Pilot-trial and modeling of a new type of pressurized entrained-flow pulverized coal gasification technology

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Abstract

A new type of pressurized entrained-flow pulverized coal gasification technology has been developed by ECUST. It is characterized with four nozzles symmetrically disposed on the upper part of a gasifier. The effects of operation conditions on gasification, N₂ as carrier gas with middle pressure superheated (MP SH) steam, CO₂ as carrier gas with MP SH steam and CO₂ as carrier gas without MP SH steam, respectively, have been tested in the pilot plant. The carbon conversion of all gasification schemes is larger than 99%. For N₂ as carrier gas, the volume fraction (Dry) of CO + H₂ is larger than 90% (v). For CO₂ as carrier gas, the volume fraction (Dry) of CO + H₂ is larger than 92% (v) with MP SH steam and larger than 95% (v) without MP SH steam.

At the same time, based on Gibbs energy minimization principle, the pulverized coal gasification system model was built. The simulation results well matched the pilot-trial data under different operation conditions. The model can be used for the design, assessment, and improvement of the entrained-flow coal gasification system.

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Keywords: Pilot-trial; Entrained-flow; Pulverized coal gasification; Gibbs energy minimization principle; Operation conditions

1. Introduction

Coal gasification is important to a lot of process industries with coal as material, such as chemical production, liquid fuel production, Integrated Gasification Combined Cycle (IGCC) power generation, and poly-generation system. As its future focus, entrained-flow coal gasification technology has been developed around the world [1] and is characterized with large-scale, high efficiency, and cleanliness.

A new type of gasifier fed with coal water slurry or pulverized coal has been developed by Institute of Clean Coal Technology (ICCT), ECUST. As shown schematically in Fig. 1, it is based on multiple opposed-impinging jets [2], with the injection of coal water slurry or pulverized coal being horizontal. Four gasification burners are disposed on the periphery of gasification chamber, each pair of which is symmetrically opposed and meets at 180°. The gasification chamber is covered by refractory brick to protect furnace wall. There are three refractory layers in this pilot gasifier. The first layer (near the gasification zone) is Cr–Al–Zr brick, the second layer is Cr–Al brick, and the third layer (near the metal wall of the gasifier) is Al₂O₃ brick. The inner dimension of the gasification chamber of this pilot gasifier is 900 mm, and the refractory is 450 mm thick.

The impinging jets have been used in the research on and the development of the industrial equipments such as coal gasifier (K-T gasifier [3] and E-GAS gasifier [3]), in which the transformation and/or the reaction processes need to be improved. The K-T gasifier adopts the entrained up-flow and is only allowed to be used under atmospheric pressure. The E-GAS gasifier employs the entrained up-flow and the two-stage gasification technology. However, the new type of gasifier of ours has four nozzles symmetrically disposed on its upper part, and can be operated under high pressure and at high temperature.
The pilot-trials of the pulverized coal gasification were performed using \( \text{N}_2 \) as pulverized coal carrier gas in December 2004 and \( \text{CO}_2 \) as carrier gas in June 2005, respectively. The results suggest that the new gasification technology is promising. The two important gasification indexes, carbon conversion and \( \text{CO} + \text{H}_2 \) volume content, were higher than those of the coal water slurry gasification. In the case of using \( \text{CO}_2 \) as carrier gas, the volume content of \( \text{N}_2 \) (inert gas) in crude syngas could be reduced significantly related to that in the case of using \( \text{N}_2 \) as carrier gas, which helps to improve the yield of some chemical syntheses such as methanol and F-T technologies, and extends the application fields of syngas such as sponge iron [4].

Based on the different carrier gas (\( \text{N}_2 \) or \( \text{CO}_2 \)) and whether the middle pressure superheated (MP SH) steam was input, three gasification schemes were studied.

