

Effect of operating conditions on gas components in the partial coal gasification with air/steam

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Abstract—With increasing environmental considerations and stricter regulations, coal gasification, especially partial coal gasification, is considered to be a more attractive technology than conventional combustion. Partial coal gasification was conducted in detail under various experimental conditions in a lab-scale fluidized bed to study the factors that affected gas components and heating value, including fluidized air flow rate, coal feed rate, and steam feed rate, gasification temperature, static bed height, coal type and catalyst type. The experiment results indicate that gasification temperature is the key factor that affects components and the heating value of gas is in direct proportion to gasification temperature. There exists a suitable range of fluidized air flow rate, coal feed rate, steam feed rate and static bed height, which show more complex effect on gas components. High rank bitumite coal is much more suitable for gasification than low rank bitumite coal. The concentrations of H₂, CO and CH₄ of bitumite coal are more than those of anthracite coal. Compounds of alkali/alkaline-earth metals, such as Ca, Na, K etc., enhance the gasification rate considerably. The catalytic effects of Na₂CO₃ and K₂CO₃ are more efficient than that of CaCO₃.

Key words: Partial Coal Gasification, Gas Components, Heating Value, Fluidized Bed

INTRODUCTION

Increasing petroleum prices, an intense demand for coal, and serious environmental pollution have led to some important questions that constrain China's economic development. It is important to develop the most effective technologies to utilize coal as an efficient and clean source of energy. The gasification technology of coal has been shown to have technical viability and environmental superiority. Compared with whole-gasification, partial-gasification does not require physical conditions such as high temperature or high pressure to convert fixed carbon to syngas completely. According to the different reaction characteristics of different components of coal, partial-gasification utilizes coal in stages. The processing equipment of partial-gasification is simpler and cheaper. Syngas with low heating value can act as fuel and chemical raw material. The residual char can be burned to produce steam, which can be used to generate electricity and supply heat, then form the offering of gas, heat and electricity. Partial-gasification can also process directional deprivation of the harmful constituents, such as sulfur, nitrogen, chlorine and mercury. Partial-gasification is the key technology of the second generation PFBC-CC [1,2]. Coal gasification in fluidized beds attracts more attention because of its enormous throughput capacity, high gasification intension, unobstructed mass transfer inside the gasifier, good applicability of coal type and little pollution to the environment [3-6]. The application of partial-gasification ranges from the Winkler process, one of the first commercial demonstrations of fluidized-bed technology, to the more recent HTW (high-

temperature Winkler) combined-cycle power-generation process.

Numerous investigations on the gasification of coal with carbon dioxide, oxide and/or steam have been conducted in a lab-scale or full-scale bed [7,8], but few about partial-gasification in a fluidized bed. And the factors that affect gas components and heating value are not enough. In order to raise the partial-gasification efficiency, it is important to study the sensitivity or the relevant significant factors, such as fluidized air flow rate, coal feed rate, steam feed rate, bed temperature, static bed height, coal type and catalyst type, to the gas components and heating value in an atmospheric pressure fluidized bed gasification boiler. This paper is also expected to be beneficial to the design and operation control of partial-gasification furnace.

EXPERIMENTAL EQUIPMENT AND RAW MATERIAL

1. Experimental Equipment

The experiment is operating on a homemade atmospheric pressure fluidized bed gasifier (Seen in Fig. 1). The gasifier design is extremely compact and flexible, and is capable of operating efficiently on a wide range of fuels, steam and air. The whole system is made up of a high temperature gas heated system, a steam generation system, a feeding system, a blower system, a gasifier furnace, a measure-control system and a sample-analyzing system, etc. The most obvious character of this equipment is that it has a jacket to heat the material. The flue gas at 1,100 °C from oil firebox heats the furnace up with the help of the furnace jacket baffle, and offers the heat for warming-bed material and furnace. The diameter of the inner tube is 100 mm, and the diameter of the outer tube is 133 mm. The vertical height from the grid plate to the furnace exit is 4.3 m. The grid plate has 18 jelsabs, 6 at inside track and 12 at outside track. There are 3 eyelets, whose diameters are 1 mm,

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distributed on every jebab. When the airflow from blower fan is heated to 450 °C by the air heater in the firebox, it will mix with the 260 °C steam which comes from the steam super-heater. The mixture

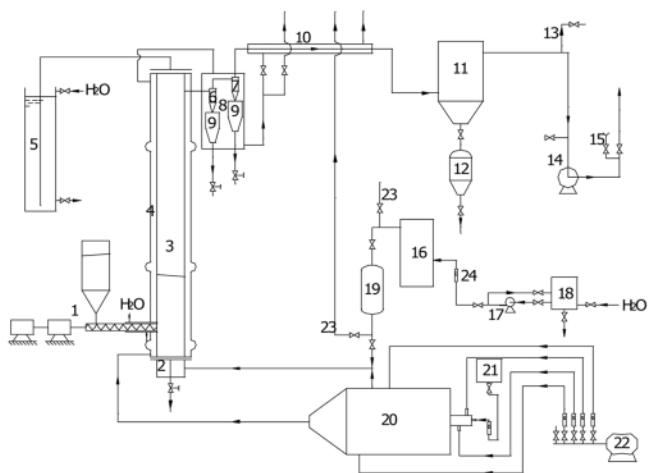


Fig. 1. Atmospheric pressure fluidized bed gasifier process.

