

Predictions of O₂/N₂ and O₂/CO₂ Mixture Effects during Coal Combustion using Probability Density Function

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A simulation model has been developed for the entrained flow coal gasification process to investigate the effect of N₂ replacement with CO₂ during combustion. Both O₂/N₂ and O₂/CO₂ gas mixtures were used, with molar fractions of oxygen varying between 10 and 100%. The conventional numerical methods and sub-models used for the pulverized coal combustion have been considered in this modeling. For reacting system, non-premixed combustion model using the Probability Density Function (PDF) for fluctuating scalars (mixture fraction) has been employed. It has been found that the O₂/N₂ environment gives higher gas temperature than O₂/CO₂ environment with the same oxygen content. On the other hand, the similar flue gas temperature and char conversion were obtained with oxygen content in the O₂/CO₂ mixture at about 30%.

1. Introduction

Coal is a major source of energy, accounting for 27% of the world energy supplies and 35% of the world electricity generation¹. It is predicted that coal will continue to play an important role in meeting the world's increasing energy demands in the foreseeable future. However, the use of coal faces several challenges. The major one is the considerable emission of CO₂, which leads to climate change and air pollution. Carbon dioxide is believed to be a "greenhouse gas" which can lead to global warming²⁻⁵. Carbon dioxide sequestration has been suggested as a means to reduce the amount of carbon dioxide being released into the atmosphere⁵. Carbon dioxide sequestration involves long-term storage of carbon dioxide in reservoirs to reduce its buildup in the atmosphere. While the advantages, disadvantages, and overall effectiveness of the sequestration strategies are still being studied. CO₂ sequestration also requires a technique to separate carbon dioxide from the stack gases found in combustion. Therefore to save the global environment and to reduce the additional cost for CO₂ sequestration, emission of CO₂ during coal gasification should be reduced.

Our research will simulate coal gasification processes giving the special attention on the CO₂ blown gasification. Coal combustion with O₂/CO₂ is one of several promising new technologies associated with mitigating the CO₂ rise in the atmosphere². In coal combustion with air, it is difficult to remove CO₂ efficiently because the CO₂ concentration in the flue gas is only about 13%. On the other hand, in coal combustion with O₂/CO₂ mixtures, the CO₂ concentration in the flue gas may be enriched up to 95% and direct CO₂ recovery therefore becomes possible without additional energy consumption³. The separated CO₂ is then recycled to the reactor inlet. This will reduce the total emission of CO₂ in the atmosphere and the total carbon conversion will be increased. Therefore, the research will contribute to clean coal technology projects using advanced power generation technologies such as Integrated Gasification Combined Cycle (IGCC).

Experimental results revealed that coal particles burned at higher mean temperatures in O₂/N₂ than in O₂/CO₂ environments at analogous oxygen mole fraction⁶. The measured volatile and char temperatures as in air (21% O₂) were attained with oxygen content in the O₂/CO₂ mixtures in the neighborhood of 30%⁶. Croiset et al.^{7,8} investigated the combustion of axisymmetric flames of two bituminous pulverized coals in a 210-kW furnace, in air as well as in O₂/CO₂ mixtures for which the oxygen mole fraction was varied from 21% to 42%. Their results showed that flame temperatures in O₂/CO₂ mixtures were similar to those in air when the oxygen mole fraction in the windbox was in the range 30–35%, depending on the particular coal. Liu et al.^{9,10} studied a high-volatile bituminous coal in a 20-kW down-fired combustor in air as well as in O₂/CO₂ mixtures and measured flame temperatures using thermocouples. They reported that replacing the N₂ in the combustion air with CO₂ resulted in a significant decrease of temperature. Specifically, coal combustion in 30%O₂/70%CO₂ produced gas temperatures matching those of coal combustion in air. Molina and Shaddix¹¹ found that particle ignition and devolatilization properties in a mixture of 30%O₂/70%CO₂ are very similar to those in air.

The aim of this study is to substitute N₂ by CO₂ during coal gasification that will maximize the syngas production. This substitution will affect the combustion characteristics of pulverized coal, the gas temperature profile, the coal burnout and the syngas concentration profile. Therefore, this work aims at obtaining basic knowledge in the gasification of pulverized coal in O₂/CO₂ versus conventional O₂/N₂ environments.