In order to determine the optimum operation conditions before the trial run of pilot plant, analyze the reliability of the pilot-trial data and design large-scale pulverized coal gasification demonstration plant, it is necessary to build an accurate model of pulverized coal gasification system. The model for the entrained-flow coal gasifier is the core of the system model, which can be divided into equilibrium model and kinetic model. The kinetic model is limited to small numbers of reactions and species with clearly defined mechanisms [5,6] and is likely to exclude kinetic data for pressurized coal gasification, which is usually embedded in CFD simulation [7]. The equilibrium model [8–11] possesses advantages on thermodynamic analysis. Using on Aspen Plus software [12], the system model of the pilot plant was developed, including a Gibbs energy minimization principle model for the pressurized entrained-flow pulverized coal gasifier. The simulation results were consistent with the pilot-trial data.

2. Experiment

2.1. Flow process diagram

In general, the pilot pulverized coal gasification system is composed of over 20 main equipments. Fig. 2 shows the functional schematic flow process diagram of pulverized coal gasification. The system can be divided into four subsystems:

(i) coal milling and drying to prepare qualified pulverized coal and satisfy grain size and humidity content;
(ii) low pressure and high pressure pulverized coal transmitting to transport pulverized coal into the gasifier stably and controllably with high solid-gas mass ratio, especially to transmit pulverized coal to four nozzles just through one pressurized feed hopper;

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(iii) the pulverized coal gasifying to achieve high performance;
(iv) crude syngas scrubbing to get clean syngas.

Subsystems (ii) and (iii) are the key parts for pulverized coal gasification compared with coal water slurry gasification. Pulverized coal with carrier gas was transmitted into nozzle’s center channel through dense pneumatic transmission. O₂ with or without MP SH steam was introduced into nozzle’s outer channel.

2.2. Raw coal and dried pulverized coal analysis

The gasification temperature of the entrained-flow pulverized coal gasifier with refractory brick lining and molten deslagging, is restricted by allowable temperature of refractory brick. The gasification temperature should be less than 1400 °C; otherwise, the refractory brick lining would be ablated rapidly. In order to satisfy the condition of the molten deslagging, it is necessary to require the ash fusion point (flow temperature) of coal below 1350 °C. The properties of raw coal and dried pulverized coal (washed BEI SU bituminous coal, Shandong Province, PR China) used for the pilot-trial are shown in Table 1.

At the same time, the particle size has a significant effect on the reactivity of coal and dense pneumatic pulverized coal transmission. The higher fineness of pulverized coal can increase coal reactivity, but it conflicts with the grinding fineness limitations set by the pneumatic pulverized coal feeding system. With increasing fineness, the fluidization characteristics of the pulverized coal may be affected by growing particle interaction force [13]. So the desired particle size distribution should be a compromise of reactivity and transport. The particle size distribution of dried pulverized coal is analyzed by laser granulometry, as shown in Fig. 3.

2.3. Operation conditions

The dried pulverized coal capacity of coal milling and drying subsystem is 1000–2000 kg/h. The pulverized coal production process can be controlled by adjusting the flow and temperature of drying gas, and the coal addition. In order to avoid the condensation of low pressure steam in mill exit pipes, the temperature of mill exit mixture should be greater than its dew-point, that is, about 25 °C.

The dried pulverized coal was introduced into the pressured feed hopper through a high pressure locker. The pressure of feed hopper is greater than the gasification pressure about 0.5–1.0 MPa. The carrier gas, N₂ or CO₂, was introduced into the feed hopper to maintain the pressure of the feed hopper and fulfill pulverized coal dense transmission. Pulverized coal mass flowmeters were equipped in transmission pipes to measure solid mass flow and solid velocity.

The coal capacity of the pilot plant is 15–45 ton/day (150–470 kg/h per transmission pipe). Gasification temper-
nature is 1250–1350 °C, and gasification pressure is 1.0–3.0 MPa. The purity of oxygen is 99.6%. The pressure of oxygen and MP SH steam is about 4.0 MPa. Based on the different carrier gas (N₂ or CO₂) and whether the MP SH steam was needed, three gasification schemes were studied. Different pulverized coal mass flow, oxygen–coal ratio and steam–coal ratio were tried in the pilot plant. Table 2 shows the operation conditions. For case 1, the oxygen–coal ratio is lower than that expected. For case 2, two nozzles are stopped and high pressure N₂ was entered into it to protect nozzles, which showed great operation flexibility and stability of this type gasifier. For the other cases, the oxygen–coal ratio and the steam–coal ratio are set mainly based on the reasonable ranges of operation conditions obtained from the simulation result.

