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2단 분류층 가스화기에서 합성가스 생성을 위한 석탄 슬러리 가스화에 대한 수치 해석적 연구

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Numerical simulation of gasification of coal-water slurry for production of synthesis gas in a two stage entrained gasifier

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Key words : gasification(가스화), entrained flow(분류층), Probability Density Function(확율밀도함수)

Abstract : Oxy-gasification or oxygen-blown gasification, enables a clean and efficient use of coal and opens a promising way to CO2 capture. The coal gasification process of a slurry feed type, entrained-flow coal gasifier was numerically predicted in this paper. The purposes of this study are to develop an evaluation technique for design and performance optimization of coal gasifiers using a numerical simulation technique, and to confirm the validity of the model. By dividing the complicated coal gasification process into several simplified stages such as slurry evaporation, coal devolatilization, mixture fraction model and two-phase reactions coupled with turbulent flow and two-phase heat transfer, a comprehensive numerical model was constructed to simulate the coal gasification process. The influence of turbulence on the gas properties was taken into account by the PDF (Probability Density Function) model. A numerical simulation with the coal gasification model is performed on the Conoco-Philips type gasifier for IGCC plant. Gas temperature distribution and product gas composition are also presented. Numerical computations were performed to assess the effect of variation in oxygen to coal ratio and steam to coal ratio on reactive flow field. The concentration of major products, CO and H2 were calculated with varying oxygen to coal ratio (0.2-1.5) and steam to coal ratio(0.3-0.7). To verify the validity of predictions, predicted values of CO and H2 concentrations at the exit of the gasifier were compared with previous work of the same geometry and operating points. Predictions showed that the CO and H2 concentration increased gradually to its maximum value with increasing oxygen-coal and hydrogen-coal ratio and decreased. When the oxygen-coal ratio was between 0.8 and 1.2, and the steam-coal ratio was between 0.4 and 0.5, high values of CO and H2 were obtained. This study also deals with the comparison of CFD (Computational Flow Dynamics) and STATNJAN results which consider the objective gasifier as chemical equilibrium to know the effect of flow on objective gasifier compared to equilibrium. This study makes objective gasifier divided into a few ranges to study the evolution of the gasification locally. By this method, we can find that there are characteristics in the each scope divided.

1. Introduction

From the perspective of energy security and environmental sustainability, highly effective uses for fossil fuel in energy industries are demanded. Power plants such as the Integrated Coal Gasification Combined Cycle (IGCC) are being developed worldwide to use coal more efficiently and cleanly[1]. For Korea, coal is an inevitable feedstock to cover

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at least 30% of the total electricity during the next 30-50 years. Recently the interest in the hydrogen society prompts a wider application of gasification technologies including IGCC. If the concerns for the environment and energy get severer, technology shift from the combustion-based to gasification based processes would be an inevitable trend in the long run[2].

Coal gasification process is grouped into fixed bed (or moving bed), fluidized-bed, and entrained bed[3]. The fixed-bed gasifiers possess the merits of high turndown ratio and mature technology, whereas the fluidized-bed gasifiers are characterized by extremely good mixing between feed and oxidizer, thereby promoting both heat and mass transfer. However, if operations are emphasized on higher fuel conversion rate and shorter solids residence time, the entrained-bed gasifier will be a better choice for the gasification reactions because the particle of feedstock in an entrained-bed gasifier is usually ground to a size of 100μm or less. As a matter of fact, another advantage of using entrained flow is the ability to handle practically any coal as feedstock. On account of the a fore mentioned advantages, the entrained-flow gasifiers have been extensively applied in the practical processes such as Shell, GE Energy, and E-gas (ConocoPhilips)[4]. Further division is also made between those containing nitrogen(i.e., air-blown) in the product gas and thos e without nitrogen(i.e.,oxygen-blown)[3]. It is known that the oxygen blown entrained flow gasifier can be operated at extremely high temperatures to ensure complete carbon conversion[5].

