

NUMERICAL STUDY OF SINGLE COAL PARTICLE COMBUSTION IN O₂/N₂ AND O₂/CO₂ ATMOSPHERES

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ABSTRACT

This paper reports a modelling work on the existing experimental data of single coal particles combustion process [1][2]. Two kinds of coal particles (a high-volatile bituminous coal / a lignite coal) are injected in a drop-tube furnace, ignited and burned in quiescent O₂/N₂ and O₂/CO₂ atmospheres with oxygen mole fraction varying from 20% to 100%. Ignition delay time, particle life time and particle surface temperature are calculated to analyse the combustion behaviour of the single coal particle. Transient simulations are carried out based on Discrete Phase Model (DPM). The predicted results are in good agreement with the experimental data and give the details of combustion process. Further, predictions of NO_x emissions are made to understand the influence of different gas atmosphere and oxygen mole fraction. This includes both thermal-NO_x and fuel-NO_x formation mechanisms. It is argued that the NO_x emissions in both O₂/N₂ and O₂/CO₂ atmospheres are the result of competitions between the maximum particle temperature and the burning time.

INTRODUCTION

Coal is widely used for energy production across the world and pulverised combustion is perhaps the most common technology for utilising coal energy. Despite serious emissions caused by coal combustion, it is still and, most probably, will continue to be amongst the main resources for power generation [3][4]. Coal combustion which involves devolatilization, char combustion and gas phase reactions is a multi-phase, multi-scale and multi-component process [2]. Types of coal and the operation condition of reactors are also the key factors in its combustion behaviour. Further, the complex chemical processes involved in coal combustion need to be explored.

Due to the increasing concerns about global warming, carbon dioxide emission by coal combustion is now a serious issue. Different methods have been developed for carbon capture and storage (CCS) to reduce the CO₂ emission from coal-fired power plants. Pre-combustion capture, post-combustion capture, oxy-fuel combustion and chemical looping are four main CCS technologies [5]. Among these, the oxy-fuel combustion is a near-zero emission technology and the most promising one for power station utilisation [5]. The replacement of air with O₂/CO₂ leads to modified distributions

NOMENCLATURE

K	[kmol/m ³ s]	Reaction rate
k	-	Reaction rate constant
c	[kmol/m ³]	Concentration
A	-	Pre-exponential factor
E	[J/kmol]	Activation energy
T	[K]	Temperature
R	-	Universal gas constant
X	-	Mole fraction
a	-	Oxygen reaction order
b	-	Temperature exponent
d	-	Reaction order
e	-	Reaction order
Special characters		
α	-	Distribution coefficient
Subscripts		
s	-	Solid
x	-	Composition of chemical element H
y	-	Composition of chemical element O
R	-	Reactant
ox	-	Oxidant

of temperature and species, as well as radiation flux, resulting from the property differences between N₂ and CO₂. This also results in changes in NO_x formation and reduction in the flame temperature [6].

Nitrogen oxides, including nitric oxide (NO), nitrogen dioxide (NO₂), nitrous oxide (N₂O) are mostly produced through the oxidation of the molecular nitrogen in air (thermal NO_x) and the organically bound nitrogen in the coal (fuel NO_x) [7]. NO_x formed from molecular nitrogen and hydrocarbon fragments resulting from the devolatilization process near the reaction zone of flame (prompt NO_x) accounts for less than 5% of the total NO_x formed [8], and is usually neglected in modelling. In a combustion system, including pulverized combustion, the formation and destruction of NO_x emissions are influenced by several factors such as fuel properties and, combustion conditions e.g. temperature of reaction and the fuel-air ratio [9]. A series of work on NO_x emissions in coal combustion was conducted by Lookwood and his co-workers in 1990s [10][11][12]. Replacing the background gas N₂ by CO₂ can limit the NO_x emissions formed at high temperatures from nitrogen of the combustion air, and then the formation of NO_x is mainly due

to the conversion of fuel-N, partially or totally into nitrogen oxides [13].

