

Numerical Simulation for Coal Carbonization Analysis of a Coke Oven Charge Using PHOENICS

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ABSTRACT

A Computational Fluid Dynamic model is analyzed for coking process in a coke oven charge using PHOENICS CFD package (Version 3.2). The model reported in this paper consists of a combination of a two-dimensional model for coke oven gas phase flowing in porous media and a one-dimensional model for semi-coke phase together with chemical reactions, heat and mass transfer between the two phases during coal carbonization in the coke oven charge. The model simultaneously calculates the transient state composition, temperature of all two phases, velocity of gas phase and porosity, density of semi-coke phase. Numerical simulation is illustrated in the predictions of evolution of volatile gases, gas flow paths, profiles of density, porosity of the coke oven charge, profiles of temperatures of the coke oven gas and the semi-coke bed. The predictions agree well with the published data on coking process.

KEY WORDS: coal carbonization; porous media; numerical simulation; PHOENICS

1 Introduction

Coking process has remained basically unchanged for over 100 years. Coal, crushed so that some 80% of the particles are <3mm diam., is charged from the top into slot-type ovens. The ovens are heated indirectly through the side walls, which are usually made of silica refractory brick. There have been many models developed for it. D. Merrick^[1~5] performed distinguished study on coking process. Besides having proposed a series of mathematical models, he meanwhile has given a good summary on the study of coking process. Yet, all these foregoing studies could not give a comprehensive understanding of this process. To date, no model has ever considered the gas-solid coupled problems in coking process. As B. Atkinson and D. Merrick^[1] pointed in their paper., coking process still needed a more advanced study for cost-effective purposes. With the advent of modern high-speed computers and CFD technology, it has become possible to construct a comprehensive mathematical model of this complicated multidimensional and multiphase process in the coke oven.

The purpose of this paper is to extend a CFD model of coking process in coke oven to give some detailed information during coking process.

2 Model Formulation

Two phases are considered in the model, they are coke oven gas (COG) and lump coal charge. A two dimensional mathematical model considers mass, momentum, enthalpy and chemical species conservation for gas phase at transient state, and a one dimensional mathematical model enthalpy conservation for the lump coal phase. The general conservation equation for COG gas phase is given in Eq. (1)

$$\frac{\partial}{\partial t}(\varepsilon_g \rho_g \Psi_i) + \frac{\partial}{\partial x}(\varepsilon_g \rho_g u_x \Psi_i) + \frac{\partial}{\partial y}(\varepsilon_g \rho_g u_y \Psi_i) = \frac{\partial}{\partial x}(\varepsilon_g \Gamma_{\Psi_i} \frac{\partial \Psi_i}{\partial x}) + \frac{\partial}{\partial y}(\varepsilon_g \Gamma_{\Psi_i} \frac{\partial \Psi_i}{\partial y}) + S_{\Psi_i} \quad (1)$$

Eq. (1) is used to represent all conservation equations of the gas phase by setting Γ_{Ψ_i} and S_{Ψ_i} to appropriate values according to the dependent variable, as listed in Table 1. Note that ε_g is the volume fraction of the gas phase.

Tab. 1 Terms used in Eq. (1). The symbol “-“ indicates this term is not applicable

Equation Name	Ψ_i	Γ_{Ψ_i}	S_{Ψ_i}
Gas continuity	1	-	$\tilde{R}_{volatile} + \tilde{R}_{moisture}$
Gas Momentum	u_x	μ_g	\tilde{F}_x
Gas Momentum	u_y	μ_g	\tilde{F}_y
Gas enthalpy	H_g	λ_g	\tilde{E}_{gs}
CH ₄ mass fraction	m_{CH_4}	D_g	\tilde{R}_{CH_4}
C ₂ H ₆ mass fraction	$m_{C_2H_6}$	D_g	$\tilde{R}_{C_2H_6}$
CO mass fraction	m_{CO}	D_g	\tilde{R}_{CO}
CO ₂ mass fraction	m_{CO_2}	D_g	\tilde{R}_{CO_2}
H ₂ O mass fraction	m_{H_2O}	D_g	\tilde{R}_{H_2O}
H ₂ mass fraction	m_{H_2}	D_g	\tilde{R}_{H_2}
NH ₃ mass fraction	m_{NH_3}	D_g	\tilde{R}_{NH_3}
H ₂ S mass fraction	m_{H_2S}	D_g	\tilde{R}_{H_2S}

Heat transfer is considered to be one-dimensional for the lump coal charge for this assumption is most accurate when simulating commercial ovens where the length/width ratio is up to 30 and height/width ratio is up to 10. [5] The charge is considered to be uniform within vertical planes. Thus the enthalpy conservation equation for the lump coal phase is given by

$$\rho_c C p_c \frac{\partial T_c}{\partial t} = \frac{\partial}{\partial x} (k_c (\frac{\partial T_c}{\partial x})) + S(t, x) \quad (2)$$

