical operating results of 12 MW poly-generation system

A performance test was made from August 2007. The test results showed that the boiler and pyrolyzer operated stably and well agreed with design value. The system can realized poly-generation of gases, tar and power from coal in an integrated production process. The operation results showed that:

- (1) CFB boiler and fluidized bed pyrolyzer could coordinately operate. Hot ash and semi-coke recycled between the boiler and pyrolyzer stably and reliably.
- (2) CFB boiler burned semi-coke from pyrolyzer stably, and also could run based on the requirements of the respective adjustment of pyrolyzer and boiler load.
- (3) The poly-generation system could produce tar and gas stably. Coal gas purification and tar recovery system operated normally.

Typical operating results were briefly shown in Table 2.

4.2 Effects of pyrolyzer temperature on product yields and compositions

Table 3 and Fig. 3 show the gas compositions at different pyrolysis temperatures. The gas was typical of

Table 2 Typical operating results of 12 MW poly-generation system

Items	Boiler	Pyrolyzer
Bed temperature (°C)	925	650
Circulating gas (m ³ /h)	40,000	2,800
Coal feed rate (t/h)	3	10.4
Gas output (m ³ /h)		1,210
Tar output (t/h)		~1
Steam output (t/h)	70	
Tar yield (%)		~10

Table 3 Typical gas compositions

Concentration (V/V, %)										H ₂ S (mg/m ³)	LHV (MJ/m ³)
H ₂	O ₂	N ₂	CH ₄	CO	CO ₂	C ₂ H ₄	C ₂ H ₆	C ₃ H ₆	C ₃ H ₈		
24.18	0.21	7.96	36.29	7.84	11.7	3.28	7.17	0.41	0.96	8,451	25

bituminous coal pyrolysis gas. H₂ and CH₄ concentrations were high, while CO content was low. The gas should have a better utilization prospect, such used as town gas or raw material of chemical synthesis processes. O₂ and N₂ were very low, indicating the 12 MW poly-generation system had excellent air-tightness. H₂ and CO increase with the pyrolysis temperature, while CH₄ changes slowly.

H₂ was mainly from the saturated hydrocarbons cracking reaction, as well as condensation of aromatic hydrocarbons. Aromatic hydrocarbons condensation occurred in the high temperature, while at lower temperatures, H₂ was mainly from saturated hydrocarbons cracking reaction. With the increase in temperature, condensation of aromatic hydrocarbons carried out quickly and promoted the growth of the H₂. While the temperature rose to a certain extent, the degree of condensation would not deepen, the H₂ growth rate gradually slow (Li et al. 1998). CO was mainly from the following two ether components: ether bridges connecting each unit of coal, which was the main sources of CO under low temperature; and the other was diarylether compounds breaking down under high temperature (Sada et al. 1992). With the temperature increasing, diarylether compounds of coal, carbonyl functional groups of tar, as well as oxygen heterocyclic ring and OH-component began to crack to produce CO, and the higher temperature, the more prone to react. Therefore, CO content increase with temperature (Yi et al. 2012).

CH₄ was slightly different from H₂ and CO. With the increase in temperature, CH₄ increased first and then reduced, when around 630 °C reaching the maximum value. CH₄ was mainly from the cracking of methyl and

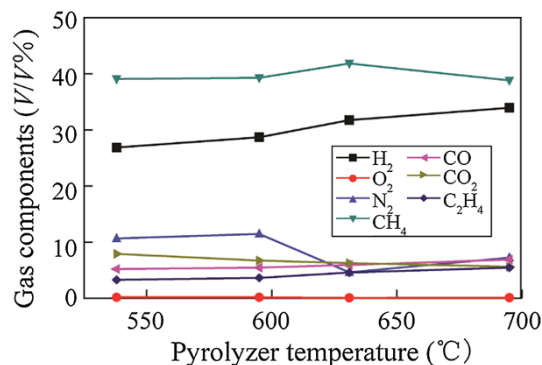


Fig. 3 Effect of pyrolysis temperature on gas compositions

methoxyl group at a relatively low temperature and decomposition of methylene connecting various units of coal at high temperature (Edwards and Smith 1980; Yi et al. 2012). With the temperature increases, more reaction occurred gradually, and the degree of reaction to in-depth, which would help to raise the CH₄ concentration. On the other hand, as the temperature increased, dehydrogenation of tar, in particular, aromatic hydrogenations, gradually increased, more tar began to form a coke. When the temperature exceeded 630 °C, dehydrogenation of aromatic hydrogenations were dominate, CH₄ concentration decreased.

