Computational fluid dynamics modeling in a fixed adsorbent layer during separation of gas mixtures

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Abstract. The paper discusses the numerical analysis results of the aerodynamic structure of a flow in the granular adsorbent layer in a pressure swing adsorption unit adsorber during atmospheric air separation and oxygen concentration. The computational experiments were carried out using one-dimensional and two-dimensional mathematical models to calculate the velocity field in the adsorbent bulk layer. While assessing the accuracy of calculating the aerodynamic structure of the gas flow in the adsorbent, it was found that the use of a two-dimensional mathematical model provides an increase in the accuracy of calculations by an average of ~1-2% compared with the one-dimensional model.

1. Introduction

To simulate the cyclic adsorption processes of gas separation and purification implemented in vertical cylindrical adsorbers with a bulk layer of the granular adsorbent, one-dimensional mathematical models are usually used where the gas flow is taken only in the axial direction along the height of the adsorbent layer. The disadvantage of this approach is that non-uniformity in the flow distribution over the adsorber cross section, the presence of stagnant zones and the incomplete use of the adsorption capacity of the adsorbent lead to a decrease in the intensity of the mass and heat transfer processes accompanying the cyclic adsorption-desorption processes in pressure swing adsorption (PSA) units, this is usually not accounted for in one-dimensional models [1–4].

The development of computer technology and special software packages for multiphysical tasks, including CFD modeling, opens up possibilities for researchers to consider complex (two and three-dimensional) problems of aerodynamics combined with mass and heat exchange. The use of two-dimensional mathematical models complicates the mathematical formulation of the problem, and also significantly increases computational costs, so the comparison of the accuracy of the solutions obtained using one-dimensional and two-dimensional models should allow to estimate the gain in the accuracy of the solution.

In recent years, works devoted to computational fluid dynamics modeling of velocity fields in an adsorber using Navier-Stokes equations have been published [5–11]. Rambabu, Muruganandam and Velu [5] present a computational fluid dynamics modeling of a vertical cylindrical adsorber during the implementation of the PSA process of separation of CO₂ and CH₄ binary gas mixture using 6-FDA granular adsorbent. Simulation studies of the influence of regime parameters (pressure, temperature and flow) on CO₂ purity in the separation process were carried out. A table was obtained, in which for different concentrations of CO₂ and CH₄ in the initial gas mixture the optimum cycle time of the plant is given, thus allowing to achieve the maximum CO₂ purity at the plant outlet.
In Zheng, Yao and Huang [6] the two-dimensional model of the process of rapid PSA air separation using granular adsorbent LiX is constructed. Simulated research of heat and mass transfer processes was carried out and the influence of regime and design parameters (two heights of dead zone, durations of pressurization and adsorption steps, product extraction) on product purity, product recovery, bed size factor and average volume product yield was established.

In Gautier et al. [7] a numerical sensitivity analysis of various physical parameters was carried out to assess the reliability of three-dimensional CFD modeling of the PSA process. It has been shown that the effective thermal conductivity of the adsorbent, mass transfer coefficient, the presence of dead zones in the adsorber, the choice of equilibrium equation (sorption isotherms) are the most important parameters for accurate modeling of the adsorption process.

Yang et al. [8] have carried out a numerical study of oxygen concentration in a PSA unit with radial-flow adsorbers and LiX granular adsorbent with the use of two-dimensional CFD-modelling. It was found that the centripetal π-flow radial adsorber provides the most uniform flow distribution in the adsorber.

Despite the importance of the presented studies, the considered works do not provide information on the increased accuracy of the obtained solutions in comparison with the traditionally used one-dimensional models and conditions (regimes) under which accuracy improves significantly.

At the same time, the development and application of a two-dimensional mathematical model for calculating the field of gas flow velocities in the vertical cylindrical adsorber will allow not only to study the aerodynamic situation, the degree of non-uniformity in the flow distribution over the adsorber cross section, but also to evaluate the effect of non-uniformity in the flow on the processes of heat and mass transfer in the granular adsorbent layer.