Treatment of Momentum Transfer

Gas-solid drag term uses Ergun's expression in an isotropic form to account for the resistances of lump coal to coke oven gas flow in x direction and y direction are shown in Eq. (3) and Eq.(4)^[7]

$$\tilde{F}_x = -\frac{C_1 \mu_g}{\rho_g} G_x - \frac{C_2}{\rho_g} G_x |G_x| \quad (3)$$

$$\tilde{F}_y = -\frac{C_1 \mu_g}{\rho_g} G_y - \frac{C_2}{\rho_g} G_y |G_y| \quad (4)$$

Treatment of Heats of Formation in Enthalpy Transfer

The enthalpy of content of the gas phase was calculated by

$$H_g = \Delta H_g^{298} + H_g^T \quad (5)$$

where

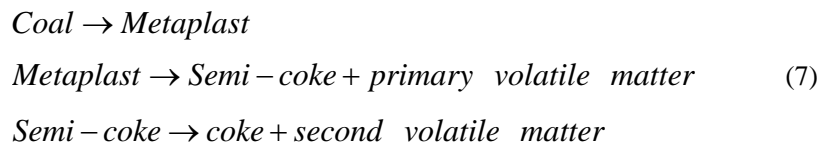
$$H_g^T = \int_{298K}^T C p_g dT_g$$

The convective gas-solid heat transfer is negligible for velocity of the gas phase is relatively small. However the volatile-release and moisture evaporation reaction enthalpy transfer sources should be included. The source is in Eq. (6)

$$\tilde{E}_{gs} = C p_g T_c \sum_i \tilde{R}_i \quad i=\text{CH}_4, \text{C}_2\text{H}_6, \text{CO}, \text{CO}_2, \text{H}_2 \dots \quad (6)$$

Release of COG

Release of volatile matter from coal charge follows the sequential scheme of first-order reactions as Eq. (7).



For any part of bulk coal charge, Composition of the volatile matter in it is defined in terms of the

following 9 species: CH₄, CO, CO₂, C₂H₆, TAR, H₂, H₂O, NH₃, H₂S. The kinetics of volatile matter release are described in the model by a system of parallel first-order reactions for which the rate constant varies with temperature according to an Arrhenius relationship^[2], which is given by

$$\tilde{R}_{t,i} = [c_i \exp(-\frac{E_i}{RT_c})](m_{0,i} - m_{t,i}) \quad (8)$$

$$m_{t,i} = \int_0^t R_{t,i} dt \quad (9)$$

The heat effect of volatile matter release on the solid lump charge is neglected in this paper for this effect is somewhat included in the calculation of the specific heat of coal charge which will be discussed in the following sections.

Boiling of moisture

Coal charge usually contains moisture. As the charge heated, the moisture near the oven walls boils. The reaction is as follows



It's assumed that at 373K in the nearest plane to the oven wall. The following equation defines the position of the boiling plane.

$$\begin{aligned} T_c &= 373K \\ (\frac{\partial T_c}{\partial x})_{xb+} &= 0 \\ (w_{moisture})_{xb-} &= 0 \end{aligned} \quad (11)$$

where x_b is the distance of the boiling plane from the coke oven wall and the suffices +,- refer to an approach from the low and high temperature sides respectively.

Boiling is represented in the model as a 'point' sink (one dimensional) of moisture and its associated latent heat. At any instant, mass and heat balance at the boiling plane therefore give respectively:

$$\tilde{R}_{H_2O} = -(k_{c,t} \frac{dT_{c,t}}{dx})_{xb-} / 3.2e6 \quad (12)$$

$$S(t, x) = \Delta H = -(k_{c,t} \frac{dT_{c,t}}{dx})_{xb-} \quad (13)$$

Gas phase properties

Tar being excluded, density of coke oven gas is calculated from the ideal gas equation, thus it is a function of temperature and pressure. Kinetic viscosity and specific heat of the gas are assumed to be constant. They are 0.8E-5m²/s and 1500J/kg^[8] respectively. In order to evaluate the thermal conductivity and mass diffusivity of the gas, for simplicity, The dimensionless numbers, Pr and

Sc, are all considered to be 1.0, which is correct for most of gases under high temperature.

Solid phase properties and modifications

For coking coal, density, thermal conductivity and voidage (also the volume fraction of COG gas phase) are the functions of temperature and composition of coal in coking process.

Effective thermal conductivity

The equation for the overall effective thermal conductivity of coking coal is combined with equations for the COG gas conduction and radiation components of heat transfer in two models of the charge thermal conductivity, one model describes particulate coal and the other semi-coke^[1].