1. Feeding system	13. Gas sampling equipment
2. Air chamber	14. Draft fan
3. Gasifier furnace	15. Gas burning equipment
4. Heating jacket	16. Electrical heating boiler
5. Anti-exploder	17. Water circulating pumps
6. Primary cyclone separator	18. Water tank
7. Secondary cyclone separator	19. Steam super-heater
8. Heating jacket	20. Oil firebox
9. Ash hopping	21. Oil tank
10. Heating jacket	22. Roots gas blower
11. Bag-type dust collector	23. Steam side road
12. Ash hopper	24. Metal flow meter

enters the gasifier furnace through the grid plate. Steam of 0.6 MPa from the electric boiler steps pressure down by the reduction valve then enters the steam super-heater to raise the temperature further. The design of the steam bypass, the adjustability of heating power of the electric boiler and the steam super-heater can ensure the accurate steam feed rate. Coal is added by a screw feeder. With some imperceptible ash, the syngas leaves the exit on the top of gasifier furnace, passes through a primary cyclone separator, a secondary cyclone separator and a bag-type dust collector in turn. The syngas is burned out in a combustion train and then vented out by a fan at last. The parameters monitored in this test include temperature, pressure or pressure drop, air flow rate, fuel feed rate and the components of the syngas, etc. The sampling port of syngas is located at the exit of the bag-type dust collector. The concentrations of H₂, CO, CO₂, and CH₄ are measured by using a GC of 1,102 type made in China. Simultaneously, a multifunction gas analyzer of NGA2000 type made in Germany is used to monitor gas components on line, which is compared with the results measured by GC.

2. Raw Material

Three types of coal chosen for this investigation are a high-grade Xuzhou bituminous coal, a low-grade Xuzhou bituminous coal and a Yangquan anthracite coal. Their particle diameter distribution can be seen in Table 1. Proximate analysis and ultimate analysis of three types of coal are shown in Table 2. The bed material used in the experiment is quartz, whose average particle diameter is about 0.362 mm.

Three types of catalyst used in the experiment include Tangshan limestone in Nanjing, Mufushan dolomite in Nanjing and pure sodium carbonate. Analytical reports of limestone and dolomite are shown in Table 3.

3. Principle of Coal Gasification

When the coal enters the gasifier, a pyrolysis process first

Table 1. The particle diameter distribution of coal

Coal type	Diameter distribution (%)				Average diameter (mm)
	0.3-0.4 mm	0.4-0.6 mm	0.6-0.8 mm	0.8-1.0 mm	
High-grade bituminous	17	19	10	54	0.63
Low-grade bituminous	24	29	19	28	0.54
Anthracite	12	26	24	38	0.61

Table 2. Proximate analysis and ultimate analysis of coal

Coal samples	High-grade Xuzhou bitumite	Low-grade Xuzhou bitumite	Yangquan anthracite
Proximate analysis	Mad %	2.72	4.42
	Vad %	30.57	25.24
	Cad %	54.09	47.44
	Aad %	12.62	22.90
	Cad %	70.40	60.36
Ultimate analysis	Had %	4.54	3.72
	Sad %	0.63	0.44
	Nad %	1.24	0.92
	Oad %	7.85	7.24
	Higher caloric heat value Qg MJ/kg	28.91	24.27
Lower caloric heat value Qd MJ/kg	27.91	23.40	25.61

Table 3. Analytical reports of limestone and dolomite

Matter	CaO	MgO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	Unit area
Limestone	51.22%	0.31%	3.76%	1.45%	0.45%	4.8081 m ² ·g ⁻¹
Dolomite	30.08%	20.38%	1.56%	0.48%	0.32%	3.2114 m ² ·g ⁻¹

happens, which includes pyrolysis and fusculation. The pyrolysis reaction, taking place in the prophase of the pyrogenation, includes the following processes. Bridge bonds in the coal's molecular structure are ruptured into free cells. Gaseous hydrocarbon forms because of the fattiness lateral chains ruptured. The functional groups, which contain oxygen, are cracked and the substances of H₂O, CO etc., come into being. Low molecular weight compounds are cracked into more volatile products.

