Numerical investigation of pyrolysis of a Loy Yang coal in a lab-scale furnace at elevated pressures

James Hart, Audai Hussein Al-Abbas & **Jamal Naser**

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Numerical investigation of pyrolysis of a Loy Yang coal in a lab-scale furnace at elevated pressures

James Hart • Audai Hussein Al-Abbas • Jamal Naser

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Abstract A computational fluid dynamics (CFD) model of the pyrolysis of a Loy Yang low-rank coal in a pressurised drop tube furnace (pdtf) was undertaken evaluating Arrhenius reaction rate constants. The paper also presents predictions of an isothermal flow through the drop tube furnace. In this study, a pdtf reactor operated at pressures up to 15 bar and at a temperature of 1,173 K with particle heating rates of approximately 105 K s-1 was used. The CFD model consists of two geometrical sections; flow straightner and injector. The single reaction and two competing reaction models were employed for this numerical investigation of the pyrolysis process. The results are validated against the available experimental data in terms of velocity profiles for the drop tube furnace and the particle mass loss versus particle residence times. The isothermal flow results showed reasonable agreement with the available experimental data at different locations from the injector tip. The predicted results of both the single reaction and competing reaction modes showed slightly different results. In addition, several reaction rate constants were tested and validated against the available experimental data. The most accurate results were being Badzioch and Hawksley (Ind Eng Chem Process Des Dev 9:521-530, 1970) with a single reaction model and Ubhayakar et al. (Symp (Int) Combust 16:427-436, 1977) for two competing reactions. These numerical results can provide useful information towards future modelling of the

A.H. Al-Abbas

Foundation of Technical Education, Al-Musaib Technical College, Babylon, Iraq

behaviour of Loy Yang coal in a full scale tangentially-fired furnace.

Abbreviations

- V Mass of volatiles
- V_f Final yield of volatiles
- kv Arrhenius rate constant
- Av Pre exponential factor
- Ev Arrhenius activation energy
- T_P Particle temperature
- Y_{1,2} Reaction yields for two reaction model
- k_{1,2} Arrhenius constants for two reaction model
- Co Mass raw coal
- Mv Mass of volatiles evolved
- HODS Higher order differencing scheme

1 Introduction

The importance of the nature of pyrolysis in the liquefaction, gasification, and combustion of coal has become more significant in recent years [1, 2]. The efficiency of the higher technology conversion methods, including pressurized entrained flow gasification and catalytic coal combustion, is expected to be enhanced through a more complete understanding of the initial pyrolysis step. Currently, there is relatively little available basic data (to allow modelling) of the reactions undergone by Australian low rank coal during pyrolysis and subsequent gasification at high pressures. An understanding of the initial pyrolysis processes and reactions that coal undergoes at high pressures will contribute to the maximisation of the efficiency of these new technologies [3, 4].

J. Hart · J. Naser (🖂)

Faculty of Engineering and Industrial Science, Swinburne University of Technology, Hawthorn, VIC 3122, Australia e-mail: Jnaser@swin.edu.au

Coal pyrolysis may be defined a set of physical and chemical processes, which break bonds within the coal structure to release and produce volatiles. The amount of volatile matter released and the proportion of each of the volatile species present depends on the pyrolysis conditions and the coal rank used in pyrolysis [5]. The products of pyrolysis are char (solid), tar (liquid), and gases. The liquid and gas phases are together known as volatiles. The volatiles are a hydrogen rich fraction that contains low molecular weight compounds such as hydrogen, oxides of carbon, methane, etc., in the gaseous phase, and higher molecular weight hydrocarbons such as light oils in the liquid phase [6].

Pyrolysis temperature determines the yield distribution of products through the primary decomposition of the coal as well as through any subsequent secondary reactions. Studies of the weight loss from coal as a function of time at various temperatures by a number of researchers including Kimber and Gray [7], Kobayashi et al. [8], Anthony et al. [9], and Suubery [10] give the same general result: a rapid rise in weight loss followed by an asymptotic levelling to an equilibrium mass of sample. In the range of temperatures typically examined in pyrolysis studies, (837 to 1,373 K [8]), the position of this asymptote increases with increasing temperature for low rank coal [8]. Other research carried out at increased heating rates and temperatures confirm this result [6–8].