For any gasification process, it is very difficult to get some detailed knowledge even by the most advanced testing instruments due to the high heating rates and short residence time. Thus, a modeling research is essential to provide a better understanding of the gasifier performance and complicated reaction phenomena in a gasifier[6]. For the past decades, simulation researches on coal reaction processes have achieved significant development. Equilibrium codes which are based on zero Gibbs energy such as ASPEN[7] or STANJAN and specialized codes for combustion and gasification such as PCGC-2[8], and commercial CFD codes like FLUENT, CFX have been used for many studies[1,5-9].

With these simulation codes, many studies have been carried out to understand characteristics of one-stage or two-stage entrained coal gasifies. Yun et al. tested coal samples to make the guidelines for IGCC using an one-stage type entrained bed gasifier[2]. Yuehong et al. conducted the process modeling using the ASPEN Plus simulator based on the new co-gasification technology[10]. Choi et al. predicted the coal gasification process of a slurry feed type numerically using the entrained flow coal gasifier[6]. Guo et al. introduced the performance of an entrained-flow gasification technology of pulverized coal in pilot-scale plant [11]. Chen investigated experimentally the partial oxidation of two different high-volatile pulverized coals in a drop tube furnace[4]. Chen et al. studied the characteristics of the 200 ton/day integrated coal gasification combined-cycle (IGCC) two-stage gasifier under various conditions, including various air ratios and coal types through comprehensive theoretical simulation[5]. Watanabe et al. developed an evaluation technique for design and performance optimization of two-stage coal gasifiers using a numerical simulation[1]. Vicente el al. used the model based on the Eulerian-Elerian concept to simulate the gasification of coal inside a two-stage entrained flow gasifer[12]. Liu et al. took the influence of turbulence into account by Gaussian PDF model for a two stage entrained flow coal gasifier[9]. Liu et al. also applied simulation to predict the gasification characteristics of an oxygen-blown gasifier[8]. Shi et al. described a CFD model of a two-stage, oxygen-blown, entrained-flow, coal slurry gasifier for use in an advanced power plant simulation[13].

The case simulated in this paper is a coal-water slurry two stage entrained oxygen-blown gasifier [13]. The effects of various operating conditions – O2/coal ratio, steam/coal ratio, temperature- on the gasifier have been numerically studied using STANJAN and FLUENT.

2. The gasifier and coal considered

The schematic of the gasifier is shown in Fig. 1. The first stage (horizontal) is chiefly a coal combustor and provides hot gases through the connection to the second stage (vertical) in which only coal slurry is injected. Most of the coal gasification process occurs in the second stage. The geometric dimensions are as follows[13]: L1 = 0.9 m, L2 = 7.0 m, L3 = 1.7 m, L4 = 0.5 m, L5 = 0.5 m, L6 = 7.6 m, D1 = 0.75 m, D2 = 1.5 m, D3 = 0.4 m and D4 = 0.4 m.