In the following pyrogenation, there is mainly a fusculation reaction. When the gasification temperature is 550-600 °C, the colloid matter solidifies to form carbocoal. If the temperature is higher, the aromatic structure will dehydrogenate and the carbocoal will turn into coal char.

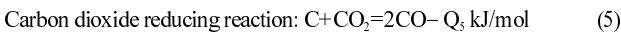
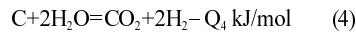
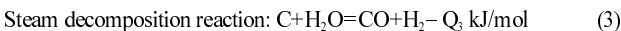
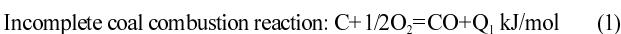
The whole equation of coal's pyrogenation is:



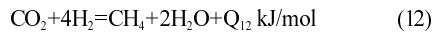
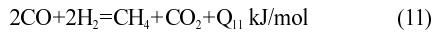
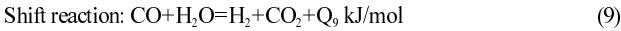
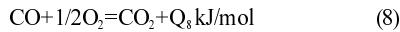
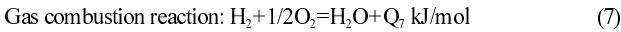
At high temperature, the subsequent gasification reactions will happen: gaseous hydrocarbon and tar have a secondly pyrogenation reaction and produce C, CH₄, H₂, CO, H₂O and so on.

After the cracking process, the coal gasification can be divided into two types: non-homogeneous gas-solid reaction and homogeneous gas reaction.

1. Non-homogeneous gas-solid reaction



2. Homogeneous gas reaction

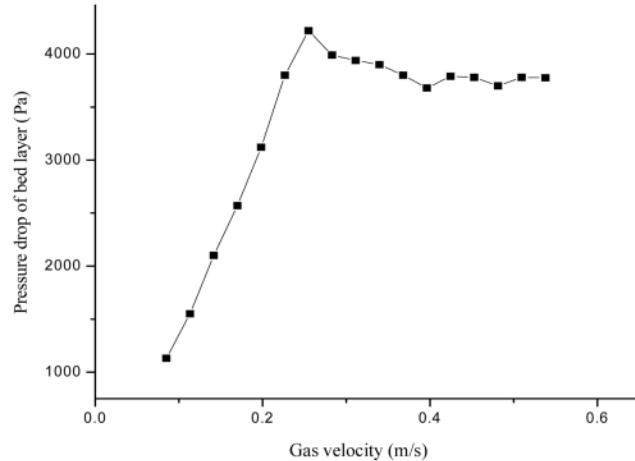
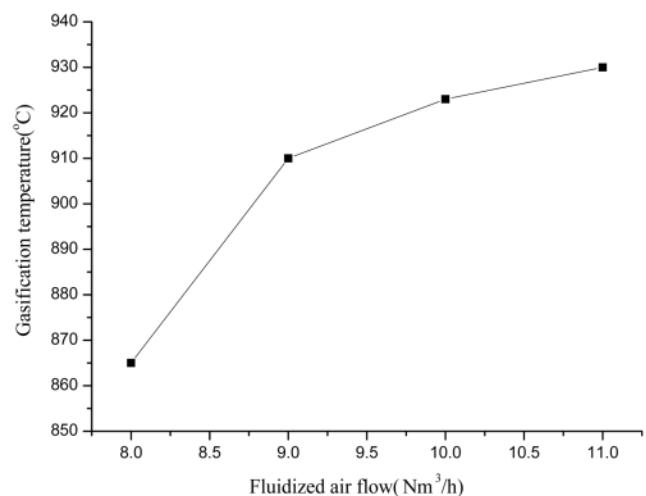


RESULTS AND DISCUSSIONS

1. Definitions

Heating value of cool syngas is a very important factor which reflects the performance of gasifier: $Q_{LHV} = (C_{H2} \times 2581 + C_{CO} \times 3018 + C_{CH4} \times 8558) \times 0.01 \times 4.1868 \text{ kJ/Nm}^3$. C_{H2} , C_{CO} , C_{CH4} represent the concentrations of H₂, CO, CH₄, respectively.

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**Fig. 2. Fluidized characteristics of the bed material.****Fig. 3. The relationship between fluidized air flow and gasification temperature.**

2. The Fluidized Characteristic of the Bed Material

The fluidized characteristic of the bed material is shown in Fig. 2. When the gas velocity exceeds 0.28 m/s, the pressure drop of bed layer almost keeps constant. At this time, the bed material is considered as the beginning of fluidization, and the critical fluidized velocity is about 0.28 m/s.

3. The Influence of Fluidized Air Flow Rate on Components and Heating Value of Syngas

When the coal feed rate, the static bed height and the steam feed rate are 4.52 kg/h, 200 mm, and 1.8 kg/h, respectively. Fig. 3 presents the effects of the fluidized air flow rate on the gasification temperature.

