Simulation and analysis of coal gasification combined with CH4 reforming

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Abstract. There is a plentiful supply of coal in China. In order to ensure clean utilization of coal, a simplified technological flowsheet (coal gasification combined with CH4 reforming) is proposed and simulating the process with Aspen Plus in this paper. Through the simulation, the process is analyzed from the thermodynamic point of view. Meanwhile, The sensitivity analysis shows that the optimum temperature of the gasifier is 1400 ° C, the optimum gasification pressure is 3.5 MPa, the optimum oxygen to coal ratio is 0.65, and the optimum water to natural gas ratio is 0.5. Under optimal process conditions, effective gas production increased by 2% and CO2 emissions decreased by 16.9%.

1. Introduction

Coal, oil and natural gas are the most extensive fossil energy sources in the 21st century. They are the main driving force for the development of human civilization and economic prosperity [1]. China is a country rich in coal resources. Thus, coal plays an important role in its energy supply for a long time. In the past 20 years, coal chemical industry has developed rapidly and coal-based gasification technology has received great attention as a clean technology. Syngas which form coal gasification can further synthesize various chemicals, which plays an important role in industrial production. However, the low carbon and high hydrogen content in coal is destined to the current status of high carbon emissions in coal chemical industry, which has caused tremendous pressure on China’s CO2 emission reduction [2-3]. At present, the development and utilization of carbon dioxide capture and storage technology are the main ways to achieve a low-carbon economy. It is a hot spot for researchers in various countries. The study found that emission reduction targets may not be achieved through carbon capture and storage technologies. In response to the above problems, some researchers have provided the idea for the joint supply. It utilizes coal and other hydrocarbon raw materials comprehensively to produce a synthesis gas with a suitable hydrocarbon ratio through the complementation of raw materials and production processes.

Song Xueping [4] et al. proposed a co-gasification process for coal and natural gas in a fixed bed, and carried out thermodynamic simulation analysis and experimental research. The experiment shows that the H2/CO of the obtained syngas can be adjusted between 1.0 and 1.5. Ouyang Chaobin et al [5-6] conducted a thermal simulation experiment on the co-gasification reactor. It is believed that the H2/CO of the syngas produced can be between 1.0 and 2.0, the effective gas content is greater than 90.0% and the residual CH4 is less than 2.0%. The design process includes a CO conversion unit to the hydrogen-carbon ratio of the syngas flexibly. Zhang yuan [7] analyzed the thermodynamics of natural gas and coal co-gasification. The calculation and analysis showed that the addition of methane could adjust the
hydrogen-carbon ratio in the synthesis gas. In addition, Guo Zhancheng [8] also studied the gasification equipment for the synthesis gas of natural gas and coal co-gasification.

Based on the above theory, at present, it is disadvantages for coal gasification to syngas that high energy consumption and high emission. This paper proposes a new process with coal gasification combined with CH$_4$ reforming, namely, coal gasification combined with CH$_4$ reforming process. which is the reason for the high energy consumption and high emission of coal gasification to syngas. The technology utilizes the coal which is multi-carbon and less hydrogen and methane which is multi-hydrogen and less carbon for material integration to achieve elemental complementation of raw materials. Through the integration of methane triple reforming in the coal gasification process, it can be realized that the energy saving and emission reduction of the syngas production process.

2. Technical principle

Coal gasification technology is an important technology to use coal cleanly. So far gasifier can be divided into three categories including fixed bed, fluidized bed, airflow bed [9]. The coal gasification process is an exothermic reaction and its main chemical reaction is shown in formula (1)–(6).

$$\text{C} + \text{O}_2 \rightarrow \text{CO}_2 \quad \Delta H^0 = -393.8 \text{kJ/mol}$$  \hspace{1cm} (1)

$$\text{CO}_2 + \text{C} \rightarrow 2\text{CO} \quad \Delta H^0 = -115.7 \text{kJ/mol}$$  \hspace{1cm} (2)

$$\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2 \quad \Delta H^0 = +131.5 \text{kJ/mol}$$  \hspace{1cm} (3)

$$2\text{H}_2 + \text{C} \rightarrow \text{CH}_4 \quad \Delta H^0 = -74.81 \text{kJ/mol}$$  \hspace{1cm} (4)

$$\text{CO} + 3\text{H}_2 \rightarrow \text{CH}_4 + \text{H}_2\text{O} \quad \Delta H^0 = +35.715 \text{kJ/mol}$$  \hspace{1cm} (5)

$$\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2 \quad \Delta H^0 = -41.5 \text{kJ/mol}$$  \hspace{1cm} (6)

As a kind of clean energy, methane also plays an important role in China's national economy. The process of producing syngas from methane is an endothermic reaction. The main chemical reaction formula is (7)–(10).

