LBM estimation of thermal conductivity in meso-scale modelling

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Abstract. Recently, there is a growing engineering interest in more rigorous prediction of effective transport coefficients for multicomponent, geometrically complex materials. We present main assumptions and constituents of the meso-scale model for the simulation of the coal or biomass devolatilisation with the Lattice Boltzmann method. For the results, the estimated values of the thermal conductivity coefficient of coal (solids), pyrolytic gases and air matrix are presented for a non-steady state with account for chemical reactions in fluid flow and heat transfer.

1. Introduction
Modelling of coupled phenomena that occur in complicated and random geometry has recently become a subject of intensive research in fields of fluid mechanics and chemical engineering. To obtain a detailed description for the number of technological processes one has to undertake the interdisciplinary research of the physico-chemical phenomena occurring in the multi-level geometry. Such approach gives the opportunity to bring information obtained through the micro-scale description to the macro-level as a closure relationship. For example: effective values of physico-chemical properties describing transport processes occurring in the representative element of volume (REV) along with modelling of coupled phenomena.

In the coal-processing industry, coking plants are widely used to obtain chemically cleaner coal (coke) with side products like benzene, ammonium hydroxide and other chemical species (generally known as coal gas) as well as tar. From the fluid thermomechanics point of view, coking occurs in a bed of granular material (coal grains) that upon heating becomes a homogeneous porous medium. During the coking process, complex phenomena occur at the level of a single grain (pore), including fluid flow through the deposit, thermal conduction and convection, chemical reactions (heterogeneous as well as homogeneous), plastic deformation and others. For accurate modelling of the coupled subprocesses above the micro level, detailed description of effective transport coefficients is essential. For this purpose more traditional CFD methods become far too expensive because of the random geometry and phenomena occurring at multiple scales. All this has to be modelled at the pore level, with additional change of the pore geometry (due to change of size and shape of coal grains) [1]. The common way for devolatilisation modelling is to provide simulation at the averaged level, usually solving the most important phenomena. A number of recent publications review the numerical tools used for simulation of the coking or pyrolysis process at the averaged level; a good example of such a description is [2]. Averaged simulation results are also used for general process description [3].
For the purpose of modeling of fluid thermomechanics we utilize the lattice Boltzmann method, recently enjoying an increasing interest as a numerical tool, especially for modeling the fluid thermo-mechanics in complex geometry. It is based on the Boltzmann equation with subsequent discretisation [4, 5]. The method has proven to be suitable for simulation of viscous and nearly incompressible fluid flow in a simple [6] and complex geometry [7, 8], as well as heat transfer [9, 10], with addition of chemical reactions [11]. In the context of practical application, Asinari et al [12] report the LBM modelling of complex phenomena occurring in fuel cells at single pore level for prediction of the overall system behaviour. Modelled process is treated here at the level of meso-scale and complex phenomena occurring at the micro-level (like transport processes inside the coal particle and related chemical reactions) are simplified. The LBM was adopted for determination of the effective heat conductivity for heat transfer in complex geometry. An example of such work is [7]; the authors use LBM to compute effective thermal conductivity of randomly created porous foams.

In this work, we present results for estimation of effective heat conductivity obtained for 2D bed of coal grains, where coal is modelled by uniform cylinders. Fluid thermomechanics as well as chemical specie evolution is modelled by LBM and a brief description of the numerical tool is provided in section 2. In the same section, simplified one specie chemical model of devolatilisation along with temperature dependence of the material properties is described. Before conclusion, results obtained for random geometry are presented.

2. Numerical model details

Chosen geometry, for this particular case, is the representative element of volume (REV) for the packed bed; this simulates coal particles in coking plants. The REV geometry requires that the size of computational domain be much smaller than the length scale present in typical devices in industry but also much larger than the smallest length in modelled phenomena (here, the diameter of the coal particle).

2.1. Lattice Boltzmann method

The Lattice Boltzmann equation, discretised in time, space (by lattice), and advection velocity ($\mathbf{e}_i$) on a regular square lattice, describes the evolution of a relevant physical field in terms of its distribution function. Description of LBM in this section is valid for D2Q9 discretisation of advection velocity; so in presented simulation approach we consider 2-dimensional lattice with 9 velocities of advection of distribution function. Evolution of density distribution function $f_i$ is described with (see [10]):

$$f_i(r + \mathbf{e}_i t + \delta t) - f_i(r, t) = -\tau_{\nu}^{-1}[f_i(r, t) - f_i^{eq}(r, t)] + Q^f_i,$$