The bed layer temperature is increased with the increase of the

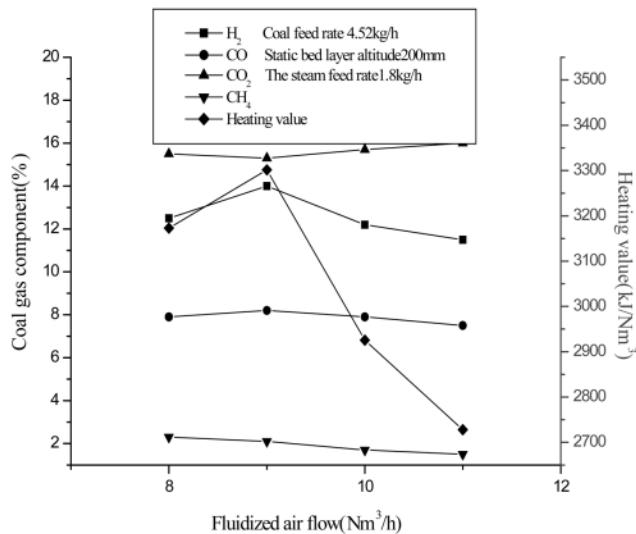


Fig. 4. The relationship between fluidized air flow and syngas components and heating value.

fluidized air flow rate. The increase of fluidized air flow rate means that the flow velocity and the quantity of oxygen in the gasifier are increased, which is in favor of making the heat transfer or mass transfer strengthen between gas and solid. So, the increase of the fluidized air flow rate favors the exothermic reactions (1), (2), (7), (8) and leads to a higher coal conversion and gasification temperature. When air flow rate is increased further, the heat dissipating capacity of the gasifier, the quantity of heat that is needed by the fluidized air flow rate and the sensible heat carried away by the syngas are also increased, which tends to making the temperature of bed layer smooth out.

From Fig. 4, it can be seen that with the increase of the fluidized air flow rate, the concentrations of H₂ and CO (V/V, same in the following chapter) is increased first and then decreased, while the concentration of CO₂ is decreased first and then increased, and the concentration of CH₄ shows decreasing trend. The heating value of the cool syngas reaches maximum when the fluidized airflow is 9 Nm³/h.

With the increase of the fluidized air flow rate, the entrainment capacity of the flow is raised, and the gasification on the dilute phase section at the top of the gasifier becomes much more acute. At the same time, the increase of gasification temperature and the increase of gasification reaction rate of carbon are in favor of making the balance point of the reaction (3) and reaction (5) shift right. The reactions of (1) and (2) can be replaced by the total combustion reaction of carbon:



Φ is a function of char-particle diameter and temperature. The ratio of mol concentrations of CO and CO₂ follows the equation:

$$\frac{C_{CO}}{C_{CO_2}} = 2400e^{-5180/(RT)} \quad (14)$$

'R' here is the gas constant (8.314 J/mol), 'T' represents the temperature of the carbon particle. The conclusion drawn from the above equation is that higher temperature is propitious to the formation of

CO. The increase of gasification temperature can also lead to prick up the secondary decomposition of these hydrocarbons and tar carrying high heating value. Above variable factors favor the increase of concentrations of H₂ and CO. While the fluidized air flow rate is further increased, the rates of the gas combustion reaction (7) and (8) will be increased. And much more combustible gas will burn, such as CO, H₂, CH₄, while the unavailable component (CO₂) is increased. Simultaneously, the gas residence time in the gasifier becomes short and the elutriation quantity of the granules is excessive. When the gasification intension is kept constant, the increase of the gas production rate necessarily makes gasification extent insufficient, while influencing the performance of the syngas. To attain a reasonable gasification effect, the heating value of the syngas and the rate of gas generation must be considered in equilibrium. That is, the ratio of air and coal must be selected suitably.

4. The Influence of the Coal Feed Rate on Components and Heating Value of Syngas

The coal feed rate's influence on the components of syngas is

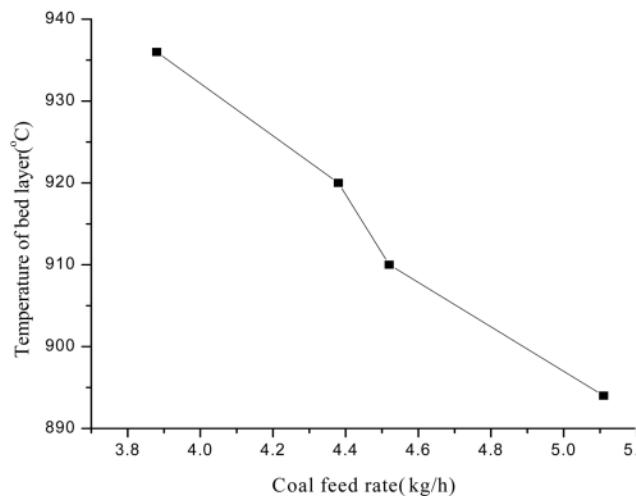


Fig. 5. The relationship between coal feed rate and temperature of bed layer.

