

Thermal analysis of coal gasification in supercritical water for power generation

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Abstract:

The ways of utilizing coal for power generation are directly combusting in air integrating with a steam cycle and gasification with O₂/air integrating with a combined cycle. Special equipment is needed for the removal of NO_x, SO_x, CO₂, PM_{2.5} and the cost is high. The technology of supercritical water gasification (SCWG) can efficiently convert coal to clean gaseous product. A novel method integrating SCWG with combined cycle for power generation is proposed. Two models, which mainly differ in heat recovery unit, are proposed, theoretically analyzed, and compared. The influence of coal-water-slurry concentration (CWSC) on the efficiencies of the models and their difference is investigated. The efficiencies of the models are increased with increasing CWSC. Implementing chemical heat recovery can raise the model efficiency by about 5.1% when the CWSC is 11.3%. The model with chemical heat recovery has an advantage over the model without chemical heat recovery in high CWSC but such an advantage is absent at low CWSC (the critical data is about 3.8%).

Keywords:

Combined cycle, Power generation, Supercritical water gasification, Thermal analysis.

1. Introduction

Coal has been used as fuel for power generation since the late 19th century [1]. Until today, coal is still the single most important fuel for power generation, and accounts for approximately 40% of the electricity generated worldwide [2–3]. The coal utilization pathway involves direct combustion in air to generate the high temperature and pressure steam required to drive the Rankine cycle in most power plants. As in R1, direct combustion of coal cannot eliminate the emission of NO_x, SO_x, CO₂, heavy metal, and PM_{2.5} [4]. Therefore, the pollutant emission reduction system is complicated, and the cost is high. Low efficiency of power generation (commonly 30%–40%) in the direct combustion pathway is another important issue for efficient coal use [5–6].



Integrated gasification combined cycle (IGCC) is promising because coal is gasified to syngas before utilization and the syngas can be used in a combined cycle to generate more electricity [7–8]. IGCC has higher efficiency of around 40%–43% low heating value, and lower emissions (about 1/10) than conventional coal-fired power generation systems [9–10]. Other advantages of IGCC over conventional coal-based power plants include product flexibility, pathway to carbon capture

and storage, higher fuel flexibility and lower water requirement [11–13]. However, critical obstacles for the commercialization of IGCC technology include higher cost (\$1500/kW) and lower reliability [12, 14]. The coal gasifier mostly operates in gas environments. Thus, special equipment is required for the purification of syngas, which often contains CO₂, H₂S, HCN, and COS. When integrated with pre-combustion technology for CO₂ sequestration, the efficiency of IGCC would decrease by 10%–14% [15–16]. Thus, a great demand for a cleaner and more efficient way to use coal is required.

Supercritical water (SCW; $T > 374\text{ °C}$ and $P > 22.1\text{MPa}$) has a single phase that has no surface tension and no liquid/gas phase boundary. SCW has a lower dielectric constant and fewer and weaker hydrogen bonds for obtaining complete miscibility with many organic compounds and gases than ambient liquid water. SCW has sufficient density for appreciable dissolving power, has diffusivity that is higher than that in liquid, and has lower viscosity to enhance mass transport. SCW provides a homogeneous and rapid reaction environment for coal gasification. N, P, S, As, Hg, and other elements in coal are deposited in supercritical water as inorganic salts to avoid the formation of pollutants, such as NO_x, and SO_x. CO₂ can easily be separate from H₂ to obtain the strong function of solubility with pressure and temperature in the critical region. Therefore, CO₂ capture and sequestration will be easy [6, 17–19].

Compared with traditional gasification, supercritical water gasification (SCWG) of coal has advantages, including higher hydrogen yield, N and S deposits as salts, easy CO₂ emission reduction, good coal adaptability, and easy energy recovery [6]. Many experiments have been performed to investigate the influences of concentration of coal slurry, gasification temperature, types of catalysts, residence time, and oxidant equivalent ratio on the product distribution [20–22].