where $Q^f_i$ is a source term, $f_i^{eq}$ represents the equilibrium state of distribution function corresponding to the averaged fields at $(r, t)$; $\tau_{\nu}$ is a non-dimensional relaxation time for fluid flow, it takes the form:

$$\tau_{\nu} = 0.5 + \frac{1}{2} \frac{\nu}{c_s^2 \delta t},$$

here $\nu$ and $\delta t$ are viscosity and time step; $c_s = c/\sqrt{3}$ is the speed of sound in the simulation and $c$ is the lattice speed of advection of distribution functions. In a similar fashion, we track the evolution of temperature with internal energy density distribution function (IEDDF) and the distribution function for chemical species concentration. Respectively for IEDDF we use $g_i$ to describe distribution function and the source term is $Q^g_i$ (in this work, it is accounted only in solid phase as change of enthalpy by chemical reactions occurs only in solid phase). Relaxation time $\tau_{\alpha,m}$ written both for fluid $f$ (gas) and solid $s$ (coal grains) in the computational domain
$(m \in \{s,f\})$ takes a form:

$$\tau_{a,m} = 0.5 + \frac{3}{2} \frac{\lambda_m}{\rho_mC_pmc^2\delta t}. \quad (3)$$

$C_{pm}$ and $\lambda_m$ are the heat capacity and heat conductivity of $m$, respectively. The evolution of chemical specie (being a mixture of gaseous products of devolatilisation) is modelled with use of LBM with additional distribution function. Because of numerical instabilities occurring close to large gradients of macroscopic fields typical of Eulerian methods, Di Rienzo et al. [11] proposed a numerical scheme for chemical species evolution. In presented approach, we track a single specie with distribution function prescribed with $\phi$ (only in fluid domain) and non-dimensional relaxation time:

$$\tau_D = 0.5 + \frac{3D_\phi}{\psi c^2 \delta t}.$$ Modifications proposed by Di Rienzo et al. [11] account for varied density field in the volume of fluid by introducing $\psi = \rho^*/\rho$ a ratio of the minimum density of fluid in the entire domain and the density in a given lattice node; $D_\phi$ is a diffusivity coefficient of the mixture and the source term is prescribed with $Q_i^\phi$.

Equilibrium distribution function for every accounted evolution equation has similar form ($f_i^{eq}$ is usually used regarding fluid flow; here, for presentation purpose, a generic form is presented):

$$f_i^{eq}(r,t) = f_i^{eq}(\beta, \mathbf{u}) = \beta \Omega_i \left( A_i \frac{B_i}{c^2} \mathbf{e}_i \cdot \mathbf{u} + \frac{C_i}{c^4} (\mathbf{e}_i \cdot \mathbf{u})^2 - \frac{D_i}{c^2} \mathbf{u} \cdot \mathbf{u} \right), \quad (4)$$

where $\mathbf{u}$ is the local fluid velocity and $\Omega_i$ are weight coefficients; for D2Q9 $\Omega_{i=0} = 4/9$, $\Omega_{i\in\{1,2,3,4\}} = 1/9$, $\Omega_{i\in\{5,6,\ldots,8\}} = 1/36$. In Eq. (4), coefficients $A_i$ to $D_i$ are not constant and their values are different for various modelled phenomena; furthermore, they can depend on discretisation scheme and directions of advection of distribution function. Full description of equations (with exact arrays of coefficients for Eqs. (1)-(4) for fluid flow and heat transfer) in 2D and 3D can be found in [10] and references therein. Modelling of the fluid mechanics (where subscript $i$ of coefficients in Eq. 4 can be removed) is done with coefficients $A = 1$, $B = 3$, $C = 4.5$, $D = -1.5$ and $\beta = \rho$. Heat transfer modelling requires that $\beta = \theta$ in Eq. (4) and for IEDDF the coefficients, generally, are not constant; namely for D2Q9 discretisation scheme:

$$\{A_i, B_i, C_i, D_i\} = \begin{cases} \{0, 0, 0, -3/2\}, & i = 0 \\ \{3/2, 3/2, 9/2, 1\}, & i = 1 - 4 \\ \{3, 6, 9/2, 1\}, & i = 5 - 9 \end{cases} \quad (5)$$

Utilized equilibrium function for chemical specie transport was first presented and validated in [11] for D2Q5; here we present results for D2Q9 where the equilibrium functions are:

$$\phi_0^{eq} = \frac{\rho Y}{g} (9 - 5\psi), \quad \phi_i^{eq} = \frac{\rho Y}{g} (\psi + 3\mathbf{e}_i \cdot \mathbf{u}), \quad i = 1, \ldots, 4, \quad \phi_i^{eq} = \frac{\rho Y}{36} (\psi + 3\mathbf{e}_i \cdot \mathbf{u}), \quad i = 5, \ldots, 9,$$

here $\phi_i^{eq}$ is the distribution function corresponding to modelled specie evolution.

