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Combined wall and thermal effects during non-isothermal packed bed adsorption

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ABSTRACT

A two-dimensional heterogeneous model was used to describe heat and mass transfer in packed bed adsorbers with a relatively low ratio between tube and particle diameter. Using packed bed the wall effect during exothermal adsorption with low tube-to-particle diameter ratio has an impact on the heat and mass flow, and consequently, on both the temperature and concentration profiles in the reactor. In this work, the calculations based on a heterogeneous two-dimensional non-isothermal model was compared with experimental breakthroughs and calculations done by other researchers. Due to the large influence of the wall, radial profiles of concentration and temperature exist. Consequently, the breakthrough of the adsorbate must be analysed not only with respect to time, but also with respect to the radial position. This paper outlines the impact of thermal and wall effects on packed bed adsorption.

1. Introduction

Physical adsorption is used as a highly selective separation method in which fluid components are removed to the surface of a solid. According to many researchers (to quota a few [1,2]), the mechanism of the isothermal adsorption and a computation of a breakthrough in packed bed with a large tube-to-particle diameter ratio, D/d_p, can be carried out with good precision. However, in a highly loaded, exothermic packed bed sorption process, when it is necessity to effectively decrease the temperature by cooling jacket, hydrodynamics and heat factors could plays an important role. Some of the authors [3–10] indicate this fact, and problem became much more complicated. Hence, for relatively small D/d_p ratio the tube wall has a considerable influence on porosity, fluid flow, heat transfer and mass transfer in the bed. Moreover, it should be pointed out that identified limitations of existing literature on modelling adsorption in highly loaded, wall-cooled packed beds refer primarily to simplifying assumptions like isothermal operation [11], specific types of adsorption equilibrium [12–14], constant pattern behaviour [13,15], neglecting the resistance of mass transfer [16,17], or the use of lumped, overall coefficients for fluid-to-particle mass transfer [3,18].

Experimental data on radial profiles of concentration and temperature are rare in literature, and typically prone to low resolution and other limitations resulting from the more or less invasive character of the measurements. To overcome this intrinsic measurement problem an optical tomography technique is developed and described in refs. [19,20].

Here the author will briefly recapitulate the model assumptions which were presented in more detail by Kwapinski et al. [21]. Then, results of calculations with experimental and numerical ones presented by Manjhi et al. [11] will be compared. Finally the additional analysis and comparison with other model simplifications will be shown and discussed. Presented calculations were made for water vapour adsorption on 4A zeolite particles in a cylindrical bed. The accuracy of numerical calculations has been confirmed by existing analytical solution for pure transport phenomena presented in ref. [21].

2. Flow maldistribution

It is important to remember that maldistribution of gas flow in the vicinity of the wall is typical for columns with a low ratio between tube and particle diameter, and that concentration and temperature profiles will not occur with respect to the axial coordinate of the bed or with time, but also with respect to the lateral (radial) coordinate, coupled with respective profiles of bed porosity, flow velocity, dispersion coefficients and effective thermal conductivity. Consideration of such profiles is an important feature of modern packed bed reactor models, as they have been presented by Winterberg et al. [22] and recapitulated by Tsotsas [23,24], so that the goal of the author's work is to continue investigate by simulating the application of respective model to adsorber operation.

From the point of view of packed bed reactor modelling, the description of transport phenomena in adsorbers was found to be usually inconsistent, or at least not up-to-date with respect to the

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Nomenclature

c	heat capacity (J/(kg K))
d	diameter (m)
D	diameter of the adsorber (m)
D	effective mass dispersion coefficient (m^2/s)
ΔH	isosteric heat (J/kg)
p	pressure (Pa)
Pe	Peclet number (–)
r	radial coordinate (m)
R	radius (m)
Re	Reynolds number (–)
t	time (s)
T	temperature (°)
u_0	superficial velocity (m/s)
u_c	superficial core velocity (m/s)
X	solids moisture content (kg/kg dry solid)
Y	gas moisture content (kg/kg dry gas)
z	axial coordinate (m)
Greek letters	
η	dynamic viscosity (Pa s)
λ	thermal conductivity (W/(m K))
Λ	effective thermal conductivity (W/(m K))
ρ	density (kg/m^3)
ψ	porosity (–)
Superscripts and subscripts	
ad	adsorption, adsorbate
ax	axial
bed	bed
c	core
dry air	dry air
eff	effective
f	fluid
in	gas inlet
p	particle
r	radial
0	initial, reference, superficial
$\bar{}$	average

choice of parameters, like the porosity profile, the effective viscosity, or the radial profiles of dispersion coefficients and effective thermal conductivities.