In this article, a novel method integrating SCWG with combined cycle for power generation is proposed. After gasification, the mixture of unreacted SCW and syngas has a large amount of sensible and latent heat. The ways to utilize the heat include the following: producing high temperature and high pressure steam to drive a Rankine cycle (with efficiency about 30%) for power generation; and using heat recovery to preheat the water before being heated to supercritical state. The produced syngas is transferred to a combined cycle for high efficiency power generation (about 60%). Because the gasification is part of the endothermal, a portion of the recovered heat is used to produce syngas for power generation of higher efficiency rather than to produce high temperature and pressure steam for power generation. Thus, implementing chemical heat recovery can improve generation efficiency. Two models based on the utilization ways are proposed, theoretically analyzed, and compared to decide which one to use in real application.

2. Introduction of the two models

2.1 Integration method

After the gasification process, the mixture of unreacted SCW and produced syngas, with a large amount of sensible and latent heat, flows out of the gasifier. The sensible and latent heat account for 30%–60% of the total enthalpy of the mixture, whereas the CWSC is 2%–10%. The ratio is higher in the lower CWSC. In IGCC, the cold gas and hot gas efficiency are 75%–88% and 85%–95%, respectively. Thus, the sensible heat of the syngas may account for 20% of the total enthalpy of coal. The sensible and latent heat of the mixture in SCWG is larger than the sensible heat of the syngas in IGCC even if the same mass of gasified coal is used. Thus, the sensible and latent heat of the mixture should be effectively used as in IGCC.

In IGCC, the produced syngas in a gasifier should be cooled down to suitable temperature for syngas cleaning. In the models proposed in this article, the produced syngas should be cooled down to the appropriate temperature to separate unreacted water and syngas. There are mainly two ways to use the sensible and latent heat, as follows:

One way to use the sensible and latent heat is to generate power. The mixture of unreacted SCW and syngas flows out of the gasifier with high temperature. The sensible heat and latent heat of the mixture can be transferred to the feed water of Rankine cycle to produce high temperature and high pressure steam and to drive a steam turbine for power generation.

The other way to use the sensible and latent heat of the mixture is to preheat the water before being heated to supercritical state. Before the water enters the gasifier, it should be heated to supercritical state. Therefore, the heat may come from coal combustion in a boiler. If the sensible and latent heat of the mixture are used to preheat the water, and the heat from coal combustion is used to further heat the water to supercritical state, the amount of combusted coal could be reduced.

SCWG of coal can be summarized into three reactions as follows:



R2 is the main reaction in the gasification process. Thus, the gasification process is part of the endothermic, and some heat is needed for the process. The ratio of the heat required and LHV of the coal is 30%–40%, whereas the CWSC is 2%–10%. Some physical heat is transferred to the chemical energy stored in the syngas through the chemical reactions. This part of heat can be provided by the recovered sensible and latent heat of the mixture of unreacted SCW and syngas.

2.2 Specific description of the two models

As seen in Figs. 1(a) and (b), the common units of the two models mainly include the following: a boiler to produce SCW; a gasifier for coal gasification under SCW conditions; a combined cycle for power generation using syngas; and a condenser to cool down exhaust water. A heat exchanger exists in each model. When the concentration of the coal slurry and the mass of coal are set, the mass of water entering the gasifier is decided.

In Figs. 1(a) and (b), h_1 is the enthalpy of SCW. h_2 is the enthalpy of the mixture of unreacted SCW and syngas; h_0 is the enthalpy of pressurized feedwater in the Rankine cycle. h_0 is the enthalpy of high temperature and high pressure steam. h_3 and h_3' are the enthalpies of cooled mixture of unreacted SCW and syngas in the models without and with chemical heat recovery, respectively. h_g and h_g' are the enthalpies of syngas in the models without and with chemical heat recovery, respectively. h_4 and h_4' are the enthalpies of exhaust water in the models without and with chemical heat recovery, respectively. h_{01} is the enthalpy of make-up water. h_6 is the enthalpy of preheated SCW. h_7 is the enthalpy of the water mixture.

2.2.1 Model without chemical heat recovery

As seen in Fig 1(a), because part of SCW is consumed in the gasification process, the mass of recycled liquid water is smaller than that of SCW entering the gasifier. Thus, make-up water is needed. After the recycled water is mixed with the make-up water, the water mixture is heated up to supercritical condition (600–700 °C and 23–25MPa) in the boiler. Then, SCW and coal are mixed in

the gasifier, and coal is gasified with a part of the SCW to syngas. The produced syngas and unreacted water flow out of the gasifier to go through a heat exchanger, thereby releasing most of the sensible and latent heat. In the heat exchange process, SCW is condensed to liquid water (about 40 °C). The syngas is separated from the condensed water and is transferred to a combined cycle for power generation, whereas the liquid water is recycled. The sensible and latent heat of the mixture of unreacted SCW and syngas are transferred to the feed water of Rankine cycle in the heat exchanger to produce high temperature and high pressure steam (about 400 °C and 25bar) for power generation.

