Two-phase Flow Modelling and Simulation of Gas Purification Column

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Abstract

In the technologies where harmful organic solvents are used it is necessary to purify the waste gases in order to ensure the protection of environment and the fulfillment of environment protecting laws. A wide-spread solution is to adsorb the undesired components on a porous fixed bed but it saturates during the operation and should be regularly regenerated resulting in an economic loss. One of the solutions to improve the process is the use of a moving bed equipment, where the regenerated particles are continuously fed to the adsorber and the saturated particles are transferred toward the regenerator providing this way a continuous operation. The objective of our research is to model and simulate the gas-solid two-phase flow using the immersed boundary method and calculate the particle movement and the adsorption process for the individual adsorber particles applying the principles of discrete element method. The surface of particles is discretized and the component transport to the surface element and the degree of saturation of surface element is calculated by a first-order adsorption kinetics. The calculation method is demonstrated on a single particle. The two-dimensional compressible Euler equations are discretized and solved by the second-order accurate TVD-MacCormack method to effectively handle the oscillations arising in the vicinity of steep spatial fronts. Furthermore, considering the physical bases of compressible gas flow, we used non-reflecting numerical boundary condition at the outflow boundary of calculation domain.

Keywords: adsorption, immersed boundary method, discrete element method, TVD-MacCormack, non-reflecting boundary condition

1. Introduction

Harmful organic solvents are used in many chemical technologies. For example, chlorinated hydrocarbons are applied during the swelling of ion-exchange resins (Kumagai et al., 2018) or gas emissions during the pyrolysis (Veksha et al., 2018). Waste gas purification is necessary for all related technologies since environment protection and sustainability are highly desired as well as decreasing the emission of greenhouse gases. A wide-spread solution of gas purification is to adsorb the undesired components on a porous fixed bed. The main operational characteristic of these units is that the fixed bed saturates during its operation and should be regularly regenerated. Improving the operational characteristic often two columns are used, while one of them operates in adsorption mode, the other one can be regenerated. Another approach is to use a moving bed equipment, where the regenerated particles are continuously fed into the adsorption column and the outgoing saturated particles are transferred into the regenerator.
equipment. This type of construction seems more effective, however it is challenging to control the two-phase flow inside both the adsorption and regenerator column, even more if temperature change plays an important role as in case of CO$_2$ capture technologies (Mondino et al., 2019). Both adsorber constructions can be investigated using models developed in the last decades in the research area of modeling gas-solid two-phase flows. However, they have to be completed with the model equations describing the component transport to the surface of adsorber particles and the diffusion toward the inside of particle.

One of the modeling approaches in modeling gas-solid two-phase flows is the direct numerical simulation (DNS) (Deen et al., 2014) which is basically coupled with the discrete particle treatment. In DNS models the Navier-Stokes equations are derived by the usual point variables and the fluid flow between the particles is fully resolved. Several DNS methods have been proposed by the authors. In the case of immersed boundary method the goal is to solve Navier-Stokes equations inside the complex calculation domain between the particles without using boundary-fitted or unstructured meshes. The flow equations are solved on a structured mesh over the entire calculation domain. The particles are virtually defined inside the flow by adding Lagrangian points on their surfaces. Then additional forcing terms are added to the Navier-Stokes equations to mimic the no slip boundary condition on these surface points. In this way a local force density is introduced to modify the flow field that realizes the particle boundaries. The motion of the particles calculated by the total surface force and torque evaluated on the surface of particles by one of the forcing approaches like direct forcing or feedback forcing (Fadlun et al., 2000). To describe the contacts between solid particles the discrete element method can be used.

The coupled application of immersed boundary method and discrete element method makes it possible to examine the gas purification process in detail. Modeling and simulation of gas-solid two-phase flow including the calculation of discrete particle motion and gas adsorption provides insight to the physical and chemical processes. By applying numerical investigation more information is available about the processes which take place in the equipment than measuring the properties of the waste gas only at the outlet. The aim of our research is to estimate the separation efficiency and the optimal operating parameters regarding both the adsorbent particles and the gas inflow. For this reason in our model the 2D Euler equation is completed with the mass balance equation of harmful organic gas component and the differential equation describing the adsorption process for the individual adsorbent particles. The applied model and simulation method can greatly help the process intensification and the tuning of operating parameters due to the better understanding of the detailed processes inside the gas purification column.

