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ABSTRACT

Underground coal gasification is a complex physico-chemical process. UCG thermodynamic equilibrium model was established and validated, and the effects of the steam-to-oxygen ratio ($R_{H2O/O2}$) and the equivalence ratio (ER) on the gasification temperature and the gaseous products were investigated. At low ER (0.08–0.14), coal is incompletely gasificated when the gasification temperature is less than the corresponding peak temperature. At high ER (0.16–0.30), coal is completely gasificated. With the increase of $R_{H2O/O2}$, the gasification temperature decreases monotonically and finally tends to approach 850°C at $R_{H2O/O2} = 4.5$. H_2 reaches high concentration at $ER = 0.18$–0.24 and $R_{H2O/O2} = 1$–3, but the change of CH_4 concentration is just the opposite. CO concentration is high at $R_{H2O/O2} < 1.5$. In order to reach high the utilization effectiveness of oxygen, $R_{H2O/O2}$ has the corresponding optimum value where the utilization effectiveness of oxygen reaches peak value at $ER = 0.08$–0.2.

Introduction

Underground coal gasification (UCG) is a process of converting coal to combustible gas at the site of the underground coal seam. Since UCG reduces the environmental pollution during coal mining and utilization (Perkins 2018; Zou et al. 2012), it is considered as a disruptive technology for carbon neutrality goals (Zou et al. 2012). However, the production process of deep underground coal gasification is invisible, and the measurement and control is difficult. The operating parameters of underground coal gasification directly affect the production safety and efficiency.

UCG is the thermochemical conversion of coal into combustible gaseous product using a gasification agent (H_2O/O_2) at high temperatures (Greg and Vairakannu 2017; Hu et al. 2021; Klebingat et al. 2016, 2018; Liu et al. 2011; Yang, Zhang, and Liu 2009), which experienced a complex physico-chemical process. The main influencing factors include as follows: temperature, coal reactivity, gasifying agents, pressure, heat loss, velocity of combustion and gas diffusion (Vivek, Mahajani, and Samdani 2019). The gasification reactions

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are driven by the high temperature formed by the coal oxidization reactions, the gasification agent (H_2O/O_2) is only injected for the gasification process, thus the injected amount of H_2O and O_2 directly influences on the gasification reaction temperature and gas product compositions. Increasing gasification temperature of UCG increases the concentrations of H_2 and CO as well as higher calorific value in the product gas (Perkins and Love 2010).

For a certain amount of coal, the gasification equivalence ratio (ER) and H_2O-to-O_2 ratio ($R_{H2O/O2}$) determine the amount of water and oxygen injected, which finally determine the gasification temperature. The gasification performance is predominantly dependent on the ER (Biswas et al. 2021; Vivek, Mahajani, and Samdani 2019). Several studies investigated the effect of ER on gasification performance. Most of those experiments were held in conventional gasifiers operating on surface (Biswas et al. 2021; Jangsawang, Laohalidanond, and Kerdsuwan 2015; Liu et al. 2013). Although many studies have investigated the effect of steam/oxygen ratio ($R_{H2O/O2}$) on UCG (Daggupati et al. 2010; Hill and Thorsness 1982; Khadse et al. 2006; Liu et al. 2012), the coupling effect of $R_{H2O/O2}$ and ER on the gas product compositions is rarely studied.

Since UCG involves several strongly coupled phenomena such as multiphase flows, reaction kinetics, heat and mass transfer, and coal spalling (Vivek, Mahajani, and Samdani 2019), modeling the UCG operation is, therefore, a complex task which requires the integration of a number of diverse processes (Andrianopoulos, Korre, and Durucan 2015; Daggupati et al. 2011; Perkins and Sahajwalla 2005). It is rather difficult to develop a single comprehensive computational model to simulate UCG. Considering the complexities involved in UCG, thermodynamic models based on the reaction equilibrium calculations are easily to estimate the product compositions in the complex UCG system, which are very important for the theoretical evaluation of limiting performance characteristics during the different phases. In the present study, a thermochemical equilibrium model is used to simulation gasification products of UCG, and the goals of the study are to analysis the effects of $R_{H2O/O2}$ and ER on the gasification temperature and the product compositions.

Model development

UCG thermodynamic equilibrium model

The detailed consideration of the UCG reaction characteristics is crucial for a reliable process model used to simulate the relevant chemical processes. Figure 1 presents the gasification processes of UCG, where the reaction mechanisms of coal gasification processes mainly consist of pyrolysis, oxidation, and reduction (Tata Sutardi 2019). Based on the reaction mechanisms of UCG shown in Figure 1, in the study of thermodynamic equilibrium model, the reaction zone is divided into four reaction zones including the drying, pyrolysis, gasification and combustion zones, as shown in Figure 2. In the process of underground coal gasification, wet coal first goes through the drying process to become dry coal, and the dry coal continues to be pyrolyzed. The reaction heat in the pyrolysis process is calculated by the combustion heat of reactants and products before and after coal pyrolysis.