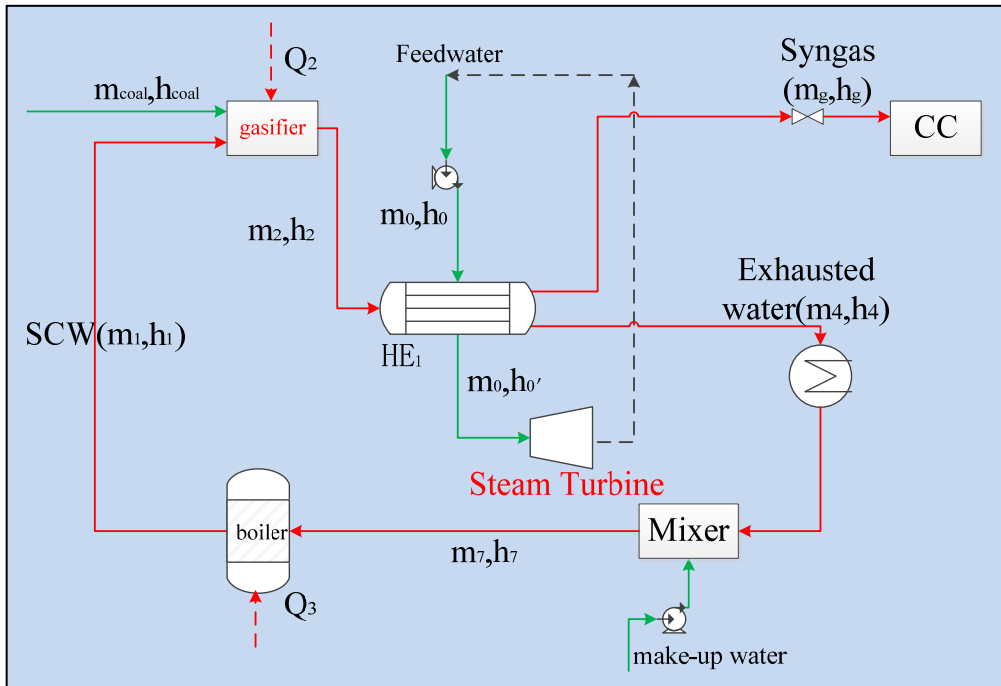


Fig. 1(a). Power generation model integrating supercritical water gasification of coal without chemical heat recovery.