2.4. Pilot-trial results

Based on material balance, three equations were proposed, as shown below. Eq. (1) is used for calculating the yield of CO + H₂, N m³ (CO + H₂)/kg coal (Dry). Eqs. (2) and (3) are used for calculating the carbon conversion, based on carbon element content in coal and combustible matter in ash, and carbon element content in coal and crude syngas, respectively. In the case of using the nitrogen as carrier gas, both Eqs. (2) and (3) are adopted. For the cases using CO₂ as carrier gas, Eq. (2) is used because the carrier gas flow is very difficult to be measured

\[
\text{Yield of CO} + \text{H}_2 = \frac{V \times (V_{CO} + V_{H_2})}{M_p \times (1 - W_M)} \quad (1)
\]

\[
\phi = \frac{W_C - W_{Ash} - W_{C,Slag}}{W_C} \times 100\% \quad (2)
\]

\[
\phi = \frac{(V_{CO} + V_{CO_2}) \times V / 22.4 \times 12}{M_p \times (1 - W_M) \times W_C} \times 100\% \quad (3)
\]

Table 3 shows the pilot-trial results. For the pilot plant, only the volume content of four main gases (CO, H₂, CO₂, N₂) in syngas was analyzed, while the other trace gases, such as H₂S, CH₄, Ar, and COS, etc., were neglected, as the total volume content of all these trace gases was about 1% and it would not have an obvious effect on the accuracy of Eqs. (1)–(5). For case 1 and case 2, the yield of CO + H₂ and the carbon conversion were very low due to the low oxygen–coal ratio. For case 3 and case 4, the values of carbon conversion obtained using different equations were close, indicating that the measurement instruments and analytic apparatus are reliable. For case 3 to case 7, the yield of CO + H₂ and the carbon conversion remained almost constant, while the CO + H₂ content in crude syngas of CO₂ as carrier gas was greater than that of N₂ as carrier gas. The system simulation results will be given a detailed explanation in terms of the effects of oxygen flow, MP SH steam flow and carrier gas flow on the gasification performance.

3. Simulations

3.1. Simulation diagram

Aspen Plus or other Process System Engineering software has been used in coal combustion and gasification field [14,15]. Fig. 4 shows the simulation diagram of the pilot plant built in Aspen Plus, including subsystems (ii), (iii), and (iv) of Fig. 1, and Table 4 shows the unit models

<table>
<thead>
<tr>
<th>Cases</th>
<th>Pressure MPa(G)</th>
<th>Carrier gas</th>
<th>Pulverized coal flow kg/h</th>
<th>Oxygen flow N m³/h</th>
<th>Oxygen–coal ratio N m³/kg</th>
<th>MP SH steam kg/h</th>
<th>Steam–coal ratio kg/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2</td>
<td>N₂</td>
<td>1503</td>
<td>753</td>
<td>0.501</td>
<td>233</td>
<td>0.16</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>N₂</td>
<td>767</td>
<td>412</td>
<td>0.537</td>
<td>135</td>
<td>0.18</td>
</tr>
<tr>
<td>3</td>
<td>2</td>
<td>N₂</td>
<td>1411</td>
<td>802</td>
<td>0.568</td>
<td>262</td>
<td>0.19</td>
</tr>
<tr>
<td>4</td>
<td>2.5</td>
<td>N₂</td>
<td>1630</td>
<td>932</td>
<td>0.572</td>
<td>285</td>
<td>0.17</td>
</tr>
<tr>
<td>5</td>
<td>2</td>
<td>CO₂</td>
<td>1295</td>
<td>745</td>
<td>0.575</td>
<td>177</td>
<td>0.14</td>
</tr>
<tr>
<td>6</td>
<td>2</td>
<td>CO₂</td>
<td>1105</td>
<td>635</td>
<td>0.575</td>
<td>239</td>
<td>0.22</td>
</tr>
<tr>
<td>7</td>
<td>2.9</td>
<td>CO₂</td>
<td>1654</td>
<td>958</td>
<td>0.579</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
and property methods adopted. All unit models came from Aspen Plus model library while the gasifier model was set specifically.

