

archives  
of thermodynamics

Vol. 34(2013), No. 2, 39–51

DOI: 10.2478/aoter-2013-0009

## Transient one-dimensional model of coal carbonization in a stagnant packed bed

SYLWIA POLESEK-KARCZEWSKA<sup>1\*</sup>  
DARIUSZ KARDAŚ<sup>1</sup>  
IZABELA WARDACH-ŚWIĘCICKA<sup>1</sup>  
ARKADIUSZ GRUCELSKI<sup>1</sup>  
SŁAWOMIR STELMACH<sup>2</sup>

<sup>1</sup> The Szewalski Institute of Fluid-Flow Machinery, Polish Academy of Sciences, Fiszerza 14, 80-231 Gdansk, Poland

<sup>2</sup> Institute for Chemical Processing of Coal, Zamkowa 1, 41-803 Zabrze, Poland

**Abstract** In the present paper, the one-dimensional model for heat and mass transfer in fixed coal bed was proposed to describe the thermal and flow characteristics in a coke oven chamber. For the purpose of the studied problem, the analysis was limited to the calculations of temperature field and pyrolytic gas yield. In order to verify the model, its theoretical predictions for temperature distribution during wet coal charge carbonization were compared with the measurement results found in the literature. In general, the investigation shows good qualitative agreement between numerical and experimental data. However, some discrepancy regarding the temperature characteristics at the stage of evaporation was observed.

**Keywords:** Carbonization; Coking process; Coal charge evaporation; Mass source; Pyrolytic gases

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\*Corresponding author. E-mail: sylwia@imp.gda.pl

## 1 Introduction

Modeling of solid fuel carbonization is one of the most significant steps in predicting and optimizing the technologies based on thermochemical conversion of solid organics. Apart from the heat and power production sector, which is focused on combustion and gasification processes, it is of general interest for chemical, coke and metallurgy industry. The ironmaking technology and the coking process itself, although known since decades, need further improvements to predict and enhance coke quality in order to meet the technical requirements and thereby improving the efficiency of the process. From practical point of view, the general aim for the coking process is to achieve the appropriate thermal conditions in the coal charge so as to obtain a fully coked product with the energy input as low as possible.

To carry out the reliable simulation of the process, it is necessary to thoroughly identify and describe the physical and chemical phenomena that occur during carbonization. Many studies have been done in order to model thermal decomposition of coal during coking process [1–3]. The mathematical modeling of coking process is not a trivial problem. The fact that the complete model needs to take into consideration both thermochemical and flow processes, that differ in time and space scale, makes the problem quite complex. Moreover, the additional difficulty is the lack or the discrepancy between the physicochemical properties of coal reported in the literature. This includes, for instance, thermal conductivity, specific heat or heat of pyrolysis [3–8].

The present article focuses on heat and mass transfer characteristics for the coking process. The numerical calculations were performed basing on the in-house program for the one-dimensional model. Obtained results of theoretical predictions regarding the temperature distribution in the coke oven chamber were compared with the experimental data found in the literature.

## 2 Model description

Due to the complexity of the coking process and its multiscale nature both in time and space, its description requires sophisticated mathematical models and long-lasting numerical calculations. A typical linear size of the coal particle in a coke oven chamber is around 3 mm, while the linear dimensions of a typical coke oven are of order from 0.5 m up to several meters. A dense

numerical grid must account for the smallest particles, and regarding the three-dimensionality of the particles and a coke oven chamber, the problem solution would need to discretize the computational area to the number of cells exceeding  $10^9$ . In addition, as it is worth to mention, perturbations in gases propagate with the velocity of an order of  $10^2$  m/s, whereas the mass flow of gases is about  $10^{-3}$  m/s, and the velocity of the heat propagation is  $10^{-6}$  m/s. Like the spatial and velocity scales, the time scales of the coking phenomena are also very large. The time of chemical reaction is of an order of  $10^{-10}$  s while the total coking process takes about 20 h. It brings the conclusion that three-dimensional transient simulation of the analysed process is beyond current computing possibilities.