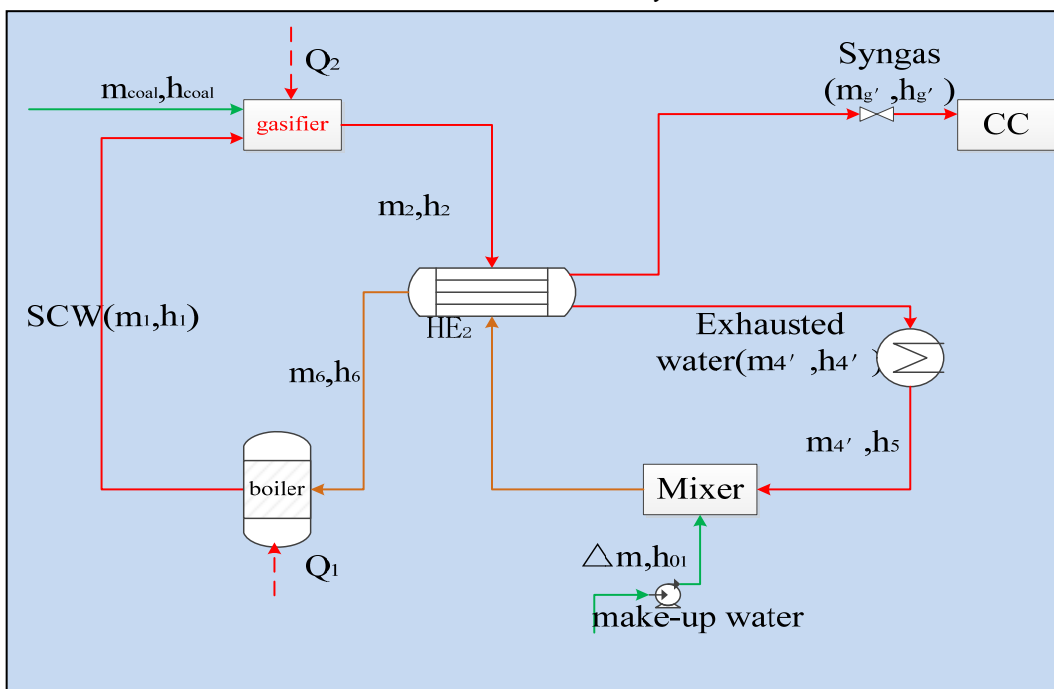


Fig. 1(b). Power generation model integrating supercritical water gasification of coal with chemical heat recovery.

2.2.2 Model with chemical heat recovery

In the model without chemical heat recovery, the water mixture directly enters the boiler. However, in the model with chemical heat recovery [Fig. 1(b)], the water mixture initially flows into the heat exchanger to absorb the highest amounts of sensible and latent heat from the mixture of unreacted SCW and syngas. Subsequently, the water mixture enters the boiler for heating to supercritical state. Then, the SCW and the coal are mixed in the gasifier to react with each other for the production of syngas under the conditions of Q_2 provided by coal combustion. As in the model without chemical heat recovery, the mixture of syngas and unreacted SCW goes through a heat exchanger, and release the highest amounts of sensible and latent heat to the water mixture for cooling to about 200 °C. The water mixture is heated up to about 400 °C. The separated syngas enters a combined cycle for power generation. The exhausted liquid water is recycled. The amount of heat released to the atmosphere when the exhaust water is cooled in the model with chemical heat recovery is larger than that in the model without chemical heat recovery because of the higher temperature of exhaust water in the model with chemical heat recovery.

3. Thermodynamics analysis of the models

To compare the two models, some assumptions are made, as follows:

- 1) The mass of coal, CWSC, the temperature and pressure of the gasifier are the same;
- 2) The temperature and pressure of feed water are 15 °C and 1bar, respectively;
- 3) The loss of heat exchangers, pipe lines, pumps power, and pressure-reducing valves are neglected.

The efficiency of the model without chemical heat recovery can be expressed as follows:

$$\eta_o = \frac{h'_0 \bullet \eta_r + h_g \bullet \eta_{cc}}{h_{coal} + h_1 - h_7 + Q_2} \quad (1)$$

The model with chemical heat recovery can be expressed as follows:

$$\eta_w = \frac{h'_g \bullet \eta_{cc}}{h_{coal} + h_1 - h_6 + Q_2} \quad (2)$$

h_{coal} represents the energy of coal as follows:

$$h_{coal} = LHV_{coal} + i_f \quad (3)$$

LHV_{coal} is the lower heating value of coal, and i_f is the sensible heat of coal.

The energy balance of the gasifier is as follows:

$$h_{coal} + h_1 + Q_2 = h_2 + h_{ur} \quad (4)$$

Where h_{ur} is the enthalpy of unreacted coal in the gasifier.

The energy balance of the heat exchanger in the model without chemical heat recovery is expressed as follows:

$$h_2 + h_0 = h_g + h_4 + h'_0 \quad (5)$$

Inserting (3), (4) and (5) into (1), results in the following expression:

$$\eta_o = \frac{h_0' \bullet \eta_r + h_g \bullet \eta_{cc}}{h_g + h_4 + h_0' - h_0 - h_7 + h_{ur}} \quad (6)$$

