Effect of Bed Diffusion and Operating Parameters on Char Combustion in the Context of Underground Coal Gasification

Ganesh A Samdani[1], Shauvik De[1], Sanjay Mahajani[*,1] and Anuradda Ganesh[1]
[1]Indian Institute of Technology Bombay
*Corresponding author: Prof. Sanjay Mahajani, Department of Chemical Engineering, IIT Bombay, Mumbai.

Abstract: Char combustion is one of the most important reactions in the process of Underground Coal Gasification (UCG). It's importance increases as it becomes the essential heat source for endothermic gasification reactions. Experiments are performed in boat reactor under near isothermal condition to demonstrate effects of heat and mass transfer and bed diffusion through ash layer. A boat reactor mimics the actual UCG conditions in a way that it has a cavity through which gas flows over the char kept in the boat. The experiments performed have demonstrated effect of bed diffusion and in addition to it experiments were also performed to investigate the effect of inlet flow rate, inlet temp, inlet gas composition. To obtain a complete overview of boat experiments and to further get insights, CFD analysis of boat reactor is performed using COMSOL Multiphysics which is a commercial finite element solver. The silent features of the model are: Navier-Stokes equation for flow through cavity, diffusion through char bed (diffusivity as a function of conversion of char), mass balance for oxygen and carbon dioxide and convective mass transfer in cavity. This model and the experimental study in together can show the effect of inlet temperate, flow rate and gas composition under the UCG conditions. The extension to this work is to get into non isothermal experimental conditions and to extend the same methodology to steam and coal gasification.

Keywords: Boat Reactor, UCG, CFD, Char Combustion

1. Introduction

Rising oil and gas prices, combined with geopolitical instability in major oil and gas producing regions, is putting coal firmly back as a reliable and cheap source of energy. Global coal demand is expected to grow steadily over the next decades, with most of the demand coming from China, India and United States [1]. Coal currently fuels 39% of the world's electricity and this proportion is expected to remain at similar levels over next 30 years. Coal gasification is a process in which solid coal is converted into gaseous products. It converts coal into carbon monoxide and hydrogen by reacting it at high temperatures with a controlled amount of oxygen and/or steam. The resulting gas mixture is called synthesis gas or syngas and is a fuel too. Coal gasification offers one of the most versatile and cleanest ways to convert the energy content of coal into electricity, hydrogen, and other energy forms. [2]

Underground Coal Gasification (UCG) is a modern technology used for in-situ gasification of coal which has the same principle reactions as the surface gasifiers. UCG process increases the coal resource available for utilization enormously by gasifying otherwise unmineable deep or thin coals under many different geological settings. Generally in UCG process, the coal block gets dried initially then pyrolysis occurs and then occurs the combustion. So char is what oxygen gets to react with. Char combustion is one of the most important reactions in the process of UCG. To study combustion reaction and effect of different parameters on the combustion rate becomes a very important part of understanding UCG, as it becomes the essential heat source for endothermic gasification reactions. [3] To understand combustion and model effect of different parameters, we need to consider a kinetics model. As pointed out by Muhammad F [4], the RPM has been used by many researchers and has been bettered to account for non-uniform pore size distributions and for high ash discards. One of the advantage of RPM is that it can represent the behaviour of a system that shows maxima in the reaction rate as well as the one that does not, and it makes out an optimal pore structure for both the systems. [4] Considering this, we decide to use RPM for kinetics determination.

As far as the effect of operating parameters on UCG is considered, Perkins [5] and Daggupati [6] have considered some of the

operating parameters and coal properties for Australian and Indian coal respectively.

2. Materials, Methods and Results

Some of the factors affecting combustion reaction are heat and mass transfer, bed diffusion through ash layer, reaction temperature, flow rate of inlet gases. Experiments were performed in boat reactor under near isothermal conditions to demonstrate effects of these parameters. A boat reactor mimics the actual UCG conditions in a way that it has a cavity through which gas flows over the char kept in the boat. (Details are given later) The actual UCG scales are too different if compared with these experiments' and this may result in actual conditions also being different but the effect of these parameters in the boat experiments can be qualitatively similar. For modeling of these boat reactor experiments, we need kinetics of combustion for the coal being used. To determine the kinetics of char combustion reaction, TGA experiments were performed, in such a way that it provides intrinsic kinetics. [7] So this kinetics determined from TGA is used for modeling of combustion reactions and bed diffusivity is added as a parameter in the model. Following is the procedure for TGA experiments and then for boat experiments.