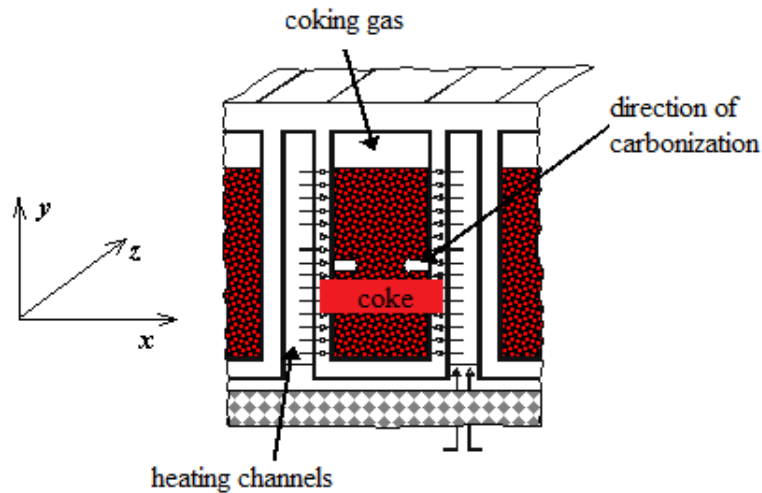


Figure 1. The schematic of coke oven chamber.

Therefore, the simplifications of the examined process seem to be necessary and desirable. Use of a one-dimensional transient model is mainly justified by the high aspect ratio of an oven chamber and, on the other hand, by the fact that it is heated from the side walls. Figure 1 shows the cross-section of the chamber, while Fig. 2 the typical temperature change in the coal/coke charge [9]. As it is clearly seen from the latter, in the first stage of the coking process the coal charge undergoes vaporization, when the moisture contained in the coal particles is released. This transition is manifested by the temperature increase from the ambient to  $100$  °C in the whole charge during first 2–3 h. Furthermore, it is generally observed that the rate of

heat transfer in the coal charge does not exceed 0.02 m/h in average. All

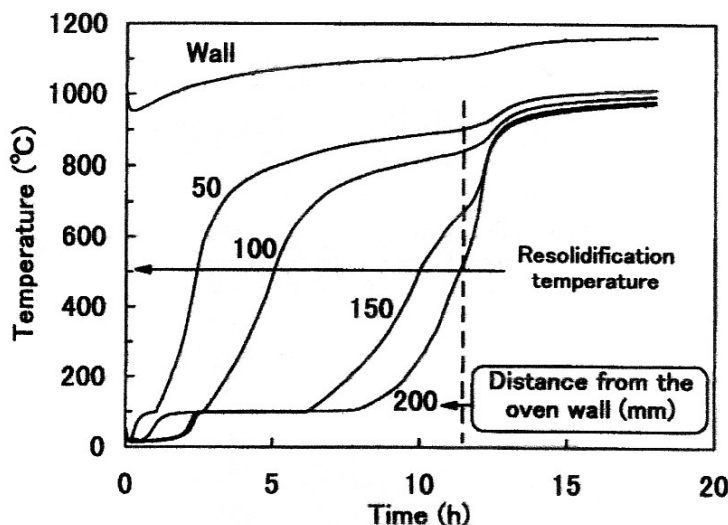


Figure 2. Typical temperature profiles in the carbonized coal charge [9].

parameters of the process mainly depend on the  $x$  direction and are constant along the  $y$ -axis. This situation results from homogeneous heating of both chamber walls, which generates relatively low temperature variations. However, some phenomena, e.g., the formation of cracks (basically in the horizontal plane) that create channels for the gas flow through the coke volume are two-dimensional at least.

The proposed model is based on the following assumptions:

- model is one-dimensional (1D),
- processes in the solid phase are considered to be nonstationary,
- flow of gases is stationary,
- pyrolysis process is dependent on current temperature and heating rates.

## 2.1 Governing equations

Considering the volume fraction of the phase being determined by  $\epsilon$  [ $\text{m}^3/\text{m}^3$ ] and introducing subscripts  $c$ ,  $g$  and  $w$ , referring to the quantities characterizing coal, pyrolytic gas and water (moisture), respectively, and following the assumptions given above the one-dimensional coal carbonization process is described by:

- mass balance equation for solid phase (coal particles)

$$\frac{\partial(\epsilon_c \rho_c)}{\partial t} = -W_{cg}, \quad (1)$$

where  $\rho$  [kg/m<sup>3</sup>] is the density,  $t$  [s] is the time and  $W_{cg}$  [kg/m<sup>3</sup>s] is the mass source term for pyrolytic gases generation;

- mass balance equation for moisture

$$\frac{\partial(\epsilon_w \rho_w)}{\partial t} = -W_{wg}, \quad (2)$$

where  $W_{wg}$  [kg/m<sup>3</sup>s] represents the mass source term for vapor;