It is very difficult for the carrier gas flow to be directly measured by the gas flow meter in pressurized feeder hopper’s inlets, because the hopper should proceed to release gas to stabilize the pressure of hopper at all time. Although the solid mass flowmeter can measure the velocity of solid phase, the gas velocity is not equal to the solid velocity due to the phase velocity difference [16]. Therefore, the carrier gas flow is deduced from the element balance, based on data in Tables 1 and 2. For the case of using N₂ as carrier gas, the carrier gas flow is calculated by using Eq. (4). For the case of using CO₂ as carrier gas, the carrier gas flow is calculated by using Eq. (5). The carrier gas flow and carrier gas–solid ratio are shown in Table 3.

\[
\text{Carrier gas flow} = V \times \frac{M_p \times (1 - W_M) \times W_N}{28} \times 22.4
\]

\[
\text{Carrier gas flow} = \frac{V \times (V_{CO} + V_{CO_2}) \times 12/22.4 - \phi \times M_p \times (1 - W_M) \times W_C}{12/22.4}
\]

The function of gasifier model (RGibbs model) is used to calculate the mass and heat balance based on the inlet and outlet streams of gasifier. The model includes 14 components, viz. H₂O, N₂, O₂, S, H₂, C, CO, CO₂, H₂S, COS, CH₄, H₃N, CHN, and Slag. Because dry coal is an unconventional stream, it should be replaced by conventional stream, composing of above 14 components. Eq. (6) describes how the dry coal is replaced by simple components, wherein \(1 - \phi\) C · C: Ash is an inert component as Slag. Parameters \(\alpha, \beta, \gamma, \delta, \iota, \phi\) can be obtained from Table 1 in terms of mass balance. Parameter \(\phi\), the rate of carbon conversion, is a key parameter for gasifier model. The rate of carbon conversion in a real gasifier depends on coal reactivity, gasifier structure parameters, and operation conditions. Ref. [8] gives an expression of carbon conversion.
based on the gasifier temperature. Parameter \( Q \) in Eq. (6), which stands for the decomposition heat of dry coal, should be set as the heat duty of the gasifier unit. Eq. (6) can be derived based on Eqs. (7)–(10). That is to say, \( Q \) can be deduced from Eq. (11).

\[
\begin{align*}
\text{CH}_a\text{N}_b\text{S}_c\text{O}_d(\text{Ash})_e \cdot (\text{H}_2\text{O})_v \\
= \phi C + \frac{x}{2} H_2 + \frac{y}{2} N_2 + \gamma S + \frac{\delta}{2} O_2 + (1 - \phi)C \\
\cdot \varepsilon \text{Ash} + \phi H_2O + Q
\end{align*}
\]

\[
\begin{align*}
\text{CH}_a\text{N}_b\text{S}_c\text{O}_d(\text{Ash})_e \cdot (\text{H}_2\text{O})_v + \left(1 + \frac{x}{2} \frac{1}{\phi} + \gamma - \frac{\delta}{2}\right) O_2 \\
= \text{CO}_2 + \left(\frac{x}{2} + \phi\right) H_2O + \frac{y}{2} N_2 + \gamma \text{SO}_2 + \varepsilon \text{Ash} + \text{HHV}
\end{align*}
\]

\[
\begin{align*}
\text{CO}_2 = C + O_2 - Q_1 \\
\frac{x}{2} H_2O = \frac{x}{2} H_2 + \frac{y}{2} N_2 - \frac{\delta}{2} Q_2 \\
\gamma \text{SO}_2 = \gamma S + \gamma O_2 - \gamma Q_3 \\
Q = \text{HHV} - Q_1 - \frac{x}{2} Q_2 - \gamma Q_3
\end{align*}
\]

The core of the Gibbs energy minimization principle is to solve a nonlinear programming problem. Refs. [17,18] give the solving process.