The effects of O₂/N₂ and O₂/CO₂ gas atmosphere on coal combustion and NO_x emissions have been investigated by a number of authors. Experiments conducted by Rathnam et al. [14] showed that the devolatilization rate was similar in either N₂ or CO₂ at temperatures lower than 1000 K, while it was higher in CO₂ than in N₂ when temperatures exceeded 1000K. These results were confirmed by Al-Makhadmeh et al. [15]. On the other side, different studies show a minor or even a negative effect of CO₂ atmosphere on coal reactivity [16][17]. Studies on NO_x emission characteristics in coal-fired O₂/CO₂ combustion have been performed recently. Li [18], Duan [19], Daood [20] and their co-workers all reported that NO emissions was much lower in oxy-fuel combustion than in air combustion experimentally. While Dhungel et al. [21] examined NO emission behaviour during oxy coal combustion in a 20 kW electrically heated furnace and concluded that the pathways of NO reduction in oxy-fuel were fundamentally similar to those in air combustion. Mackrory et al. [22] investigated oxy-fuel combustion in a multi-fuel flow reactor (MFR) and found that oxy-fuel combustion can result in lower NO_x emissions than air combustion independent of the reduction of recycled NO_x, which was different from previous studies. Shao et al. [23] conducted a more detailed research about NO emissions in varied gas conditions. They found that the average NO_x concentration increased along with increasing oxygen concentration, and the maximum value and peak shape of the NO_x concentration curve changed with temperature. Specifically, the average NO_x concentration increased from 823 K to 923 K and declined from 923 K to 1223 K. NO_x emissions were measured to compare in a single particle combustion by Zhou et al [24]. They reported that the temperature produced little influence on the conversion of fuel-N, volatile-N and char-N to NO_x for anthracite.

Computational fluid dynamics (CFD), which can effectively integrate turbulent mixing, combustion chemistry, heat transfer and their interactions together has become a very powerful method to understand and simulate the combustion of coal. Kuffa et al. [25] successfully modelled coal gasification in a fluidized bed reactor. Oevermann et al. [26] established a model for simulating wood gasification in a fluidised bed reactor by using the Euler–Euler approach. Useful CFD analyses of oxy-fuel combustion have been successfully performed and reported in literature as well. CFD simulations of lab-scale oxy-coal combustion were carried out by Chui et al. [27] to assist with future pilot scale oxy-fuel combustion experiments and burner scale-up. Wang et al. [28] conducted a comprehensive CFD simulation of a propane-fueled, oxygen-enriched, turbulent, non-premixed jet flame. Zhou and Moyeda [29] performed CFD simulations for process evaluation of oxy-fuel combustion with flue gas recycle in a conventional utility boiler. However, fewer research focused on the whole combustion process of single coal particles, e.g. ignition characteristics, particle temperature, NO_x emissions. Moreover, attention on coal combustion under varied gas atmosphere need to be paid.

This paper aims at developing a combustion model considering the different gas–solid behaviours, heat transfer and thermal conversion processes for single coal combustion by

using Discrete Phase Model. The work is also concerned with formation of NO_x emissions when burning different kinds of coal under varying oxygen concentration with both N₂ and CO₂ as the background gas.

FUEL PROPERTIES

The proximate and ultimate analysis of the two kinds of coal is shown in Table 1 [1]. The diameter of particles is defined as 80 μm when carrying out numerical simulations as the size of 75-90 μm was used in the experiments [1].

Table 1 Coal Analysis Data

Fuel	PSOC-1451 Bituminous	DECS-11 Lignite
<i>Proximate analysis (received)</i>		
Moisture (%)	2.5	13.2
Volatile (%)	33.6	48.6
Fixed Carbon (%)	50.6	29.8
Ash (%)	13.3	8.4
<i>Ultimate analysis (dry basis)</i>		
C	71.9	66.2
H	4.7	4.0
O	6.9	18.6
N	1.4	0.9
S	1.4	0.7
Ash	13.7	9.6
Heating value dry fuel (MJ/kg)	31.5	25.7

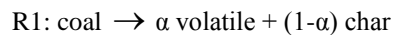
COMBUSTION MECHANISMS

Coal Combustion

Four chemical processes were considered during coal particles combustion: devolatilization, volatile (represented by CH_xO_y) combustion, char (represented by C_(s)) oxidation and other gas phase reactions.