Generally speaking, UCG has the characteristics of large reactant particle size, long gasification time and high gasification pressure. Unlike conventional on surface coal gasification (entrained flow coal gasification, bubbling fluidized bed, etc.), the reactants involved in UCG are large coal particles (20–30 mm) rather than pulverized coal (about 75
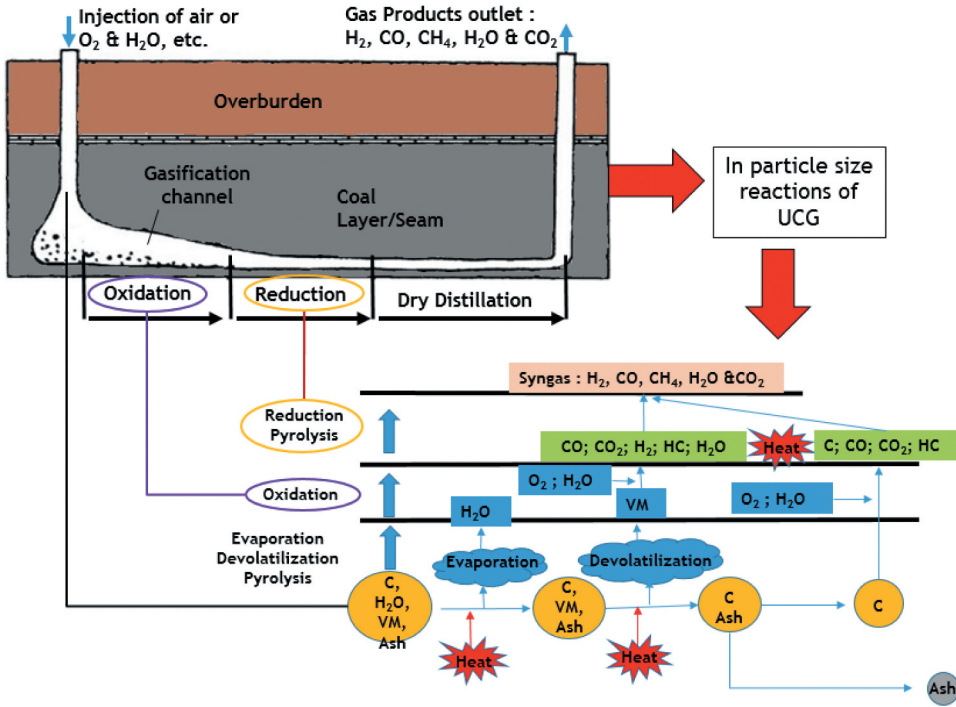
Figure 1. Process illustration of coal particle gasification model and UCG (Tata Sutardi 2019).
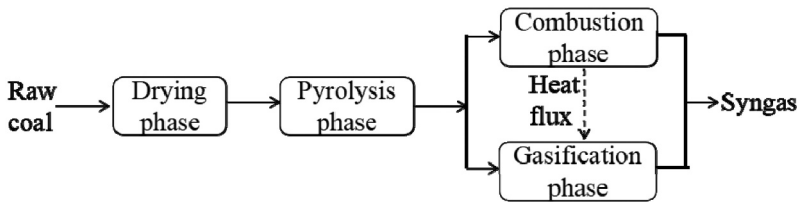

Figure 2. The process flow diagram of coal gasification in UCG.

μm) (Xi et al. 2016). Especially, the large-scale gasification chamber of UCG makes the residence time of coal gasification is long. It is generally believed that the long reaction time and high pressure can make UCG process reach chemical reaction equilibrium (AlNouss et al. 2020; Andrianopoulos, Korre, and Durucan 2015; Gregg and Edgar 1978). Thus, the thermodynamic equilibrium model is selected to simulate the effects of the $R_{H2O/O2}$ and *ER* on underground coal gasification.