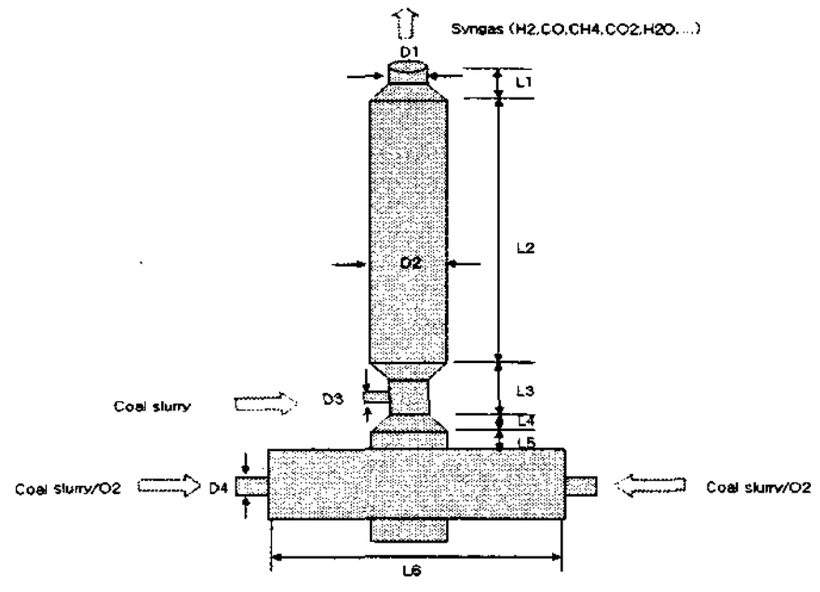

Fig. 1 Schematic of a two stage coal slurry entrained flow gasifier[13]

Results of proximate analysis and ultimate analysis are listed in Table 1[13,14], and the operating conditions used in the simulations are listed in Table 2.

Table 1 Coal properties

Type	Type: Illinois #6	
LHV	LHV : 23.0 MJ/kg	
Proximate Analysis (wt%)	Fixed Carbon	44.19
	Volatile	34.99
	Ash	9.70
	Moisture	11.12
Ultimate Analysis (wt%)	Carbon	63.75
	Hydrogen	4.50
	Nitrogen	1.25
	Chlorine	0.29
	Sulfur	2.51
	Oxygen	6.88

Table 2 Summary of test conditions

Feed Rates(kg/s)	Coal	20.16 (1 stage)
		6.113 (2 stage)
	Oxygen/Coal(kg/kg)	0.2-1.5
	Steam/Coal(kg/kg)	0.3-07
Inlet Gas Properties	Coal slurry	400K
	Oxygen	280K

3. Numerical model

In entrained-flow gasification, the coal particle flow chiefly follows the gas flow and the gasifier is typically in a dilute flow regime, in which the volume occupied by the particles and the particle-particle interactions are negligible. A criterion often used in CFD for dilute flow is that the particle volume fraction is less than 10%. In this case, a discrete phase method (DPM) can be applied to model the particle flow. Also the ratio of the mass flow rate of solids to that of the gas is less than or equal to one, which is required for ensuring the stability of DPM calculations.

The following physical and chemical processes were included in the present model: (1) turbulent flow of gas and mixing of gaseous reactants, (2) entrainment of coal particles and their turbulent dispersion, (3) coal devolatilization, volatile combustion, and heterogeneous char reactions including combustion and gasification, (4) convective and radiative heat transfer among coal, char and ash particles.

The gas-phase combustion was modeled using the fast chemistry mixture fraction approach. The Discrete Ordinates model was used as the radiation model[15]. Using DPM, the particle trajectories, alone with mass and energy transfer to particles, were computed with a Lagrangian formulation, but the effect of particle on the gas and particle was not considered.

3.1. Gas phase model

The gas phase is assumed to be a steady state, reacting, continuum field that can be described by general conservation equation as follows:

$$\frac{\partial(\rho u_i \varphi)}{\partial x_i} = \frac{\partial}{\partial x_i}(\Gamma \frac{\partial \varphi}{\partial x_i}) + S_\varphi$$

where u is the fluid phase velocity and ρ is the fluid phase density. φ refers to any quantity of mass, velocity, turbulent kinetic energy (k)andturbulentkineticenergydissipationrate (ε),gas enthalpy(h),and mixture fractions (f_i). S_φ is the source term[9]. The mixture fraction, denoted by f_i ,is defined as the local mass fraction o fburnt and unburnt fuel stream elements (C,H,etc.) in all the species. It can be written in terms of the atomic mass fraction as

$$f = (Z_i - Z_{i,ox}) / (Z_{i,fuel} - Z_{i,ox})$$

where, Z_i is the elemental mass fraction for some element, i .The subscript 'ox' denotes the value at the oxidizer stream inlet and the subscript 'fuel' denotes the value at the fuel stream inlet[15].