2. Model development and numerical solution

A two-dimensional flow model is set to model the compressible gas flow in the gas purification column shaped as a rectangular channel (Figure 1a). The inlet boundary is located at the bottom of left sidewall while the outflow boundary defined at the top of the column.
The two-dimensional flow model consist of the continuity equation for the non-harmful gas components, the mass balance equation for the harmful gas component which is to be adsorbed, the two momentum and the energy balances:

\[ \frac{\partial \rho_g}{\partial t} + \frac{\partial (m \rho_g)}{\partial x} + \frac{\partial (n \rho_g)}{\partial z} = 0 \]  

(1)

\[ \frac{\partial c}{\partial t} + \frac{\partial (m \rho c)}{\partial x} + \frac{\partial (n \rho c)}{\partial z} = R \]  

(2)

\[ \frac{\partial m}{\partial t} + \frac{\partial (m^2 \rho + p)}{\partial x} + \frac{\partial (n m \rho)}{\partial z} = F_x \]  

(3)

\[ \frac{\partial n}{\partial t} + \frac{\partial (mn \rho)}{\partial x} + \frac{\partial (n^2 \rho + p)}{\partial z} = F_z \]  

(4)

\[ \frac{\partial \rho E}{\partial t} + \frac{\partial (m \rho E + p)}{\partial x} + \frac{\partial (n \rho E + p)}{\partial z} = 0 \]  

(5)

where \( \rho_g \) is the mass density of the mix of non-harmful gas components, \( c \) is the component mass density of the harmful gas component, \( \rho \) is the total mass density \((\rho = \rho_g + c)\). \( m \) and \( n \) are the components of the mass velocity vector in \( x \) and \( z \) direction. \( R \) is the loss term describing the mass flow rate of harmful gas component to be adsorbed, \( p \) is the pressure (calculated according to the ideal gas law), \( F_x \) and \( F_z \) are the force densities introduced according to the immersed boundary method and \( \rho E \) is the sum of the inner and kinetic energy.

The adsorption of harmful gas component is calculated for the individual adsorber particles. The surface of particles is discretized and the component transport to the surface element \( (c_S) \) and the degree of saturation of related particle volume element is calculated by the following first-order adsorption kinetics (Eq(e)):

\[ \frac{\partial c_s}{\partial t} = k \cdot ds \cdot (c - c_0) \]  

(6)
where $c_s$ [kg/m$^3$] is the harmful gas component concentration in the particle volume element, $k$ [m$^{-2}$·s$^{-1}$] is the mass transfer coefficient, $ds$ [m$^2$] is the area of surface element, $c$ [kg/m$^3$] is the mass density of the harmful gas component in the gas phase and $c_0$ [kg/m$^3$] is the equilibrium mass density of harmful components on the solid surface, which is calculated as $c_0=bc_s$ (Bird et al., 2002), where $b$ is a constant.

Eq(6) supposes the development of a homogeneous concentration profile in the individual particle volume element of the adsorber particle after the component transport to the surface element.

The source term $R$ in Eq(2) is calculated by Eq(7). For a given grid point of gas phase it is the sum of component rates that absorbed through the surface elements (n) located in the vicinity of calculation cell $i,j$.

$$R_{i,j} = - \sum_{n=1}^{N} k \cdot ds \cdot (c_{i,j} - c_{0,n})$$

Eqs (1)-(5) are solved by the TVD-MacCormack second order accurate scheme (Yee, 1989) in order to get physically admissible solutions. In this scheme the two steps MacCormack's scheme is completed with a third step by introducing a dissipation term to avoid any unphysical oscillation in the vicinity of strong gradients in the numerical solution. At the outflow boundary, taking into account of the physical bases of compressible gas flow, non-reflecting numerical boundary condition is applied. The application of non-reflecting boundary condition at outflow boundary instead of constant physical boundaries prevents reflections of the unsteady compression and expansion waves back into the calculation domain. In our simulation study a generalized method of non-reflecting outflow boundary condition specification, developed by (Thompson, 1990) is applied. All the calculations are carried out in a self-developed program in MATLAB environment.