Tar was the important liquid products of poly-generation system. As shown in Fig. 4, the tar yield changed with the temperature markedly. When the temperature of the pyrolyzer was 550–600 °C, the maximum tar yield reached 11 %. As the temperature continued to rise, the tar yield reduced. This was in good agreement with those obtained by Ralph J work (Tyler 1980). Table 4 represented the results of proximate and ultimate analysis of tar, the atomic ratio of H/C of tars from coal pyrolysis was higher than that from the parent coal. As temperature increased, the atomic ratio of H/C declined, the similar results were found by Edwards and Smith (1980).

At the beginning of coal pyrolysis, easier pyrolysis components of coal decomposed first. With the rising of temperature, more difficult fractions of coal cracking occurred, and high temperature speeds up the rate of coal pyrolysis. Tar yield got a marked increasing. When the pyrolysis temperature reached 550–600 °C, tar yield reached the maximum, followed by the tar yield reducing

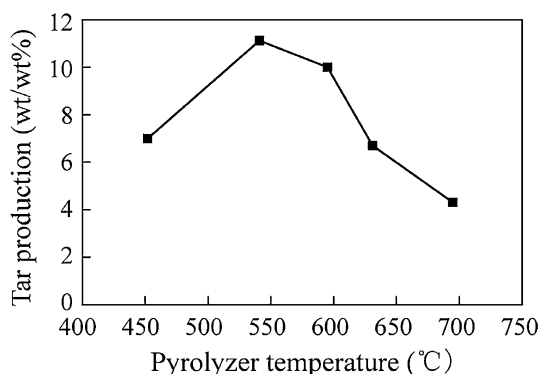


Fig. 4 Effect of pyrolysis temperature on tar yields

Table 4 Proximate analysis and ultimate analysis of tar

Pyrolyzer Temperature (°C)	Proximate analysis %				Qnet,ar (MJ/kg)	Ultimate analysis %				
	M _{ad}	A _{ad}	V _{ad}	F _{Cad}		C _{ad}	H _{ad}	N _{ad}	S _{t,ad}	O _{ad}
600	1.08	4.75	69.58	24.59	34.73	82.62	5.42	1.43	0.44	4.26
650	0.96	6.24	64.56	28.24	33.6	81.39	4.26	0.44	0.46	6.25

with the temperature. It is mainly due to the following aspects: (1) with the temperature increasing, on the one hand, more tar yielded, and on the other hand, part of tar cracked to generate gas, so that the tar yield declined. (2) Pyrolysis atmosphere. With the pyrolysis temperature increasing, H₂ concentration increased, as shown in Fig. 3. The existence of H₂ reduced the polymerization of free radicals, reduced the combination opportunity for free radical to generate tar (Zhu et al. 1998).

4.3 Influence of temperature on tar cut fractions

Table 5 shows the influence of temperature on tar cut fractions. With the temperature increasing, >360 °C cut fractions gradual increased. So heavy components increased and took the main shares among the tar cut fractions, which making tar “heavy”, and the quality declined. Wang et al. had confirmed that the main yields of tar cut fractions at the temperature below 300 °C were phenol and its alkyl-substituted homologs from C1 to C3.

Table 6 shows the hydrocarbon compositions of tar. It indicated that alkanes accounting for about 15 %, aromatic hydrocarbons accounts for about 50 %, compared with the rest of colloid. With the temperature increasing, alkanes in

Table 5 Cut fractions of tar from different pyrolysis temperatures

Pyrolyzer temperature (°C)	The percentage (%) of every distillation range (°C)				Recovery percent (%)
	<170	170–300	300–360	>360	
540	4.14	5.72	25.09	57.5	92.45
600	6.77	4.33	19.5	62.49	93.09
630	8.31	6.33	15.5	61.3	91.44
700	7.24	4.18	11.26	69.59	92.28

Table 6 Hydrocarbon Compositions of tar

Compositions (%)	Temperature (°C)	
	540	600
Alkane	15.2	14.6
Aromatic hydrocarbon	52.9	51.7
Colloid	31.9	33.7
Total	100	100