3.2. Particle phase dispersion

The equation of motion of a particle is following:

$$\frac{du_p}{dt} = F_D(u - u_p) + \vec{g}\frac{(\rho_p - \rho)}{\rho_p} + \vec{F}$$

where, F_D is the drag force per unit particle mass, u_p is the particle velocity, ρ_p is the particle density and $\vec{F}$ is the additional force. The dispersion of particles due to turbulence in the fluid phase is predicted using the stochastic tracking model in which random velocity fluctuations based on turbulence intensity and eddy lifetime are added on top of the particle instantaneous velocities. The heat transfer from the continuous phase to the discrete phase is computed by examining the change in thermal energy of a particle as it passes through each control volume. This heat exchange appears as a source or sink of energy in the continuous phase energy balance during any subsequent calculations of the continuous phase flow field. The mass transfer from the discrete phase to the continuous phase is computed by examining the change in mass of a particle as it passes through each control volume.

This mass exchange appears as a source of mass in the continuous phase continuity equation and as a source of a chemical species[13].

3.3. Coal reactions

3.3.1 Devolatilization model

Two-competing-rates model proposed by Kobayashi is used for kinetic devolatilization rate as following[15],

$$R_1 = A_1 e^{-(E_1/RT_p)}$$

$$R_2 = A_2 e^{-(E_2/RT_p)}$$

where, R_1 and R_2 are competing rates that may control the devolatilization over different temperature ranges, T_p is the particle temperature, A_1 and A_2 are Arrenhenius type pre-exponential factor, E_1 and E_2 are activation enegy, and R is universal gas constant.

The two kinetic rates are weighted to yield an expression for the devolatilization as

$$\frac{m_v(t)}{(1-f_{w,o})m_{p,0}-m_a} = \int_0 (\alpha_1 R_1 + \alpha_2 R_2)\exp(-\int_0 (R_1+R_2)dt)dt$$

where, $m_v(t)$ is the volatilized mass at time t, $m_{p,0}$ is the initial particle mass, $f_{w,0}$ is the mass fraction of volatile material and m_a is the ash content in the particle, α_1 and α_2 are yield factors.

3.3.2 Char gasification model

After the volatile component of the particle is completely evolved, a surface reaction begins, which consumes the combustible fraction of the particle. The surface reaction mechanism is assumed to be one step, irreversible reaction.

$$\text{Char(s)} + 0.5\ O_2\ \text{(g)} \rightarrow \text{CO(g)}$$

For heterogeneous surface reaction rate, in this work, the kinetics/diffusion-limited model (Field, 1969; Baum and Street, 1971) is used in which the reaction rate is determined by combining the diffusion rate with the kinetic rate. The diffusion rate is:

$$D_0 = C_1 \frac{[(T_p + T_\infty)/2)]^{0.75}}{d_p},$$

and the kinetic rate is:

$$\Re = C_2 e^{-(E/RT_p)},$$

which, through the parameters C_2 and E ,incorporates the effects of chemical reaction on the internal surface of the char particle and pore diffusion. The resulting equation is

$$\frac{dm_p}{dt} = -\pi d_p^{\,2} \frac{\rho R T_\infty Y_{o_2}}{M_{w,o_2}} \frac{D_0 \Re}{D_0 + \Re}$$

where, C_1 is the mass diffusion limited rate constant, C_2 is the kinetics limited rate pre-exponential factor, T_∞ is the local fluid temperature, d_p is the particle diameter, m_p is the instantaneous mass of the particle, Y_{O_2} is the local mass fraction of oxygen in the gas and M_{w,O_2} is the molecular weight of oxygen.[16].