$$\text{CH}_4 + \text{H}_2\text{O} = \text{CO} + 3\text{H}_2 \quad \Delta H^0 = +206 \text{kJ/mol}$$  \hspace{1cm} (7)

$$\text{CH}_4 + \text{CO}_2 = 2\text{CO} + 2\text{H}_2 \quad \Delta H^0 = +247 \text{kJ/mol}$$  \hspace{1cm} (8)

$$\text{CH}_4 + \frac{1}{2}\text{O}_2 = \text{CO} + 2\text{H}_2 \quad \Delta H^0 = -36 \text{kJ/mol}$$  \hspace{1cm} (9)

$$\text{CO} + \text{H}_2\text{O} = \text{CO}_2 + \text{H}_2 \quad \Delta H^0 = -41 \text{kJ/mol}$$  \hspace{1cm} (10)

A large amount of heat is released from the gasification process. The hydrogen carbon ratio is relatively low in the synthetic gas. The methane conversion process is an endothermic reaction, which consumes a large amount of heat to maintain the reaction. The synthesis gas produced by the conversion has a relatively high hydrogen to carbon ratio. The heat released by the gasification process is used as the energy required for natural gas conversion to achieve energy complementation, which can greatly reduce fuel consumption. In addition, achieving complementarity between elements not only reduces the investment in subsequent converters but also reduces the amount of carbon dioxide emissions in the converter.

3. Coal gasification combined with CH$_4$ reforming process simulation

In this paper, Aspen Plus is used to simulate the process of coal gasification combined with CH$_4$ reforming and the subsequent system analysis. The process of coal gasification and CH$_4$ reforming to syngas consists of multiple units. This paper mainly analyzes the combined gasification process. The selected model is a SHELL coal gasification process which is widely industrialized in China.

3.1. physical methods

When calculated with Aspen Plus software, it will involve both conventional and unconventional components. The conventional components include CO, H$_2$, CH$_4$ etc. and the unconventional components include pulverized coal, coke, ash, and the like. The applicable system of RK-soave equation is a mixture of nonpolar or weak polar components such as hydrocarbons and light gases like CO$_2$, H$_2$S and H$_2$. Especially suitable for high temperature and high pressure conditions like hydrocarbon processing, supercritical extraction and so on [10-11]. In this paper, the coal gasification
process simulate is carried out under high temperature and high pressure conditions. The components produced by gasification are mostly light gases. Therefore, the physical property method adopts RK-Soave.

3.2. Model establishment

The modeling of SHELL powder coal gasification process uses RStoic, Mixer, Gibbs, CIChng and other reactor. The Gibbs reactor simulates the coal gasification process and the methane reforming process. Since the Gibbs reactor cannot handle unconventional component coal, the coal is pretreated into conventional components and ash by the RStoic reactor firstly. The heat generated is supplied to Gibbs reactor by the decomposition. Its simulation flow chart is shown in Figure 1.

![Figure 1](image)

**Figure1.** coal gasification combined with CH$_4$ reforming simulation flow chart.

3.3. Simulation parameters

According to the above model, the coal gasification combined with CH$_4$ reforming process is simulated. The operating parameters during the simulation are shown in Table 1 below. Elemental analysis and industrial analysis of raw coal are shown in Table 2.

**Table 1.** Gasifier operating parameters [12]

| Pressure | Temperature | O$_2$ (T/℃) | H$_2$O (T/℃) | Coal (T/℃) | CH$_4$ (T/℃) |
|----------|-------------|--------------|--------------|-------------|--------------|
| 3.5Mpa   | 1500℃      | 25           | 25           | 80          | 25           |

**Table 2.** Industrial analysis and elemental analysis of raw coal

| Index | Industrial analysis | elemental analysis |
|-------|---------------------|--------------------|
| Date  | M$_{ad}$ | A$_{ad}$ | V$_{daf}$ | F$_{C+}$ | C | H | O | N | S |
|       | 10.21   | 9.32    | 32.76    | 57.92   | 74.39 | 4.07 | 10.67 | 0.73 | 0.82 |

According to the above simulation conditions, the composition of the gasification furnace after combined gasification is shown in Table 3:

**Table 3.** Gas composition of the gasifier

| Composition | H$_2$ | CO | CO$_2$ | CH$_4$ | H$_2$O | N$_2$ | H$_2$S | NH$_3$ | COS |
|-------------|-------|----|--------|--------|--------|-------|--------|--------|-----|
| Date/mol    | 0.2671| 0.4499 | 0.0638 | 0.1678 | 0.0473 | 0.0020 | 0.0019 | 0.00004 | 0.0001 |
4. Influence and optimization of process variables on co-gasification

In order to obtain a higher content of effective gas (CO+H\textsubscript{2}) and reduce CO\textsubscript{2} emissions, the parameters of the coal gasification and methane reforming process were analyzed by the sensitivity analysis module in Aspen Plus to obtain the optimal process parameters.