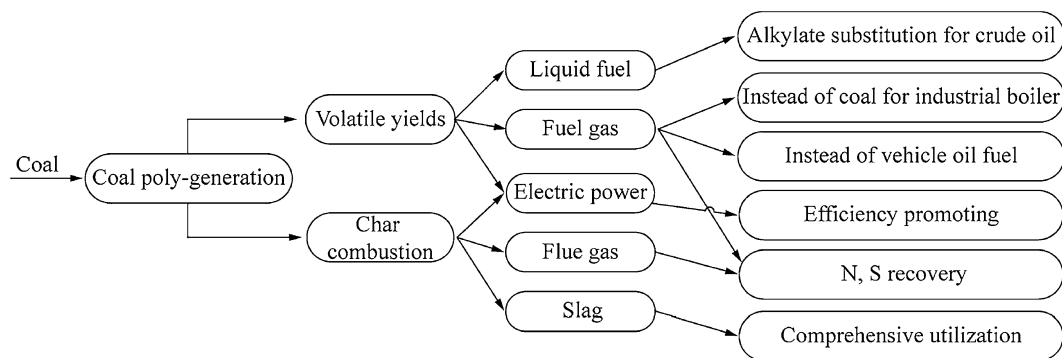


Fig. 5 Application prospect of the new poly-generation system

tar decreased slightly, while the aromatic components increased slightly.

5 Application prospect

Coal staged conversion poly-generation system converted coal into gas, tar, electricity and heat. Gas can be converted into natural gas or chemical product and tar was also converted into liquid fuel by hydro-refine process. Steam can be used for power generation, heat supply and refrigeration. What's more, N-containing and S-containing pollutants in fuel gas and flue gas were recovered to produce yields of sulfuric acid and calcium nitrite respectively, ash was refined to obtain precious metals and then used as construction materials. In general, coal poly-generation achieved comprehensive utilization of coal resource and nearly zero emission of pollutants. The application prospect of poly-generation system was presented in Fig. 5.

At present, the total capacity of thermal power unit in China attained 765 GW, which consumed about 1.8 billion tons coal for power generation in 2011. The volatile of total 1.8 billion tons coal is equal to natural gas of 271.3 billion cubic meters of which scale was 23 times larger than that of the West to East Pipeline Project or crude oil of 0.22 billion tons (the quantity of China oil imported was 0.25 billion tons in 2011). The yield of all coal ash will achieve about 0.45 billion tons and can produce 1.1 billion tons cements with 32.5 MPa strength grade (the amount of cement achieving 2.06 billion tons in 2011) or generate Al_2O_3 product of 90 million tons (the number of electrolytic aluminium was 18.06 million tons in 2011) and 20 million tons ferric oxide. The recovery of sulfuric acid will reach 40 million tons (apparent sulfur consumption of China was 13.52 million tons) and about 30 million tons products of calcium nitrite or ammonium nitrate.

If all coal-fired power plant in the whole China are retrofitted into poly-generation system, the investment of system will increase by 10 %–30 %. If fuel gas from poly-

generation system is utilized through highly efficient gas turbine to form partial gasification combined cycle power generation, the average power plant efficiency will increase 6.4 %. The poly-generation system can also decrease emission of SO_2 and NO_x over 50 %. So the poly-generation system has ability to increase the power plant efficiency and decrease pollutant emission.

Now the industrial boiler is a big problem as it's huge number, low efficiency and high pollution. If all coal fired industrial boiler would be substituted for fuel gas from the poly-generation system, it could increase efficiency of industrial boiler from 65 to 85 %, which will save 100 million tons coal every year, and decrease the emission of SO_2 , NO_x , $\text{PM}_{2.5}$ and CO_2 greatly.

In summary, the poly-generation system can supply multi-energy like gas, oil, heat and electricity and will be strategically important for energy-saving and emission reduction. So it is a promising clean and high efficiency coal technology to electric power generation, which will increase the coal utilization value and decrease the pollutant emission greatly.

6 Conclusions

A new coal staged conversion poly-generation system combined coal combustion and pyrolysis has been developed for clean and high efficient utilization of coal. Through pyrolysis, coal converted into gas, tar and semi-coke. The process temperature affected the relative distribution of three products. At high temperature, up to about 550–600 °C, the tar yield reached the maximum value. The system can realize coal multi-product generation and has a great potential to increase coal utilization value.

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