It is well known that near the wall is a higher porosity which leads to higher velocities in this region of the tube (Fig. 1). For this reason, the lower tube-to-particle diameter ratio, the more pronounced the deviation from uniform flow (plug-flow) assumption in a model could be. The increase of flow velocity in the vicinity of the wall is accompanied, due to continuity, by a decrease in the middle of the bed, which was one of the subjects of numerous studies [4,6,21,22,25–28]. The velocity profile is calculated by means of the extended Brinkman equation

$$\frac{\partial p}{\partial z} = -f_1 u_0(r) - f_2[u_0(r)]^2 + \frac{\eta_{\text{eff}}}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u_0}{\partial r}\right) \tag{1}$$

with the effective viscosity according to Giese et al. [26]

$$\frac{\eta_{\text{eff}}}{\eta_{\text{f}}} = 2.0\exp\left(3.5\times 10^{-3}Re_0\right) \tag{2}$$

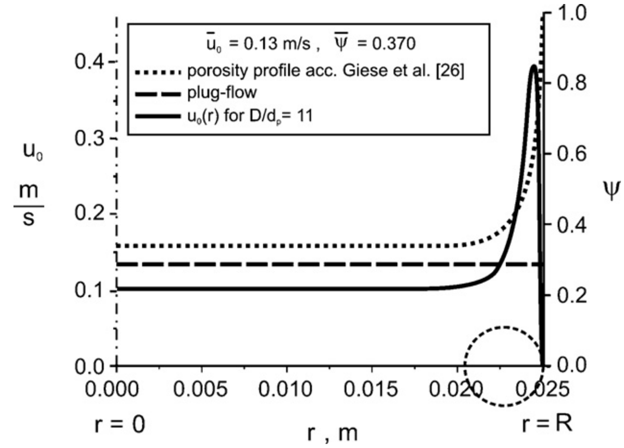

Fig. 1. Profiles of bed porosity and velocity in packed bed for d_p = 4.75 mm, average velocity 0.13 m/s and average porosity 0.370.

the D'Arcy and Forchheimer factors

$$f_1 = 150\frac{\left[1-\psi(r)\right]^2}{\left[\psi(r)\right]^3}\frac{\eta_{\text{f}}}{d_{\text{p}}^2} \tag{3}$$

$$f_2 = 1.75\frac{\left[1-\psi(r)\right]}{\left[\psi(r)\right]^3}\frac{\rho_{\text{f}}}{d_{\text{p}}} \tag{4}$$

In the computation of the flow profile the author used an effective viscosity instead of fluid viscosity, as most authors do. As it was showed in previous publications [4,5], the assumption made in Eq. (1) for fluid viscosity is not realistic.

3. Thermal effect

First models for a fixed-bed reactor which take into account the non-isotropic character of the fixed-bed were formulated in ref. [29] and also in ref. [26]. A two-dimensional model for isothermal adsorption, with a constant effective dispersion coefficient, was proposed in ref. [27], while in ref. [30] was developed a model with the effective dispersion coefficient as a function of flow velocity. Bart et al. [5] presented solutions for non-isothermal adsorption with maldistribution in fixed-bed for small changes of temperature and constant overall transport coefficient, Mhimid [31] provides calculations for desorption.

The adsorption process is often exothermic, hence temperature and concentration profiles migrate through the porous media (one for the solid and one for the fluid), due to the transport process between two phases. Two-phase, with partial differential set of two-dimensional axial symmetric Cartesian equations model has to be solved along with the mass energy balance (Eqs. (5) and (6)).

