Numerical Investigation on Kinetic Parameter of Coal Devolatilization Reaction by Identifying Ignition Delay Parameter

Tata Sutardi, Manosh C. Paul, and Nader Karimi

Abstract – Identifying the difference of ignition delay time between bituminous and lignite coals is the main aim of this research. Finding the reaction mechanisms affecting this behaviour is a key part of the investigation. Seven reaction mechanisms are used to represent coal combustion at a particle level, and this paper principally focuses on the devolatilzation process reaction for predicting the ignition delay time of coal particle combustion. Two types of coal namely PSOC 1451 and PSOC 1443 are examined numerically, and results are compared with the experimental data. Existing kinetic parameters for the devolatilization reaction R1 **(Coal → Coalvolatile + char) underestimates the ignition delay time which is largely influenced by the value of the pre-exponent factor (*A*) of** R1**. Results giving the best agreement with the experiment are obtained with A= 3.12 x 10^5 and 9.36 x 10^7 for PSOC 1451 and PSOC 1443, respectively.**

Keywords: Combustion, devolatilization, coal particle simulation, kinetics parameter.

I. INTRODUCTION

Among all the different types of fossil fuels, coal has the largest global reserve according to the study reported in 2016 [1]. By sharing 29.2% (consisting of 27.5% hard coal, and 1.7% lignite) of the global Prime Economic Contribution (PEC), coal was the second most important energy resources in 2015 after the crude oil [2]. Further, coal accounted for 40% of the total electricity generation in 2012 and also predicted to be the highest contributor until 2025 [3].

Based on the types, coal is classified as lignite, sub-bituminous, bituminous and anthracite. These varieties descended from the origin of coal formation: the creation of peat or partial-decomposed plant materials [4]. Increased heat and pressure from overlaying strata produced higher rank coal. Lignite, a brown-black coal with high-moisture, high ash contents and low heating value, is the lowest rank coal. The higher rank coal is sub-bituminous, and then bituminous coal which has a higher heating value, less moisture and ash content than other coal types [5]. Many studies have been performed to investigate the combustion behaviour for each coal type [6], since each of them has the uniqueness in their performance of combustion. The recent studies exhibit this topic in various manners, either through experiment [7] or numerical simulation [8], with an aim of better understanding as well as characterising the processes of coal utilization.

The characteristic of ignition delay time is an important parameter for designing coal combustion systems. It has significant roles in the prevention of spontaneous ignition and in the production of stable flame [9]. Experimental study of Levendis et al. [5, 10] on coal particle combustion reported that the lower rank coal has the shorter ignition delay time (t_{id}) compared to a higher rank coal [5]. This result generally agrees well with the studies of several other authors [9, 11, 12], and further indicates that the ignition delay time increases from a lower to higher rank coal. Additionally, Ref [13] reported that the lignite coal is more reactive than other types of coal.

The ignition delay is a lapse of time between the injection of coal to a combustion chamber and when the combustion process begins. However, devolatilization reaction of coal initiates the process of combustion [14, 15], therefore potentially links with the ignition delay. Other study also mentioned that the coal volatile is typically responsible for the flame ignition and thus has large impacts on the overall combustion characteristics [16]. More specific on the process of devolatilization, these studies [16-19] are most relevant. Generally, two different methods for determining the devolatilization process of fuels were implemented: constant temperature or at constant heating rate [20]. The study of Levendis et al. [5, 10], as aforementioned, is in line with the constant heating rate and at almost homogenous temperature.

Numerical study of bituminous coal particle combustion has been performed recently in our group [21-23]. The numerical model has been validated with the experimental study of Levendis et al. [5], specifically based on the results of the ignition delay time (t_{id}), char burnt out time (t_{char}), maximum temperature of coal volatile combustion (T_{cv}), and maximum char temperature (T_{char}) [23, 24]. This paper particularly aims at the investigation of devolatilization reaction and how it influences the ignition delay time. A comparison of the ignition delay time will also be made between bituminous and lignite coals, representing respectively a high and low rank coal since they have significant different chemical compositions. Result expected could give better understanding of the devolatilization reaction for further modelling application.