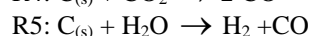
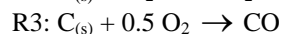
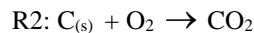
The particles are heated up and consequently they release moisture (drying process) at first, and then releasing volatiles rapidly (devolatilization process).

In this study, volatiles release is described by the single rate model. It assumes that the rate of devolatilization is first-order dependent on the amount of volatiles remaining in the particle and employs global kinetics. The reaction and its rate constant are:



$$k_d = A \exp(-E/RT)$$

Char formed during devolatilization process is consumed by heterogeneous processes of combustion and gasification and its combustion yields carbon monoxide (CO) and carbon dioxide (CO₂). Heterogeneous reactions can also include the char-H₂O reaction.



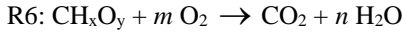
The combustion rate of char is assumed to be limited by the chemical kinetics because the only reactive species that is included in the gas phase is O₂. Then, the reaction rate K (kmol/m³s) is defined as

$$K = k_a c_{c(s)} c_{o_2}$$

and k_a is the reaction rate constant given by the Arrhenius type relation:

$$k_a = AT^b \exp(-E/RT)$$

In the gas phase reactions, as the detailed chemical species in the volatile are not completely understood due to the complexity of the chemical structure of coal, for simplicity in this study, it was generally treated as a single species which varies depending on the type of coal and comprising carbon, hydrogen and oxygen (CH_xO_y) in a ratio determined from the ultimate analysis of coal. For the two kinds of coal (PSOC-1451, DECS-11) used in this study, the volatile gas species are represented as $CH_{2.99}O_{0.24}$ and $CH_{1.48}O_{0.43}$ separately. The homogeneous reaction related to the volatile is as followed:

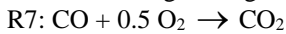


Thus, equating the numbers of atoms of each element in the reactants to the number in the products gives:

$$m = 1 + x/4 - y/2$$

$$n = x/2$$

And the carbon monoxide (CO) is oxidised to CO_2 according to the following homogeneous reaction:



The reaction rates are given as

$$K = AT^b \exp(-E/RT) [c_R]^d [c_{O_2}]^e$$

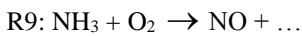
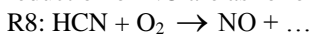
Six chemical reactions are considered totally and a summary of the kinetics data used in the present combustion modelling and the values of d and e are provided in Table 2.

Table 2 Kinetic constants

Reaction NO.	Kinetic parameters			d	e	Ref.
	A	E	b			
R1	3.12E+05	7.4E+07	-	-	-	[30]
R2	4.4	1.62E+08	1	-	-	[30]
R3	1.33	1.47E+08	1	-	-	[31]
R4	0.002	7.9E+07	0	-	-	[32]
R5	0.052	1.33E+08	0	-	-	[32]
R6	1.30E+11	1.26E+08	-	0.5	0.5	[33]
R7	2.119E+11	2.027E+08	-	0.2	1.3	[30]

NO_x Formation

The nitrogen forms NO_x emissions via intermediates HCN and NH_3 . The reactions related to NO formation and the reduction of NO are as follows:



The reaction rates are given by De Soete [34] as

$$K_8 = A_8 X_{HCN} X_{O_2}^a \exp(-E_8/RT)$$

$$K_9 = A_9 X_{NH_3} X_{O_2}^a \exp(-E_9/RT)$$

$$K_{10} = A_{10} X_{HCN} X_{NO} \exp(-E_{10}/RT)$$

$$K_{11} = A_{11} X_{NH_3} X_{NO} \exp(-E_{11}/RT)$$

The kinetic constants and the oxygen reaction order are shown in Table 3.

In this study, it is assumed that the fuel nitrogen is distributed evenly between the volatile and the char [35]. And the assumptions for partition of volatile nitrogen via the intermediates are: 50% HCN, 10% NH_3 and 40% NO [36].