$$\psi(r)\frac{\partial Y}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left[D_{\text{r}}(r)r\frac{\partial Y}{\partial r}\right] + D_{\text{ax}}(r)\frac{\partial^2 Y}{\partial z^2} - u_0(r)\frac{\partial Y}{\partial z} - \left[1-\psi(r)\right]\frac{\partial X}{\partial t}\frac{\rho_{\text{p}}}{\rho_{\text{f}}} \tag{5}$$

$$\{\psi\rho_{\text{f}}c_{\text{f}} + \left[1-\psi\right]\rho_{\text{p}}c_p\}\frac{\partial T}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left[\Lambda_{\text{r}}(r)r\frac{\partial T}{\partial r}\right] + \Lambda_{\text{ax}}(r)\frac{\partial^2 T}{\partial z^2} + -u_0(r)\rho_{\text{f}}c_{\text{f}}\frac{\partial T}{\partial z} - [1-\psi(r)]\Delta H_{\text{ad}}\rho_{\text{p}}\frac{\partial X}{\partial t} \tag{6}$$

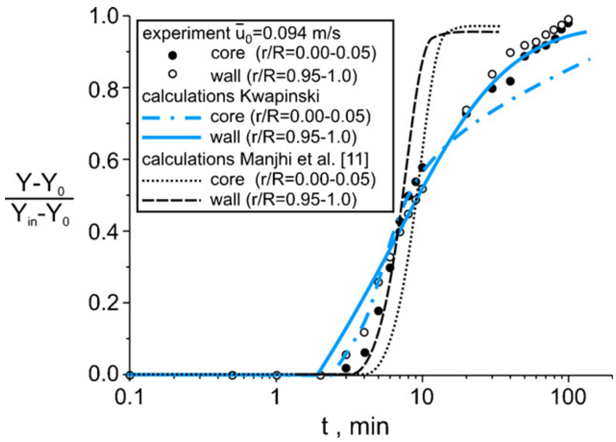

Fig. 2. Experimental and calculated breakthrough curves for the wall and core region of the adsorber.

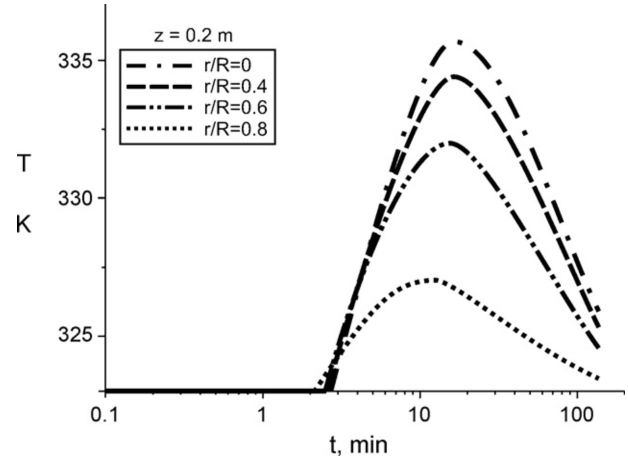

Fig. 3. Calculated temperature profiles for selected regions of the adsorber.

These balances are couple by last change of adsorbate in time and have to be solved simultaneously. Partial differential equations (for mass and energy) contain dispersion terms in axial and radial direction were described in detail in ref. [21]. There is also a briefly described method of numerical solution and a more detailed model validation. This set of equations with adsorption equilibrium is called the "complete model". A numerical solution of the model, shows adsorption breakthrough as well as temperature profile at every location inside the column.

Manjhi et al. [11] experimentally investigated concentration profiles in a tubular adsorber with the help of tomographic technique. The concentration measurements were made at the outlet of the adsorber (a couple of millimetres above packed-bed). The moisture concentration was measured. The breakthrough was presented as an average value from the core (axial) region for $r/R = 0.0–0.05$ and at the vicinity of the wall for $r/R = 0.95–1.0$.

In Fig. 2, the results of tomographic measurements compared with calculations made in ref. [11], and also using non-isothermal model made by author of this publication are presented. The measurements are for water vapour adsorption at the 4A zeolite in the packed-bed cylindrical tube for the $D/d_p = 11$ (where average $d_p = 4.75$ mm) and average superficial velocity of 0.094 m/s. Necessary data on single-particle adsorption kinetics and equilibria have been determined separately by means of a magnetic suspension balance, this was taken from earlier publication [32].

Manjhi et al. [11] used lattice Boltzmann three-dimensional isothermal model to describe experimental data. However, the water vapour adsorption at 4A zeolite is exothermic, and the temperature in the core region can rise by 10 °C. Those researchers had to modify the equation in the model by some factors to get calculated curves crossing corresponding experimental data in 50% of the ordinate. However, in ref. [11] assumed the importance of implementing maldistribution flow in their model, it is not enough to give the nature of non-isothermal adsorption. They noticed that fact and hope to address this aspect in the near future.