Macroscopic values (density $\rho$, velocity $\mathbf{u}$, temperature $\theta$, heat flux $\mathbf{q}$ and concentration $Y$) are obtained by integration of respective distribution function [7]:

$$\rho = \sum_i f_i, \quad \mathbf{u} = \rho^{-1} \sum_i f_i \mathbf{e}_i, \quad \theta = \sum_i g_i, \quad \mathbf{q} = \sum_i \mathbf{e}_i g_i, \quad Y = \sum_i \phi_i \quad (6)$$
2.2. Convection
Model of free convection by Guo et al presented in [6] is used here to account for density variation in fluid caused by local temperature gradient in a field of gravity force $g$. Coupling of the fluid flow and heat transport by convection is done by introducing the source term:

$$Q_f^i = 3\Omega_i\alpha\delta t\rho(\theta - \langle\theta\rangle)\frac{e_i\cdot g}{c},$$

where $\alpha$ is heat expansion coefficient for air; $\theta$ and $\langle\theta\rangle$ are local and averaged (in the whole domain) temperature, respectively.

2.3. Pyrolysis gas release
In presented approach, a mixture of gases is modelled; in recently presented works detailed investigation has been taken into account with additional evolution of tar (see [14]). This type of analysis, at the moment, is far too expensive for physically sound simulation at the meso-scale.

For the purpose of simplified modelling, we use an empirical correlation known from literature, in details presented in [15]; for a grain $S$ the total amount of released gas is described by equation:

$$Q_S = \varepsilon\delta t V_S [(\langle\rho\rangle - \rho_0 g(\theta))] f(\varepsilon_S^{\text{vol}}, \theta),$$

where $\varepsilon$ is the porosity (here the fluid volume fraction), $V_S$ and $\varepsilon_S^{\text{vol}}$ for grain $S$ are the lattice volume and the volume fraction of volatiles, respectively; $\langle\rho\rangle$ is the density averaged over the volume of fluid. The source term in Eq. (1) $Q_f^i(r) = \sum_i\Omega_i Q_S$.

The reaction rate of degasification of mixed gases in Eq. (7) is represented by the Arrhenius type equation:

$$f(\varepsilon_S^{\text{vol}}, \theta) = a(\varepsilon_S^{\text{vol}}) \exp[-b(\varepsilon_S^{\text{vol}})/\theta];$$

where the activation energy $b(\varepsilon_S^{\text{vol}})$, as well as the frequency factor $a(\varepsilon_S^{\text{vol}})$, known from the Arrhenius equation, have a simple low-order polynomial form and correspond to different gases which are released at the given temperature range.

The empirical function in Eq. (7) has been proposed in [15]:

$$g(\theta) = 1 - 0.1a_Y - 0.1 \begin{cases} a_Y \text{erf}(b_Y \theta - c_Y), & \text{if } \theta \leq 573K \\ (d_Y\sqrt{\theta} - e_Y)[1 + \text{erf}(f_Y\theta - g_Y)], & \text{otherwise}. \end{cases}$$

It corresponds to change of density due to heating and coupled phenomena like gas release.

2.4. Change of material properties
During the pyrolysis solid and fluid properties change rapidly; change of temperature along with elemental composition has a significant impact on the solid phase. Modelling of technological processes like devolatilisation requires introducing simplification for density and heat capacity for solid ($\rho_S, C_p,S$) that depends on distribution of elements in coal grain. In presented study, for the solid phase two properties are functions of temperature (namely, density and heat capacity) and for fluid the viscosity. Figure 1 presents change of these properties in function of temperature along with its influence on the relaxation time in Eq. (3,2).

2.5. Further details of numerical procedure
Arithmetic mean of grain diameter is $d = 4.8 \text{ mm}$ (Sauter mean diameter is 4.9 mm). Domain is constructed as the REV element, with the diagonal of about 80 medium-sized obstacles. Obstacles (coal grains) are chosen by selecting a random position and diameter, from the uniform and Gaussian distributions, respectively. Domain is discretized by 200 $\times$ 400 nodes.
In the preliminary examination, we choose that left boundary is a heated wall and the bottom wall is adiabatic. This is done by simply introducing no-slip wall of constant temperature (for heated wall) and zero heat flux (for adiabatic wall). At right boundary of the domain symmetry plane is imposed. In LBM, this condition is achieved by assigning to the unknown distribution functions a value of populations leaving the numerical domain, with the symmetry taken into account. For the top boundary-free outlet is implemented by Gradd boundary scheme, see [13]. In case of the heat transfer and specie evolution, zero flux boundary is implemented.