Gibbs free energy minimization method is a widely used method for thermodynamic analysis of reaction systems. In the study, based on the theory of Gibbs free energy minimization, the composition of syngas at the reaction equilibrium is calculated, and the detailed calculation formula were provided in Refs (Hu et al. 2021; Jangsawang, Laohalidanond, and Kerdsuwan 2015; Otto and Kempka 2020). It is well-known that HSC Chemistry software is a powerful software that is accessible to analyze the equilibrium product composition on basis of the Gibbs free energy minimization algorithm (Hu et al. 2021; Kumabe et al. 2007;

Rani et al. 2017), which has been designed for the calculation of various kinds of chemical reactions and equilibrium parameters. In this paper, HSC Chemistry® software is employed in order to facilitate the simulation of the chemical processes. When we enter the data of the raw materials, amounts of compounds, and reaction conditions of almost any chemical process, the program provides the resulting amounts of the products.

Since the overall UCG process is strongly exothermic, but the heat loss to the surrounding strata in UCG is difficult to estimate and depends on the properties of the dry rock above the coal seam (Kasani and Chalaturnyk 2017). In the study, the drying and pyrolysis process are instantaneous and no heat loss. The drying stage and pyrolysis stage of underground coal gasification can be completely carried out at high temperature. Figure 3 shows the method of thermodynamic equilibrium calculation by HSC Chemistry®. The input data if gasifing agent is provided in the Table A1 of Appendix A.

The gasification performance is predominantly dependent on their effective proportion called equivalence ratio (ER) (Biswas et al. 2021). Equivalence ratio is commonly used to indicate quantitatively whether a fuel oxidizer mixture is rich, lean or stoichiometric. The equivalence ratio is defined as (Biswas et al. 2021):

$$\text{Equivalence Ratio}(ER) = \frac{(\text{Oxygen}/\text{Fuel})_{\text{Actual}}}{(\text{Oxygen}/\text{Fuel})_{\text{Stoichiometric}}} \tag{1}$$

In the study, UCG of Santanghu coal seam is simulated, and the proximate and ultimate analysis of the coal is presented in Table 1. Oxygen and water are used as the gasification agent in the feed stream. The operating gasification pressure is 8 MPa, which is the hydrostatic pressure for an 800 m deep coal layer. All results are presented as a function of the injected H_2O/O_2 mass ratio (0.6–6) and gasification equivalence ratio ($ER = 0.08$–0.3).
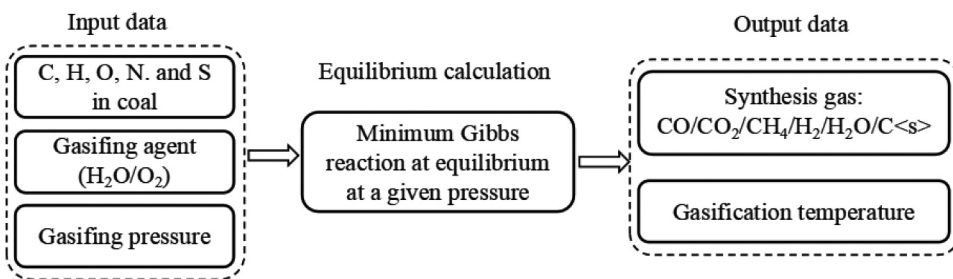

Figure 3. Schematic of the thermodynamic equilibrium calculation for UCG.

Table 1. Characteristics of santanghu coal.

Proximate analysis/%, air dry basis				Ultimate analysis/%, air dry basis				
M	A	V	FC	C	H	O	N	S
2.56	6.30	29.46	61.68	70.82	3.95	15.34	0.26	0.77

Model validation of UCG thermodynamic equilibrium model

In order to validate the model prediction performance of the final product gas composition in UCG, the model simulations were performed to validate the model performance using Alberta field trials data of UCG in a deep coal seam (depth of 1400 m). The operating gasification pressure was 11.5MPa, the gasification agent flow rate was 245 t/h (O_2 45t/d, H_2O 200 t/d), coal consumption was 118 t/d, and the proximate and ultimate analysis of coal seam coal core was presented in Ref (Kasani and Chalaturnyk 2017; Swan 2012).

Figure 4 shows the product gas composition results of simulation using UCG thermodynamic equilibrium model comparison with the field trials data. The high correlation of the field trials and modeling results indicates the reliability of the chemical process models developed. Specifically, the dry product concentrations of CH_4 and CO simulation using the models developed (36.4% and 4.2%) are comparable to the field trials results (37.0% and 5.0%) (Kasani and Chalaturnyk 2017).

Simulation results and discussion

The effects of H_2O-to-O_2 ratio and gasification equivalence ratio on reaction temperature

The injected amount of H_2O and O_2 directly influences on the gasification temperature and product compositions. For a given amount of coal, *ER* and $R_{H2O/O2}$ determine the amount of water and oxygen in the gasification agent, and finally determine the gasification temperature.