For particulate charge, the model is given by Eq. (14). The model combines the thermal conductivity of coal charge (k_0), the conductivity of the gas phase (k_1), the effective thermal conductivity of radiation (k_2) and the thermal conductivity of moisture (k_3).

$$k_c = w_{moisture} k_3 + (1 - w_{moisture}) / (e' / (k_1 + k_2) + (1 - e') / k_0) \quad (14)$$

where

$$k_0 = (d_s / 4511.0)^{3.5} T_c^{0.5}$$

$$k_1 = 7.45(10^{-5}) T_c$$

$$k_2 = 2.28(10^{-10}) T_c^3$$

$$e' = 1 - (1 - \varepsilon_g)^{1/3}$$

$$k_3 = 0.6$$

After resolidification at the critical temperature, the effective thermal conductivity of coke charge becomes

$$k_c = (1 - \varepsilon_g) k_0 + \varepsilon_g k_4 \quad (15)$$

where

$$k_4 = 4.96(10^{-5}) T_c$$

Specific heat

Published experimental measurements related the specific heats during thermal decomposition are based on the total heat required for carbonization to various temperatures. Heats of reaction can be assumed negligible. In this paper, the model of specific heat of coal and coke is given by Eq.(16), which is proposed by D.Merrick^[3]

$$Cp_{daf} = (R/a)[g_1(380/T_c) + 2g_1(1800/T_c)] \quad (\text{Jkg}^{-1}\text{K}^{-1}) \quad (\text{daf basis}) \quad (16)$$

Where $1/a = \sum_1^5 y_i / u_i$ ($u_i=12,1,16,14,32$), y_i is the mass fraction of C,H,O,N,S of the coal

charge on daf basis. The meaning of function g_1 could be referred to literature [8]. If ash and moisture in coal are included, the specific heat is given by

$$Cp_c = \sum_1^3 w_i Cp_i \quad (i=\text{ash, daf coal, moisture}) \quad (17)$$

Voidage of coke-oven charge

An important factor affecting flow resistances is the charge voidage fraction available for gas flow. The approach adopted in the present model was to assume that, in terms of the resistance to the gas flow predicted by the Ergun equation, all of the charge can be regarded as a bed of particles of constant diameter 0.002 m. The differences in flow resistance assumed to exist are reflected by variations in the effective porosity with position in the charge. The variation in porosity was chosen arbitrarily to correspond to a minimum value in the plastic layer. The values are polyfitted using the data based on literature [5].

$$\varepsilon_g = \max(0.1, -3.43958 + 0.024T_c + 5.20E - 5T_c^2 + 4.52E - 8T_c^3 + 1.36E - 11T_c^4) \quad (18)$$

In the above equations from (6) to (18), some are functions of the daf-basis composition of coking coal. The daf composition depends on the coking time for the occurrence of chemical reactions. And thus these compositions are evaluated by mass balance with coking process going on.

Boundary Conditions

Gas flow boundary conditions

Boundary conditions on the gas flow are expressed in terms of the flow rate and can be of two types:

1. At a surface open to the atmosphere, the pressure is fixed to zero.
2. The boundary condition of the centre line of the oven (the line $x=0$ in this paper) is set as symmetry boundary conditions.
3. For the lump solid phase, the temperature of the coke-oven wall is fixed to 1473K and for the gas phase, the coke-oven wall is adiabatic.

3 Method of Solution

The model is solved numerically using PHOENICS. All calculation is performed using a Cartesian grid of 215 y-directional and 22 x-directional divisions. The size was found to be acceptable by grid dependence test. Grids layout is illustrated in Fig. 1.

4 Results and Discussions

The calculation uses geometrical and operational data from a coke oven of inner volume 25.2m^3 operating at a charge of some 17 ton. Operational data and bulk solid properties are described in Table 2.

Tab. 2 Data for calculation

<u>Coal Specification (wt%, daf basis)</u>				
C	H	O	N	S
87.6	5.1	5.3(diff)	1.4	0.6
<u>Charge conditions</u>				
Moisture content (%)	Volatile matter (%)	Ash yield (%)	Dry bulk density (kg/m ³)	Temperature (k)
4.4	33	11.0	600.0	298
<u>Oven Specification</u>				
Oven Width(m)	Oven height(m)	Oven length(m)		
0.45	4.3	13.0		

The coking process lasts some 13~14 hrs. With the model proposed above, it's possible to describe the whole process. Initial condition of simulation is that at the start instant, the temperature of every grid in the geometry is 373K, the gas composition is 100% water vapor with velocity magnitude being 0.0m/s. And initial composition of the coal is listed in table 2. These assumptions are nearly 2h after charge.