4.1. Influence of gasifier temperature on co-gasification process

Given the situations that other conditions remain unchanged, when the gasifier temperature changes from 1000 to 1600 °C, the change trend of the gasification outlet temperature, the mole fraction of effective gas and CO\textsubscript{2} are shown in Figure 2. Figure 2 shows that the outlet temperature, the mole fraction of effective gas and CO\textsubscript{2} from the gasifier do not change significantly with the increase of gasification temperature before 1300°C. With the increase of gasification temperature, the outlet temperature of the gasifier and the mole fraction of the effective gas increased rapidly, and then there is not change significantly. The molar fraction of CO\textsubscript{2} decreased rapidly and then there is not change significantly. Through the above analysis, it was determined that the optimum gasification temperature of the gasifier is 1400 °C.

![Figure 2](image_url)

**Figure 2.** Effect of gasifier temperature on gasification result.

4.2. Influence of gasification pressure on co-gasification process

Given the situations that other conditions remain unchanged, the operating pressure of the gasifier changes from 1 to 7 MPa. Then the gasifier outlet temperature, the mole fraction of effective gas and CO\textsubscript{2} are shown in Figure 3. Figure 3 shows that the outlet temperature of gasifier rises with the increasing pressure, but the amplitude is not large. The mole fraction of syngas decreases slightly with increasing pressure, nevertheless, CO\textsubscript{2} increases slightly. It can be seen that the pressure has little effect on the composition of the syngas. It is deemed that the main reason why the outlet temperature rises of gasifier is that coal gasification is a process of increasing volume. However, from a thermodynamic point of view, pressurization is unfavourable to reaction equilibrium and the ash causes conversion rate of coal is reduced. However, increasing the pressure can increase the concentration of reactants and products, thereby increasing the gasification reaction rate and intensity, resulting in an increase in reaction temperature. Through the above analysis, it was determined that the optimum gasification pressure of the gasifier is 3.5Mpa.
4.3. Effect of oxygen to coal ratio on co-gasification process

Given the situations that other conditions remain unchanged, when the oxygen-coal ratio is increased from 0.55 to 1, the change trend of the gasification outlet temperature, the mole fraction of effective gas and CO$_2$ are shown in Figure 4.

Figure 4 shows that the outlet temperature of gasifier increases along with the change of Oxygen-coal ratio. This just shows the combustion reaction is becoming increasingly intense in the gasifier. The mole fraction of effective gas can be added over oxygen-coal ratio but CO$_2$ decreases accordingly. The principal reason for increasing content of effective gas is that the reactions play a leading role which are semi-coke and O$_2$ generate CO, semi-coke and water vapor generate H$_2$. Therefore, the mole fraction of effective gas increases while the mole fraction of CO$_2$ decreases. Through the above analysis, it was determined that the optimum Oxygen-coal ratio is 0.65.
4.4. Influence of water and natural gas feed ratio on co-gasification process

Given the situations that other conditions remain unchanged, when the water-methane ratio is increased from 0.5 to 1.4, the change trend of the gasification outlet temperature, the mole fraction of effective gas and CO\(_2\) are shown in Figure 5. Figure 4 shows that the outlet temperature of the gasifier and the mole fraction of the effective gas decrease with the increase of water-methane ratio, but the mole fraction of CO\(_2\) increases gradually. The main cause of temperature reduction is that the reactions of methane with water and CO\(_2\) all are endothermic reaction. The reason for the decrease in the effective gas generation rate is that CO in the effective gas reacts with water to form CO\(_2\) and H\(_2\), which is also a cause of an increase in the carbon dioxide content. Through the above analysis, it was determined that the optimum Water-natural gas ratio is 0.5.

![Figure 5. Effect of water and methane ratio on gasification result.](image)

In summary, under the optimal process parameters of the coal gasification combined with CH\(_4\) reforming process, the gas composition of the gasifier outlet gas is shown in Table 4. Compared with the results in Table 3, the effective gas production is increased and the CO\(_2\) emissions are reduced, achieving the purpose of optimization.

| Composition | \(H_2\) | CO  | CO\(_2\) | CH\(_4\) | H\(_2\)O | N\(_2\) | H\(_2\)S | NH\(_3\) | COS |
|-------------|--------|-----|---------|---------|--------|-------|--------|--------|-----|
| Date/mol    | 0.235  | 0.497 | 0.053   | 0.180   | 0.031  | 0.002 | 0.0021 | 0.00004 | 0.00021 |

5. Conclusions

In this paper, the coupling process of coal gasification and methane reforming is simulated by Aspen Plus software. At the same time, the process parameters are analyzed and optimized accordingly. Through analysis, it is found that the increase of gasification pressure has little effect on the export parameters; the increase of oxygen-coal ratio is beneficial to the gasification process; the reduction of water to methane ratio is beneficial to the gasification process. Under the optimal process parameters, the production of effective gas increased by 2% and the emission of CO\(_2\) decreased by 16.9%, which has guiding significance for the actual production process of chemical industry.
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