The energy balance of the heat exchanger in the model with chemical heat recovery is as follows:

$$h_2 + h_7 = h_g' + h_4' + h_6 \quad (7)$$

Inserting (4), and (7) into (2) results in the following:

$$\eta_w = \frac{h_g' \bullet \eta_{cc}}{h_g' + h_4' - h_7 + h_{ur}} \quad (8)$$

The difference between η_w and η_o is used to compare the models, as follows:

$$\begin{aligned} \Delta\eta = \eta_w - \eta_o &= \frac{h_g' \bullet \eta_{cc}}{h_g' + h_4' - h_7 + h_{ur}} - \frac{h_0' \bullet \eta_r + h_g \bullet \eta_{cc}}{h_g + h_4 + h_0' - h_0 - h_7 + h_{ur}} \\ &= \frac{\eta_{cc} \left(1 - \frac{h_4' - h_4 + h_0}{h_0'}\right) - \eta_r \left(1 + \frac{h_4' - h_7 + h_{ur}}{h_g}\right)}{\left(1 + \frac{h_g}{h_0'} + \frac{h_4 - h_0 - h_7 + h_{ur}}{h_0'}\right) \bullet \left(1 + \frac{h_4' - h_7 + h_{ur}}{h_g}\right)} \\ &= \frac{\eta_{cc}(1 - \lambda) - \eta_r(1 + \beta)}{(1 + \alpha + \gamma)(1 + \beta)} = f(\eta_{cc}, \eta_r, \alpha, \beta, \gamma, \lambda) \end{aligned} \quad (9)$$

We have taken the assumption of $h_g = h_g'$ in (9) because the sensible heat of the syngas is much smaller than the lower heating value of the syngas.

In (9),

$$\alpha = \frac{h_g}{h_0'} \quad (10)$$

$$\beta = \frac{h_4' - h_7 + h_{ur}}{h_g} \quad (11)$$

$$\lambda = \frac{h_4' - h_4 + h_0}{h_0'} \quad (12)$$

$$\text{and } \gamma = \frac{h_4 - h_0 - h_7 + h_{ur}}{h_0'} \quad (13)$$

The α represents the ratio of chemical and physical energies of the mixture consisting of SCW and syngas because h_g is mainly the lower heating value of syngas, and h_0' denotes most of the sensible and latent heat of the mixture. β represents the ratio of energy not efficiently used in the model with chemical heat recovery and chemical energy of the mixture. γ represents the ratio of energy not efficiently used in the model without chemical heat recovery and physical energy of the mixture. The λ represents the ratio of the difference between the heat released to the atmosphere in the model with chemical heat recovery and the heat released to the atmosphere in the model without chemical heat recovery and physical energy of the mixture flowing out of the gasifier. From (9), we can see

that $\Delta\eta$ is a function of η_{cc} , η_r , α , β , γ , and λ . $\Delta\eta$ increases with decreasing α , β , γ , and λ . α , β , γ , and λ are related to gasification temperature, pressure, CWSC, respectively, i.e.,

$$\alpha, \beta, \gamma, \lambda = g(T, P, C) \quad (14)$$

where T, P, and C are gasification temperature, pressure, CWSC, respectively. From (9), we can see

that, if η_{cc} , η_r , β , and λ satisfy $\beta < \frac{\eta_{cc} \cdot (1 - \lambda)}{\eta_r} - 1$, η_o will be smaller than η_w , thereby indicating that

the model with chemical heat recovery has an advantage over the model without chemical heat recovery.

4. Model parameters and calculation conditions

To further compare the models, the influences of gasification temperature, pressure, CWSC, and the efficiencies of the combined cycle and Rankine cycle on the performance of the integrated models should be considered. In this article, only the influence of CWSC on the efficiencies of the models is discussed under condition of certain gasification temperature, pressure, η_{cc} and η_r .

Experiments on the supercritical water gasification of Hongliulin coal in a fluidized bed system are carried out in State Key Laboratory of Multiphase Flow in Power Engineering (SKLMF). The proximate and ultimate analyses of Hongliulin coal are listed in Table 1.

Table 1. Ultimate analysis and proximate analysis of Hongliulin coal.

Ultimate analysis, wt%		Proximate analysis, wt%	
C _{ar}	74.29	M _{ar}	2.79
H _{ar}	4.69	A _{ar}	6.84
O _{ar}	9.26	V _{ar}	33.19
N _{ar}	1	FC _{ar}	57.18
S _{ar}	1.12	LHV, MJ/kg	25.4

The experimental results are taken from [23] and rearranged in Figs. 2(a) and 2(b), whereas the working conditions of the experiments are listed in Table 2. The gasification temperatures and pressures of different working conditions are about 660 °C and 25MPa, respectively. The experimental results can be considered as the function of CWSC.

Table 2. Working conditions of different experiments.