2. Numerical Models

2.1 Modeling geometry and operating conditions

A schematic geometry considered for this simulation is shown in Fig. 1. The proximate and ultimate analysis of coal considered in this simulation is given in Table 1. The inlet boundary conditions are summarized in

Table 2. Although the inlet boundary conditions do not completely simulate the actual industrial gasifier, the purpose of the present work is only to study the combustion characteristics inside the coal gasifier. The coal is assumed to inject with a velocity of 15m/s and 10m/s along x and y axis respectively.

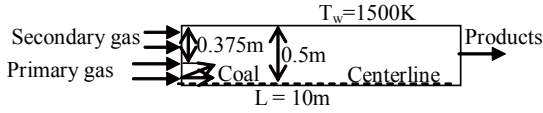


Fig. 1 Schematic view of gasifier geometry.

Table 1 Proximate and ultimate analysis of bituminous coal (Indonesia)

Proximate analysis (wt %)		Ultimate analysis (wt% d.a.f)	
Moisture	4.74	C	74.3
Volatile matter	38.72	H	5.25
Fixed carbon	48.55	O	10.1
Ash	7.98	N	1.85
LCV(MJ/kg)	29.4		

Table 2 Inlet boundary condition for modeling study

Operating condition	Value
Primary gas flow rate	6.748e-3 kg/s
Primary gas temperature	300K
Secondary gas flow rate	39.73e-3 kg/s
Secondary gas temperature	400K
Coal flow rate	4.0e-3 kg/s
Coal inlet temperature	300K

2.2 Mathematical

Under certain assumptions, the thermo chemistry can be reduced to a single parameter: the mixture fraction. The mixture fraction, denoted by f , is the mass fraction that originated from the fuel stream. In other words, it is the local mass fraction of burnt and unburnt fuel stream elements (C, H, etc.) in all the species (CO_2 , H_2O , O_2 , etc.). The approach is elegant because atomic elements are conserved in chemical reactions. The mixture fraction is a conserved scalar quantity and can be written in terms of the atomic mass fraction as¹²⁾:

$$f = \frac{Z_i - Z_{i,ox}}{Z_{i,fuel} - Z_{i,ox}} \quad (1)$$

where Z_i is the elemental mass fraction for 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.

Non-premixed modeling involves the solution of transport equations for one or two conserved scalars (the

mixture fractions). Equations for individual species are not solved. Instead, species concentrations are derived from the predicted mixture fraction fields. Interaction of turbulence and chemistry is accounted for with an assumed-shape Probability Density Function (PDF). The Probability Density Function, written as $p(f)$, can be thought of as the fraction of time that the fluid spends in the vicinity of the state f and mathematically can be expressed as,

$$p(f)\nabla f = \lim_{T \rightarrow \infty} \frac{1}{T} \sum_i \tau_i \quad (2)$$

where T is the time scale and τ is the amount of time that f spends in the ∇f band.

The probability density function $p(f)$, describing the temporal fluctuations of f in the turbulent flow, can be used to compute averaged values of variables that depend on f . Under the assumption of chemical equilibrium, all thermo chemical scalars (species fractions, density, and temperature) are uniquely related to the mixture fraction(s). Density-weighted mean species mass fractions and temperature can be computed as:

$$\bar{\phi}_i = \int_0^1 p(f)\phi_i(f)df \quad (3)$$

In non-premixed combustion, turbulence slowly convects/mixes fuel and oxidizer into the reaction zones where they burn quickly. In such cases, the combustion is said to be mixing-limited and the complex chemical kinetic rates can be safely neglected. A turbulence chemistry interaction model, based on the work of Magnussen and Hjertager¹³⁾, called the eddy dissipation model employs for this simulation. The net rate of production of species i due to reaction r , $R_{i,r}$, is given by the smaller (i.e., limiting value) of the two expressions below:

$$R_{i,r} = v'_{i,r} M_{w,i} A \rho \frac{\varepsilon}{k} \min \left(\frac{Y_R}{v'_{R,r} M_{w,R}} \right) \quad (4)$$

$$R_{i,r} = v'_{i,r} M_{w,i} A B \rho \frac{\varepsilon}{k} \frac{\sum_P Y_P}{\sum_j v'_{j,r} M_{w,j}} \quad (5)$$

where:

A is an empirical constant equal to 4.0

B is an empirical constant equal to 0.5

3. Results and discussion

3.1 Combustion species study

The main combustion species involved in combustion reaction are N_2 , O_2 , H_2 , CO , CO_2 , CH_4 , H_2O , C_2H_6 , C_2H_4 and solid carbon $\text{C}(s)$. The concentration profiles for some major species are shown in the Figs. 2 and 3. From these figures we see that CO and H_2 concentration decrease as the reaction proceed. This is because the general reaction of CO and H_2 with O_2 produce CO_2 and

H₂O, respectively. CH₄ and solid carbon also get oxidized to form carbon dioxide if sufficient O₂ is provided. Fig. 4 also shows the concentration profiles of H₂ and CO in the region of secondary flow area. The profile is different in this area because of relatively lower temperature than the central zone. The high inlet species concentrations of H₂ and CO in the center zone are due to the presence of coal volatile source in this region.

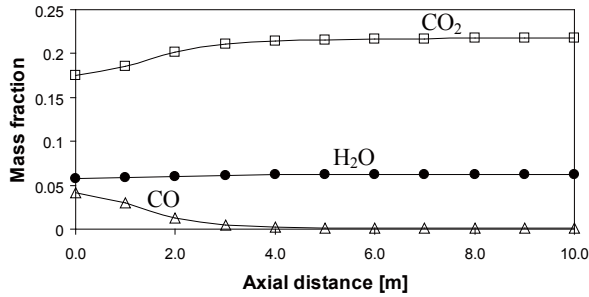


Fig. 2 Centerline concentration profiles of CO, CO₂ and H₂O for conventional O₂/N₂ mixtures.

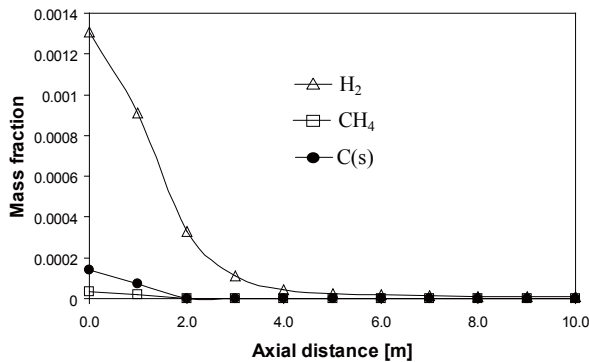


Fig. 3 Centerline concentration profiles of H₂, CH₄ and Carbon solid C(s) for conventional O₂/N₂ mixtures.

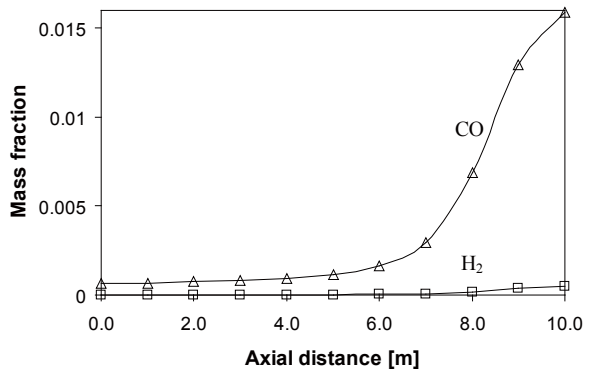


Fig. 4 Concentration profiles of H₂ and CO for conventional O₂/N₂ mixtures at a distance 0.25m from the centerline.

3.2 Temperature profile comparison

The centerline temperature profile has been investigated both for O₂/N₂ and O₂/CO₂ mixtures with different O₂ mole fraction. The comparisons are shown in Fig. 5. The results show that coal combustion with O₂/CO₂ mixtures gives a significant decrease in combustion gas temperatures. And, coal combustion in 30%O₂/70%CO₂ can produce similar gas temperature profiles to those of coal combustion in air. This calculated result shows a good agreement with the study of Liu et al.⁹. It has also been found that after 2.0 m from the inlet of the reactor, the temperature profiles become almost constant.