- mass balance equation (stationary) for gas phase (pyrolytic gases)

$$\frac{\partial(\epsilon_g \rho_g v_g)}{\partial x} = W_{cg} + W_{wg}, \quad (3)$$

where  $v_g$  [m/s] is the velocity of pyrolytic gases released from coal;

- stationary momentum balance for gases

$$\frac{\partial(\epsilon_g \rho_g v_g v_g)}{\partial x} = -\frac{\partial(\epsilon_g p_g)}{\partial x} - \frac{\mu_g \epsilon_g v_g}{k}, \quad (4)$$

where  $k$  [m<sup>2</sup>] is the bed permeability and  $\mu_g$  [kg/(m s)] is gas dynamic viscosity;

- energy balance for the packed bed

$$\rho c_v \frac{\partial T}{\partial t} + \frac{\partial(\epsilon_g \rho_g c_{vg} v_g T)}{\partial x} = -\frac{\partial q}{\partial x} + W_{cg} u_{cg} + W_{wg} u_{wg}, \quad (5)$$

where the second term on the left side represents the convection of pyrolytic gases;  $T$  [K] denotes bed temperature,  $u_{cg}$  and  $u_{wg}$  [J/kg] are, respectively, the heat of pyrolysis and the heat of vaporization, whereas  $q$  [W/m<sup>2</sup>] is the heat flux density. Symbols  $\rho$  [kg/m<sup>3</sup>] and  $c_v$  [J/kgK] without additional subscripts are the average (values effective) for coal charge (bed), density and specific heat, respectively, and are defined as follows:

$$\rho = \epsilon_c \rho_c + \epsilon_g \rho_g + \epsilon_w \rho_w,$$

$$c_v = \frac{1}{\rho} (\epsilon_c \rho_c c_{v,c} + \epsilon_g \rho_g c_{v,g} + \epsilon_w \rho_w c_{v,w});$$

- evolution equation for heat flux density,  $q$ ,

$$q + \theta \frac{\partial q}{\partial t} = -\lambda \frac{\partial T}{\partial x}, \quad (6)$$

where  $\theta$  [s] is the nonzero time required for the system to achieve the equilibrium state. Coefficient  $\lambda$  [W/(mK)] is the effective thermal conductivity of coal charge expressed by

$$\lambda = (\epsilon_c \lambda_c + \epsilon_g \lambda_g + \epsilon_w \lambda_w) + \lambda_{rad} = \lambda_{cond} + \lambda_{rad},$$

where  $\lambda_{rad}$  represents thermal conductivity resulting from radiation [10, 11], and in addition,

- the equations for production of chemical gas mixture components

$$\frac{dY_i}{dt} = k_i (Y_{0,i} - Y_i), \quad (7)$$

where  $i$  represents the basic gas mixture components, such as H<sub>2</sub>O, H<sub>2</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, BTX, H<sub>2</sub>S, NH<sub>3</sub>. Parameter  $k_i$  [1/s] is the release rate for  $i$ th component,  $Y_{0,i}$  and  $Y_i$  [kg/kg] are the mass fractions of  $i$ th gas mixture component, the maximum and the current (i.e., for given time/temperature) value, respectively.

Mass source term for gases is determined basing on the relationship

$$W_{cg} = -k_c \epsilon_{c,0} \rho_{c,0} (Z - Z_e), \quad (8)$$

where  $Z$  [kg/kg] is the pyrolysis progress being defined as the ratio of coal sample mass decreasing in result of thermal decomposition to its initial mass,  $Z = m_c/m_{c,0}$ . Parameter  $k_c$  [1/s] is the rate of carbonization. Subscript 0 means the initial value and  $e$  refers to the pyrolysis performed for very low heating rates and therefore assumed to be near equilibrium.

## 2.2 Description of moisture evaporation

In the conventional coke oven charging system the moisture content of coal does not exceed 10% wt. The wet coal charged into the coke oven chamber is immediately exposed to high heating rates and in result the moisture evaporates. Generally, as it is proved by the industrial- and laboratory-scale

measurements, the coal charge is being dried in the coke oven center, reaching temperature of 100 °C in time less than 2–3 h [9, 15, 16]. Considering the high value of heat of vaporization,  $u_{wg} = 2.2$  MJ/kg, it becomes clear that too high moisture content decreases the efficiency of the process due to the energy input demand higher in comparison to the process when dry coal charge is carbonized. As it was shown for instance in [15], where the temperature variations in the coke oven chamber center for wet and pre-heated charge were presented, the moisture content of ca. 8% wt prolongs the total coking time by up to nearly 2 h.