II. MODEL DEVELOPMENT

A. Reaction Mechanisms

Computational model of coal particle combustion in a drop tube furnace (DTF) was developed in the previous study [21-23]. The physical geometry of the DTF is illustrated in Fig. 1 [5]. It is represented by a cylindrical shape geometry as illustrated in Fig. 1(a), with the inlet diameter of 7 cm, and the hot wall furnace length of 25 cm from the inlet. The coal particle injection starts from the centre of the inlet. The axi-

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symmetric model with a grid distribution used for the simulation can be seen in Fig. 1(b).

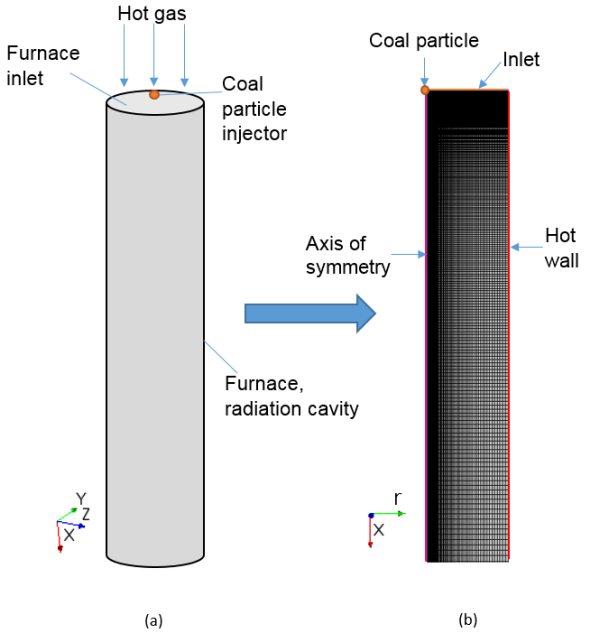

Fig. 1.An illustration of the geometry model, (a) cylindrical shape and (b) axi-symmetric model with grid

The simulation procedures are applied based on the experimental study of [5], and the reaction mechanisms are defined as shown in TABLE I [25].

TABLE I
COAL COMBUSTION AND GASIFICATION REACTIONS [25]

No	Mechanism	Enthalpy (kJ/mol)
R1	Raw coal → YY Coal volatile + (1-YY) Char	
R2	C + O_2 → CO_2	-393
R3	C + 0.5O_2 → CO	-111
R4	C + CO_2 → 2CO	+172
R5	C + H_2O → CO + H_2	+131
R6	Coal Volatile + O_2 → CO_2 +H_2O + N_2	
R7	CO + 0.5O_2 → CO_2	-283

B. Governing Equations

The mechanisms of coal particle conversion / interaction with gas inside the reactor are described through the several equations as follows [26].

The continuity equation of raw coal component in particle is described as

$$\frac{dm_{cp}}{dt} = -R_{cp}\,, \tag{1}$$

where the net rate for raw coal consumption is given by

$$R_{cp} = k_1 \alpha_{cp} m_p\,. \tag{2}$$

And the rate of production for coal volatile is described as

$$R_{cv} = k_1 YY \alpha_{cp} m_p\,. \tag{3}$$

Then, the reaction rate is represented by the Arrhenius equation, as follows

$$k_1 = AT^{\beta} exp\left(\frac{-E_a}{R_c T}\right). \tag{4}$$

Particle and gas reactions begin after the volatile fraction of raw coal particle completely evolved. This is a heterogeneous reaction, and the reaction rate is determined by combining the effect of the Arrhenius rate and diffusion coefficient. The model of particle rate consumption is then determined by

$$R_p = \frac{dm_p}{dt} = -\frac{k_m k}{k+k_m} \emptyset C_g M_w A_p\,, \tag{5}$$

where,

$$k_m = \frac{(S_h)(D_m)}{d}\,. \tag{6}$$

The reaction rate between gases (i.e. homogeneous reactions) is a function of the composition and rate constant, given by the expression:

$$R_j = R_{i,kin} = -k_j \prod_{all\ reactants} \left(\frac{\rho Y_i}{M_i}\right)^{p_{ij}}. \tag{7}$$

The equation of motion for the particle is defined as,

$$m_p \frac{d\overline{u_{i,j,k}^{p}}}{dt} = \sum \bar{F}. \tag{8}$$

The effect of gravity force is included in this simulation since these forces influence the parameter of investigation.