2.1 Char preparation

Indian lignite coal, obtained from Gujarat Industries Power Company Limited, INDIA (GIPCL) is used to prepare char for experimentation. The coal properties are shown in Table 1.

Table 1 Coal properties

	Dry basis	
Proximate Analysis	Volatile matter	44.92%
	Fixed Carbon	46.61%
	Ash	8.47%
Ultimate Analysis	Carbon	40.594%
	Hydrogen	5.672%

The char prepared is ground to the size range of 90 -150μm.

2.2 Kinetic Determination

The TGA experimental procedure for studying the reaction kinetics of combustion is as follows: The char sample is heated from ambient temperature to $110^{\circ}C$ and is maintained for 30 min to dry the sample and flush out air from the TGA chamber. Then the temperature is increased to the specified reaction temperature and combustion of the char is carried out at the reaction temperature until all the char is consumed.

Resistance due to inter-particle diffusion of gas in char can be disregarded as we are using very small (i.e.<150 micron) size of char particle. Effect of bed diffusion resistance has been disregarded as the experiments are performed with monolayer of char at the top of TGA pan, below which alumina is filled. Reactions were performed at 3 different temperature. Reactions order with respect to oxygen was assumed to be one (so, no experiments with different flow rates/partial pressures). To get the structural parameter value of the char BET experiments were performed. The analysis of the char surface area and other parameters was used to find out value of psi. Values of parameters from these experiments are as following:

BET surface area (S_0) = 179.16 m^2/g

$$\psi = \frac{4\pi L_0(1-\varepsilon_0)}{S_0^2} = 3.8485$$

Pre exponential factor (K_0) = 0.2 $\times$ 10^8 (sec^{-1})
Activation energy (Ea) = 116.7 kJ/mol

2.3 Boat Reactor Experiments

2.3.1 Experimental Setup

The boat reactor set up consists of a cylindrical quartz tube (length 0.8 m and diameter 0.05 m), and a rectangular quartz boat is placed inside the quartz tube. Boat dimensions are: length 0.15m, breadth 0.035m and thickness 0.005m. There are three heaters on the top and another three on the bottom of quartz tube. The quartz tube is completely insulated from outside so that no heat is transferred out from the walls of the tube. However it can be heated by using the heaters at top and bottom and the heat also goes out with

the hot gases getting out. Reactor setup has internal thermocouple for temperature sensing flow controller for flow metering, and an online GC for gas analysis (Fig 1).
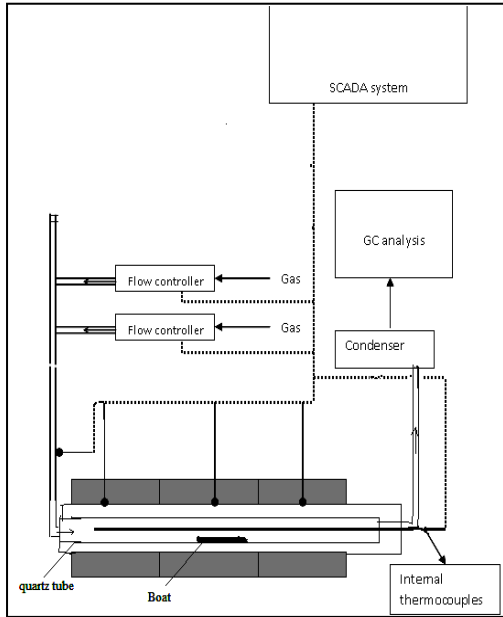

Figure 1 Boat reactor setup

2.3.2 Experimental procedure

1. Boat is initially filled with char (char is prepared by the procedure as discussed earlier).
2. Char sample is heated from ambient temperature to $110^o C$ in inert atmosphere of nitrogen for 30 min to dry the sample and to flush out air (flow rate: 75 ml/min).
3. Temperature is steadily increased to $500^0 C$ by heaters at top and bottom.
4. Once steady state is reached, feed is switched over from nitrogen to oxygen.
5. Combustion of the char is carried out at the reaction temperature until all the char is consumed. Complete conversion is ensured by analyzing the outgoing gases with gas chromatography.
6. While the experiment is going on, outlet gas is continuously analyzed with GC to obtain the outlet gas composition at various times till char gets converted completely.