The experimental results show (Fig. 2) that for a certain period of time moisture concentration in the core region is higher than in the wall region. This observation was confirmed by author's calculations using complete non-isothermal model and the explanation of this is the fact of fastest temperature growth (the temperature of the wall of the adsorber reactor were constant) in axial region cause periodically smaller adsorption capacity in this region and influence on the efficiency of the process. In this time, maldistribution effect is overlapped by reduction of adsorption capacity in the core cause by high increase of temperature. It should be highlighted that in the model there is no fitting coefficient to adjust experimental data.

All calculations for the next figures were made for the same conditions as presented for Fig. 2.

In Fig. 3, calculated temperatures profiles for selected regions of the adsorber are presented. According to the calculations non-isothermal model calculations are based on the equilibrium curves presented in ref. [32]. A maximum change of temperature of up to 13 K was observed. Temperature is gradually going down with a increasing of radius to a wall temperature which was assumed constant at 323 K. This high grow of temperature suggest that there may be a big difference between breakthrough for isothermal and non-isothermal conditions. Fig. 4 confirm this conclusion, the comparison reveals completely different position and slope of the breakthrough curves. For the non-isothermal model, the breakthrough is faster, because of growing temperature of a bed which decreases it capacity. However, letter on the slope is going down because the bed is cool down (it may be seen in Fig. 3). Consequently, the bed was fully saturated later than in the case of isothermal conditions. In case of isothermal operation, near-wall breakthrough is always faster than the breakthrough in the centre of the adsorber. In addition, for the assumption of plug-flow there is one average breakthrough laying in-between those two for maldistribution model and representing average concentration in adsorber. In the non-isothermal case, the character of curves is also completely different.

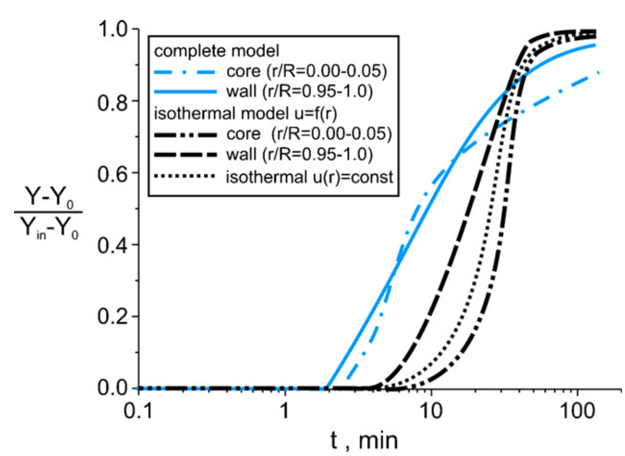

Fig. 4. Calculated breakthrough curves for core and wall region of the adsorber, for non-isothermal and isothermal model, and isothermal plug-flow model.

4. Wall effects

By wall effect in packed-bed can be call all consequences that are not visible for a high D/d_p ratio. Discussion concerning wall effect in isothermal and non-isothermal adsorption using the model that is presented above was published in ref. [21]. In this publication, there are additional example and studies showing this effect. In complete model the effective axial conductivity and radial conductivity is described by Eqs. (7) and (8), respectively were the thermal conductivity of a bed is a function of radius,

$$\Lambda_{ax}(r) = \lambda_{bed}(r) + \frac{Pe_0}{2}\lambda_f \tag{7}$$

$$\Lambda_r(r) = \lambda_{bed}(r) + K_1 Pe_0 \frac{u_c}{\bar{u}_0} f(R-r)\lambda_f \tag{8}$$

with the function

$$f(R-r) = \begin{cases} \left(\dfrac{R-r}{K_2 d_p}\right)^2 & \text{for} \quad 0 \le R-r \le K_2 d_p \\ 1 & \text{for} \quad K_2 d_p < R-r \le R \end{cases} \tag{9}$$