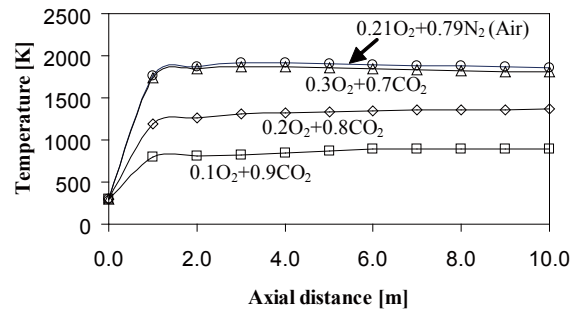


Fig. 5 Centerline temperature profiles for different combustion media (air, various O₂/CO₂ mixtures).

3.3 Species concentration profile comparison

In air and 0.3O₂/0.7CO₂ environments, the centerline concentration profile for CO and H₂ are shown in Figs. 6 and 7, respectively. It has been found that the tendencies of concentration profile are same for both conditions. The deviation for inlet composition for CO and H₂ is due to different mixture fraction present during coal devolatilization. However, the final concentrations become same if the reactor length is high. From the Figs. 6 and 7, we found that if the reactor length is above 4.0 m then both environments give similar outlet concentration. After this limit the concentration profile also become almost constant.

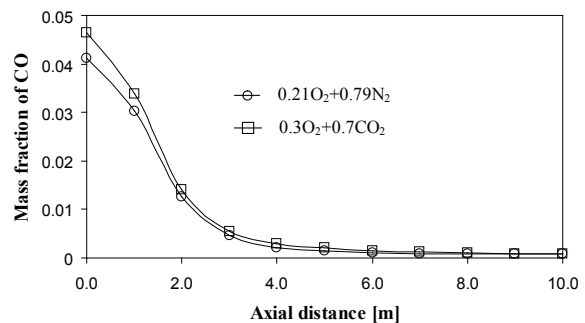


Fig. 6 Centerline concentration profile of CO for O₂/N₂ and O₂/CO₂ mixtures.

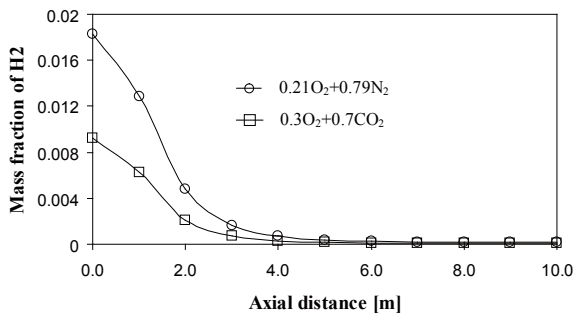


Fig. 7 Centerline concentration profile of H₂ for O₂/N₂ and O₂/CO₂ mixtures.

3.4 Char conversion

Char conversion during coal combustion has been studied by changing the mole fraction of O₂ in O₂/CO₂ mixtures. The results are shown in the Fig. 8. This result confirms that the combustion of char can be completed in an O₂/CO₂ atmosphere in the mixture of 0.3O₂/0.7CO₂. This shows a good agreement with the study of Wang et al.¹⁴⁾ confirming that the combustion of pulverized coal can be completed in a CO₂/O₂ atmosphere over a range of CO₂ to O₂ mole ratios between 2.23 and 3.65. The conventional O₂/N₂ mixture gives 97.36% char conversion during combustion. It also indicates that 0.21O₂/0.79N₂ and 0.3O₂/0.7CO₂ mixtures give similar char conversion. But, 0.2O₂/0.8CO₂ (analogous O₂ content with air) mixtures give only 64.89% char conversion.