In the reacting flow, the changes of pressure, temperature, velocity, and species concentration are the results of the interaction among the fluid flow, molecular transport, heat transfer and chemical reaction. In order to consider these effects on the simulation models, a set of mathematical modelling, which consists of the Navier–Stokes, mass continuity, species mass conservation and energy conservation equations, is developed.

C. Boundary Conditions and Results

In the previous study, a type of bituminous coal namely PSOC 1451 was used [22-24]. Another type of coal (PSOC 1443) representing a lignite coal is included in this study. The chemical properties of these coals are presented in TABLE II. [5].

TABLE II.
COAL CHEMICAL COMPOSITIONS [5]

	PSOC 1451	PSOC 1443
Proximate Analysis as received		
Moisture (%)	2.5	18.6
Volatile matter (%)	33.6	50.3
Fixed Carbon (%)	50.6	13.7
Ash (%)	13.3	17.4
Ultimate Analysis (dry basis)		
Carbon (%)	71.9	56.8
Hydrogen (%)	4.9	4.1
Oxygen (%) (by diff.)	6.9	15.8
Nitrogen (%)	1.4	1.1
Sulphur (%)	1.4	0.7
Sodium (%)	0.06	0.04
Ash (%)	13.7	21.4
Heating value dry fuel (MJ/kg)	31.5	23.0

The initial boundary conditions were taken from the experimental study of [5, 27]. The furnace was heated up with hot air before the injection of the coal particle. The inlet condition was set as a velocity inlet, with an initial

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temperature of hot air of 1200K, and at the same time, the furnace wall temperature was set at 1400K. The inlet air with a velocity of 0.045 m/s was injected through the furnace's inlet until the flow became fully developed. Additionally, to accommodate the full development region, the furnace wall was extended to 75 cm and it was set as an isolator. The coal particle combustion simulations are conducted under a quiescent gas condition (inactive flow) and the quiescent gas condition is set by turning off the gas flows a few seconds prior to the particle injection.

The temperature profile of coal particle - PSOC 1451 is obtained and compared with that of the experiment study [23].

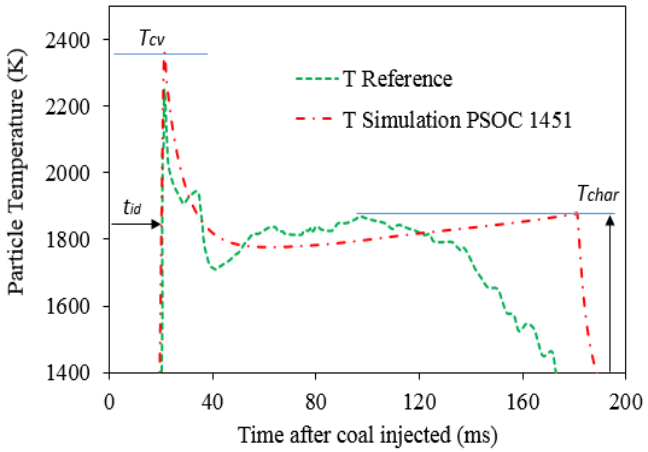

Fig. 2. Result of simulation compare to experimental

Fig. 2 shows the parameters of t_{id}, T_{cv}, and T_{char} for results of simulation and experiments. It is reported in the previous study [23, 24] that they indicate the good agreement between the experimental and simulation result. This agreement meets when the simulation used the set of kinetic parameter form each reactions of TABLE I, which is taken from several sources. These kinetic parameter value can be seen in TABLE III.