The activation energy of each species in COG is listed in table 3. Also in table 3 is the final yield of each species in COG which is evaluated with the method in literature [2]. The frequency factor in Eq. (8) is 3300.0 for all species in COG.

Tab. 3 Parameters values used in Eq. (8)

	CH ₄	C ₂ H ₆	CO	CO ₂	TAR	H ₂	H ₂ O	NH ₃	H ₂ S
E/(kJ/mol)	60.98	48.79	48.79	48.19	57.42	62.05	37.42	50.57	57.82
M ₀ /(kg/kg coal)	29.75	4.93	7.31	3.34	109.93	5.09	9.22	0.20	0.74

Heat Transfer and Temperature profiles in charge

The calculated temperature history profiles of the charge are shown in Fig. 2. In this and subsequent figures, only half of a vertical cross-section of the coke oven is shown. The model reproduces the main features of the measured profiles and shows reasonable agreement with data on coke-oven charge temperature histories. However the source term in Eq. (2) includes only the heat source of water boiling. The ignorance of reaction heat of volatile matter releases leads to temperature at the charge centerline increases more rapidly than the measured and the data reported^[1].

Fig.3 shows the history of the change of voidage of the charge. it has a close relation with the COG gas flow pattern which will be discussed in the following section. As shown in Fig. 3, the

voidage fraction changes much more rapidly near the coke oven wall than in the center.

Evolution of COG

The predictions are illustrated as COG paths and y- direction velocity profiles in Figure 4 for the different stages from 2h to 10h after charging. At the beginning of coking (2 hours after charging), COG escapes the coke oven from top surface of charge around the central line and little gas escape along the wall. At time from 3h to 4h after charging, the gas flow is clearly divided into two parts: One flow of gas cross the 'cool' side of the charge (near the center line). Most of COG escapes from this region. And the other, a small part of COG escapes from the channel at the wall. From 5h after charging on, the flow pattern of gas still changes. The main COG-crossing region is slowly moves from the region near the center line to the channel at the wall. Before the stage at 6h after charging, the magnitude of y-directional velocity increases quickly and after that instant, the magnitude of y-directional velocity decreases slowly. After the coking time reaches 10h., there is scarcely any COG evolving from the charge. The main changes then are resolidification, contract of semi coke and charge temperature increasing.

The reason of the COG flow pattern can be explained in terms of the voidage distribution. COG produced mainly in plastic layer will tend to move towards the nearest open surface the top of the oven and the channel at the wall. The area of low voidage region and hence high resistance between the COG and water vapor producing region and the surface impedes this natural movement of the gas. However, points in the plastic layer are near two open surfaces, viz. the top and the wall, and it's reasonable to expect a large portion of COG to cross the plastic layer here than elsewhere.

Fig. 5 shows the pressure drop in the coke oven at different coking stage. Pressure drop of the coke oven somewhat reflects the COG producing rate. From Fig. 5, it clearly shows that at time form 5h to 6h after charging, the pressure drop reach its maximum and hence, in this period, the charge release a large amount of COG.

Investigation the composition of COG shows that at initial stage, the main component of COG is water, which occupies 90 %(mass fractions). The two other main compositions (H_2 and CH_4) are small. The gas flow pattern at 5h after charging is illustrated in Fig. 4. In this stage, COG mainly escapes from the channel at the wall while there is only a very small COG flow near the center line. In this stage, the components of COG are mainly H_2 and CH_4 , Fig. 6 illustrated the gas phase mass fractions of methane at different coking stages, and other gas components could also be investigated in a similar way. Indicated from Fig. 7, at time from 2h to 3h after charging, moisture in the coal charge evaporates quickly and thus the main composition in the gas phase is water vapor. While at 5h~6h after charging, other COG species releases vehemently. CH_4 mass fraction in some regions in the oven at 3h is higher than at 4h and 5h. This is due to the producing rate of COG. As illustrated in Fig. 2, in time from 3h the coal temperature is low except at region near the wall.

At last, the temperature of COG is investigated. The temperature of COG has a close relation to the temperature of coal charge. The gas temperature profiles at different stages of coking process are illustrated in Fig. 8. Gas temperature in the coke oven at 3h is much higher than those at the following stages such as (4h, 5h and 6h). This is due to the evolving rates of COG at corresponding coking stages and the ignorance of convective heat transfer between gas and coal.

5 Conclusions

A CFD model to describe COG flow pattern and coal carbonization behavior in the coke oven has been developed using PHOENICS CFD package. For typical operating conditions, the model predicts that the major chemical reactions and physical structures modification within the charge. It's expected that this model is an appropriate base from which to include full multiphase interaction as well as chemistry.