7. The same operation is repeated at $550^{\circ}C$ and $600\ ^{\circ}C$ for inlet flow rates of 75, 100 and 125 ml/min.

Generally for any gas solid reaction and so for coal combustion reaction, reaction regimes are controlled by either chemical reaction or inter-particle diffusion or mass transfer inside the coal particle (intra-particle diffusion) or mass transfer from the surrounding gas to the external surface (bed diffusion) or all or some in combination. The relative significance of these steps, which are dependent on temperature, coal/char properties and flow dynamics around the particle and around the coal seam should be considered to model these kind of processes. Unlike the normally carried out combustion of coal at high temperatures with relatively porous chars where the effect of intra-particle diffusion and chemical reaction is negligible and external diffusion is considered to be the controlling step; in UCG, all the steps are important.

As far as the experiments are considered following are the conditions: These experiments are performed at low temperature and relatively higher flow rates so that the limitations because of transfer of gas and heat from the bulk gas to the external layer of the particle bed (external mass and heat transfer) can be neglected. To avoid problems of pore diffusion of CO_2 (if any) and effective conduction of heat through the porous particles (i.e. diffusion into the interior of the char particles) particle size of char used is very small. (<150 micron). As it is not possible to avoid bed diffusion of gases and heat in the current setup used for experiment, is was used as modeling parameter.

2.3.3 Results of Experiments
As explained above the performance of boat reactor experiments depends on char properties, temperature, flow dynamics, bed diffusion i.e. height of bed. So these were considered as parameters affecting the process. As described In the experimental procedure, outlet gas is analysed using gas chromatography which represents the performance of the experiment with particular combination of operating conditions and bed height. As one cannot
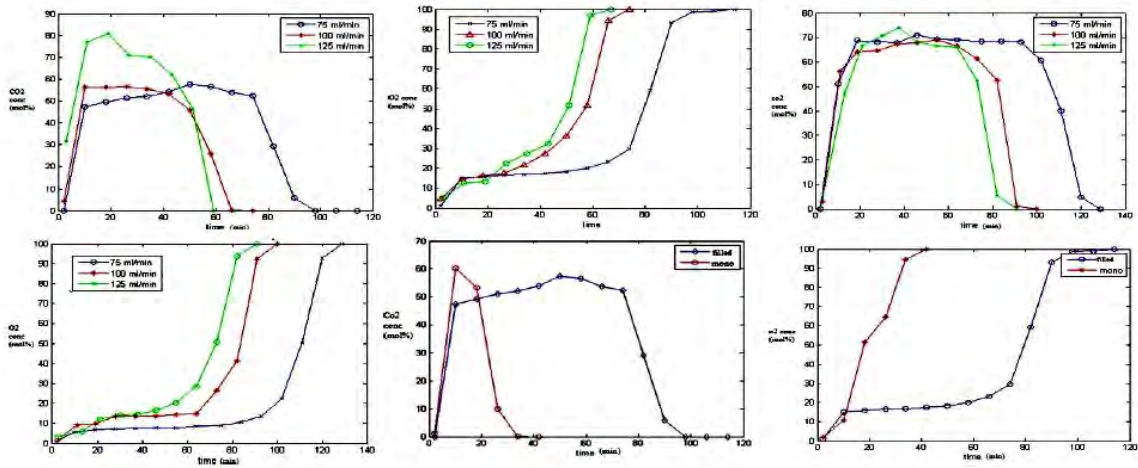
Figure 2 Concentration of CO2 and O2 vs. time at 500 and 550˚C respectively (fig. a, b, c, d), comparison of monolayer and complete filling at 500˚C, 100 ml/min (fig. e, f)

measure weight of char as a function of time in the current experimental setup, no data for conversion of char is presented. In all the experiments, it is observed that the outlet gas composition mainly comprises of CO_2 and unreacted oxygen. The product gas profiles for gases at different temperatures are plotted. Each of the mole percentage profiles in figure 2(a, b, c and d) shows profiles of three different inlet flow rates of oxygen. The results show that concentration of CO_2 initially increases and reaches the maxima then the concentration keeps almost constant value for some time and finally it falls drastically.

These concentration profiles clearly show the difference in outlet gas composition during course of reaction for different flow rates. A set of experiment is also performed using a single (mono) layer of char in place of the boat fully filled with char; the results when compared with results of fully filled char show (Figure 2-e and 2-f) clearly shows the effect of ash layer diffusion during combustion reaction.