The aim of this work is to build a two-dimensional mathematical model of the aerodynamic flow structure in PSA adsorber units and assess the influence of the adsorber design parameters (the adsorber height \( L \) and diameter \( D_a \), fitting diameter \( d_f \), shape and size of internal cavities), mode variables (gas flow velocity \( u \) and pressure \( P \)), and the adsorbent particle diameter \( d_p \) on the uniform distribution of the gas flow in the granular adsorbent layer. The list of accepted symbols and abbreviations has presented in table 1.

| Symbol     | Description                                      |
|------------|--------------------------------------------------|
| \( D_a \)  | adsorber inner diameter, m                       |
| \( d_f \)  | fitting diameter, m                              |
| \( d_p \)  | adsorbent particle diameter, m                   |
| \( d_e \)  | diameter of the equivalent channel between       |
|            | adsorbent particles, m                           |
| \( G^m \)  | gas flow velocity at the inlet to the adsorber, m/s |
| \( L \)    | height (length) of the adsorber, m               |
| \( L_1 \)  | height (length) of the diffuser part, m          |
| \( P \)    | pressure, \( \times 10^5 \) Pa                  |
| \( P_i \)  | adsorber inlet pressure, \( \times 10^5 \) Pa    |
| \( \alpha \) | angle of the diffuser part of the adsorber, °     |
| \( \mu \)  | dynamic viscosity, Pa×s                         |
| \( \rho \) | gas flow density, kg/m³                         |

2. The mathematical description of the oxygen enrichment process

The design scheme of the adsorber fragment and its main characteristics as a simulation object are shown in figure 1. Since the adsorber is an axisymmetric cylindrical object, in order to reduce the required computational resources, the velocity field will be considered in its left half assuming that the distribution in the right half is mirror relative to the axis of symmetry (figure 1). The gas-air flow with the velocity \( u \) enters the adsorber with a diameter \( D_a \) and a height \( L \) through the lower fitting and is diverted through the upper fitting (fitting diameter \( d_f \)). The height of the adsorbent layer with a particle diameter \( d_p \) is \( L-2L_1 \) and the angle of the diffuser part is \( \alpha \).
The velocity field \( \mathbf{u} \) and pressure \( P \) in the adsorber are described using the Navier–Stokes equations [12]:

\[
\rho \left( \frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) = -\nabla P + \mu (\nabla \mathbf{u} + (\nabla \mathbf{u})^T) - \frac{2}{3} \mu \nabla \cdot \mathbf{u},
\]

where \( \rho \) is the gas flow density, kg/m\(^3\); \( \mu \) is the dynamic viscosity, Pa\(\cdot\)s. These equations are supplemented by the corresponding initial equations (2):

\[
\mathbf{u}(t=0) = \mathbf{u}_0,
\]

and boundary conditions for the adsorber walls:

\[
\mathbf{u}(\partial \Omega) = 0,
\]

and the adsorber inlet:

\[
\mathbf{u}(z=0) = \mathbf{u}(t).
\]

When constructing a two-dimensional model of the gas-air mixture flow in the granular adsorbent layer (figure 1), the problem of determining the distances between particles in the layer arises. This distance can be calculated as the equivalent diameter of the channel formed by adjacent particles (figure 2).

The diameter of the equivalent channel \( d_e \) is determined from the value of the surface area of the hatched region (figure 2),

\[
d_e = \sqrt{\frac{4 \cdot 0.16r_p^2}{\pi}} = 0.45r_p = 0.225d_p.
\]

Figure 1. The design scheme of the adsorber as a simulation object.

Figure 2. Calculation scheme for determining the diameter of the equivalent channel between the adsorbent particles, \( r_p = d_p/2 \).
In PSA processes, a velocity jump occurs when the controlled valves are opened (closed) at the initial moment of implementing the adsorption or desorption stages [13, 14]. Using a special test bench [15], the dynamics curves of the gas-air flow velocity were obtained at the adsorber inlet in the PSA unit (figure 3), which were then used as the boundary condition (4).

![Figure 3](image)

**Figure 3.** Dynamics of the air velocity at the adsorber inlet at $P_{in}=3\times10^5$ Pa, $d_p = 0.001$ m and values: (a) $G_{in}$: 1 – $4.5\times10^{-5}$ m$^3$/s, 2 – $9\times10^{-5}$ m$^3$/s, 3 – $18\times10^{-5}$ m$^3$/s, 4 – $36\times10^{-5}$ m$^3$/s; (b) $l$ – $9.6\times10^{-5}$ m$^3$/s, 2 – $19.5\times10^{-5}$ m$^3$/s, 3 – $38.7\times10^{-5}$ m$^3$/s, 4 – $77.4\times10^{-5}$ m$^3$/s.