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Nomenclature

C_p : specific heat (J/kg.K)

D : diffusivity ($1/m^2.s$)

d_s : mean particle size of coal charge (m)

C_1, C_2 : constant

\tilde{E} : heat source (W/m^3)

E : activation energy (kJ/mol)

\tilde{F} : drag force (N/m^3)

G : mass velocity of gas phase (kg/s)

H : enthalpy for gas (J/kg)

k : thermal conductivity of coal charge ($Wm^{-1}k^{-1}$)

$m_{0,i}$: final yield of species I of volatile matter (kg/m^3)

$m_{t,i}$: yield of species I of volatile matter at instant t (kg/m^3)

P : pressure (pa)

\tilde{R} : rate of release of COG component i

R : gas constant

T : temperature of gas or coal (k)

t : time (s)

u : x directional velocity component of gas phase (m/s)

v : y directional velocity component of gas phase(m/s)

w : component mass fraction in coal charge

x : x directional coordinate (m)

y: x directional coordinate (m)

Greek Symbols

Ψ :general dependent variable in Eq. (1)

Γ_{ψ} :diffusion coefficient for ψ in Eq.(1)

μ :viscosity (Pa/s)

λ : thermal conductivity of gas

ρ : density (kg/m³)

ε :volume fraction of gas, demensionless

Subscripts

g: gas

c: coking coal

i: COG gas speciest

t: instant

x: x direction

y: y direction

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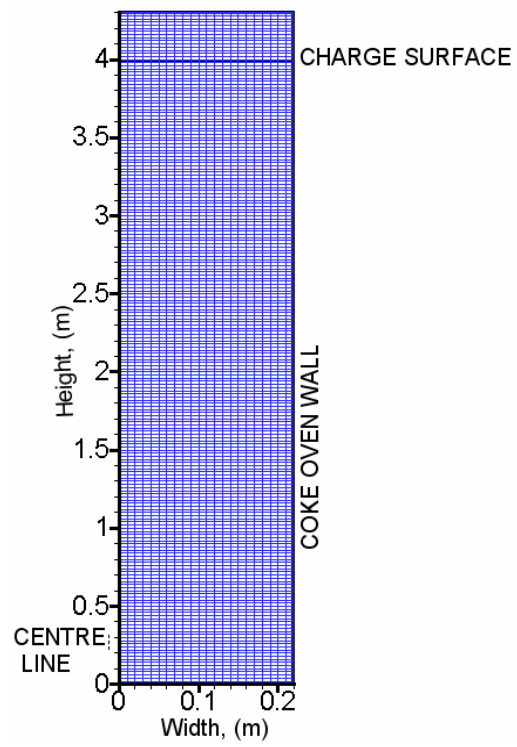