Figure 5 shows the influence $R_{H2O/O2}$ and *ER* on gasification temperature. At a given *ER*, when *ER* is in the range of 0.08–0.14, the temperature first increases and then decreases with the increase of $R_{H2O/O2}$, and there is a peak value of temperature (see Figure 5a). With the increase of *ER*, the temperature peak values increase, but the corresponding $R_{H2O/O2}$ decreases from 1.5 to 1.0. When *ER* is in the range of 0.16–0.3, the gasification temperature decreases monotonically (see Figure 5b). Many studies considered gasification temperature as an independent factor to study UCG (Andrianopoulos, Korre, and Durucan 2015; Hu
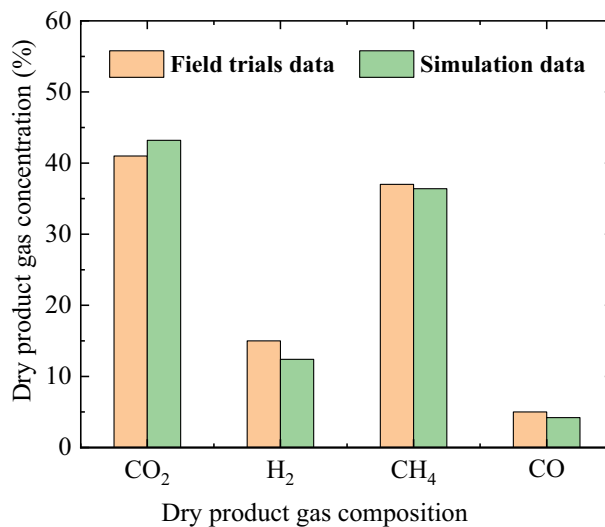

Figure 4. The comparison of product gas simulation results and the field trials data.

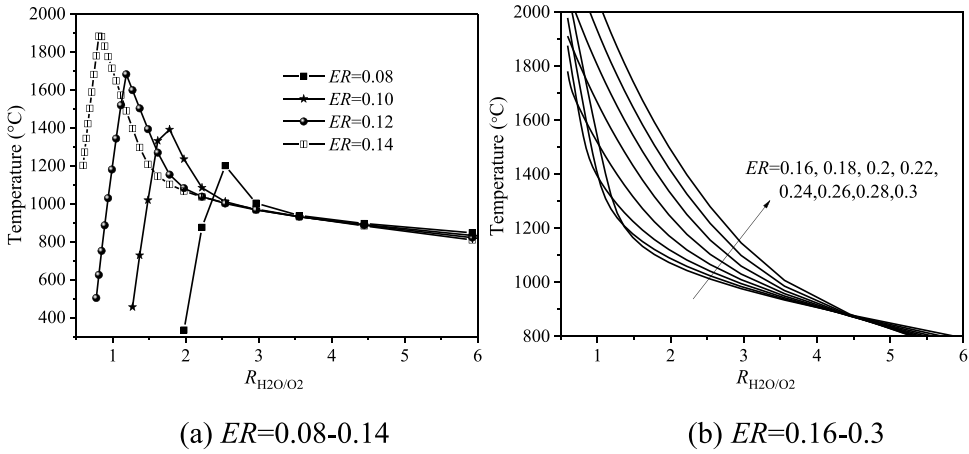(a) *ER*=0.08-0.14   (b) *ER*=0.16-0.3

Figure 5. Effect of steam-to-oxygen ratio and equivalence ratio on gasification temperature.

et al. 2021; Klebingat et al. 2018), but above results indicate that ER and $R_{H2O/O2}$ have significant influences on gasification temperature, and the gasification temperature cannot be used as an independent factor to influence UCG.