Working condition	A	B	C	D	E
CWSC, wt%	2.0	3.8	6.6	8.4	11.3

From Fig. 2(a), we can see that with increasing CWSC, the gas fraction of H₂ decreases and that of CH₄ increases. Higher CWSC facilitates reaction R4 to produce more methane and to lower the gas fraction of hydrogen. The hydrogen yield and carbon gasification efficiency decrease with increasing CWSC, which can be seen from Fig 2(b). Coal is not fully gasified under high CWSC condition. The enthalpy of the unreacted carbon will be calculated based on the energy balance of the gasifier.

Aspen 11.1 is used for calculation. Peng-Robinson property method is used in this article. Peng-Robinson equation of state is accurate for systems up to 278 bar [24].

5. Results and discussions

The efficiencies of the models with or without chemical heat recovery are illustrated in Fig.3. η_w and η_o increase with increasing CWSC. To explain the results above, the influences of CWSC on α , β , γ ,

λ are discussed first. The results are illustrated in Fig. 4. In this analysis, the mass of coal is set at 1 kg in different CWSCs. α increases with increasing CWSC. The heat released from the mixture flowing out of the gasifier to produce high temperature and high pressure steam sharply decreases with increasing CWSC, whereas the enthalpy of the syngas decreases more gently, i.e., h_g decreases more gently than h_o' . This phenomenon causes the increasing characteristic of α , as shown in (10). β is decreased with increasing CWSC. When T_4 is set, h_4 is in direct proportion to the mass of water flowing out of the gasifier. However, with increasing CWSC, the mass of unreacted water dramatically decreases under conditions involving a low decrease in syngas enthalpy. As shown in (11), β has a decreasing characteristic. γ is small in all CWSCs. γ increases more sharply in high CWSC than in low CWSC mainly because a higher quantity of unreacted coal is present in high CWSC.

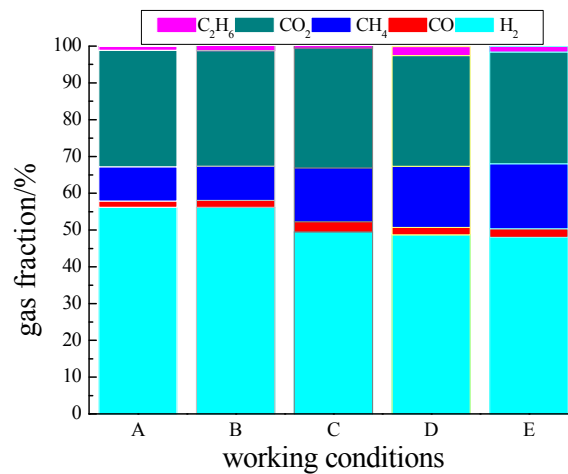


Fig. 2(a). Gas fractions under different working conditions.

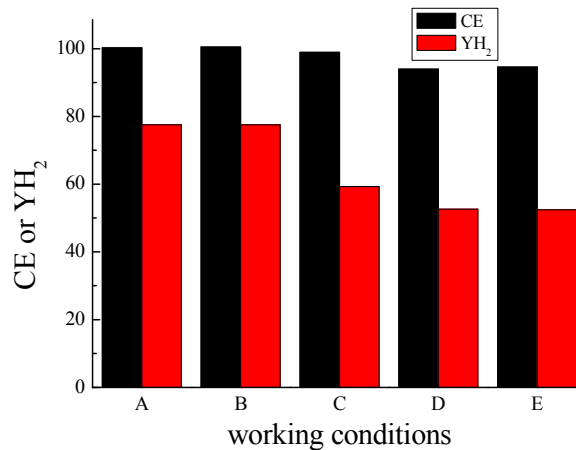


Fig. 2(b). CE or YH₂ under different working conditions.