4. Results and discussion

4.1. Comparison with previous study

Fig. 2 is the diagram of comparing with Shi et al[13]. made results which are the same assumption as current study except the interaction model between chemistry and turbulence. As shown in Fig. 2, concentrations of synthesis gas at the outlet are similar. This means that use of Non-premixed model instead of Finite rate/eddy dissipation model for more flexible and convenient study is reasonable. There have been several studies by Non-premixed model for entrained gasifiers[1,6,12].

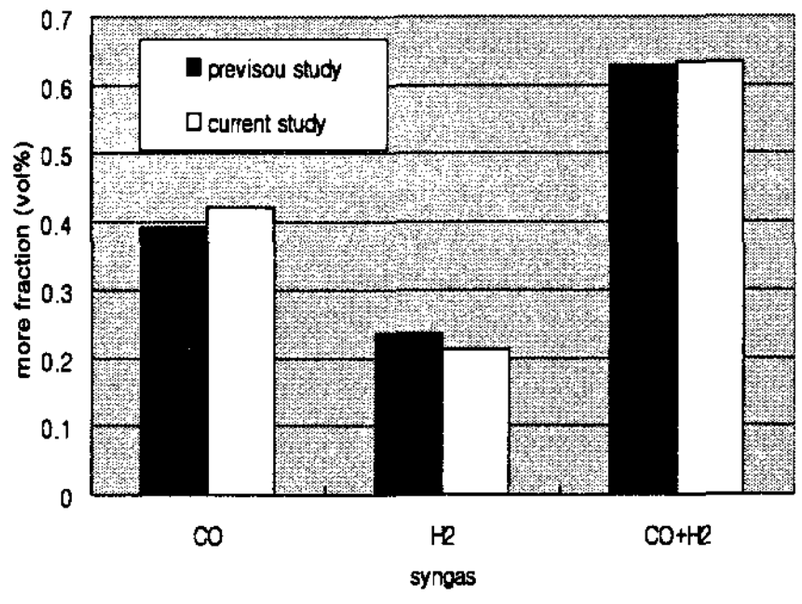

Fig. 2 Comparison of mole fraction of synthesis gas

4.2. Effect of O2/Fuel in feedstock

Fig. 3 shows the plotting of volume averaging temperature, CO and H2O with O2/Fuel ratio. When the mass fraction of O2/Fuel is increased from 0.6 to 0.8, temperature is increased rapidly. It is thought that the increase in O2 favors exothermic combustion reaction and leads to more combustion reaction. Then, there exists a turning

point on the increasing tendency of the reactor temperature with increasing O2, as shown in Fig. 3. In this study, it was found that the turning point appears in the range where the H2 concentrations reach maximum[10].

Fig. 3 also shows the concentration of synthesis gas with O2/Fuel ratio. The concentrations of CO and H2 are very low until an O2/Fuel ratio around 0.5-0.6. However, there is change of this trend from the ratio. As it can be seen in Fig. 3, the reason is thought due to the result of rapid gasification in the range of 800-1000K.

From the Fig. 8, it is also shown that the maximum of synthesis gas is formed in the range of O2/Fuel ratio 0.8-1.0. The reason is that rich O2/Fuel ratio prompts the combustion and makes little gasification. Therefore, it is important to select the O2 flow rate where the O2/Fuel ratio is around the optimum region for the richer synthesis gas. In this work, it is concluded that oxygen should be controlled so that the O2/Fuel ratio may be within the range of 0.8-1.0.