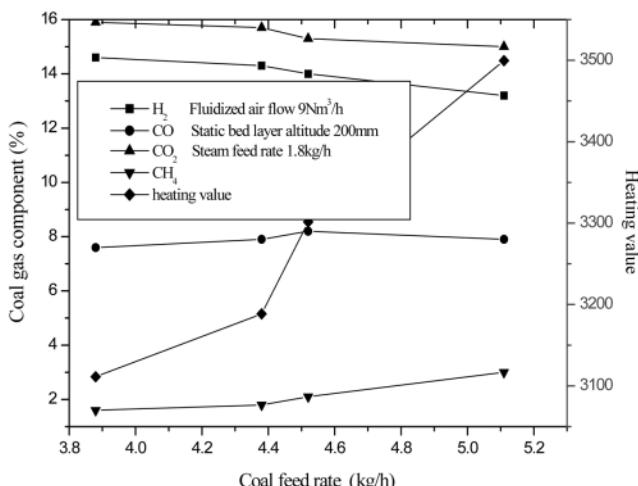


Fig. 6. The relationship between coal feed rate and syngas components and heating value.

complicated. With the increase of coal feed rate, the temperature of the bed layer is decreased gradually (seen in Fig. 5), the concentration of CO is increased firstly and then decreased, the concentrations of H_2 and CO_2 are decreased, the concentration of CH_4 is increased and the heating value of syngas is increased (seen in Fig. 6).

The increase of the coal feed rate means the increase of absolute quantity of coal, which attends the reaction. At the presupposition that the quantity of gasification agent (air and steam) keeps a constant, the gas production rate per unit coal is decreased, and the concentration of CO is increased accordingly. But it also bring the negative effect of the decrease of bed layer temperature, which causes the coal gasification velocity decreased, the gasification extent not enough, and the concentration of CO decreased. Although the increase of coal feed rate arises more coal to crack into more H_2 , the decrease of the bed layer temperature makes the equilibrium point of reaction (9) shift right and is not favor of the reaction (3) and (4). Considering the above two factors, the absolute quantity of H_2 is increased but limited. Because of the increase of syngas amount, the concentration of H_2 is inversely decreased in a small degree. The concentration of CH_4 is in inverse proportion to the coal feed rate, because CH_4 is not produced by methanation but by cracking the volatile matter in the coal and the reactions (10), (11) and (12) need existence catalyst [9,10].

5. The Influence of Steam Feed Rate on Components and Heating Value of Syngas

With the increase of steam, much more steam attends the reactions (3) and (4). As reactions (3) and (4) are endothermic, and the quantity of heat, which is needed to raise the temperature of steam, is increased, the temperature of bed layer shows decreasing trend (seen in Fig. 7).

The components and the heating value of syngas change with respect to the steam feed rate as presented in Fig. 8. When the quantity of steam keeps in lesser, the decomposition reaction of steam mainly takes place in reaction (3). At this time, the increase of steam is favor of the producing of H_2 and CO, and the increase of H_2 is more obvious than that of CO. The possible reason is that the decrease of bed layer temperature makes the equilibrium point of the reaction (9) between CO and H_2O shift right. When the steam feed

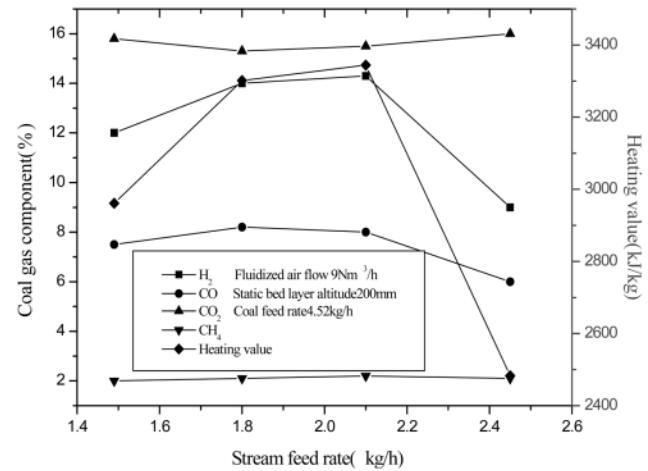


Fig. 8. The relationship between steam feed rate and syngas components and heating value.

rate rises, much more steam attends the reaction (4). As a result, the concentration of H_2 keeps on increasing, CO decreasing and CO_2 increasing. If the quantity of steam becomes much higher, the bed layer temperature falls considerably and the residence times of gas and solid inside the furnace become shorter. At this point, the excess steam is not favor of gasification, which shows that the decrease of the concentration of H_2 , CO and the heating value of syngas.