which includes slope parameter $K_1 = 1/8$, and dumping parameter

$$K_2 = 0.44 + 4\exp\left(-\frac{Re_0}{70}\right) \tag{10}$$

where the Reynolds number

$$Re_0 = \frac{\bar{u}_0 \rho_f \bar{d}_p}{\eta_f} \tag{11}$$

and the molecular Peclet number for heat transfer

$$Pe_0 = \frac{\bar{u}_0 \rho_f c_f \bar{d}_p}{\lambda_f} \tag{12}$$

The thermal conductivity of the packed bed without fluid flow is calculated from the correlation provided by Zehner and Schlünder [33] as a function of local porosity. For the packed bed of zeolite particles the thermal diffusivity was measured experimentally described in ref. [34]. The decrease of Λ_r towards the wall (Fig. 5) correlates with the increase of porosity near the wall (Fig. 1), both calculations were made for the same conditions. This and the damping of cross-mixing that is due to the presence of the wall are reflected in the $\Lambda_r(r)$ function $f(R-r)$. Maximal value of Λ_r (Eq. (8)) is only about 2.5 times smaller than Λ_{ax} (Eq. (7)), hence the radial effective conductivity influence on the process of heat migration. This can be see in Fig. 6 where assumed in the calculations that the $\Lambda_r = 0$. There is a big difference in a shape of the breakthrough curves compared with the complete model. Assuming that there is no radial element in the energy equation, the core of the bed is cooled much slower and the adsorption capacity is smaller due to a higher temperature of a bed.

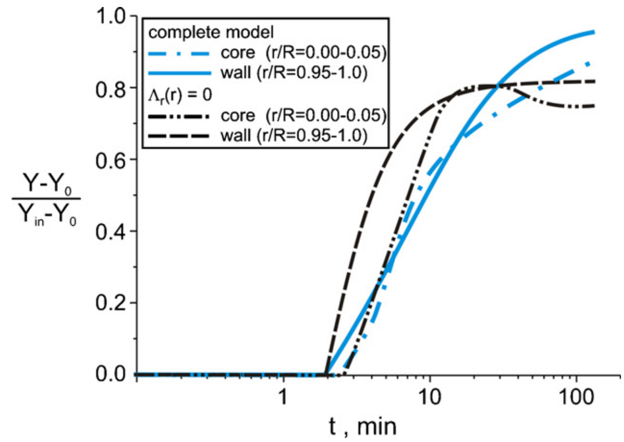

Fig. 6. Calculated breakthrough curves for core and wall region of the adsorber, for non-isothermal complete model and for the case when there is no radial element in energy equation.

In Fig. 7, profiles of water moisture concentration in gaseous phase are presented for different time of adsorption at the end of the column ($z = 0.2$ m). The shape of curves confirms observation and conclusions from Fig. 2, that there are periods when adsorption is higher in the core than near the wall and opposite. And it mostly depends on the temperatures in the bed which changes with time and is the highest for $r = 0$, as is shown in Fig. 8. For the adsorption with such strong thermal effect it is difficult to decuple thermal from wall effect. However such a trial will be presented in the next publication.

One more example in Fig. 9 shows breakthrough curves in the core and near the wall of adsorber calculated for different D/d_p ratios using a complete model for the conditions. The average porosity for smaller particles is smaller, however the thermal conductivity of a bed were assumed the same, for the reason not to additionally differentiate those two cases. However, smaller spherical particles cause higher capacity of the adsorber, which is the reason breakthrough starts growing a bit later than for the bed contain bigger particles. For such conditions, beside smaller maldistribution effect for higher D/d_p ratio the thermal effect cause even a bigger difference between wall and core concentrations.

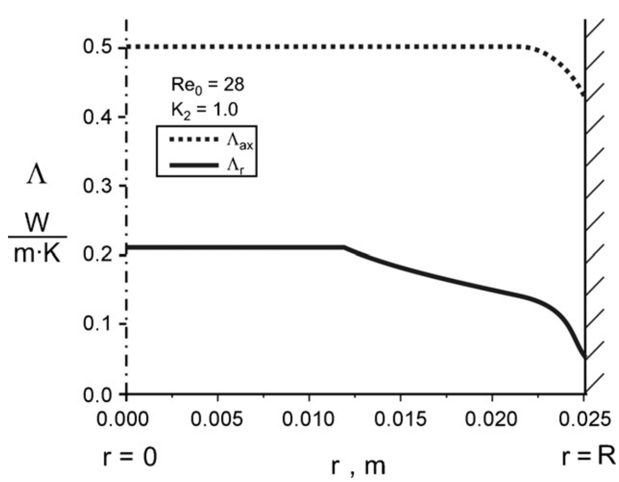

Fig. 5. Radial profiles of axial and radial effective thermal conductivities.

