Second African Conference on Computational Mechanics – An International Conference – AfriCOMP11
January 5 – 8, 2011, Cape Town
A.G. Malan, P. Nithiarasu and B.D. Reddy (eds)

STEADY STATE CFD MODELLING OF CARBON DIOXIDE RELEASE AND METHANANOGENESIS IN COAL HEAPS

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SUMMARY

The Kyoto Protocol and its related instruments have increased awareness of the phenomenon of global warming. The two main so-called greenhouse gases deemed to cause this effect have been identified as CO_2 and CH_4. Other greenhouse gases include N_2O, hydrofluorocarbons, perfluorocarbons and SF_6, although these occur to a lesser extent than the former two. This has led to efforts to quantify and curb the release of these two main constituent greenhouse gases. CO_2 is released through various biological, natural and industrial processes and the burning of fossil fuels. Methane is also classified as a fossil fuel leading to CO_2 release but can also be spontaneously released to the atmosphere from the earth's crust due to release of trapped methane or methanogenic processes. The global warming potential of methane is 21 times greater than that of CO_2. The coal stockpiles and dumps from mining activities are difficult to control sources of great amounts of both CO_2 and CH_4 due to spontaneous oxidation. The aim of this study is to highlight the capability of CFD in capturing the chemical kinetics leading to the formation of both CO_2 and CH_4 in coal heaps.

Key Words: oxidation, spontaneous combustion, conduction, convection

1. INTRODUCTION

The stockpiling and dumping of high grade and low grade coal respectively presents major environmental and safety problems. Coal naturally reacts with atmospheric oxygen to produce carbon monoxide, carbon dioxide and heat. If the amount of heat generated is equal to the amount of heat dissipated then equilibrium conditions persist and the temperature is stabilized. On the other hand, if less heat is dissipated in comparison with heat generated, the temperatures tend to rise. This occurs if there is some mechanism preventing free heat flow, such as stockpiling fresh material on an existing dump which has already attained thermal equilibrium, thus inhibiting heat flux through the under-surface. A steady build-up of temperature from ambient conditions to about 230ºC will result in thermal runaway or combustion, ultimately causing hazardous fires to break out. The coal dump is an interesting example of a reacting system which involves flow driven by natural convection coupled with chemical reaction in a porous medium. A model set-up for CFD simulations can then be able to predict phenomena such as flow regimes, temperature increase, as well as emissions of gases such as CO_2 and CH_4.

2. LOW TEMPERATURE OXIDATION

The chemical reaction between coal and oxygen at low temperatures is complex. Generally, the following three types of processes are believed to occur (Carras and Young, 1994):

- physical adsorption followed by chemical adsorption, which leads to the formation of coal oxygen complexes and oxygenated carbon-species; and
- oxidation in which the coal and oxygen react with the release of gaseous products, typically carbon monoxide (CO), carbon dioxide (CO_2) and water vapor (H_2O).

The first two stages involving sorption process are relatively fast and lead to an initially high rate of oxidation, while all sorption sites are being occupied by oxygen molecules, thus it is neglected as a short-term reaction. The main reaction in the longer term is the direct burn-off and the chemical reaction between coal and oxygen is simplified as (Yuan & Smith 2000)

$$\text{Coal} + O_2 \rightarrow CO_2 + 0.1CO + \text{heat} \qquad (1)$$

The detailed chemical structure of coal is not clear and varies with the rank and origin of coal. According to experimental data (Smith and Lazzara, 1987), one mole of coal reacting with one mole of oxygen generates one mole of carbon dioxide and roughly 0.1 mole of carbon monoxide plus heat at the early stage of coal oxidation.

The dependence of the rate of oxidation on temperature and oxygen concentration can be expressed by

$$Rate = A\cdot[O_2]^n \exp(-E/RT) \qquad (2)$$

where

A is the pre-exponential factor,
E is the apparent activation energy that is the energy needed to initiate a chemical reaction,
R is gas constant,
n is the apparent order of reaction,
T is the absolute temperature and
$[O_2]$ is the oxygen concentration.

The complete set of reactions with kinetic parameters, which also lead to methanogenesis in the coal dump can be classified in Table 1 (Mann et al., 2004)

Table 1

Reaction	**Name**	**A (s^{-1})**	**E (kJ/mol)**	**ΔH (kJ/mol)**
$xC_{(s)} + yO_{2(g)} \rightarrow aCO(g) + bCO_{2(g)}$	Oxidation	2×10^6	57.4	-300
$H_2O_{(g)} + C_{(s)} \rightarrow CO_{(g)} + H_{2(g)}$	gasification	810	147	131
$H_{2(g)} + C_{(s)} \rightarrow CH_{4(g)}$	hydrogenation	0.0061	80.4	-74.85
$H_2O_{(g)} + CO_{(g)} \rightarrow CO_{2(g)} + H_{2(g)}$	Water-gas shift	3.23×10^7	49.4	-41.7
$CO_{(g)} + 3H_{2(g)} \rightarrow CH_{4(g)} + H_2O_{(g)}$	methanation	0.26	66.5	-357.8
$CO_{2(g)} + C_{(s)} \rightarrow CO_{(g)}$	Boudouard	525	247	283.0

3. MODELLING

The geometry is modelled as a 2D trapezoid porous medium coal heap whose base length is 500m and height 100m and angle of incline 45º. Permeability was $10^{-7}m^2$ and porosity 0.3. Wind speed was also a constant at 3 ms^{-1}, ambient temperature was kept at 300K and ambient humidity at 2%.