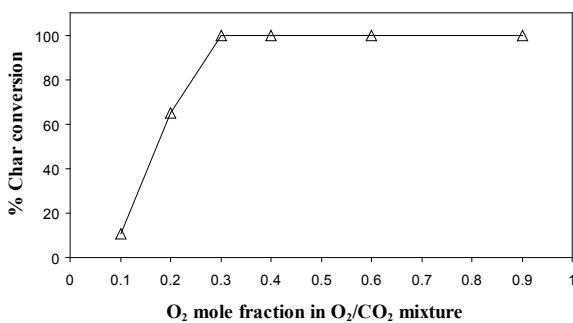


Fig. 8 Char conversion with different O₂ mole fraction in O₂/CO₂ mixtures.

3.5 Prediction of CO and H₂ production

From sections 3.2 and 3.3, we know that after 4.0 m reactor length the temperature and concentration do not change with length. Therefore, we predicted the product concentration for CO and H₂ at a length of 5.0 m by changing the O₂ mole fraction in O₂/CO₂ mixtures. The results are shown in Figs. 9 and 10. It has been found that the concentration of CO and H₂ reduce with increasing O₂ contents in the O₂/CO₂ mixtures. The concentrations become almost zero at a mixture of 0.4 O₂/0.6CO₂.

However, with a mixture of 0.2O₂/0.8CO₂ which has analogous mole fraction of O₂ in air, gives significant amount CO and H₂. On the other hand, conventional O₂/N₂ mixture gives a mass fraction of 0.0014 and 0.00038 for CO and H₂, respectively.

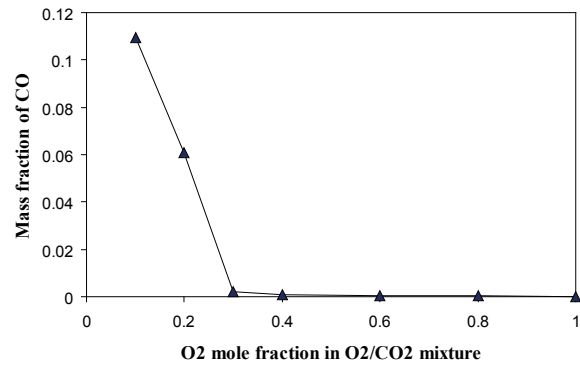


Fig. 9 Outlet concentration of CO with different O₂ mole fraction in O₂/CO₂ mixtures.

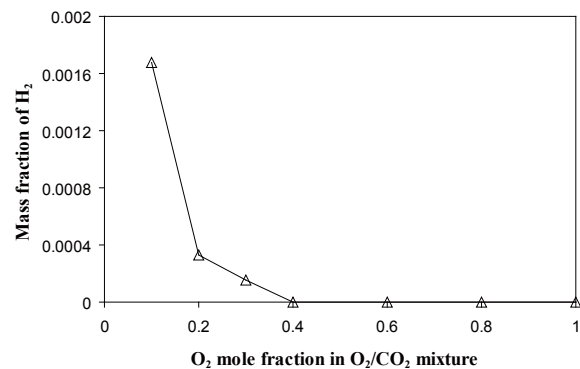


Fig. 10 Outlet concentration of H₂ with different O₂ mole fraction in O₂/CO₂ mixtures.

4. Conclusions

A number of coal combustion modeling using mixture fraction approach has been carried out with different combustion media (air, various O₂/CO₂ mixtures). From this study, we can conclude as follows:

- O₂/CO₂ combustion mixture with same O₂ content in air results in significant decrease in gas temperature (1859.45K to 1363.97K) and the char conversion (97.36% to 64.89%).
- To maintain a similar temperature profile and CO and H₂ concentration profile for coal combustion in air and for coal combustion in O₂/CO₂, the oxygen concentration in O₂/CO₂ mixture has to be kept around 30%.

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Nomenclature:

- $V'_{i,r}$ = Stoichiometric coefficient for reactant i in reaction r (dimensionless)
 $V''_{j,r}$ = Stoichiometric coefficient for product j in reaction r (dimensionless)
 $M_{w,i}$ = Molecular weight of species i (kg/kgmol)
 $M_{w,j}$ = Molecular weight of species j (kg/kgmol)
 Y_P = Mass fraction of any product species, P (dimensionless)
 Y_R = Mass fraction of any reactant species, P (dimensionless)
 ρ = Density (kg/m³)
 k = Turbulent kinetic energy (m²/s²)
 ε = Turbulent dissipation rate (m²/s³)

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