TABLE III
THE KINETIC PARAMETER VALUE

No	Kinetic parameters			Ref.
	A **(unit vary)**	***Ea*** **(j/kmol)**	***β***	
R1	3.12E+05	7.40E+07	0	Alganash et.al [28]
R2	0.002	7.90E+07	0	Alganash et.al [28]
R3	85500	1.40E+08	0.84	Watanabe et.al [29]
R4	4.4	1.62E+08	1	Alganash et.al [28] & Silaen [30]
R5	1.33	1.47E+08	1	Alganash et.al [28], Silaen [30], Howard [31]
R6	2.12E+11	2.03E+08	0	Alganash et.al [28]
R7	1.30E+11	1.26E+08	0	Alganash et.al [28], Howard [31]

The set of kinetic parameters outlined in TABLE III provides an important information for this study. This information can be used to identify the reaction rate of each species based on the reaction defined. Reaction rate has an effect on the time of chemical species reacting and forming to be a new species as products [21]. Therefore, this study associates the ignition delay time and kinetic rate of reaction, and moreover on the reaction rate of devolatilization. The results from the experimental study from Levendis at al. [5], showed the ignition delay time of bituminous coal (PSOC 1451), t_{id} is ~20 ms after coal injection, while for lignite coal (PSOC 1443), t_{id} is ~10 ms [5]. The ignition delay of lignite coal is shorter than that of bituminous coal. This difference is investigated through the numerical simulation, by correlating the kinetic reaction of devolatilization process and the ignition delay time.

III. MODEL APPLICATION FOR THE LIGNITE COAL

By using the same procedures, the combustion model of lignite coal (PSOC 1443) in the DTF reactor is developed and the ignition delay time between the results of simulation and experiment is assessed. The devolatilization reaction process is simulated initially with the kinetic parameters of R1 in TABLE III. The model simulation allows the process of devolatilization to be simulated either by including or excluding the process of combustion of coal volatile species. Therefore, the devolatilization process can be simulated independently from the other reactions, or even simultaneously with the other reactions. For identification, the simulation process of PSOC 1443 (lignite coal) combustion with the kinetic parameters in TABLE III, is named as Simulation A. Other simulations named accordingly as Simulation B, C and D are developed as a part of the investigation to identify the effects of the kinetic reaction of devolatilization on the ignition delay. The simulation results of the model devolatilization process for each simulation can be seen in Fig. 3.

Fig. 3 shows the process of devolatilization in terms of the coal volatile fraction profile. Fig. 3(a) presents the devolatilization process without volatile combustion while (b) with combustion. The devolatilization process of Simulation A lasts between ~20 and ~40 ms, with the most rapid coal volatile release occurring at ~30 ms as seen in Fig. 3(a). If it is performed with combustion, as in Fig. 3(b), the peak of coal volatile profile occurs also at ~30 ms, but then it goes down, which indicates its burning out. However, the coal volatile combustion initiates the combustion of coal particle, so at the time when the most rapid combustion occurred, the temperature of the coal particle increases rapidly and initiates its burning. The period between the particle injection and the particle start burning is the ignition delay time. Therefore, the ignition delay of Simulation A is determined as ~30 ms after the coal injection. But this result does not agree with the experiment [5, 32], and therefore Simulation B, C and D are developed by systematically increasing the pre-exponent factor (A). Note that the reactor condition is same for each simulation (heat rate and temperature), so the activation Energy (E_a) and temperature exponent (β) are assumed to be the same. The value of the pre-exponent factor of Simulation B, C, and D, is increased 10, 100 and 300 times that of Simulation A, respectively. Finally, the results indicate that the best fit result of the ignition delay time is that obtained by Simulation D. The Simulation D takes for ~10ms, which agrees well with the ignition delay time for the lignite coal PSOC 1443 in the experiment [5]. It thus further indicates that the kinetic parameter value of Simulation D is suitable for the lignite coal combustion.