Fig. 1 22×215 grids used for the calculation

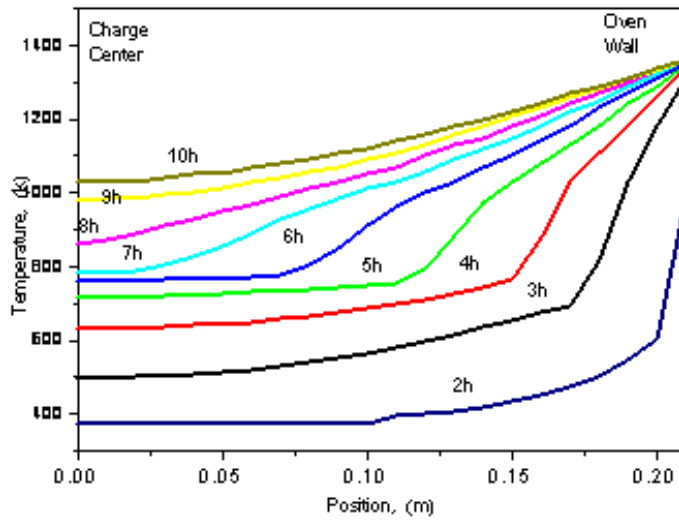


Fig. 2 History of charge temperature profiles

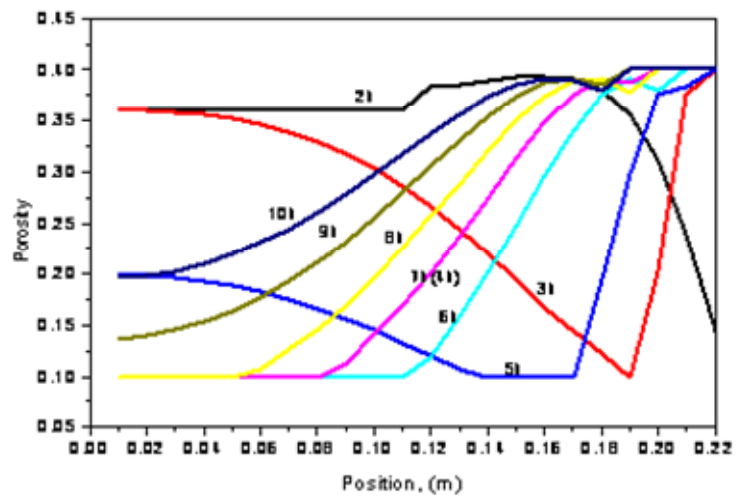


Fig. 3 History of charge porosity profiles

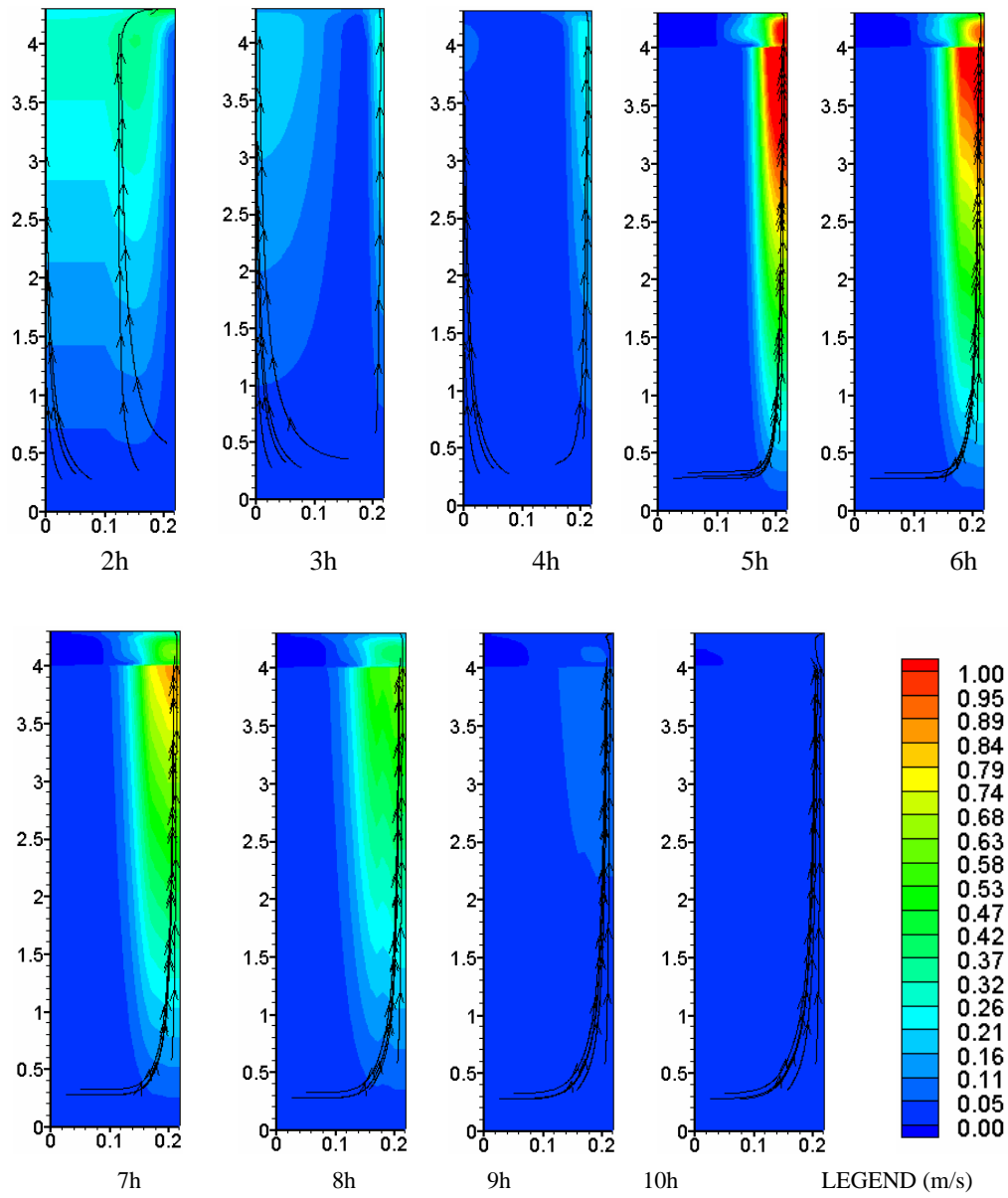


Fig. 4 Y-directional velocity contours and streamlines patterns at different coking stage