3.2. Comparison between experimental and simulation results

Table 5 displays the simulation results obtained from the data in Tables 1 and 2, and the carrier gas flow in Table 3. For case 1, \( \phi = 0.848 \); for case 2, \( \phi = 0.912 \); for case 3–7, \( \phi = 0.99 \). The bias of simulation result related to experiment result is little. In general, the simulation results well matched the experiment result Table 3.

3.3. Analyses of operation conditions

3.3.1. Effect of pressure on gasification results

It was found that the pressure (in pilot-trial range) had little effect on the carrier gas–solid ratio from Table 3. The carrier gas–solid ratio was assumed to be constant for the study of the effect of pressure on gasification. Figs. 5 and 6 show the effects of pressure on the gasification results, using the same operation conditions as shown in Table 2. With the increase of gasification pressure, the gasification temperature and the volume content of CO and H\(_2\) varied only to a very limited degree. Therefore, the effect of pressure on the gasification results can be neglected. The water shift reaction (\( \text{CO} + \text{H}_2\text{O} \leftrightarrow \text{CO}_2 + \text{H}_2 \)) was the most important reaction that influenced the equilibrium state of gasification reaction system, but

<table>
<thead>
<tr>
<th>Cases</th>
<th>Gasifier temperature (^\circ\text{C})</th>
<th>Crude syngas flow (\text{Nm}^3/\text{h})</th>
<th>Crude syngas composition, v% (Dry)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1145</td>
<td>2920</td>
<td>57.16</td>
</tr>
<tr>
<td>2</td>
<td>1128</td>
<td>1710</td>
<td>53.38</td>
</tr>
<tr>
<td>3</td>
<td>1321</td>
<td>3116</td>
<td>58.8</td>
</tr>
<tr>
<td>4</td>
<td>1345</td>
<td>3565</td>
<td>59.18</td>
</tr>
<tr>
<td>5</td>
<td>1300</td>
<td>2720</td>
<td>65.4</td>
</tr>
<tr>
<td>6</td>
<td>1277</td>
<td>2370</td>
<td>62.53</td>
</tr>
<tr>
<td>7</td>
<td>1345</td>
<td>3359</td>
<td>71.7</td>
</tr>
</tbody>
</table>
this reaction was characteristics of no volume change before and after reaction. That is to say, the pressure has little effect on the equilibrium state of the gasification system.

3.3.2. Effect of operation conditions on gasification results

There are a lot of factors having effects on the gasification results, including coal reactivity, the structure of gasifier and nozzle, and the operation conditions. Oxygen–coal ratio, steam–coal ratio, carrier gas–solid ratio, and gasification pressure are the four main operation conditions. Gasification pressure has little effect on gasification results, but it determines the gasifier dimension. The gasification pressure is set at 2.0 MPa. With N₂ as carrier gas, carrier gas–solid ratio will affect the N₂ volume content (inert gas) in crude syngas. With carbon dioxide as carrier gas, CO₂ can act as auxiliary gasification agent simultaneity.

Fig. 7 shows the effects of operation conditions on gasification temperature. The carrier gas–coal ratios of Fig. 7a–c are the same as those in cases 4, 5, and 7 in Table 3. In order to meet the requirement that slag with the molten state can be discharged from the gasification chamber, gasification temperature should be greater than coal fusion point. Considering the allowable temperature of refractory brick, the gasification temperature is limited in the range of 1250–1400 °C. The grid area of Fig. 7 satisfies gasification temperature limitation and carbon conversion > 99%, and the outer of the grid area cannot satisfy all these terms simultaneously. Fig. 7 indicates that the oxygen–coal ratio and the steam–coal ratio, or the oxygen–coal ratio and the CO₂–coal ratio should be adjusted simultaneously in order to satisfy the conditions of gasification temperature, carbon conversion and the (CO + H₂) volume content in syngas. The oxygen–coal ratio is sensitive to the gasification temperature. Consequently, it can be used to adjust the gasification temperature. However, if the steam–coal ratio or the CO₂–coal ratio is not adjusted simultaneously with the oxygen–coal ratio, the carbon conversion is not possibly able to reach 99% although the gasification temperature can satisfy operation conditions. This is because the gasification agents in the gasification reaction system are not enough.