NUMERICAL METHOD

The basic geometry used in this study is taken from Khatami et al [1,2]. An axisymmetric computational domain is shown in Figure 1. The coal particles are injected through the origin.

Table 3 Reaction parameters

Reaction NO.	Kinetic constants	
	A	E
R8	1.0E10	2.8E+08
R9	4.0E06	1.33E+08
R10	3.0E12	2.51E+08
R11	1.8E08	1.13E+08
<i>Oxygen reaction order</i>		
Oxygen mole fraction		<i>a</i>
$X_{O_2} \leq 4.1E-03$		1
$4.1E-03 \leq X_{O_2} \leq 1.11E-02^2$		-3.95-0.9ln X_{O_2}
$1.11E-02 \leq X_{O_2} \leq 0.03$		-0.35-0.1ln X_{O_2}
$X_{O_2} \geq 0.03$		0

The numerical simulations are performed using an Euler-Lagrange model. The weighted-sum-of-gray-gases model (WSGGM), single-rate devolatilization model and multiple-surface-reactions combustion model (e.g., char- O_2 , char- CO_2 , and char- H_2O) are used for the modelling. The flow field was solved using the SIMPLE algorithm for velocity-pressure coupling. Gravity is enabled as well when running these numerical simulations. NO_x formation is performed with thermal NO and fuel NO sub-models. The simulations are run on ANSYS FLUENT.

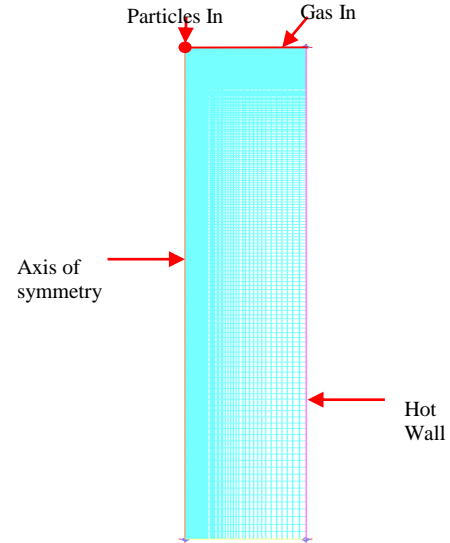


Figure 1 2D axisymmetric geometry with mesh

The coal particle combustion experiments are conducted under a quiescent gas condition (inactive flow, i.e., no injector flow and no furnace flow) and quiescent gas condition is set by turning off the gas flows a few seconds prior to the particle injection [2]. While running numerical simulations, gas injection stops when enabling DPM to inject coal particle. The operating conditions refer to the conducted experiments in ref. [1,2], and are shown in Table 4. Some assumptions are made when running

simulations: 1) the coal particles are regarded as spherical in shape and 2) the gas phase are treated as an ideal-gas mixture.

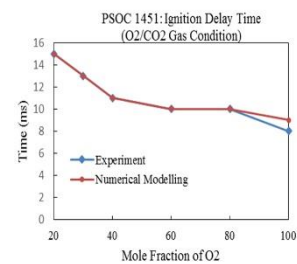
Table 4 Operating Conditions

Parameters	Values
Wall Temperature	1400 K
Temperature of injecting gas	1200 K
Velocity of gas	4.55 cm/s
Diameter of particles	80 μm
Initial temperature of particles	1200 K
Velocity of particles	15 cm/s

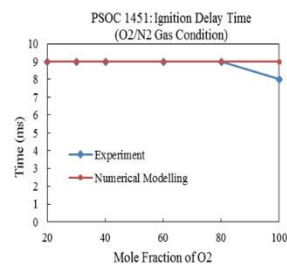
RESULTS

Validation of the Numerical Simulation

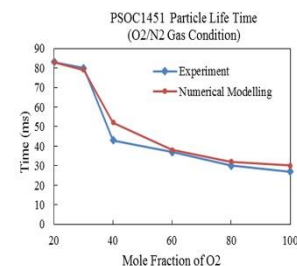
Particle ignition delay time, particle life time (sum of particle ignition delay and burnout time) and particle temperature of the two kinds of coal (PSOC-1451 and DECS-11) under quiescent O_2/N_2 and O_2/CO_2 atmospheres with oxygen mole fraction varying from 20% to 100% were measured in the experiments of Khatami et al. [1][2]. Numerical simulations of coal particles combustion were run and data was obtained to compare with experimental one.