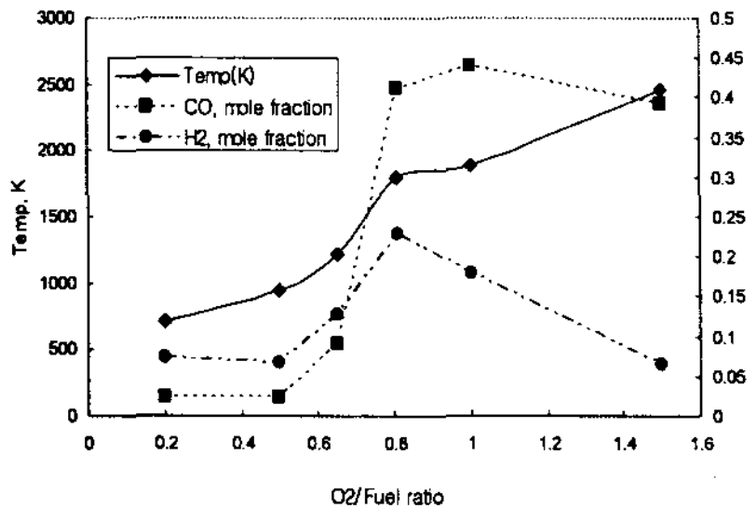

Fig. 3 Influence of O2/Fuel ratio on Temperature

In order to present the evolution of gasification in this gasifier, Fig. 4 gives five sets of ranges which show characteristics of operating region. Fig. 5 shows the cross-sectional averaged temperatures at defined cross-sections with O2/Fuel ratios.

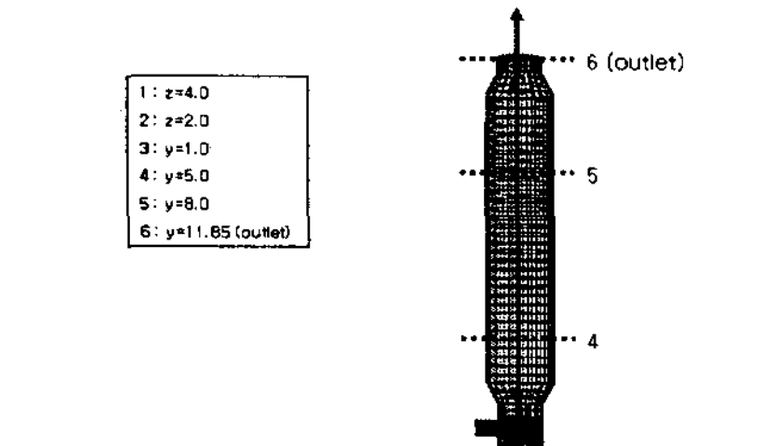

Fig. 4 Classification of tracked path for study of temperature characteristics

As presented in Fig. 5, the maximum temperature is encountered in the region 1-2, which is due to complete combustion in the region. In the region 2-3, gas temperature decreases sharply, especially, at the ratio 0.82. Fig. 6 shows that the decrease of temperature in the region 2-3 leads to a reduction of CO concentration.

The lower O2/Fuel ratio(<0.5) lead to less than 800K except in the region 1-2. As shown in Fig. 6, the CO has decreased profiles when the temperature is less than 800K.

According to Zhao et al.[10], gasification that proceeds at temperature above 873K causes the kinetic barriers to be largely minimized, and the gaseous mixtures leaving many types of gasifiers are found to approach equilibrium. These simulation results are in good agreement with the suggestion of Zhao et al, as shown at the region 4-5 and 5-6 in Fig. 6.