6. The Influence of Gasification Temperature on Components and Heating Value of Syngas

In order to study the influence of bed layer temperature on gasification process solely and ensure a constant residence time, other gasification parameters are kept constant. Different temperatures of bed layer are achieved by adjusting justly the temperature and quantity of flue gas in the heating jacket and initial temperature of the fluidized flow air. Fig. 9 shows the variation trend of the components of syngas and the heating value according to the bed layer temperature.

The temperature of bed layer is one of the main factors affecting the components of syngas. If the temperature of the bed layer is high-

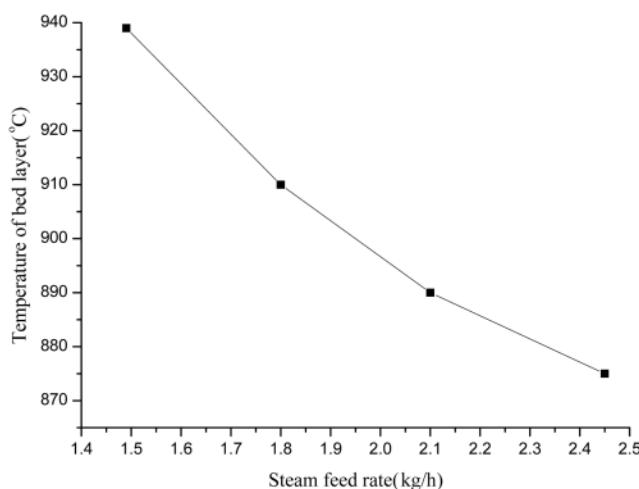


Fig. 7. The relationship between steam feed rate and temperature of bed layer.

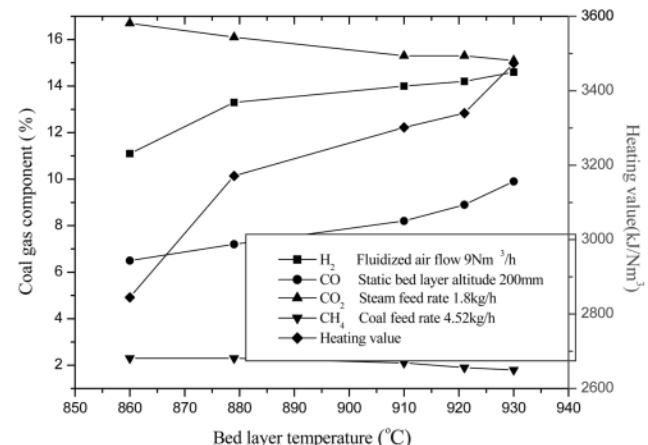
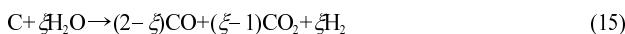


Fig. 9. The relationship between bed layer temperature and syngas components and heating value.

er, much more carbon atoms' energies will exceed the activation energies needed in the gasification, and much more effective collision will happen between carbon atoms and oxygen atoms or steam molecule. So the reaction rate of gasification is accelerated and the gasification intension is strengthened. According to the conclusion drawn by Fang, when the temperature of the bed layer increases 20-30 °C, the reaction rate of gasification will be doubled [11]. Simultaneously, the increase of temperature of bed layer also promotes a gaseous secondary reaction among the volatile components. H₂ comes from the pyrolysis reaction of hydrocarbons components and the condensation reaction of aromatic ring. CO comes from the pyrolysis reaction of hydroxyl functional group and oxygen heterocyclic ring in the tar molecule. Above-mentioned variety factors bring the increase of H₂ and CO in the syngas.

Although the increase of bed layer temperature accelerates the rate of reaction (4), the equilibrium constant of reaction (3) is increased faster with the increase of bed layer temperature than that of reaction (4) [11]. Steam decomposition reactions (3) and (4) can be summarized as:



ξ is experimentally determined to be in the range 1.1-1.5 and is found to decrease with increasing temperature [4]. So if other parameters remain invariable, the increase of CO₂ in syngas is slower than CO. The phenomenon of the increase of CO and the decrease of CO₂ is the representation of the equilibrium point of reaction (9) shifting left and the equilibrium point of CO₂ reducing reaction (5) shifting right. The decrease of CH₄ in syngas is the result of the methane's second decomposing, which is promoted by the bed layer temperature. In a word, the heating value of syngas is proportional to the temperature of bed layer.