Anthony et al. [9] and Suuberg et al. [10] performed experiments under a wide range of heating rates (from 270 to 10^4 K s⁻¹) utilising an electrically heated wire grid reactor. They determined that no heating rate effects occurred in their experiments. The authors suggested that when pyrolysis experiments are carried out in different types of reactors, factors such as changes in the mode of mass transport out of and away from the hot char surface, which affects secondary reaction processes be considered. As a result, the heating rate is not the overwhelming variable affecting pyrolysis.

Tsai [11] conducted some ways to decouple these processes by developed mathematical models to estimate the time temperature history of pyrolysis coal particles. Results of the numerical calculations suggest that the temperature of coal particles heated mainly by convection, follows that of the surrounding gas if their size is less than 100 μ m. The heating rate of these particles was shown to be controlled primary by the heating rate of the carrier gas, and is dependent upon the mixing and thermal diffusivity of the gas and the coal feed rate.

Brown coal fired boilers are typically large and complex in their behaviour. It is desirable to have an accurate CFD model of such a furnace in order to economically investigate design changes for more efficient operation. A major part of any CFD modelling in these furnaces is the coal combustion model, which consists of two parts; predicting pyrolysis rates, or the rate at which volatiles are driven from the coal particles being burned, and the char combustion reaction rates. Together with the homogeneous gas phase reactions, these make up a CFD model of combustion of coal particles.

It is often difficult to validate a CFD model against data from a large furnace, firstly because good quality data is often not available, and secondly because there are many factors effecting combustion behaviour in a furnace, such as aerodynamic effects from burner geometry [12–14], radiation heat transfer [15, 16] and complex single phase [17] and multi-phase chemical kinetics [18–21]. To have confidence in a CFD model, it is useful to be able to check the accuracy of each part of the model separately, and then bring them all together in a large furnace model.

Modelling combustion and pyrolysis in the pdtf gives the opportunity to test the reaction kinetics of these two phenomenons without the added difficulty of complex geometry and aerodynamic flow conditions. The pdtf was designed for experimental modelling of coal combustion, and the same advantages were exploited in the present CFD modelling. In the modelling of the present pdtf we are able to predict the pyrolysis reactions of coal particles in a relatively simple geometry.

To the best of our knowledge, there is little modelling research work conducted on the pyrolysis process of the Loy Yang Australian coal in a pdtf reactor at high pressures. The present work is a CFD simulation of the experimental investigation carried out by Marney [6]. He investigated devolatilization of a Loy Yang coal in a pressurised drop tube furnace, under nitrogen atmosphere to eliminate the possibility of combustion. The paper also presents the simulation of isothermal flow through the drop tube furnace. The predicted results are validated against the available experimental data.

2 CFD model geometry

The pdtf can be thought of as consisting of two section; an upper section where the bulk of the fluid is heated and passed vertically through a flow straightener. The straightener flow then moves to the lower section of the furnace, the injector section. Here particles are injected with a small amount of carrier gas into the main stream, where they are heated by the surrounding fluid and undergo pyrolysis.

Shown in Fig. 1 is a diagram of the CFD geometry used to model the flow straightener section, and Fig. 2 shows the injector section. The geometry and operating conditions used in the present study are same as those used in the experimental investigation of Marney [6]. Including both

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sections in the one model was judged to be wasteful of computational resources, as nothing that happens in the injector section affects the flow straightener, and so it was decided to model the flow straightener separately and use the results of this model as inlet data for the injector section. To accomplish this, all data such as velocity, pressure, turbulent properties (k and ε) etc. at the same vertical height as the inlet to the injector section model were written out to a file, which was then read in and used as inlet data for the lower section. This provided significant savings in computational time for the model, as well as allowing more grid points to be used in each section.