3. CFD analysis of the Boat Experiment

Boat experiments' results depends on the flow dynamic around the reaction area i.e. char inside boat reactor, so we need to perform a flow dynamics study for analysis of these results. This analysis is performed using COMSOL multiphysics software. COMSOL is based on finite element method (FEM) solvers. For the modelling exercise, flow of gases in the quartz tube is assumed to be governed by incompressible Navier Strokes equation. Mass transport is by convection and diffusion in tube and only diffusion in boat. Experiments were performed at nearly isothermal conditions, so heat transport is not considered. Figure 3 shows the geometry used for modelling the Boat Experiment.
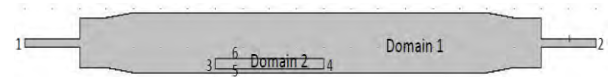

Figure 3 Schematic diagram of reactor in Boat experiment for CFD analysis

Governing equations are momentum balance and mass balance:

$$\rho\frac{\partial \mathbf{u}}{\partial t} + \rho(\mathbf{u}\cdot\nabla)\mathbf{u} = \nabla\cdot[-p\mathbf{I}+\tau] + \mathbf{F}$$

[Navier Strokes equation]

$$\frac{\partial c}{\partial t} + \mathbf{u}\cdot\nabla c = \nabla\cdot(D\nabla c) + R$$
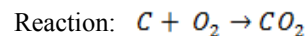
[Mass transport equation in quartz tube]

$$\frac{\partial c}{\partial t} = \nabla\cdot(D\nabla c) + R$$

[Mass transport equation in boat]

3.1 Assumptions:
- Incompressible laminar flow
- Inlet gas is pure oxygen
- Flow of gas in the channel,
- Diffusion and combustion in the boat.
- 2-D geometry modelled.

Reaction: $C + O_2 \rightarrow CO_2$

Intial conditions and boundary conditions are as shown in table 2 and 3 respectively.

Table 2 Initial Conditions

domain	velocity	O_2	CO_2	char
1	0	Zero concentration		NA
2	NA	Zero concentration		c_cinit

Table 3 Boundary Conditions

Boundary	velocity	O_2	CO_2
1	u_in	c_o2_fluxin	0
2	outlet	convective flow	
6	wall	continuity	
all other		Insulation (wall)	

3.2 Solution strategy

For the simulations, the geometry was meshed and then a steady state velocity profile is obtained which is fixed and used for the unsteady state simulation for transport of other species. The mesh quality and quantity is as following: minimum element quality: 0.6689, average element quality: 0.9909, triangular elements: 151779. This is fairly good quality of mesh. Meshing is dense near and inside the boat and also at inlet and outlet. This is prepared to take care of stiffness of the equations in those areas. BDF(backward difference formula) solver is used for unsteady state solutions and for linear problems, a direct solver (PERDISO) is used.

3.3 Results of CFD Modeling

Results thus obtained after the simulation are discussed in this section. Figure 4 shows the steady state velocity profile, which is kept same throughout the simulation.
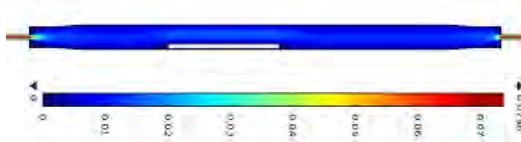

Figure 4 Steady state velocity profile

Figure 5 shows the evolution of char concentration in boat with time. It can be seen that initially the boat is fully filled with char. With time, as char reacts to form CO_2, its concentration decreases. The CFD results show that the char concentration decreases in a layer wise manner. The top surface first gets in contact with reactive oxygen, so it gets consumed and ash formation occurs. Then oxygen diffuses through the ash layer and gets in contact with char below it and further combustion takes place. The char in the top layer, (and so in all the layers), if near the inlet, reacts earlier than the char away from the inlet, resulting diffusion of oxygen through the ash layer. Finally total char gets consumed to form ash.

Figure 6 show the CO_2 and O_2 concentration profiles at the exit of the quartz tube. These profiles qualitatively match with the profile obtained from boat experiments. Initially CO_2
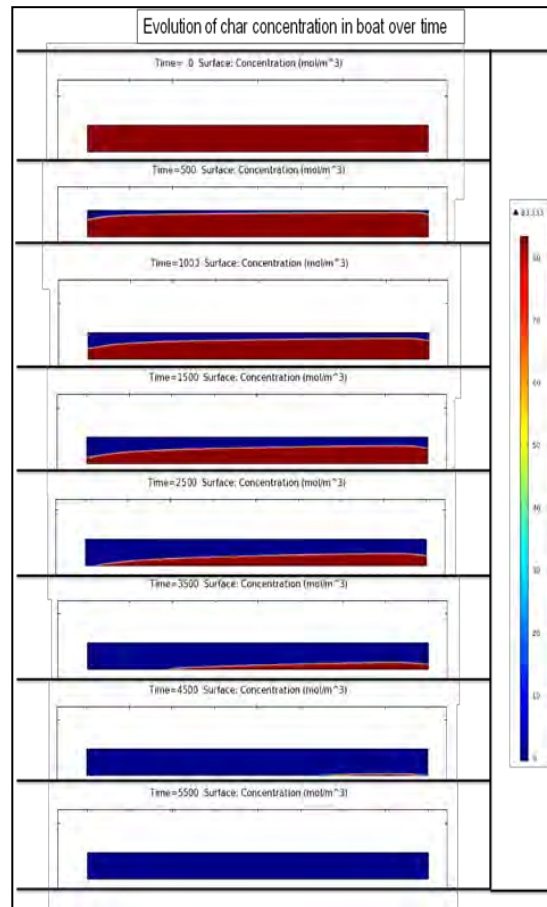