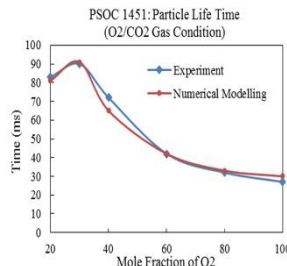
(a) Ignition Delay Time O_2/N_2



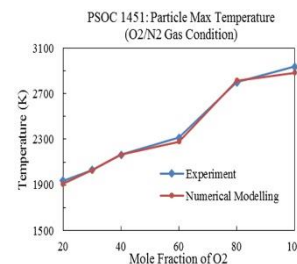
(b) Ignition Delay Time O_2/CO_2



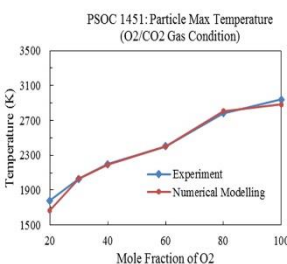
(c) Particle Life Time O_2/N_2



(d) Particle Life Time O_2/CO_2

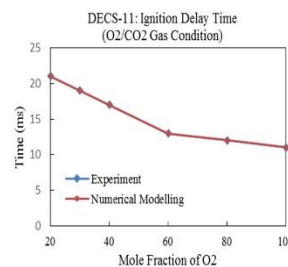


(e) Particle Temperature O_2/N_2

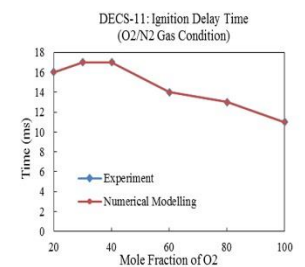


(f) Particle Temperature O_2/CO_2

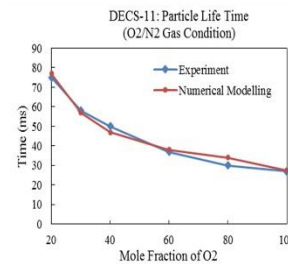
Figure 2 Compared particle ignition time, particle life time and particle temperature versus varied oxygen concentration under both N_2 and CO_2 background gas for coal PSOC-1451.



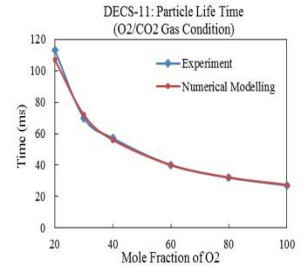
(a) Ignition Delay Time O_2/N_2



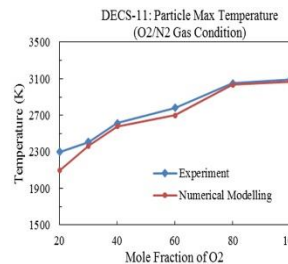
(b) Ignition Delay Time O_2/CO_2



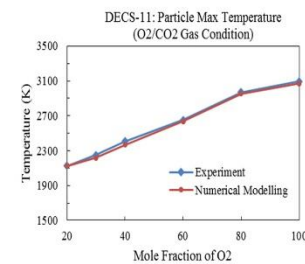
(c) Particle Life Time O_2/N_2



(d) Particle Life Time O_2/CO_2



(e) Particle Temperature O_2/N_2



(f) Particle Temperature O_2/CO_2

Figure 3 Compared particle ignition time, particle life time and particle temperature versus varied oxygen concentration under both N_2 and CO_2 background gas for coal DECS-11.

Figures 2(a) and 2(b) show a comparison between the simulated particle ignition delay time and the corresponding experimental data for coal PSOC-1451. The figures show that, apart from the pure oxygen case, the numerical results in other cases spot on the experimental data. Figures 2(c), 2(d) and Figures 2(e), 2(f) give the information of particle life time and particle temperature versus varied oxygen concentration under both N_2 and CO_2 background gas coal PSOC-1451. Once again, it is shown that the numerical results match the experimental data well.