Note: 1) CE: carbon gasification efficiency, $CE = \frac{\text{the mass of carbon in gaseous products}}{\text{the mass of carbon in feedstocks}}$

2) YH₂: hydrogen yield, $YH_2 = \frac{\text{mole of produced hydrogen}}{\text{the mass of feedstocks}}$

λ , is nearly maintained at a constant level. The heat released from the mixture in the heat exchanger

is mainly from two aspects, as follows: sensible heat of the syngas, and sensible and latent heat of unreacted water. The former is much smaller than the latter. Only sensible and latent heat of unreacted water is considered, and h_0 is neglected, as shown in (12).

$$\lambda' = \frac{h_4' - h_4}{h_{2uw} - h_{4uw}} = \frac{m_{uw} \bullet (\overline{h_4'} - \overline{h_4})}{m_{uw} \bullet (\overline{h_{2uw}} - \overline{h_{4uw}})} = \frac{\overline{h_4'} - \overline{h_4}}{\overline{h_{2uw}} - \overline{h_{4uw}}} \quad (15)$$

m_{uw} is the mass of unreacted water, h_{2uw} is the enthalpy of unreacted water with the temperature of the gasifier, h_{4uw} is the enthalpy of unreacted water with the temperature of T_4 , and enthalpies (represented with the symbol ' $\overline{\quad}$ ') are corresponding specific enthalpies. From (14), we can see that λ' is a constant. Thus, the sensible heat of the syngas and h_0 leads to the small change of λ' .

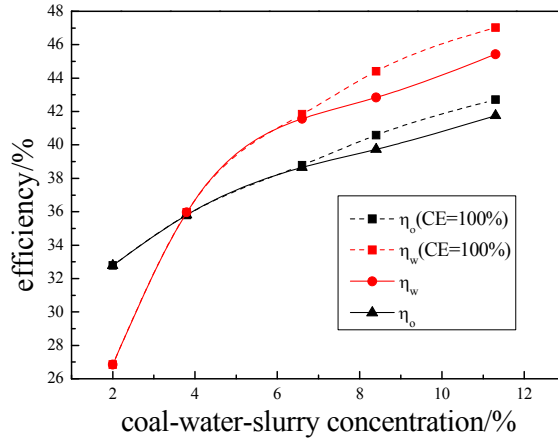


Fig.3. The influences of CWSC on efficiencies of the models.

Therefore, for η_0 :

$$\eta_0 = \frac{h_0' \bullet \eta_r + h_g \bullet \eta_{cc}}{h_g + h_4 + h_0' - h_0 - h_7 + h_{ur}} = \frac{\eta_r + \frac{h_g}{h_0'} \bullet \eta_{cc}}{1 + \frac{h_g}{h_0'} + \frac{h_{ur} + h_4 - h_0 - h_7}{h_0'}} \square \eta_{cc} - \frac{\eta_{cc} - \eta_r}{1 + \alpha} \quad (16)$$

α increases with increasing CWSC and η_0 increases with increasing α . Thus, η_0 increases with increasing CWSC. Because α represents the ratio of chemical energy and physical energy of the mixture, a larger percentage of energy will be transferred in the combined cycle for higher efficiency power generation than in the Rankine cycle to obtain higher model efficiency when α increases. For η_w :

$$\eta_w = \frac{h_g' \bullet \eta_{cc}}{h_g' + h_4' - h_7 + h_{ur}} \square \frac{\eta_{cc}}{1 + \beta} \quad (17)$$

η_w increases with decreasing β and with increasing CWSC. From a previous analysis, we know that β represents the ratio of energy that is not efficiently used in the model with chemical heat recovery and chemical energy of the mixture. A larger percentage of energy is used in combined cycle for power generation than the percentage directly discharged to the atmosphere, thereby leading to higher model efficiency. However, η_w decreases sharply in low CWSC. More heat is released to the atmosphere at low CWSC than at high CWSC, which causes the low efficiency of the model with chemical heat recovery.

In Fig.3, the dashed lines represent the efficiencies of the two models with or without chemical heat

recovery, whereas CEs are assumed to be 100%. When the unreacted coal is considered in the energy balance of the gasifier, the efficiencies of the models decrease. When CWSC is at 11.3%, the efficiency of the model with chemical heat recovery increases by 3.7%, whereas the CE is assumed to be 100%.