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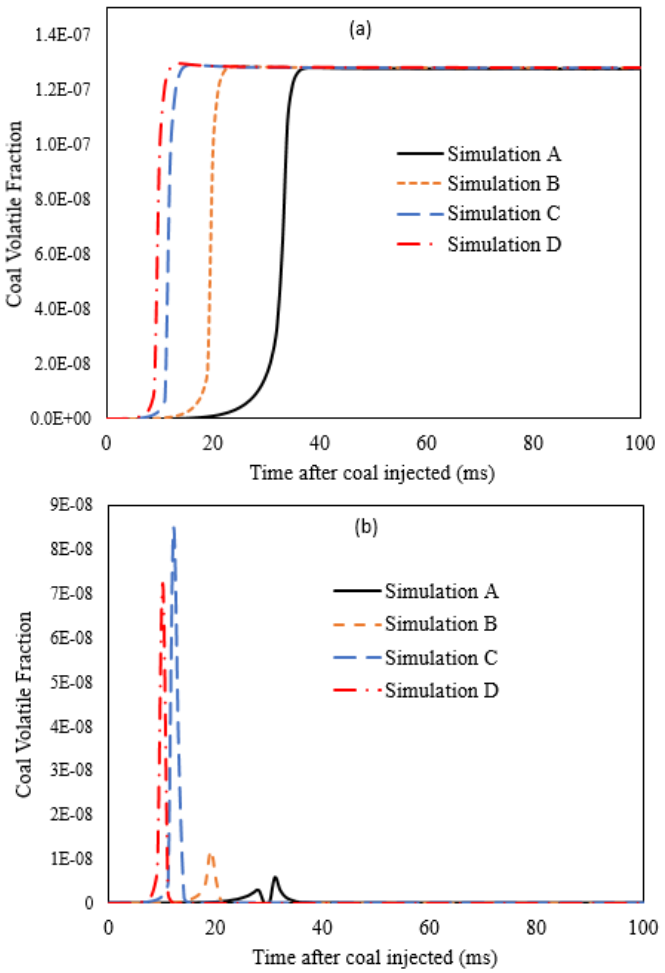

Fig. 3. Coal volatile release for each condition (a) without combustion, and (b) with combustion

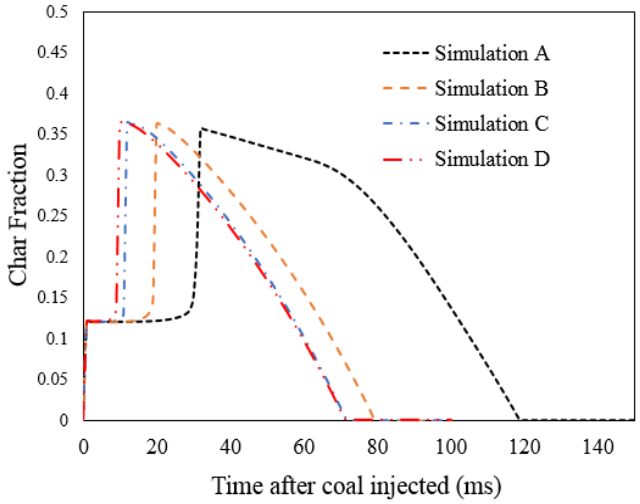

Fig. 4. The char profile based on pre-exponent factor of variation

Fig. 4 shows the char fraction profile of coal combustion of each simulation. Initially the volatile release, at the rapid increment of char fraction, indicates the rapid release of coal volatile from the coal particle. At this condition the combustion starts, and the period of ignition delay occurred. The combination of coal volatile and char profile can also be used to further validate the simulation results with the experiment. A comparison of the temperature profile of coal particle is presented in Fig. 5.

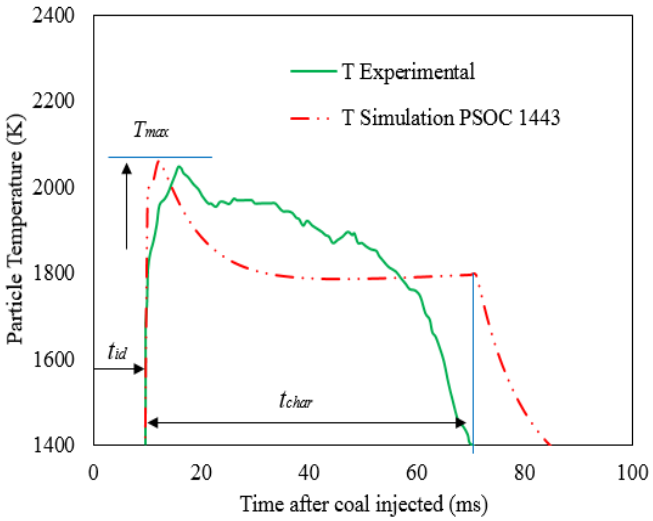

Fig. 5. The comparison of Simulation and experimental result

The maximum temperature, ignition delay time and total burnt out time are compared with the experimental data in Fig. 5. These show having a good agreement between them, thus further confirms that the value of kinetic parameter identified for the lignite coal – PSOC 1443 combustion is realistic.