The arrow on the images indicated the flow direction at the inlet. The flow straightner also shows the plane on which data are written out to be used as inlet data for the injector section. Geometry was also used by CRC researcher Mey Manickham [22], for a previous project. This project aimed to predict the velocity profiles in the drop tube furnace, in order to check estimates of particle residence time. No combustion modelling was completed at that stage. Figure 3 shows the schematic diagram of the drop-tube furnace with gas temperature thermocouple positions.

In the experimental rig [6] particles were sampled at various heights in the furnace, each height corresponding to a certain residence time, weighted and then a graph of weight loss versus residence time was constructed. As the surrounding fluid was inert nitrogen, the only weight loss experienced by the particles is from loss of volatiles, so in this way an Arrhenius model of the devolatilization reaction could be made.

The semi-implicit method for pressure linked equations (SIMPLE) was implemented for computing the



Fig. 1 Diagram of flow straightener (velocity contours (m/s))



Fig. 2 Diagram of injector section

combination between the pressure and velocity. For the convergence, the normalized absolute residuals for all the variables were limited to be less than 10^{-4} . A grid independency test is used, in this numerical study, by testing three different grid systems (126,000, 252,000, and 504,000 cells). However, the results of the grid independence test were very marginal (within 1 % for all grid systems), and hence the numerical solution was not sensitive to the number of cells. Therefore, the 252,000-grid system was selected to validate the predicted results against the experimental data.

3 Mathematical models used

The three-dimensional governing equations of mass conservation, momentum, and energy transport equations have been solved in the Cartesian tensor form:

Mass conservation equation

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_i} \left(\rho \, u_i \right) = 0 \tag{1}$$

• Momentum conservation equation

$$\frac{\partial}{\partial t}(\rho u_i) + \frac{\partial}{\partial x_j}(\rho u_i u_j) = -\frac{\partial P}{\partial x_i} + \frac{\partial}{\partial x_j}(\tau_{ij})_{eff} + \frac{\partial}{\partial x_j}\left(-\overline{\rho u'_i u'_j}\right)$$
(2)

Energy transport equation

$$\frac{\partial}{\partial t} \left(\rho E\right) + \frac{\partial}{\partial x_i} \left[u_i(\rho E + P)\right] = \frac{\partial}{\partial x_j} \left(k_{eff} \frac{\partial T}{\partial x_j} + u_i(\tau_{ij})\right) + S_{\varphi}$$
(3)

where P is the pressure of fluid, u_i and u_j are the fluctuating velocity components in the ith and jth directions,

Fig. 3 Schematic diagram of the drop-tube furnace with gas temperature thermocouple positions, all dimensions are given in mm [6]



Bottom of the furnace tube

respectively, while term S_{ϕ} represents the appropriate source of the variables ϕ .

The general purpose CFD software package CFX was used for all CFD modelling. Fluid flow is modelled using the Navier–Stokes equations, utilising the k- ϵ model for turbulence, and the SIMPLEC algorithm for velocity– pressure coupling. A simple hybrid differencing scheme is used for all variables.

The coal combustion models used in CFX consists of an Arrhenius reaction for pyrolysis and char combustion, and has the choice of either Mixed-is-Burnt or Eddy Break-up for the homogeneous gas phase reactions. As the surrounding fluid is nitrogen, this prevents any reactions apart from devolatilization from occurring, and so the other reactions are not important in this investigation.

Pyrolysis can be modelled by either a single Arrhenius reaction based on the work of Badzioch and Hawksley [1], or two competing Arrhenius reactions based on Ubhayakar et al. [2]. Either one must use constants taken from literature, based on experimental investigations of pyrolysis rates, however the two competing reaction model has two separate reactions with different yields, one for low temperature and one for high temperatures, Eqs. (4) and (5) mathematically describe these reactions. The dominant reaction depends on the local temperature experienced by the particle, as well as its temperature history. The rate of production of the volatile is given by the first order reaction as follows:

$$\frac{dV}{dt} = K_{\nu}(V_f - V) \tag{4}$$

where *V* is the product of volatiles that have already released from unit mass of pulverized coal at time t, V_f is the ultimate product of volatiles and K_v is the rate constant given by the Arrhenius form as: $A_v \exp\left(\frac{-E_v}{T_p}\right)$, where T_p is the temperature of coal particle, A_v and E_v are the preexponential factor (s⁻¹) and the activation temperature (K) constants, respectively, that are determined experimentally for the particular coal. These factors are usually obtained from the proximate analysis of the coal.