Because the operation conditions of case 1 and case 2 in Table 2 locate at the outer of the grid area, both gasification temperature and carbon conversion are very low. For the cases of using N₂ as carrier gas and CO₂ as carrier gas with MP SH steam input, as shown in Fig. 7a and b,

![Fig. 7. The effect of operation conditions on gasification temperature: (a) N₂ as carrier gas with MP SH steam input; (b) CO₂ as carrier gas with MP SH steam input; (c) CO₂ as carrier gas without MP SH steam input.](image-url)
the gasification temperature can be adjusted through controlling oxygen–coal ratio and steam–coal ratio simultaneously. For the case of using CO₂ as carrier gas without MP SH steam input, as shown in Fig. 7c, the gasification temperature can be adjusted through controlling oxygen–coal ratio and CO₂–coal ratio simultaneously. In addition, sufficient CO₂ as the auxiliary gasification agent can be input by decreasing solid–gas ratio of pulverized coal transmission, or injecting some CO₂ into oxygen flow.

Fig. 8 shows the effect of operation conditions (grid area in Fig. 7) on syngas composition. The volume content of CO + H₂ in crude syngas with CO₂ as carrier gas is greater than that with N₂ as carrier gas, because CO₂ is not only a carrier gas but also a gasification agent, which facilitates the gasification reactions, whereas N₂ is an inert gas in gasification system. The volume content of CO and H₂ in crude syngas varies with operation conditions and gasification schemes. When the quantity of steam and/or CO₂ is

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Fig. 8. The effect of operation conditions on syngas composition: (a) N₂ as carrier gas with MP SH steam input; (b) CO₂ as carrier gas with MP SH steam input; (c) CO₂ as carrier gas without MP SH steam input.
excessive in gasification system, both the steam–coal ratio and the CO₂–coal ratio have an influence on the ratio of H₂/CO and the volume content of (CO + H₂) in syngas through a water shift reaction. When CO₂ is carrier gas, CO + H₂ volume content (Dry) is >92% with MP SH steam and is >95% without MP SH steam. CO₂ can act not only as carrier gas but also as an auxiliary gasification agent that functions in a similar way as MP SH steam. This makes the volume content of inert gas (N₂) very low. The crude syngas composition can be adjusted by the quantity of auxiliary gasification agent.

3.3.3. Reasonable ranges of operation conditions

For a specific coal type, such as coal type in Table 1, it is found that the reasonable operation ranges of some operation parameters vary with gasification schemes. The operation conditions should satisfy maximal productivity of CO + H₂ and minimum material consumption. Meanwhile, flow fluctuation and operation flexibility should also be considered. Table 6 shows the reasonable ranges of operation conditions for these three gasification schemes. The MP SH steam consumption and CO₂ consumption should be controlled reasonably.

4. Conclusions

This paper has reported a new type of pressurized entrained-flow pulverized coal gasification technology. The pilot-trial results of three different gasification schemes show very low raw material consumption and very high yield CO + H₂. The carbon conversion of all gasification schemes is >99%. In the case of using N₂ as carrier gas, CO + H₂ content (Dry) is >90%. When CO₂ is carrier gas, CO + H₂ content (Dry) is >92% with MP SH steam and is >95% without MP SH steam. CO₂ can act not only as carrier gas but also as an auxiliary gasification agent that...
functions in the same way as MP SH steam. This makes the volume content of inert gas (N₂) very low. Due to the gasification temperature limitation of refractory brick, it is necessary to replace refractory brick with water cooling wall in order to satisfy wider coal type in the future.

It is difficult to build a detailed gasification model, which includes coal reactivity, the structure of the gasifier and the nozzle, and the operation conditions. The free energy minimization principle model of the gasifier is an optional simulation method. The carbon conversion should be given as a model parameter; otherwise, the simulation results would give the equilibrium carbon conversion. The carbon conversion can be obtained from the pilot-trial data, TGA, etc. The simulation results well matched the pilot-trial data, indicating that the equilibrium model can be used to model the entrained-flow gasifier. Depending on given coal type and gasification scheme, this model can be used to determine reasonable ranges of operation conditions accurately for designing, running, and improving an entrained-flow gasifier.

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