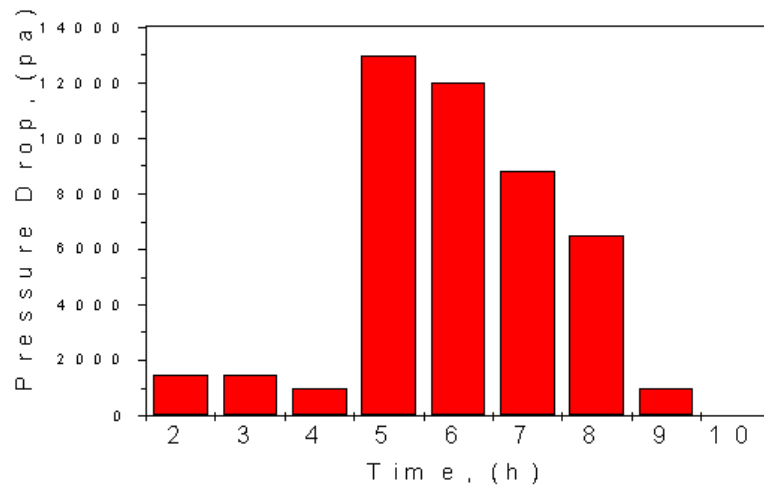


Fig. 5 Pressure drop from the bottom of coke oven to the charge surface at different stages