7. The Influence of Static Bed Height on Components and Heating Value of Syngas

The formation characteristic and heating value of syngas with static bed height is shown in Fig. 10. As the bed layer static altitude is increased, the volatile matter in the broiling layer is cracked more strongly and the concentrations of H₂ and CO are increased, whilst the concentration of CH₄ is decreased (seen in Fig. 10). As the bed height is below to the coal feed port, a part of volatile matter and

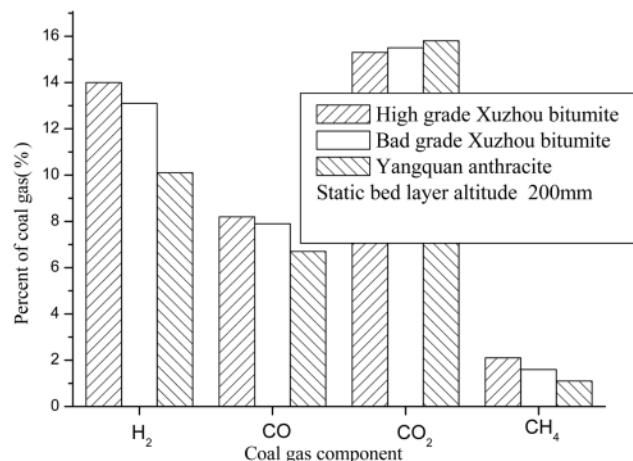


Fig. 11. The relationship between coal type and syngas components

small grain separate out of coal before the coal mixes with the bed material. In addition, the gasification ability in suspended space is much lower than that in dense phase space. So the concentration of CH₄ is more than 4% in the syngas. When the bed layer static altitude reaches 400 mm, the concentrations of H₂ and CO are inversely decreased because compared with the inner diameter of gasification furnace, the altitude of bed material is too high and the bed material surges acutely to prevent the valid gasification of coal. Gasification operation is difficult if the static bed height exceeds 400 mm.

8. The Gasification Characteristics of Different Types of Coals

As an organic mixed mineral carrying many impurities, coal's components, ash fusion temperature, activity, cohesiveness and free-swelling index can all affect the gasification and its efficiency greatly. Therefore, the gasification characteristics of three types of coal were studied in the experiment. On the whole, if all of the gasification parameters are the same, the combustible components (H₂, CO, CH₄) in syngas coming from bitumite coal are more than those from anthracite coal, and the combustible components in syngas coming from high rank bitumite coal are more than those from low rank bitumite coal (seen in Fig. 11). The reason is explained as follows: the higher coal rank, the higher activity energy and the higher temperature when thermal decomposition begins to happen. The char gasification reaction rate becomes faster with coals of progressively lower rank and the reactivity is somewhat more sensitive to coal quality at atmospheric pressure [12]. In addition, the more contents of moisture and volatile matters in coal, the looser coal's structure, the bigger unit surface-area of the residual char, which has abundant transitional pores and macroscopic holes. Then the gasification agents can be easy to diffuse to the reaction surface.

9. The Influence of Catalyst Type on Syngas Components

It is commonly believed that alkali/alkaline-earth metal compounds are dependable and effective catalysts for gasification. During the first stage of gasification, while the carbon is being heated, reduction of the alkali/alkaline-earth metal compounds and subsequent oxidation to metal oxide/carbonate invariably takes place. Much more dispersion efficiency is attained by physical mixing of metal oxide/carbonate with carbon.

The CaO derivated from limestone's decomposing has the catalysis to coal's gasification (X is CaO) [13]:

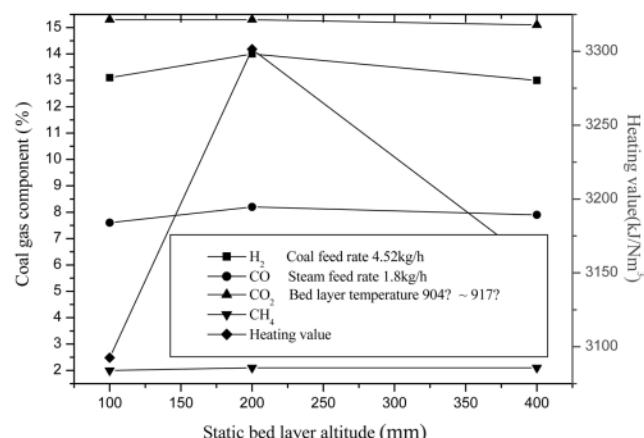


Fig. 10. The relationship between static bed layer altitude and syngas components and heating value.