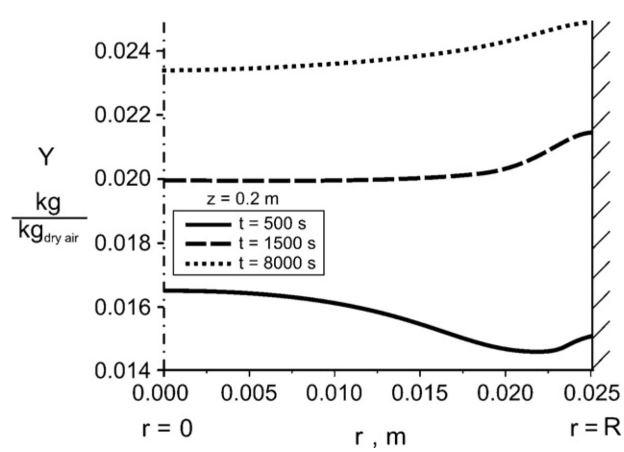

Fig. 7. Radial profiles of water moisture concentrations for different time of adsorption at the outlet of the column.

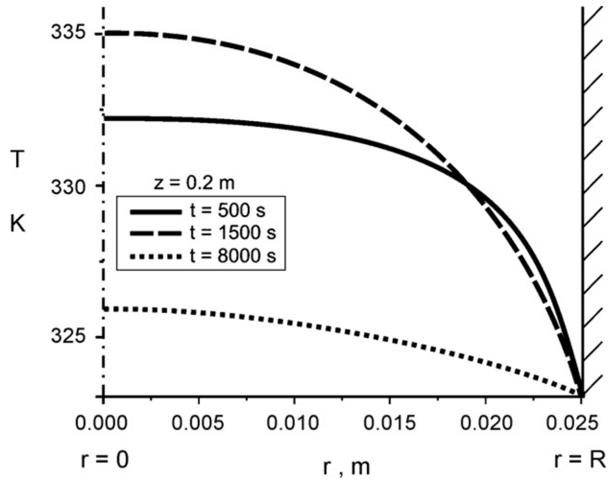

Fig. 8. Radial profiles of temperatures for different time of adsorption at the outlet of the column.

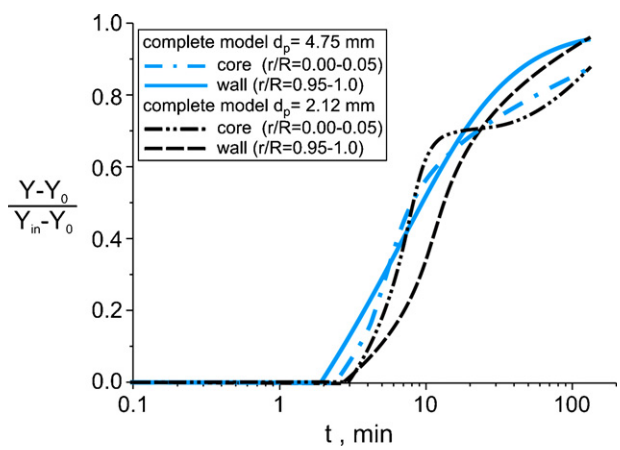

Fig. 9. Calculated breakthrough curves in the core and near the wall of adsorber calculated for different diameters of the particles in a bed.

5. Summary

A two-dimensional model of coupled heat and mass transfer in packed beds should be used to simulate non-isothermal adsorption under high wall and thermal effects, to avoid oversimplifying assumptions. Hydrodynamics and thermal effect were confirmed to have a strong impact on the adsorption process and process efficiency in a packed bed reactor. In case of a relatively small D/d_p ratio in the reactor, the wall effect problems increase considerably. It is necessary to model the phenomena, taking into account the choice of parameters, like, increase the porosity and the flow velocity near the tube wall, or the radial profiles of dispersion coefficients and effective thermal conductivities. The wall and thermal effect in non-isothermal adsorption interact and overlap in a complex way.

The results measured at the outlet of a tube packed with zeolite particles during the adsorption of water vapour from air were compared with the transient concentration and temperature fields calculated numerically by means of the model. The agreement has been found to be satisfactory.

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