In order to calculate the flow field around the adsorber particle and the interaction between solid particle and gas-phase the immersed boundary method is used. The flow equations are solved on a structured and stationary mesh over the entire calculation domain and the solid adsorbent particles occur with a virtual boundary (Figure 1b). As the flow field around the particle is fully resolved, the size of the calculation grid should be approximately one order of magnitude smaller than the size of the particles, which is set in our simulation to 2 mm in diameter like a usual activated carbon adsorber particle. Lagrangian points (red dots in Figure 1b) are defined equidistantly around the particle. The distance (ds) is tuned to the Eulerian grid size and equal to 0.0002 m. Extrapolating between the two grid point locations are carried out by a distance-based weight function (Figure 1c), in our case a simple triangle function.

The calculation algorithm takes all the Lagrangian grid points of virtual particle and looks for the Eulerian grid points in a certain distance limit. In Figure 1b the orange colored circle shows the distance limit and the blue dots are the Eulerian grid points that influence the given Lagrangian grid point. The degree of influence of Eulerian grid points is a function of their distance to the Lagrangian grid point. The gas flow is forced to bypass the virtual solid body by the application of direct forcing method in which an extra force density terms are added to the momentum Eqs (3) and (4). The extra term balances the momentum equation such a way that the modified flow field realizes the particle boundary.
3. Results and discussion

The dynamic model given by Eqs (1)-(5) was solved by TVD-MacCormack scheme using non-reflecting outlet boundary condition. The inlet harmful gas component concentration was set to 2 kg/m$^3$. The non-harmful gas was considered as air. The inlet gas velocity was set to 10 m/s. The mass transfer coefficient $k$ and constant $b$ in the equations were equal to 1x10$^8$ m$^2$.s$^{-1}$ and 1.1. The time step of numerical solution was determined in each iteration step according to the Courant-Friedrich-Lewy criteria, and was changing around 2x10$^{-7}$ s. At simulation time 5x10$^{-3}$ s the velocity field of the gas phase around the particle is shown in Figure 2a and the concentration field of harmful gas component show in Figure 2b. The concentration of harmful gas component in the particle volume elements around the particle is shown in Figure 2c giving a picture of the saturation state of the particle surface. The saturation state around the particle shows a distribution due to the dynamically changing concentration values of those gas phase elements which interact the surface of adsorber particle. Figure 2d shows the dynamic change of saturation at six highlighted surface points. It can be seen that surface points located at the different points of the particle surface are saturated by different velocity depending on the concentration of interacting gas-phase elements.

![Figure 2](image)

Figure 2. a) Velocity field of the gas phase around the particle. b) Concentration field of the gas phase around the particle. c) Degree of saturation around the adsorbent particle after 2x10$^4$ timesteps. d) Saturation curves at specific surface points of the adsorber particle.

As it is seen from the simulation results the model is able to calculate the saturation curves of particle surface points and can provide information about the necessary residence time of particles in the purification column at given operating parameters and this way it can help in the optimal design of column.

4. Conclusions

Adsorption process is modeled for a single particle by a first order adsorption kinetics with the fully resolved dynamic gas flow around the particle. The gas flow around the
motionless adsorber particle is modeled by immersed boundary method, and the gas-solid interaction is calculated regarding the velocity field and the harmful component adsorption. The adsorber material is considered a layer on the carrier particle in our case, in which the diffusion is fast, therefore homogenous concentration field is assumed. However, the main achievement of this study is to calculate the saturation state of the adsorber layer resolved for the small segments of particle surface. This knowledge helps to define the overall saturation time of particle more accurately, and could also help in other process development issues.

Motion and collision of particles is not calculated in the current work, however the next step in improving our detailed model is to populate the gas purification column with more particles, which can interact with the gas flow and each other. Using our extended model it will be possible to simulate the whole purification column and the operating parameters can be designed more efficiently.

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