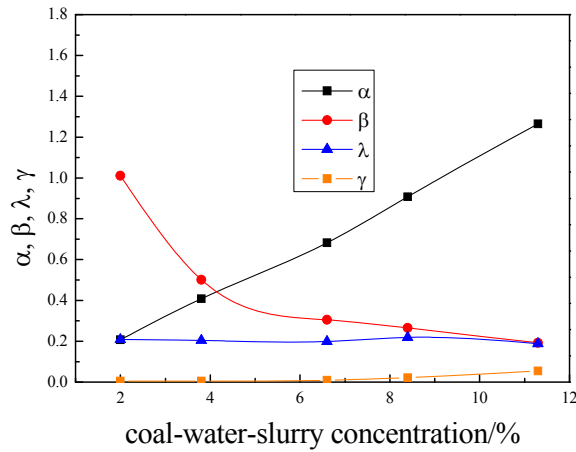


Fig.4. The influences of CWSC on α , β , γ , and λ .

The influence of CWSC on $\Delta\eta$ is illustrated in Fig. 5. In low CWSC, $\Delta\eta < 0$, which mean that the model with chemical heat recovery has no advantage over the model without chemical heat recovery. The sensible heat of exhaust water in the model with chemical heat recovery dramatically increases with decreasing CWSC. As seen in Fig.4, the energy not efficiently used in the model with chemical heat recovery is equal to the enthalpy of the syngas when the CWSC is 2%, thereby leading to negative $\Delta\eta$ from (9). However, when CWSC is larger than 3.8%, then $\Delta\eta > 0$, which mean that the model with chemical heat recovery has an advantage over the model without chemical heat recovery. This phenomenon is due to the much lower energy that is not efficiently used in the model with chemical heat recovery at higher CWSC.

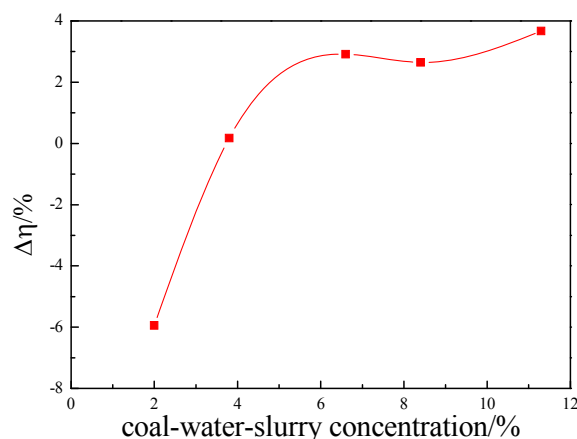


Fig. 5. The influences of CWSC on $\Delta\eta$

The sensible and latent heat of the mixture of unreacted SCW and produced syngas account for 30%–60% of the total enthalpy of the mixture while the CWSC is 2%–10%, and the ratio is higher at lower CWSC. If the heat is used to produce high temperature and high pressure steam, and the

syngas is transferred to a combined cycle for power generation, the generating efficiency of the model without chemical heat recovery will depend on the ratio of enthalpy of the syngas and the sensible heat and latent heat of the mixture, i.e., α . When α increases, more energy is transferred to a combined cycle for higher efficiency (about 60%) power generation than to Rankine cycle for lower efficiency (about 30%) power generation, thereby leading to higher model efficiency. This can be seen in (16) from the perspective of mathematics.

For the model with chemical heat recovery, if a larger percentage of energy is transferred to a combined cycle for power generation rather than released to the atmosphere, i.e., β , the efficiency of the model will be larger, as seen in (17) from the perspective of mathematics.

If a large amount of energy is released to the atmosphere, the performance of the model with chemical heat recovery will worsen. From Fig.4, α increases, and β decreases with increasing CWSC. Meanwhile, λ is nearly kept constant, and γ is always small. Thus, the efficiencies of the two models increase with increasing CWSC. The model with chemical heat recovery has advantage over the model without chemical heat recovery at high CWSC, but such advantage is absent at low CWSC (the critical data is about 3.8%), as seen from Fig. 5.

6. Conclusions

SCWG can cleanly and efficiently realize the utilization coal. A novel concept integrating SCWG of coal with a combined cycle for power generation is proposed in this article. Two models, which mainly differ in heat recovery unit, are proposed, theoretically analyzed, and compared. The influence of CWSC on the efficiencies of the models and their difference is studied. The efficiencies of the models are increased with increasing CWSC. Implementing chemical heat recovery can raise the model efficiency by about 5.1% when the CWSC is 11.3%. The model with chemical heat recovery has an advantage over the model without chemical heat recovery at high CWSC but such an advantage is absent at low CWSC (the critical data is about 3.8%).

Acknowledgments

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