Since the gasification temperature is determined by UCG reactions, in order to analyze the complex influences of ER and $R_{H2O/O2}$ on gasification temperature at $ER = 0.08–0.14$, the gasification products are provided in Figure 6. As known from Figures 5a and 6, with the increase of $R_{H2O/O2}$, before reaching the temperature peak (see Figure 5a), the concentrations of CO and CH_4 increase, but the residual carbon (C<s>) decreases (see Figure 6). At $ER = 0.08–0.14$, due to the relatively low amount of oxygen in gasification agent, the heat released by oxidation reaction of coal is little, the gasification temperature is low, and char is hard to completely gasified, which results in the presence of residual carbon (C<s>) in the gasification
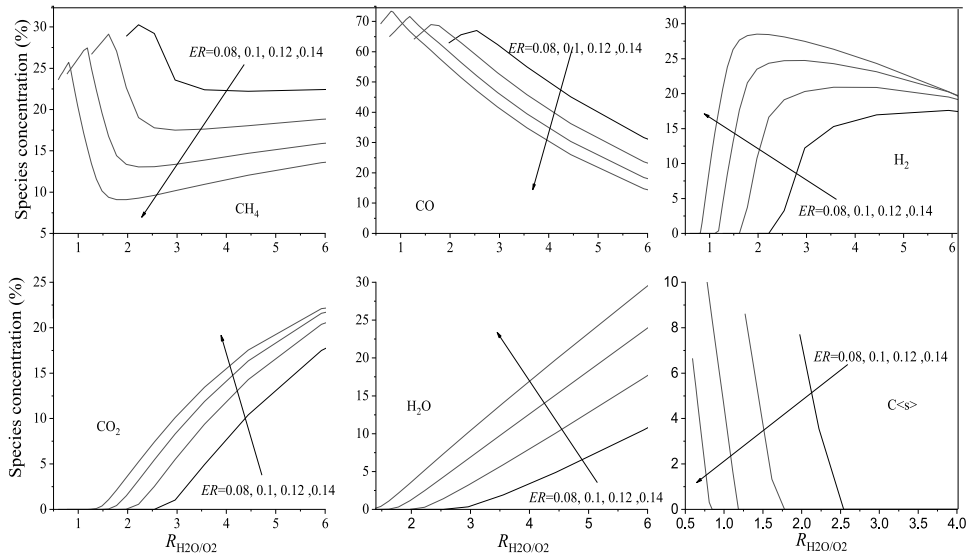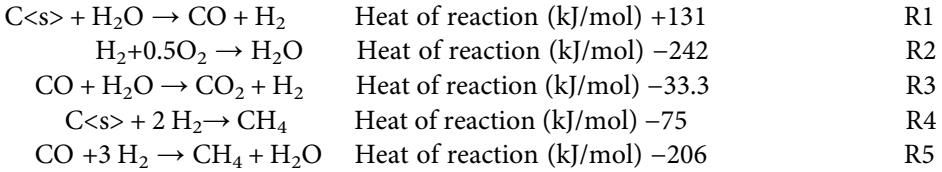

Figure 6. Effect of $R_{H2O/O2}$ and ER on gasification equilibrium products at ER=0.08–0.14.

products (see Figure 6). In this condition, increasing $R_{H2O/O2}$ is in favor of the following reactions:

$$C<s> + H_2O \rightarrow CO + H_2 \qquad \text{Heat of reaction (kJ/mol) } +131 \qquad \text{R1}$$
$$H_2 + 0.5O_2 \rightarrow H_2O \qquad \text{Heat of reaction (kJ/mol) } -242 \qquad \text{R2}$$
$$CO + H_2O \rightarrow CO_2 + H_2 \qquad \text{Heat of reaction (kJ/mol) } -33.3 \qquad \text{R3}$$
$$C<s> + 2\,H_2 \rightarrow CH_4 \qquad \text{Heat of reaction (kJ/mol) } -75 \qquad \text{R4}$$
$$CO + 3\,H_2 \rightarrow CH_4 + H_2O \qquad \text{Heat of reaction (kJ/mol) } -206 \qquad \text{R5}$$

Above reactions can promote char gasification and release heat, and the temperature increases and reaches a peak value (see Figure 5a). Therefore, before reaching the temperature peak (see Figure 5a), increasing $R_{H2O/O2}$ promote C<s> reaction, which make the concentrations of CO and CH_4 in the gasification products increase. With the increase of ER, the temperature peak value increases and the corresponding ER decreases (see Figure 5a).

For $ER = 0.08–0.14$, after reaching the temperature peak (see Figure 5a), the corresponding residual carbon is zero in the gasification products (C<s>, see Figure 6), which means coal is completely gasificated. When $R_{H2O/O2}$ further increases, the excess water steam acts as a heat sink and only reduces the gasification temperature (see Figure 5a), thus the temperature decreases and finally tends to approach (850°C) at $R_{H2O/O2} = 4.5$ (see Figure 5a).

For $ER = 0.16–0.3$, the amount of oxygen supplied is relative high in the gasification agent, more heat is released by the oxidation reactions, thus coal gasification temperature is high enough to completely gasify coal. In the condition, increasing $R_{H2O/O2}$ means that a higher water content of injected fluid leads to a lower gasification temperature (see Figure 5b). This can be explained by the heat absorption of the steam gasification reaction and the energy required for evaporation (Nourozieh et al. 2010)

The effects of H₂O-to-O₂ ratio and gasification equivalence ratio on gasification products