The local heat balance for generation of steam, moving across the surface area  $A$  (evaporation front) during the time period  $dt$  may be expressed as

$$Q_{wg} = q_{wg} dt A = m_w h_{wg} , \quad (9)$$

where  $m_w$  [kg] stands for mass of water,  $h_{wg}$  [J/kg] represents the enthalpy of the phase change (evaporation), and  $q_{wg}$  [W/m<sup>2</sup>] is the heat flux density for evaporation. Equation (9) after introducing the volume of vaporized water  $dV = A dx_w$  and rearrangement, yields the volumetric vapor mass flow rate

$$\frac{q_{wg}}{dx_w h_{wg}} = \frac{m_w}{dt dV} = W_{wg} . \quad (10)$$

In the investigated coking case it might be assumed that evaporation takes place under constant pressure, and thus  $h_{wg} = u_{wg}$ . Continuing, it follows that the evaporation occurs locally until  $q$  in Eq. (5) is less than  $q_{wg}$  defined by Eq. (10). Otherwise, the heat input is used for heating up dry charge.

### 3 Problem solution

To solve studied problem, the mixed explicit-implicit numerical scheme was applied. Equations for mass balances for each phase, as well as the momentum balance equation for gas were solved explicitly, whereas for calculations of heat transfer the implicit Crank-Nicolson algorithm was used, [12]. The finite-difference representation for the energy balance equation, Eq. (5), takes the following form:

$$\begin{aligned}
 (\rho c_v)_i^n \frac{T_i^{n+1} - T_i^n}{\Delta t} + \frac{(\epsilon_g \rho_g c_{vg} v_g T)_E^n - (\epsilon_g \rho_g c_{vg} v_g T)_W^n}{\Delta x} = \\
 - \left[ \frac{q_E^{n+1} - q_W^{n+1}}{2\Delta x} + \frac{q_E^n - q_W^n}{2\Delta x} \right] + (W_{cg} u_{cg})_i^n + (W_{wg} u_{wg})_i^n , \quad (11)
 \end{aligned}$$

where  $i$  denotes the cell number, and  $n$  and  $n + 1$  are the time steps, and subscripts  $E$  and  $W$ , denote east and west cell boundary, respectively. The heat flux densities at the cell boundaries in the time step  $n + 1$ ,  $q_E^{n+1}$ , and  $q_W^{n+1}$ , are obtained using the finite-difference approximation for the Eq. (6)

$$\frac{q_E^{n+1} - q_E^n}{\Delta t} + \left(\frac{\lambda}{\theta}\right)_E^n \left[ \frac{T_{i+1}^{n+1} - T_i^{n+1}}{2\Delta x} + \frac{T_{i+1}^n - T_i^n}{2\Delta x} \right] = -\frac{q_E^{n+1} + q_E^n}{2\theta_E^n}, \quad (12)$$

$$\frac{q_W^{n+1} - q_W^n}{\Delta t} + \left(\frac{\lambda}{\theta}\right)_W^n \left[ \frac{T_i^{n+1} - T_{i-1}^{n+1}}{2\Delta x} + \frac{T_i^n - T_{i-1}^n}{2\Delta x} \right] = -\frac{q_W^{n+1} + q_W^n}{2\theta_W^n}. \quad (13)$$

Accounting for the symmetry of analyzed problem, the numerical simulation was performed for half width of the coke oven chamber with the symmetry condition employed at the center line.

## 4 Results and discussion

The numerical calculations for coal charge having the moisture content at the level of 3% were performed to verify the model predictions with the experimental data reported by Nomura *et al.* [9]. The temperature profile at the wall, presented in their work, was used as a boundary condition for the numerical simulation. The width of coke oven charge was set to 0.4 m. As it was mentioned above, the determination of the source/sink term being introduced to the balance equation, Eq. (1), requires the equilibrium pyrolysis progress  $Z_e$  and the devolatilization rate to be known. The first one was defined by temperature-dependent function approximating the thermogravimetric data for slow pyrolysis case (i.e., for low heating rates), whereas the second was expressed by the formula [14]

$$k = A \exp\left(-\frac{e_a}{T - T_0}\right), \quad (14)$$

where  $A$  and  $e_a$  are defined by functions dependent on coal volatile content  $Y_v$

$$A = -1107 + 2.1862Y_v - 2.0396Y_v^2, \quad (15)$$

$$e_a = 2221.8 + 22939.31Y_v - 713.05Y_v^2. \quad (16)$$



In the studied case the thermogravimetric data for Budryk coal having the volatile content  $Y_v$  at 31.18% were taken into consideration. The details regarding the data approximation for slow pyrolysis are presented elsewhere [13]. The physico-chemical properties used for calculation are summarized in Tab. 1.