For two competing reactions, the rate of production of volatile can be written n the following expression:

$$\frac{dV}{dt} = (Y_1k_1 + Y_2k_2)C_0$$
(5)

where k_1 and k_2 are constants of the same type as for the single reaction model, while Y_1 and Y_2 represent the reaction yields for two reaction model, and C_0 is the mass raw coal.

CFX code requires coal composition to be set, and this takes the form of a proximate analysis, part of which is the mass fraction of volatiles available in the coal particle. This mass fraction multiplied by the enhancement of volatiles sets the maximum weight loss achievable by the particle, as there can be no weight loss from char combustion in an inert atmosphere. The enhancement factor accounts for the higher volatiles loss experienced under rapid heating conditions. The Loy Yang coal modelled here had a volatile mass fraction of approximately 0.45 from the proximate analysis, with a yield enhancement factor of 1.3, increasing the total fraction of volatiles available to 0.70. The coal particles contained approximately 10 percent moisture and 1.0 percent ash. The proximate and ultimate analysis of coal particle (Loy Yong coal particle), used in the present study are summarized in Table 1.

The particle of the coal is generally assumed to be spherical in shape in which each group of identical non-interacting particles termed parcels. This method operates by introducing the particle parcels in the flow domain and is tracked through the computational grid used. The size of the inlet particle is kept constant in this simulation with a mean diameter of 50 (μ m).

4 Comparison of fluid flow predictions with experiment

Isothermal flow predictions were undertaken to validate the aerodynamic of the model against physical measurements. These measurements were taken from a cold flow model of the pdtf, originally used to find velocity profiles in the furnace to estimate residence time more accurately. The cold model has exactly the same dimensions as the combustion furnace, allowing comparison with the data.

Firstly the flow straightener section of the model was modelled, using the required flow rate (≈ 90 L/min). The inlet data for the injector section was written out and used as inlet data for the lower section. A comparison of the predicted velocity profiles with the experimentally obtained velocity profiles [23] is shown in Fig. 4.

Individual points show experimental data at different distances downstream of the particle injector, solid lines show the CFD predictions of velocity at the same planes. It can be seen from this figure that the CFD model accurately predicts the lopsided nature of the flow in the pdtf, caused by the geometry of the flow straightener, and the minimum velocity in the centre of the tube 20 mm from the injector tip. This is important as it proves we have an accurate fluid flow model in the region of the injector tip, which is where most of the reaction occurs. The CFD model is shown to be overly diffuse in the region between the bulk fluid and the weaker jet from the injector. This is a region of high shear, which is known to be poorly predicted by turbulence models. A higher order differencing scheme (HODS) was also implemented to see if false diffusion effected the prediction, these predictions are illustrated by the individual points overlaying the solid lines of the predictions at 20 and 50 mm. The fact that using a higher order scheme made absolutely no difference to the predictions shows that false diffusion due to the grid and differencing scheme had no effect.

5 Comparison of mass loss predictions with experiment

With the fluid flow model validated, it is then possible to model the mass loss of coal particles due to pyrolysis. In the experiments [6], the investigations of Australian low-rank coals were obtained at residence times up to approximately 2.5 s, operating pressures up to 15 bar, at a reaction temperature of 1,173 K with particle heating rates of approximately 10^5 K s⁻¹, in an entrained flow reactor.