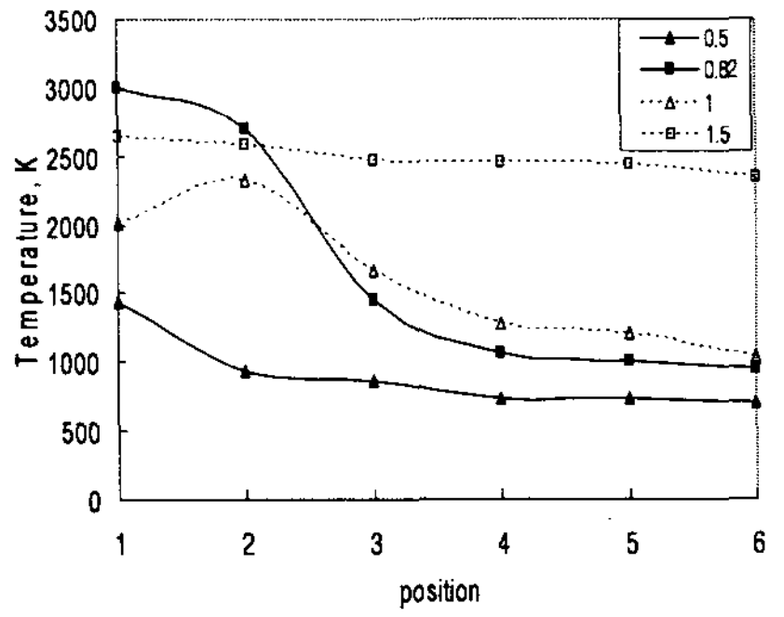

Fig. 5 Trend of temperature at various tracked position on various O2/Fuel ratios

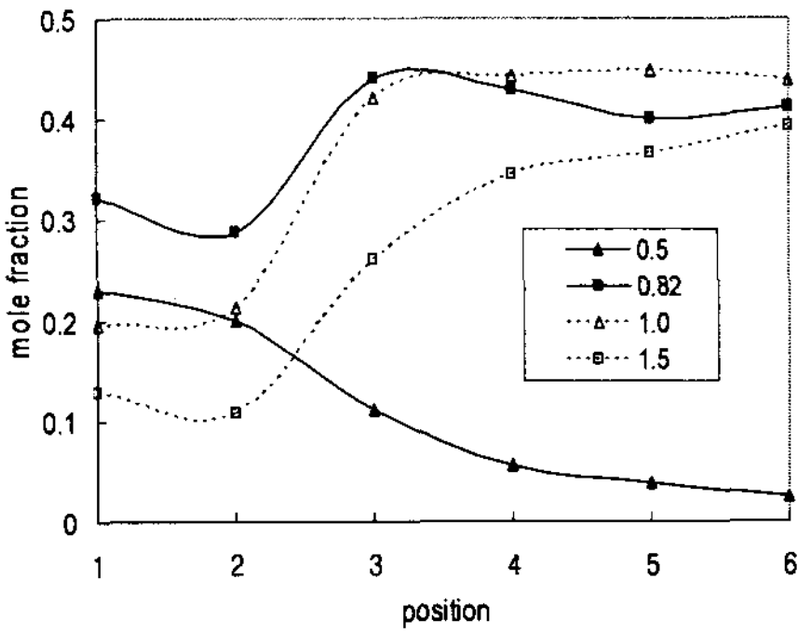

Fig. 6 Trend of CO at various tracked position on various O2/Fuel ratios

4.3. Effect of Steam/Fuel ratio in feedstock

Fig. 7 shows the increase of steam leads to reduction of synthesis gas at the outlet of the gasifier. This is due to the fact that the increased steam steals some heat and decreases the reactor temperature, as shown in Fig. 8, hindering the endothermic reactions of steam reforming[10]. Although there is the water-gas shift effect due to

increasing steam, the counter-effect of temperature seems to be bigger. In case of CO, the maximum range is 0.4-0.5.

Therefore, steam may be an approximate method to adjust the reactor temperature for its effect on the synthesis position, but there is a limited point. In this work, it is concluded that steam should be controlled so that the O2/Fuel ratio may be within the range of 0.4-0.5.