Table 1. Properties of coal charge assumed for calculation.

Parameter	Data
$\epsilon_g$	0.45
$\theta$	900.0 s
$\rho$	$\rho = \rho(T)$ , [10]
$\lambda_{cond}$	$\lambda = 0.0031 \exp(0.063T)$ , basing on data from [1]
$c_{v,c}$	Merrick, [4]
$c_{v,g}, \rho_g, \lambda_g$	data for air, [17]

Figure 3 shows temperature distribution in the wet coal charge undergoing carbonization. The total time of coking process in this case is less than 18 h. The comparison between experimental data and numerical results for temperature profiles in two distances from the coke oven wall, at  $x = 0.2$  m (the oven center) and at  $x = 0.1$  m was presented in Fig. 4.

It is clearly seen that experimental data and model predictions are in good qualitative agreement. The discrepancy is observed in the region of evaporation. The time for the total duration of charge drying was predicted to be much longer than it was indicated by the measurements (see Figs. 4 and 5). However, the predicted temperature increase within the coal charge following the evaporation converge with the experimental data.

It is worth to point out the variations in velocity of thermal front during the process. Taking into consideration the model predictions, it is nearly 0.04 m/h for temperatures below 100 °C, about 0.01 m/h during the main process stage which is between 100 and 800 °C. At the end stage, i.e., after 13th hour, when the devolatilization in the coal charge is being completed, the heat transfer speeds up to 20 cm/h.

Figure 6 shows the volumetric mass source for pyrolytic gases within the coal charge in the coke oven chamber of given geometry. It might be seen, as expected, that its intensity is moving forward along the coke oven chamber with the heat wave (temperature increase) and achieves its highest value (0.024 kg/m<sup>3</sup>s) when the whole charge volume achieves the temperature of 1000 °C.

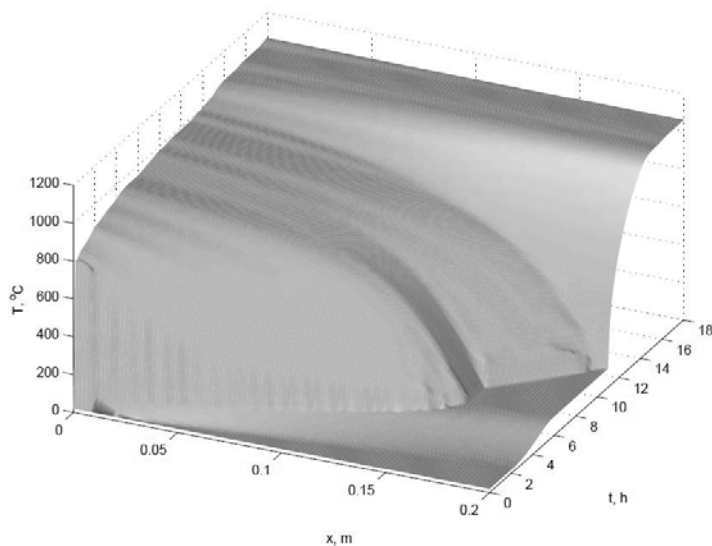


Figure 3. Temperature predictions for wet coal charge ( $\epsilon_w = 3\%$ ).

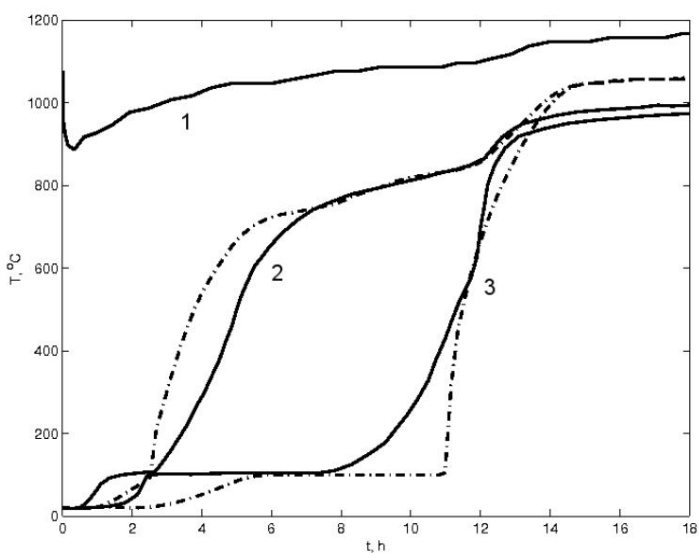


Figure 4. Temperature profiles for wet coal charge ( $\epsilon_w = 3\%$ ): 1 – oven wall, 2 – 0.1 m from the wall, 3 – centerline; solid line (experimental [9]), dashed line (model predictions).