Figures 3(a)-3(f) present the same compared information of coal DECS-11. The ignition delay time is spotted nicely according to Figures 3(a) and 3(b). Figures 3(c)-3(f) show that the numerical results of particle life time and particle temperature match the experimental data only with slight errors.

It is clear from the results shown in Figures 2 and 3, that there is a good agreement between the results of numerical simulations and experimental data. Hence, it can be concluded that the numerical model is valid.

Evolution of Chemical Species Concentration

Burning of a single particle in the experiments [1][2] resulted in very small amount of chemical species. Experimental measurement of these amounts is extremely hard. Yet, without knowing the evolution of chemical species concentration, our knowledge of combustion process will be incomplete. To address this, numerical simulations can be utilised to predict the details of the single coal combustion process. The mass-averaged mole fraction of selected species volatile (hv_vol), H₂O, CO, and H₂ versus time after particle release of coal DECS-11 particle burning in 20% O₂ and 80% N₂, 100% O₂, 40% O₂ and 60% CO₂ gas atmosphere are shown in Figure 4-6.

When the coal particles combust in O₂/N₂ gas atmospheres (Figure 4), the volatile appeared since the coal particle was injected into the reactor and the amount kept increasing till the peak. Further, the volatile reacted with oxygen to produce H₂O and CO₂. When all the volatile was consumed completely, the amount of H₂O became stable. The species CO, H₂ appeared when the coal particle was ignited. The amount of H₂ kept increasing during the coal combustion process and then became stable when the coal particle burned out. The amount of CO began decreasing when the rate of consuming CO was higher than producing CO, and finally all CO turned to CO₂. The changing trends of these species meet the combustion mechanism well.

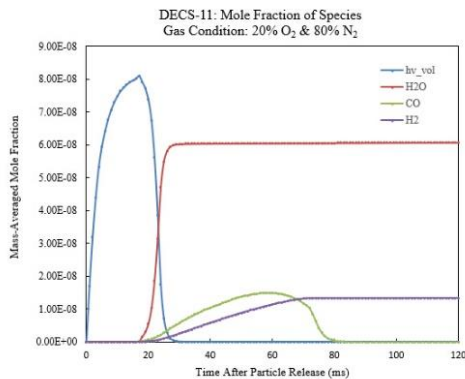


Figure 4 Mass-averaged mole fraction of selected species (hv_vol, H₂O, CO, H₂) versus time after particle release of coal DECS-11 particle burning in 20% O₂ and 80% N₂ gas atmosphere.

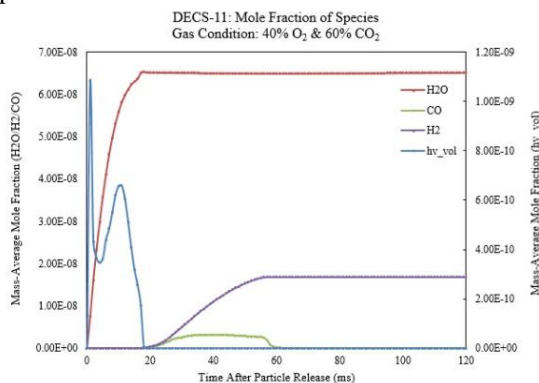


Figure 5 Mass-averaged mole fraction of selected species (hv_vol, H₂O, CO, H₂) versus time after particle release of coal DECS-11 particle burning in 40% O₂ and 60% CO₂ gas atmosphere.

When the coal particles burn in O₂/CO₂ gas atmospheres, the amount of the volatile firstly accumulated to one peak until the volatile reacted with oxygen and some was consumed, and then the release of volatile and depletion of volatile continued until volatile was completely consumed. During the competitive process of formation and consumption of volatile, the second peak, in Fig. 5, formed. For other species, H₂O kept increasing until the volatile combust totally, and H₂ appeared when the particle was ignited and reached maximum when the particle burnt out, and CO produced during coal combustion reacted with oxygen and formed CO₂ at last.