As mentioned above, coal can be completely gasificated at $ER = 0.16–0.3$, thus the analysis of gasification product gases is very meaningful for practical operation of UCG under the condition of the high gasification equivalence ratio. Figure 7 shows the effects of ER and $R_{H2O/O2}$ on the concentrations of gasification product gas species (CO, H_2, CH_4, and CO_2) at $ER = 0.16–0.3$ and $R_{H2O/O2} = 0.3–6$, where coal is completely gasificated and there is not residual carbon (C<s>). As shown in Figure 7, H_2 reaches high concentration at $ER = 0.18–0.24$ and $R_{H2O/O2} = 1–3$, but the change of CH_4 concentration is just the opposite. CO concentration is high at $R_{H2O/O2}<1.5$, the concentration decreases with the increase of ER and $R_{H2O/O2}$, but the change of CO_2 concentration is just the opposite. $R_{H2O/O2}$ has significant impact on the production of CO_2, and CO_2 reaches high concentration at $R_{H2O/O2}>4$.

As shown in Figure 4b, at $R_{H2O/O2}<1.5$, the gasification temperature is high (above 1200°C), which shifts the reverse water-gas shift reaction R6 to left the right side and the Boudouard reaction R7 to the right side, results in a higher concentration of CO (see Figure 7c):
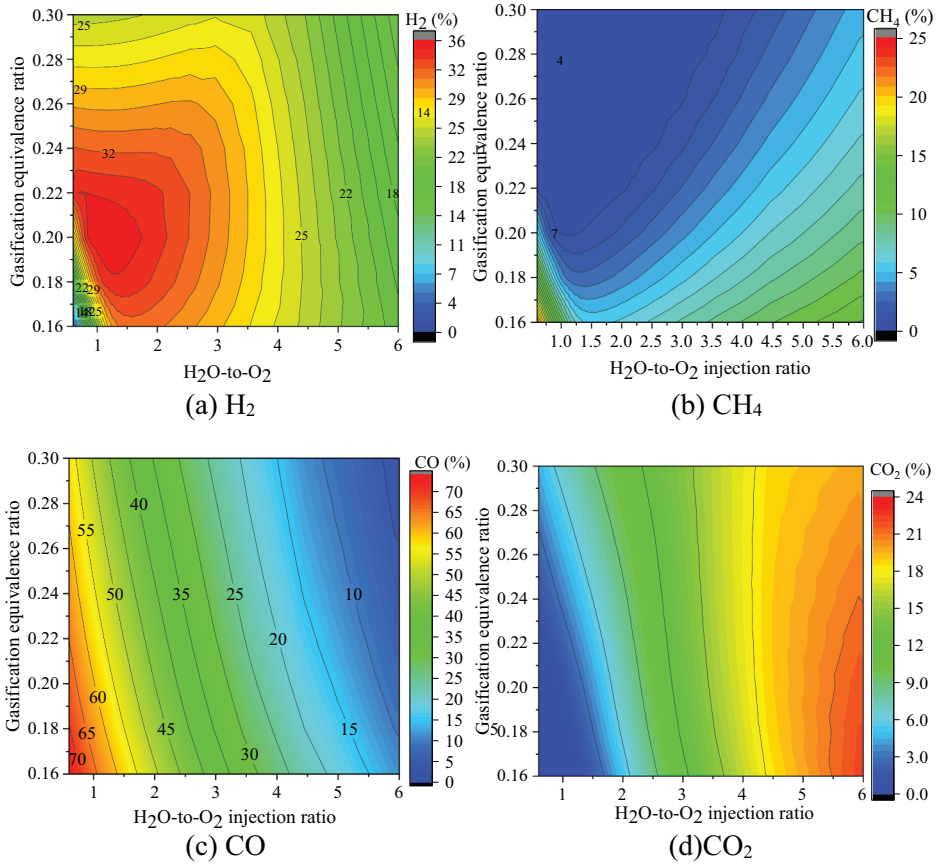
$$CO + H_2O \leftarrow CO_2 + H_2 \qquad \text{R6}$$
$$C<s> + CO_2 \rightarrow 2CO \qquad \text{R7}$$

Figure 7. Effects of $R_{H2O/O2}$ and *ER* on product gas concentrations at *ER*=0.16–0.3 and $R_{H2O/O2}$ =0.3–6.

By increasing $R_{H2O/O2}$, more H_2 is produced by R1 and finally reaches a maximum. At $R_{H2O/O2}$ >3.5, the gasification temperature drops below 1000°C, decreasing reaction temperature is conducive to the production of CH_4 according to the chemical equilibrium model, which makes the reaction of R3 shifts to the right side and results in a high composition of CH_4 (see Figure 7d). At $R_{H2O/O2}$>4.5, the gasification drops below 850°C (see Figure 5), the reaction R5 is hard to proceeds to the right side and results in a low CO concentration and high CO_2 concentration (see Figures 7a & 7d).