Figure 5 Evolution of char concentration

profile goes to maxima as the upper layer of char is fully exposed to oxygen. Once the upper layer is reacted, CO_2 has to diffuse through growing ash layer to react with char. This results in decrease in overall rate and so a decrease in CO_2
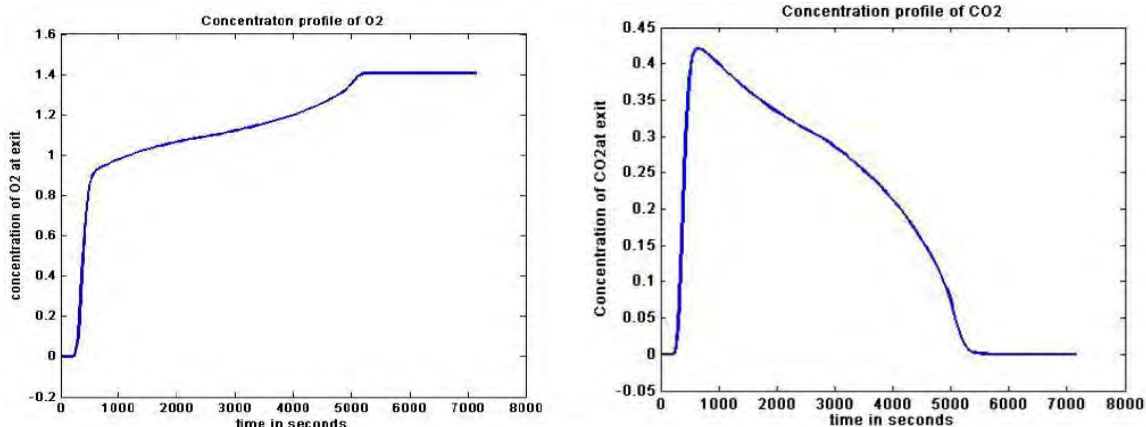
Figure 6 Oxygen and carbon dioxide concentrations at the exit

concentration. Finally char get completely converted and at that time, CO_2 concentration at outlet is zero and O_2 concentration at outlet is equal to the inlet O_2 concentration. The oxygen concentration at the outlet is much less initially, further it increases at a slower rate due to reaction with char in boat. The rate of increase of O_2 concentration at outlet increase with time, which signifies char layer diffusion resistance.

4. Discussions and Future Work

An extensive experimental programme is carried out to evaluate effect of different operating parameters and bed diffusivity/bed height on the char combustion in UCG like condition. A complete kinetic modeling for char combustion experiments is also started. For this kinetic modeling, monolayer combustion experiments are performed and intrinsic kinetic parameters are estimated using random pore model. In boat reactor set up the effect of operating parameters and bed diffusivity effects are studied. Effect of temperature is in increase in rate of the reaction as temperature increased and so the CO_2 concentration peak is higher and char get converted faster in case of high temperatures as compared to the cases with lower temperature. This is because of the Arrhenius form of K_0. The flow rate effects are also similar to those of temperature. As one increases the inlet flow rate, two things happen: 1st is the change in flow dynamics and 2nd is the change in concentration of the reactant gas in the quartz. Both of these effects favor the reaction i.e. it increase rate of reaction and it can be observed from the concentration profiles. One can see the higher peak and early drop in the concentration of CO_2.

The modeling of the boat experiment using COMSOL provided an insight of the experiments. The velocity profile of gas can be realistically shown using the simulations. The mass transfer mode (convection and diffusion model) gives a better idea about the concentration profile of oxygen in the channel and through the bed. Modeling also gives a fair idea about the char layer conversion and the results are similar to experimental findings. The outlet gas profiles are also similar in nature to that we obtained from the experiments. The simulation results still need to be matched for all experimental conditions and the CFD model needs to be validated.

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