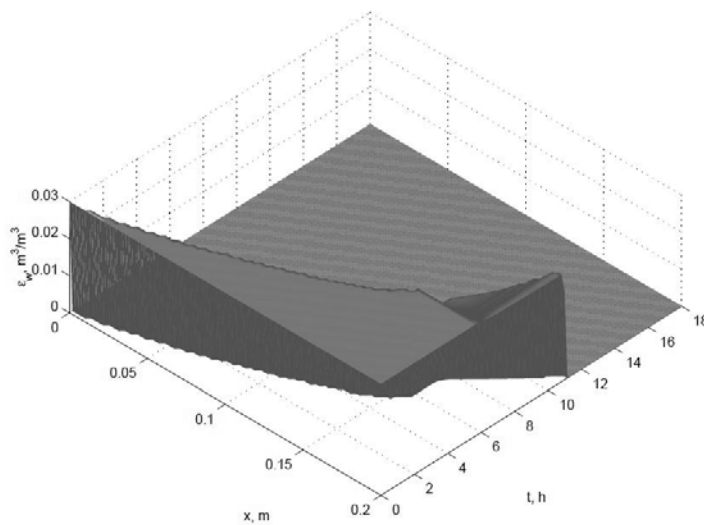


Figure 5. Change in the moisture content within the coal charge during coking – model predictions.

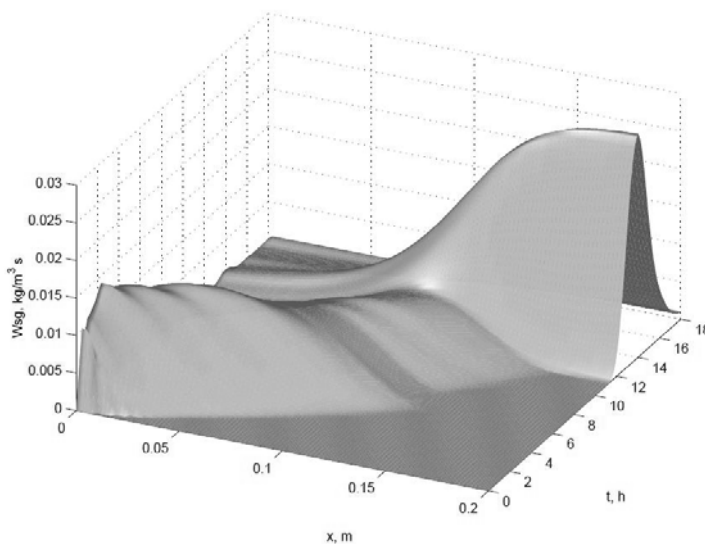


Figure 6. Gas generation within the coal charge during coking – model predictions.

## 5 Final remarks

From practical point of view, numerical simulations of coal carbonization seems to be of general importance in predicting the thermal behavior of

coal charge and in result, the quality of the product (coke) itself. Since the quality of the coke, basically its strength and porosity, and the process effectiveness is strongly affected by temperature distribution in the coal charge, there is a large interest in developing the reliable model helpful in estimation of basic process parameters and requirements for the input coal charge. The performed analysis shows that temperature predictions obtained using the proposed herein model are to a large extent convergent with experimental data. The disagreement with the measurement data yielded for the temperatures below 100 °C representing the stage of evaporation, which is proved to have considerable effect on thermal process characteristics, indicates on the need for further model improvement. To complete the description the model should also account for the effect resulting from the condensation of vapor entering the parts of charge being at the temperature less than 100 °C. This is the task for the nearest future. Nevertheless, the developed model might be considered as promising tool for application in simulating thermochemical processes of solid fuels, such as coal carbonization, and not only.

**Acknowledgements** The work presented in the paper has been performed within the National Project POIG.01.01.02-24-017/08 “Smart Coke Plant Meeting the Requirements of Best Available Techniques” cofinanced by the European Regional Development Fund (ERDF) and supported by Strategic Program “Advanced Technologies for Energy Generation” co-financed by the National Centre for Research and Development.

*Received 8 March 2013*

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