At a given $R_{H2O/O2}$, the concentrations of CO and CH_4 decrease with increasing *ER*. Increasing *ER* is conducive to CO and CH_4 oxidation, and the gasification temperature increases (see Figure 5). For H_2 and CO, at $R_{H2O/O2}$>4.0, *ER* has insignificant impact on the concentrations of H_2 and CO, the concentrations are mainly dependent on $R_{H2O/O2}$, and the concentrations decrease with increasing $R_{H2O/O2}$.

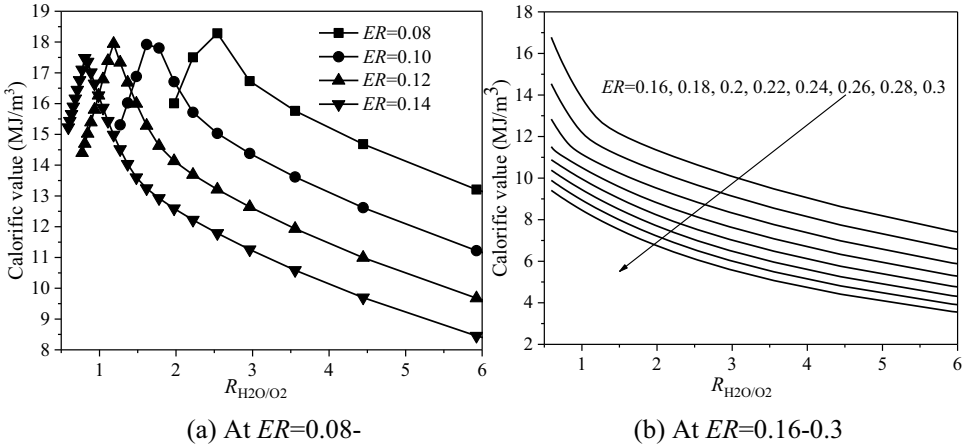(a) At *ER*=0.08-   (b) At *ER*=0.16-0.3

Figure 8. Effects of *ER* and $R_{H2O/O2}$ on calorific value of gasification gaseous products.

The effects of H$_2$O-to-O$_2$ ratio and gasification equivalence ratio on the calorific value of gaseous products

Figure 8 shows the effect of $R_{H2O/O2}$ and *ER* on the calorific value of gasification gaseous products, the variation trend of the calorific value with $R_{H2O/O2}$ is similar to that of temperature (see Figure 5).

At low gasification equivalence ratio (*ER* = 0.08–0.14), the calorific value of the gasification gaseous products has a peak value at a given *ER*. Due to the low *ER*, the gasification temperature is too low to completely gasificate coal (see Figure 6), increasing $R_{H2O/O2}$ can promote C<s> gasification by the reactions of R1-R5, and then the calorific value increases with the increase of $R_{H2O/O2}$. However, when $R_{H2O/O2}$ further increases, the water content of gasification agent is too high, the excess water acts as a heat sink and only reduces the temperature (see Figure 5). Therefore, at low *ER* (0.08–0.14), in order to obtain gaseous products with high calorific value, there exists an optimum $R_{H2O/O2}$ (see Figure 8a).

At high gasification equivalence ratio (*ER* = 0.16–0.3), oxygen content and gasification temperature are enough high to completely gasificate coal. Increasing $R_{H2O/O2}$ only results in the excess water acting as a heat sink, which makes the gasification temperature decrease. With the increase of $R_{H2O/O2}$ and *ER*, the concentrations of CO_2 and H_2O in gaseous products increase, but the concentrations of CO and H_2 decrease (see Figure 4), which results in the decrease of the gaseous product calorific value (see Figure 8b).

The effect of H$_2$O-to-O$_2$ ratio and gasification equivalence ratio on the oxygen utilization effectiveness

For UCG by oxygen and water used as gasification agent, it is hoped that the minimum amount of oxygen is consumed to produce the most useful syngas, this not only requires high calorific value of syngas, but also produces more amount of syngas, thus the utilization effectiveness of oxygen is an important indicator:

$$X_{O2} = \left(Q_g \times F_g \right) / F_{O2} \tag{2}$$

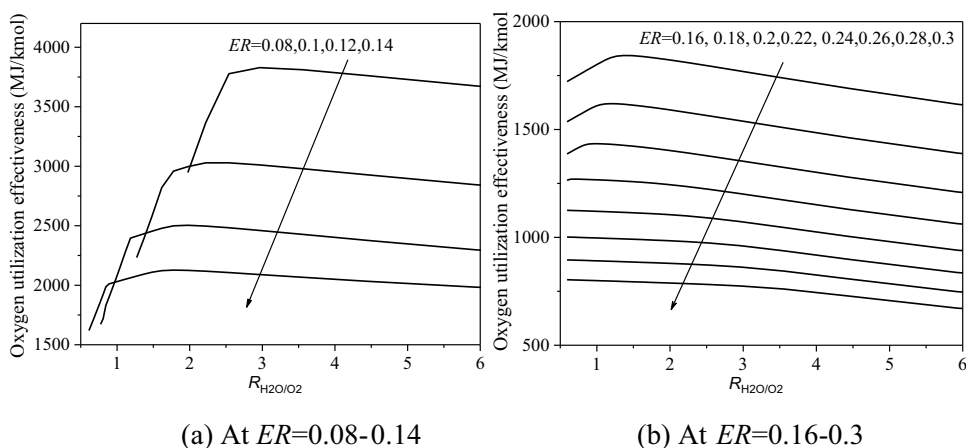(a) At ER=0.08-0.14   (b) At ER=0.16-0.3

Figure 9. Effects of ER and $R_{H2O/O2}$ on the utilization effectiveness of oxygen.

where, X_{O2} is the utilization effectiveness of oxygen, MJ/kmol; Q_g is the calorific value of syngas, MJ/kmol; F_g is the amount of syngas, kmol; F_{O2} is the amount of oxygen used as gasification agent, kmol.

Figure 9 shows the variation of utilization effectiveness of oxygen with ER and $R_{H2O/O2}$. The gasification equivalence ratio has great influence on the utilization effectiveness of oxygen, and decreasing ER makes the utilization effectiveness of oxygen rapidly increases (especially at $ER = 0.08–0.14$. The results indicates that, in the UCG, the ER is a key parameter, it directly influences gasification temperature and the gasification chemical reactions.

At low ER (0.08–0.14), the utilization effectiveness of oxygen is significantly higher as compared that at high ER (0.16–0.3) owing to the high calorific value of gaseous products. With the increase of $R_{H2O/O2}$, the utilization effectiveness of oxygen first increases, reaches a peak value and then decreases, which is similar to the variation of calorific value mentioned above (see Figure 8a). In practical production, although the high calorific value syngas is obtained at low ER (0.08–0.14), char is possibly incompletely gasified due to the low gasification temperature (see Figures 5a and 6), and the low gasification temperature easily causes gasification production termination. At $ER = 0.22–0.3$, the utilization effectiveness of oxygen monotonously decreases with the increase of $R_{H2O/O2}$. When ER is in the range of 0.08–0.2, $R_{H2O/O2}$ has the corresponding optimum value, where the utilization effectiveness of oxygen reaches the corresponding peak value.

Considering the reliability and safety of the practical underground coal gasification operation, it should be avoided to occur the large fluctuation of gasification temperature, the component concentration of gaseous products, gaseous product flowrate owing to the fluctuation of gasification agent parameters (the flowrate and steam-to-oxygen ratio). Therefore, based on above analysis, the operating ranges of ER and $R_{H2O/O2}$ can select 0.16–0.24 and 1.5–2.0, respectively.

Conclusions

UCG thermodynamic equilibrium model was established and validated, and the effects of $R_{H2O/O2}$ and ER on the gasification temperature and the concentrations of the gaseous products were investigated. The major conclusions are as follows:

(1) ER is a key operating parameter of UCG, it directly influences gasification temperature and the gasification chemical reactions. At low ER (0.08-0.14), coal is incompletely gasificated when the gasification temperature is less than the corresponding peak temperature. At high ER (0.16-0.30), coal is completely gasificated. With the increase of $R_{H2O/O2}$, the gasification temperature decreases monotonically and finally tends to approach 850°C at $R_{H2O/O2}$ = 4.5.

(2) H_2 reaches high concentration at ER = 0.18-0.24 and $R_{H2O/O2}$ = 1-3, but the change of CH_4 concentration is just the opposite. CO concentration is high at $R_{H2O/O2}$<1.5, which decreases with the increase of ER and $R_{H2O/O2}$. $R_{H2O/O2}$ has significant impact on CO_2 formation, and CO_2 reaches high concentration at $R_{H2O/O2}$>4.

(3) ER has significant influence on the utilization effectiveness of oxygen. Especially, when ER is less than 0.16, decreasing ER makes the utilization effectiveness of oxygen rapidly increases. At ER=0.08-0.2, $R_{H2O/O2}$ has the corresponding optimum value, where the utilization effectiveness of oxygen reaches the corresponding peak value.

Disclosure statement

No potential conflict of interest was reported by the authors.

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