TABLE IV
COMPARISON RESULT

PSOC 1443	Max Temperature (K)	Ignition delay (ms)	Total Burn out (ms)
Experimental	2000	10	72
Deviation [5]	93	-	15
Simulation	2042	10	71

IV DISCUSION

Increasing the pre-exponent factor (*A*) of R1 300 times the value of the previous study provides a good correlation for the ignition delay with the experimental result. This result also agrees with some other studies that stated that the coal volatile burn out time is a function of the coal type [20, 33, 34]. During the rapid devolatilization, bituminous coals are known to produce an abundance of light and heavy hydrocarbons while lignite produces mostly CO, CO_2, H~O, H_2 and light hydrocarbon gases [18]. This also supports the reason of the ignition delay time for lignite coal which is shorter than the bituminous coal.

When the combustion is represented by its char profile, as seen in Fig. 4, it can be seen that the ignition delay as the time after coal injection to the time at the char fraction increases rapidly. After the coal volatile burnt out, the char reaction occurred which is indicated by the decreasing fraction. The coal burn out time is achieved from the time at coal injected to the time at the char fraction burnt out.

The kinetic parameter of devolatilization reaction (R1) has important role to simulate the ignition delay time of coal combustion. There is a different value of kinetic parameter of R1 for bituminous and lignite coal. In this model, the value of kinetic parameter lignite coal is higher than the value of kinetic parameter bituminous coal. It is affirming the role of devolatilization process, resulting in the ignition delay of coal

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combustion. In this model simulation, the difference between the bituminous and lignite coals on the kinetic reaction is the value of kinetic parameter of R1.

VI CONCLUSION

The single coal particle model of combustion has been developed to investigate the effect of devolatilization reaction on the ignition delay of bituminous and lignite coal combustion.

Based on the numerical investigation, the ignition delay of coal combustion is most affected by the devolatilization reaction.

It has been identified the best fit kinetic parameter for PSOC 1451 and PSOC 1443 coal, and these value can be considered for further investigation of these types of coal.

NOMENCLATURE

Roman Symbol

A	Pre- exponential factor (unit vary)
A_p	Surface area of particle (m^2)
C_g	Reactant gas concentration (kmol/kg)
C_i	Concentration of species (kg/m^3)
D_m	Diffusion coefficient (m^2/s)
E	Energy sources (J)
E_a	Activation Energy (J/kmol)
F	External force (N)
g	Gravity (m/s^2)
G_k	Generation of turbulence kinetic energy due to buoyancy
M_i	Molecular weight of species *i*
M_w	Molecular weight of solid reactant
R_c	Gas universal constant (J/kmol K)
Y_i	Mass fraction of species *i*
k	Kinetic energy dissipation
k_i	Kinetic rate coefficient for *i*
k_m	Mass transfer coefficient
m	Mass fraction
R_i	Rate exponent of reacting species
h	Enthalpy (kJ/kg)
J_i	The flux of species *i*
S_m	Source of mass (kg)
S_h	Sherwood number
T	Temperature (K)
YY	Mass stoichiometric coefficient
M	Mass of particle (kg)
p	Pressure (Pa)
r	Radial displacement (m)
$C_{\varepsilon 1};\ C_{\varepsilon 2}$	Model constant
t	Time (s)
x	Axial displacement (m)
u	Velocity (m/s)
t_{id}	Ignition delay time
t_{cv}	Coal volatile burnt out time
t_{char}	Char burn out time
T_{cv}	Maximum temperature coal volatile combustion (K)
T_{char}	Maximum temperature char combustion (K)

Greek Symbol

α_i	Mass fraction of coal/particle component
β	Temperature exponent
τ_{ij}	Stress tensor
$\emptyset$	Ratio of stoichiometric of solid and gas
ρ	Density (kg/m^3)
ρg_i	Gravitational body force
μ	Viscosity (kg/m.s)
σ	Turbulent Prandtl number
δ	Kronecker delta

Subscript

p	Particle
c	Coal component
i, j	Species or phase
t	Turbulent
vm	Volatile matter
k	Turbulent kinetic energy (m^2/s^2)
ε	Turbulent dissipation rate (m^2/s^3)

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