For simplicity fluid-solid interface is treated as no-slip wall with LBM schemes known from literature: the bounce-back (see [13]) and He scheme for heat transfer (see [9]). In case of the fluid flow, additional source term at the boundary is implemented to account for mass increase due to gas release. As the initial condition we impose zero concentration of the chemical species in the volume of the fluid; during heating of the obstacle, chemical compounds are released and transported to the surface of grain. Solid grains are simplified by 0D model, that is: release of gaseous product and change of internal energy are uniformly distributed along surface nodes (for fluid and chemical specie evolution) and nodes of solid grain (for heat transfer).

3. Results and effective thermal conductivity estimation

The purpose of this work is to calculate effective transport coefficient for geometry of packed grains of coal accounting for phenomena occurring during the coking process. A number of accompanying phenomena play an important role in heat transfer through the modelled geometry. Important ones are not implemented in proposed model; namely solid grain Some dilatation occurring during heating as well as radiation are still under implementation.

Resulting pressure in the numerical domain along with the velocity map are presented in Fig. 2 (left and right, respectively). Due to heating, release of gaseous products along with convection plays a major role in fluid flow through the deposit. Decrease of solid density (due to release of gases and dilatation) results in the increase of the fluid pressure (which linearly influence the density in the LBM). Increase of the fluid pressure is slightly altered by convection. Although the gas release has a local effect on the fluid flow, the mass transfer is mainly influenced by convection phenomena; see right plot of Fig. 2, where the source term from chemical reactions in Eq. (1) is neglected.

Figure 3 presents an instant for heat transfer. Change of density lets the cold fluid to penetrate the deposit of coal grains, what (in the modelled geometry) slows down the heat transfer; on the other hand, although gas release improves the heat transfer (by release of hot gas from the grain volume), the chemical reaction is treated as the endothermic process which
Figure 2. Density and velocity profiles averaged over fluid nodes in slices parallel to the heated wall (left plot); map of velocity profile with a few isolines of temperature in the trimmed computational area.

(Uniformly in the volume of grain) reduces the temperature through a negative source term in the evolution equation for IEDDF. This phenomenon has its influence on heat flux what appears as the local maximum of mean heat flux value. This intensification of heat transfer appears only locally in a range of temperatures where chemical reactions occur.

Figure 3. Averaged heat flux and temperature profiles. For comparison, the heat flux component parallel to the heated wall is also plotted. Averaging is done over fluid and solid nodes in the slices parallel to the heated wall.

The effective thermal conductivity coefficient is estimated by [7]:

\[ k_{\text{eff}} = \frac{L \int q_x dY}{\Delta \theta \int dY} \]
where $q_{\parallel x}$ is heat flux projected on direction perpendicular to heated wall and $Y$ is the area of the slice; $\Delta \theta$ is the temperature difference between adjacent slices of domain where heat flux is calculated. Resulting $k_{\text{eff}}$ is presented in Fig. 4. Although presented results refer to unsteady state of the simulation, calculated heat conductivity is nearly constant (with negligible deviations). At this stage, it is difficult to distinguish areas of numerical domain where modelled phenomena: convection, chemical reactions, etc. play a crucial role in heat transfer and estimation of the effective heat conductivity.

![Figure 4](image.png)

Figure 4. Estimated value of effective thermal conductivity and temperature difference along the numerical domain.

4. Conclusion and future work

Presented results show good agreement with the qualitative predictions concerning the influence of the subsequent phenomena in the coking process. Implemented phenomena allows to predict influence of the fluid flow, release of gases on heat transport in random, two dimensional geometry of the bed of coal particles. Described investigation, to the best knowledge of the author, is a first approach to meso-scale modelling of coking process in REV geometry.

The paper presents detailed analysis and first results; in order to achieve a good agreement some additional phenomena have to be considered; the most important seems to be a thermal dilatation, because it plays a major role for fluid flow and heat transfer phenomena. Also radiation is under considered to implementation to check its influence in heat transfer for this modelling approach. The future work will be oriented toward quantitative comparison of results with other known from literature simulations.

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