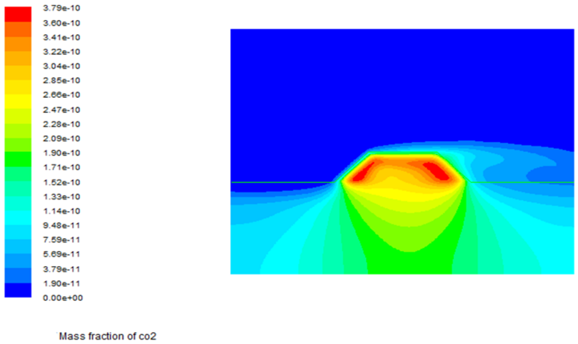

Fig 1. Contours of mass fraction of CO_2 in coal heap at steady state.

The steady state distribution of CO_2 is depicted in Fig. 1. The main oxidation reaction is dependent on O_2 content and this is greatest closer to the walls due to diffusion from atmosphere and this is where most CO_2 is formed and diffuses to the atmosphere. The lower part of the computational domain also depicts CO_2 percolation into the ground.

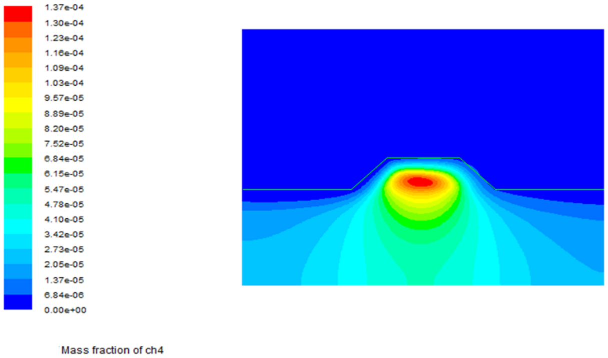

Fig. 2 Contours of mass fraction of CH_4 in coal heap at steady state

When the concentration of oxygen is low, temperatures are high and there is sufficient moisture content, the conditions for the formation of methane can be met. This is depicted in Fig 2, where methanogenesis occurs at an epicentre right in the middle of the dump, where low O_2 concentration and high temperatures would be expected. This is supported by Fig. 3, which depicts the temperature profile of the coal heap at steady state and in which the high temperatures are experienced at the centre of the heap. The temperatures reach a maximum of about 1190K, which is consistent with reports from actual coal heaps, where platinum probes for temperature measurement have been known to melt under the intense heat produced deep inside the heap.

For the purposes of quantifying carbon dioxide release, Fig 4 shows a track of carbon dioxide concentration at the upper right hand wall of the domain, which is actually modelled as an exit boundary condition. Fig. 4 is actually a transient simulation over a few days; integration of this curve over the time period in conjunction with the mass flow rate will quantify the amount of CO_2 release over the period.

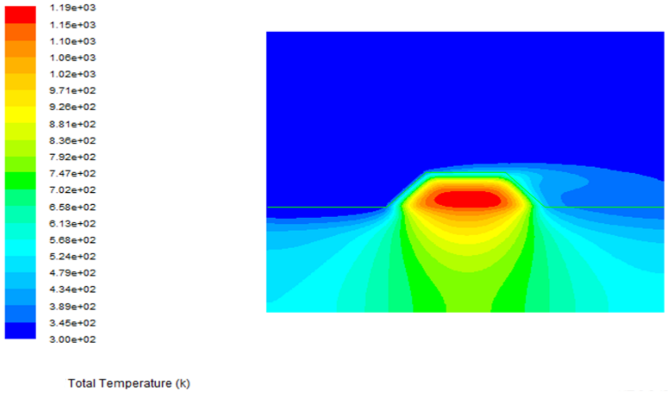

Fig. 3 Temperature profile of coal heap at steady state.

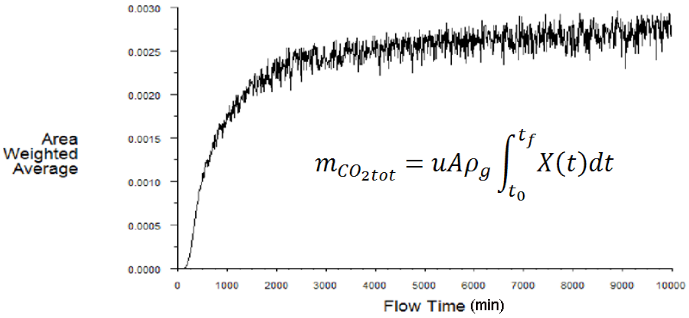

Fig. 3 CO_2 tracking at the domain exit

4. CONCLUSIONS

CFD as a tool can be used to qualify and quantify emissions from coal heaps resulting from low temperature oxidation. Methane is formed at the epicentre of the coal heap where extremely high temperatures and low O_2 concentrations can be expected. Quantities of CO_2 and CH_4 released can be used in either mitigation strategies or carbon credit trade.

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