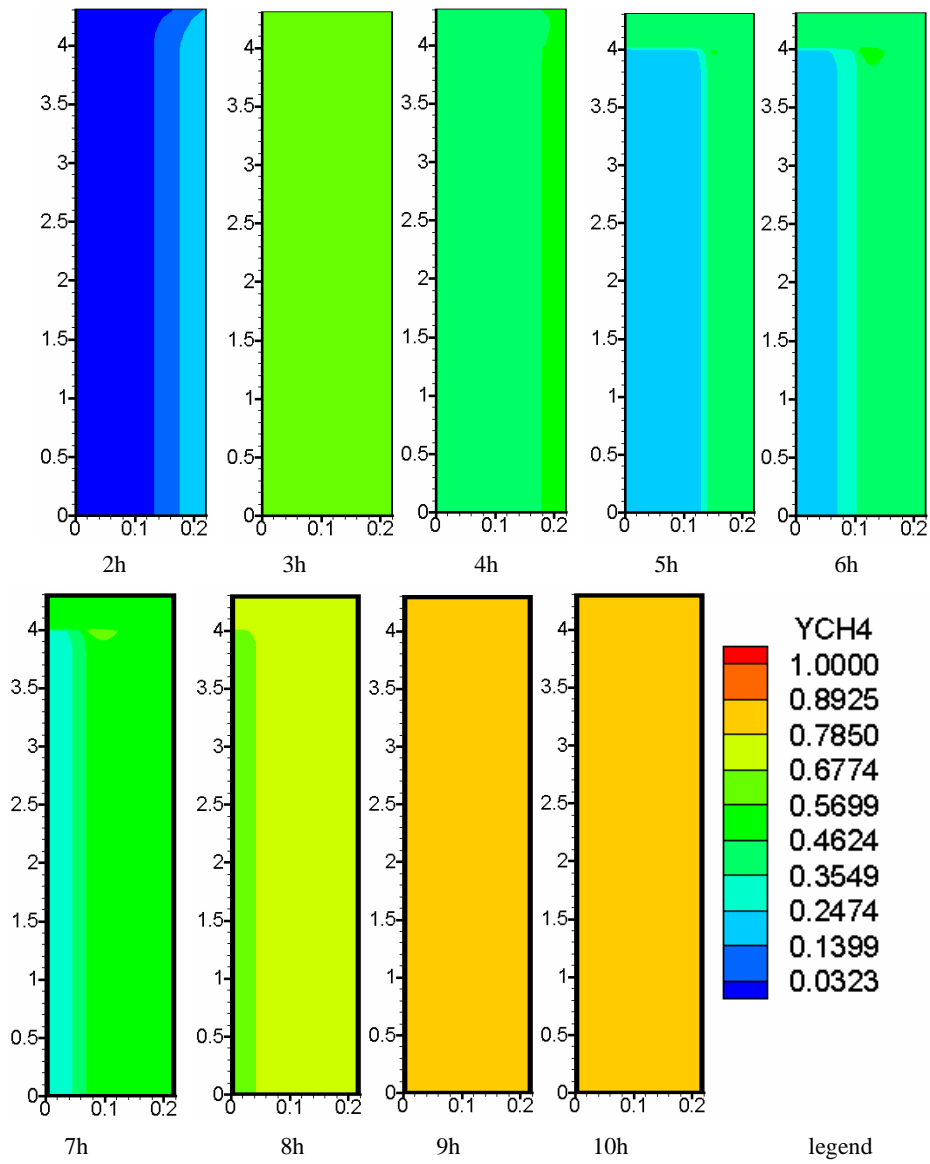


Fig. 6 CH₄ gas phase mass fraction at different stages of coking process

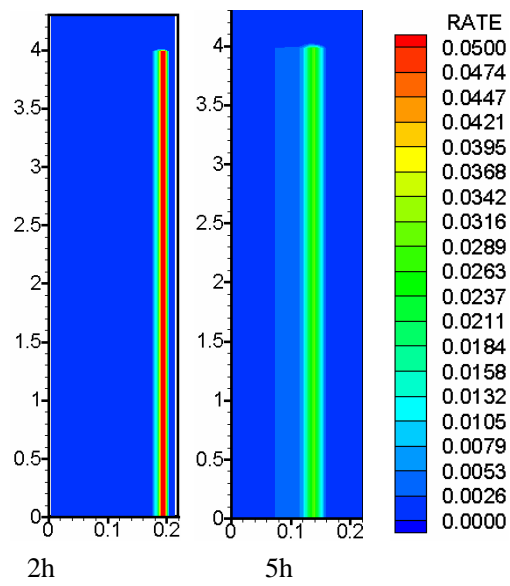


Fig. 7 water-evaporating rate at different coking stages

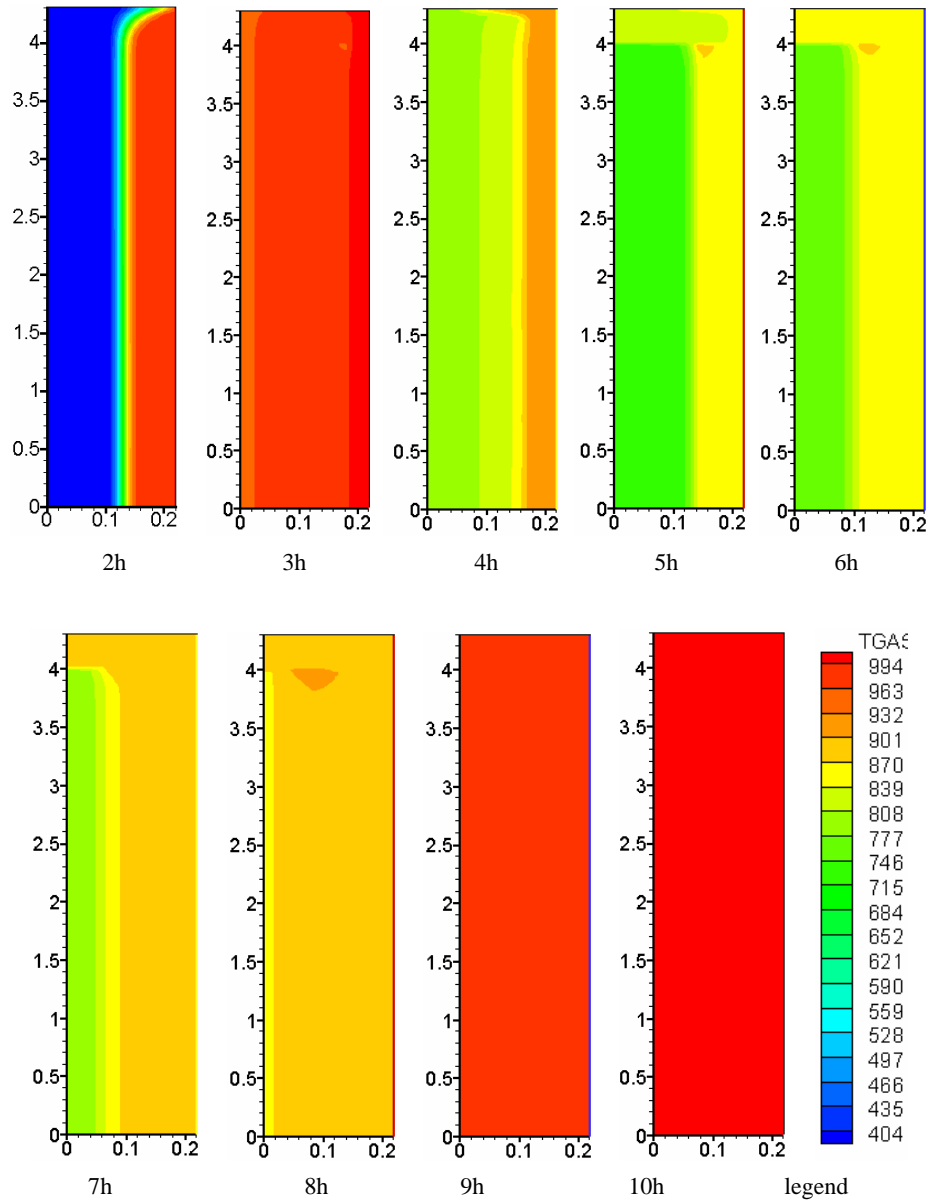


Fig. 8 Gas temperature profiles at different stages of coking process