In the experiments of Khatami et al [37], the volatile and char burnt sequentially (two-mode combustion) or overlapped (one-mode combustion) under different gas atmosphere. Nonetheless, the full reproduce of the combustion mode need to be further explored.

NO_x formation

NO_x concentration was not measured during experiments. Based on the valid numerical model, the predictions of NO emissions of both two kinds of coal in different oxygen mole fraction gas atmosphere were done by numerical simulations. Results are presented in Figure 6 and Figure 7. The two kinds of coal shared the same trends for all runs.

As only one single particle is burned in each case, the NO_x emissions produced by fuel-NO_x is much lower than thermal-NO_x. Therefore, the NO_x concentration is lower when coal particles burned under O₂/CO₂ gas conditions than O₂/N₂ gas conditions.

When the coal particles combust in O₂/N₂ gas conditions, the NO_x concentration increases when the mole fraction of oxygen increases from 20% to 30% and 40%. However, when the mole fraction of oxygen exceeds 50%, the NO_x concentration decreases when the mole fraction of oxygen increases. While the coal particles combust in O₂/CO₂ gas conditions, the NO_x concentration keeps decreasing when increasing the mole fraction of oxygen. NO_x emissions formed from fuel-NO_x are greatly influenced by fuel temperature and burnout time. Therefore when the particle life time decreases and particle temperature increases along with increscent oxygen concentration, the NO_x concentration descends when combusting in O₂/CO₂ gas conditions.

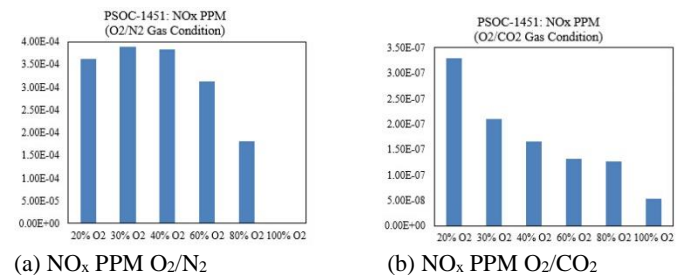


Figure 6 NO_x PPM for coal PSOC-1451 at T = 200 ms (Time After Particle Release).

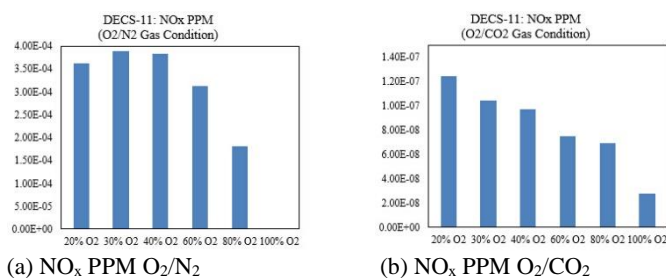


Figure 7 NO_x PPM for coal DECS-11 at T = 200 ms (Time After Particle Release).

CONCLUSIONS

Combustion of single coal particles (PSOC-1451 and DECS-11) was modelled numerically, existing experimental data were used to validate the simulation. The predicted results for the two kinds of coal featured a good agreement with the experimental data in terms of the particle ignition time, particle life time and particle temperature versus varied oxygen concentration under both N₂ and CO₂ background gas. Further, detailed changing trends of gas species were investigated in this study. Hence, the established model worked successfully and could be a useful predictive tool for coal combustion.

The NO emissions of both two kinds of coal in different oxygen mole fraction gas atmosphere were predicted. The two kinds of coal shared the same trends for all conducted simulations. The NO_x concentration was lower when coal particles burned under O₂/CO₂ gas conditions than O₂/N₂ gas conditions. When the coal particles combust in O₂/N₂ gas conditions, the NO_x concentration increased along with increasing oxygen concentration when the mole fraction of oxygen below 40%, and decreases when the mole fraction of oxygen exceeds 60%. On the other hand, the coal particles combusted in O₂/CO₂ gas conditions, the increase of the mole fraction of oxygen led to the decrease of NO_x concentration.

ACKNOWLEDGEMENT

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