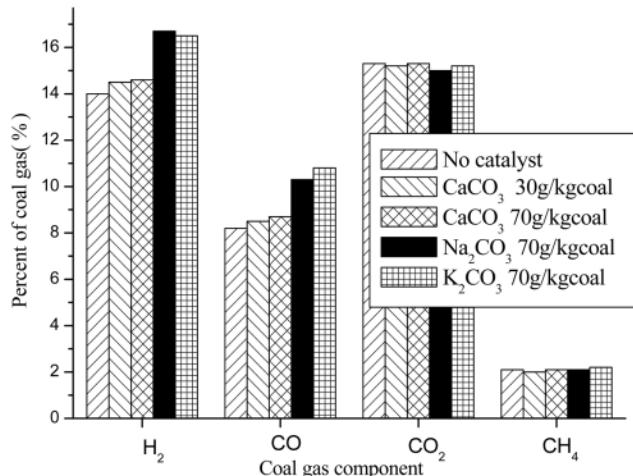
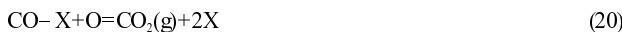


Fig. 12. The relationship between catalyst and syngas components.



When CaCO_3 is added into raw coal, the concentrations of H_2 and CO in syngas will be increased accordingly. The more CaCO_3 in coal, the more concentrations of H_2 and CO (Fig. 12). As the decomposition reaction of limestone produces some CO_2 , the concentration of CO_2 in syngas is little changed. It is known that the compounds of Na and K also exhibit catalytic properties in coal gasification [14]. So when sodium carbonate and potassium carbonate are added to coal, the performance of syngas will be improved obviously. From Fig. 12, it can be seen that the catalytic capability of sodium carbonate or potassium carbonate is much stronger than that of CaCO_3 .

When limestone in raw coal reaches 3%, the concentrations of H_2 and CO are increased 0.5% and 0.3%, respectively. When limestone in raw coal reaches 7%, the concentration of H_2 and CO is only increased 0.1% and 0.2%, respectively. The weakening of CaO catalytic capability shows that the increase of catalyst improves the reaction velocity and the gasification condition. On the other hand, the attachment degree of catalyst on the surface of coke increases. After the catalyst on the surface of coke reaches its maximum quantity of monolayer disperse, the redundant catalyst is devoid of good distribution. Then the redundant catalyst prevents the gasification agent from diffusing to the interior of char and syngas to transfer, and restricts the reaction velocity to increase more to a certain extent. So there is an optimal quantity of the catalyst.

CONCLUSIONS

From the experiment of partial coal gasification, the following conclusions are obtained:

1. With the increase of fluidized air flow rate, the bed layer tem-

perature is increased, the concentrations of H_2 and CO are increased first then decreased, the concentration of CO_2 is decreased first then increased and the concentration of CH_4 is decreased. The heating value of cool syngas reaches max when the fluidized air flow rate is 9 Nm^3/h . If the fluidized air flow rate keeps on increasing, the increasing trend of bed layer temperature becomes smooth. To attain a reasonable gasification effect, the heating value of the syngas and the rate of gas generation must be considered in equilibrium. That is, the ratio of air and coal must be selected suitably.

2. When the feed rate of coal is changed, the gasification temperature and components of syngas are changed. With the increase of coal feed rate, the temperature of the bed layer is decreased gradually. The coal feed rate's influence on the components of syngas is complicated. If the coal feed rate is excessive, the gasification profundity will be not enough and the performance of syngas will be degraded. But if the coal feed rate is too little, the gasification capability will be superfluous and the performance of syngas will be degraded too.

3. With the increase of steam, the temperature of the bed layer is decreased gradually. When the quantity of steam is small, steam is in favor of the increase of H_2 and CO . While the steam keeps on increasing, much more steam reacts with carbon to produce more CO_2 and H_2 . So the concentration of H_2 keeps on increasing, the concentration of CO begins to decrease, and the concentration of CO_2 begins to increase. When the steam becomes superfluous, gasification extent will not become enough and the performance of syngas will be degraded too.

4. The bed layer temperature is one of the main factors affecting the components of syngas. The higher the temperature of bed layer, the more concentrations of H_2 and CO and the less concentration of CO_2 . The decrease of CH_4 in syngas is a result of the methane's second pyrogenation, which is caused by the increase of bed layer temperature. The heating value of syngas is proportional to the bed layer temperature.

5. When the bed layer static altitude is increased, the volatile matter in the broiling layer is cracked much more strongly, and then the concentration of H_2 and CO is increased and the concentration of CH_4 is decreased. The combustible components (H_2 , CO , CH_4) in syngas coming from bitumite coal are more than that from anthracite coal, and high rank bitumite coals are more suitable for gasification than low rank bitumite coals.

6. The alkali/alkaline-earth metal compounds such as Ca, Na and K exhibit catalytic properties in coal gasification. The catalytic capability of sodium carbonate or potassium carbonate is much stronger than that of CaCO_3 . The more CaCO_3 in raw coal, the more concentrations of H_2 and CO in syngas. After the quantity of catalyst reaches a specific value, the catalytic capability becomes saturated.

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