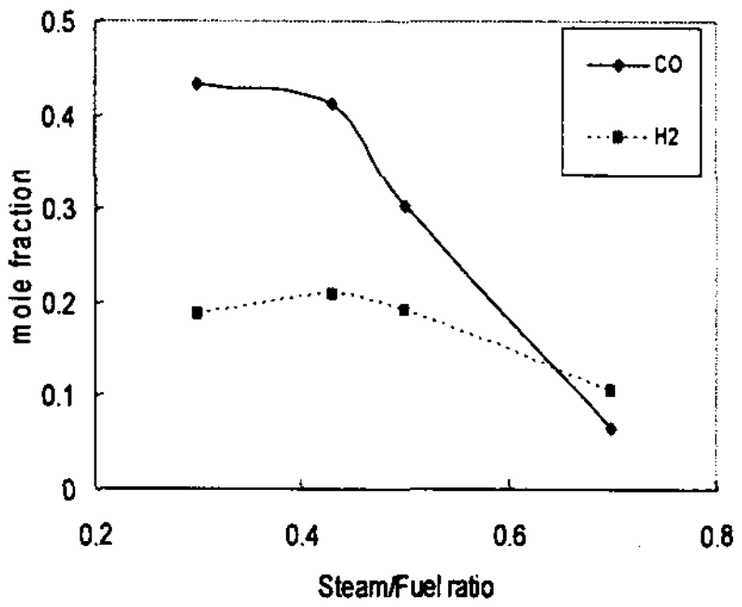

Fig. 7 Influence of Steam/Fuel rate on syngas mole fraction

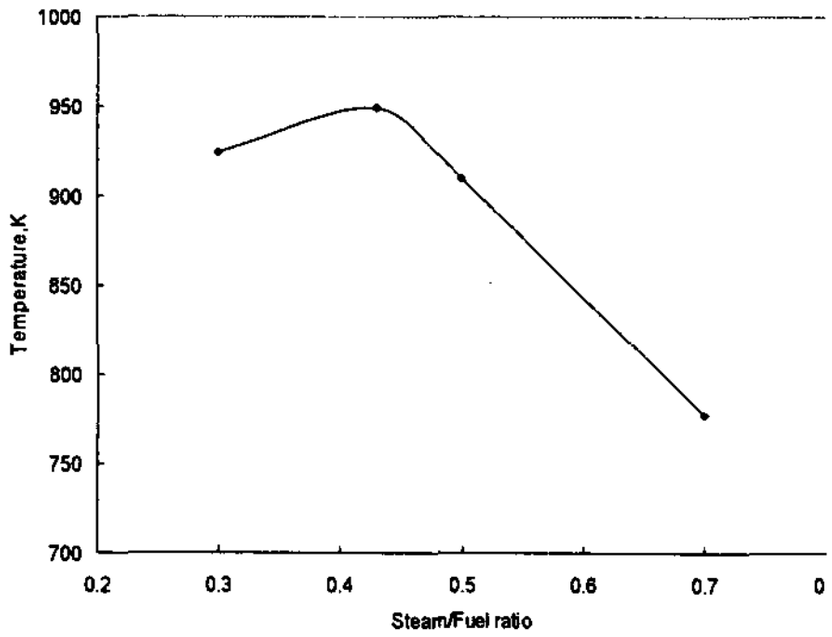

Fig. 8 Influence of Steam/Fuel rate on temperature

5. Conclusion

A comprehensive computer simulation model has been developed and applied to the simulation of a coal water-slurry two stage entrained flow gasifier recently developed for the IGCC process. In this study, we have used non-premixed model, which assume the local composition is defined from the mixing of the inlet steams and coal off-gas. The simulation results are in good agreement with the previous work, in which the only difference model is the interface model of turbulence and chemistry. Based on the above models, we have studied the effect of the temperature, O2/Fuel ratio, and Steam ratio on the product of synthesis gas respectively. According to our studies, the high values of CO and H2 were obtained when the oxygen-coal ratio was between 0.8 and 1.2, and the steam-coal ratio was between 0.4 and 0.5.

We have also divided the gasifier into a few regions and studied the characteristics of gasification evolution of the coal water-slurry two stage entrained flow gasifier. This approach shows the characteristics of the staged gasification.

For the considering the mixing effect of gasifier, we have compare the result of CFD with one of chemical equilibrium calculation at the outlet of gasifier. Through it, we can find the effect of mixing on the synthesis gas at outlet of gasifier is little in case the O2/Fuel ration is more than 0.8.

Acknowledgement

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