Coal particles were introduced to the flow, and their pyrolysis reaction was modelled using several different reaction rates from literature, using both single and two competing reactions. The results of weight loss versus residence time are shown in Fig. 5. Experimental data is shown as discrete points, CFD mass loss predictions are shown as continuous lines. The mass loss has been normalised to the total mass of the particle, 70 % being the maximum mass loss achievable through loss of volatiles, as explained in Sect. 3.

Several rate constants were used, some specific for lignite coals, others more generic in their applicability. The default rate parameters used in CFX for both the single reaction and two competing reaction models were found to

Table 1 The physical properties (proximate and ultimate analysis) of Loy Yang coal particle

Proximate analysis (wt%, as received)			VM (% d.a.f)	Ultimate analysis (wt%, d.a.f.)				HHV (MJ/kg)	
Ash	Combustibles	Moisture Fraction		С	Н	0	Ν	S	
1.0	88.8	10.2	45.0	69.9	5.4	23.1	0.6	1.0	20.9



Fig. 4 Comparisons of velocity profiles between the predicted results and experimental data for drop tube furnace at different locations from the



Fig. 5 Comparisons between the predicted results and experimental data for the particle mass loss versus particle residence time at two different reaction schemes

give better results. These constants are from Badzioch and Hawksley [1] and Ubhayakar et al. [2], respectively. Hart and Naser's constants [23] also give a reasonable fit, however they cause the reaction to proceed to fast when compared to Badzioch and Hawksley [1] above. Table 2 shows the Arrhenius rate constant used in each pyrolysis model. The particle radiation feature of CFX was also implemented for Hart and Naser's constants [23], however it was found not to improve the prediction significantly. Two reaction rate constants of Anthony et al. [9], along

injector tip

Table 2 Arrhenius reaction constants used in this stud	y
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Model	Pre-exponential fact	or (s^{-1})	Activation temperature (K)		
	First reaction	Second reaction	First reaction	Second reaction	
Badzioch and Hawksley [1]	2.0×10^{4}	N/A	5,941	N/A	
Ubhayaker [2] 2 Reaction	3.7×10^{5}	1.46×10^{13}	8,852	30,189	
Anthony [9] 2 Reaction	60.7	283.0	4,534	5,592	
Yeasmin [24]	6.0	N/A	1,479	N/A	
Kobayashi [8]	6.6×10^{4}	N/A	12,600	N/A	
Hart and Naser [23]	1.0×10^{7}	N/A	8,850	N/A	

with the single reaction constants from Yeasmin [24], and Kobabyashi et al. [8] significantly under predict the time taken to release volatiles from the coal.

Of particular interest is the poor prediction given by Yeasmin's model [24], as this model was constructed from experiments using similar coal pyrolysed in the pdtf, leading to the expectation that it would give the best results. The model used by Yeasmin [24] was, however, constructed for a wide range of temperatures and elevated pressures, possibly accounting for its inaccuracy.

6 Conclusions

By modelling pyrolysis in a pdtf reactor, it is possible to determine the most suitable rate constants for a Loy Yang coal. It was found that the best constants to use are Badzioch and Hawksley [1] and Ubhayakar et al. [2] Both single reaction and two competing reaction models were used, each gave slightly different results, however it is not possible to tell from one set of experimental data whether the two competing reaction model gave more accurate results. The single reaction model would be the most appropriate for a coal combustion model as it requires slightly less computation, and as long as heating rates for the coal are high, as found in industrial boilers, then one reaction describes the pyrolysis accurately enough.

As a result of this work, one can be reasonably confident that CFD in general is capable of predicting the pyrolysis behaviour of brown coal. This is important for future work where the coal combustion model (including pyrolysis) will be used to model combustion in a large brown coal tangentially fired furnace.

7 Directions for future work

Char combustion reaction rates are determined in a similar way to pyrolysis rates. There is assumed to be an Arrhenius reaction of the char particle with surrounding oxidant. This same experimental apparatus and CFD model will be used in a similar way to determine the most appropriate char combustion reaction rates to use in the coal combustion model. Coupled with the pyrolysis model, this provides a well validated coal combustion model.

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