3. Results and discussions
The numerical studies using the mathematical models (1)–(4) included determining the effect of the flow rate of the initial mixture $G_{in}$ (relative to the adsorber volume $V_a$), the height (length) of the diffuser part $L_1$ (relative to $L$ for different $G_{in}/V_a$), values $\alpha$ for various $G_{in}/V_a$, the ratio $d_f/D_a$ for various $G_{in}/V_a$, as well as the effect of $d_p$ on the uniform distribution of the flow in the adsorber. The changing ranges of variables are presented in table 2.

Fragments of the velocity field in the adsorber at different times (obtained by solving the problem (1)–(4) by the finite element method) for one of the numerical experiments are introduced in figure 4.

![Figure 4](image)

**Figure 4.** Fragment of the velocity field in the adsorber, $G_{in}/V_a = 0.25$; $L_1/L = 0.083$; $\alpha = 30^\circ$; $D_a = 0.034$ m; $L = 0.24$ m; $d_p = 0.001$ m; $L_1 = 0.02$ m; $d_f = 1/4$at: (a) $t = 1$ s; (b) $t = 3$ s; (c) $t = 4$ s; (d) $t = 6$ s.
Table 2. Initial data for a computational experiment.

| No. | $G_{in}/V_a$ | $L_i/L$ | $\alpha$ (º) | $D_a$ (m) | $L$ (m) | $d_p$ (m) | $L$ (m) | $d_f$ (m) | $u_z$, $z = 0$ (m/s) |
|-----|--------------|---------|--------------|-----------|---------|-----------|---------|-----------|------------------|
| 1   | 0.250        | 0.042   | 0            | 0.034     | 0.240   | 0.001     | 0.010   | 1/4'      | $l$, Figure 3a   |
| 2   | 0.034        | 0.044   | 0.292        |           |         |           |         |           | $l$, Figure 3    |
| 3   | 0.510        | 0.042   | 0.034        | 0.240     |         |           |         |           | 2, Figure 3      |
| 4   | 0.034        | 0.044   | 0.292        |           |         |           |         |           | 2, Figure 3b     |
| 5   | 1.010        | 0.042   | 0.034        | 0.240     |         |           |         |           | 3, Figure 3a     |
| 6   | 0.034        | 0.044   | 0.292        |           |         |           |         |           | 3, Figure 3b     |
| 7   | 2.020        | 0.042   | 0.034        | 0.240     |         |           |         |           | 4, Figure 3b     |
| 8   | 0.034        | 0.044   | 0.292        |           |         |           |         |           | 4, Figure 3b     |
| 9   | 0.250        | 0.083   | 0            | 0.034     | 0.240   | 0.001     | 0.020   | 1/4'      | $l$, Figure 3a   |
| 10  | 0.021        |         |              |           |         |           | 0.005   |           |                  |
| 11  | 0.510        | 0.083   | 0.020        |           |         |           |         |           | 2, Figure 3a     |
| 12  | 0.021        |         | 0.005        |           |         |           |         |           |                  |
| 13  | 0.250        | 0.083   | 30           | 0.240     | 0.001   | 0.020     | 1/4'    |           | $l$, Figure 3a   |
| 14  |              | 45      |              |           |         |           |         |           |                  |
| 15  | 0.510        | 0.083   | 30           |           |         |           |         |           | 2, Figure 3a     |
| 16  |              | 45      |              |           |         |           |         |           |                  |
| 17  | 0.250        | 0.083   | 0            | 0.034     | 0.240   | 0.001     | 0.020   | 1/8'      | $l$, Figure 3a   |
| 18  |              |         |              |           |         |           |         | 1/2'      |                  |
| 19  | 0.510        | 0.083   |              |           |         |           |         | 1/8'      | 2, Figure 3a     |
| 20  |              |         |              |           |         |           |         | 1/2'      |                  |
| 21  | 0.510        | 0.083   | 0            | 0.034     | 0.240   | 0.002     | 0.020   | 1/4'      | 2, Figure 3a     |
| 22  |              |         |              |           |         |           | 0.0005  |           |                  |
| 23  |              |         |              |           |         |           | 0.004   |           |                  |

The analysis of velocity fields presented in figure 4 shows that the gas flow velocities in the center of the adsorber and at its walls are equalized even before the time $t = 3$ s is reached, and the flow is established (the difference in velocities in the center and at the walls of the adsorber disappears) during the first 10-15 adsorbent layers. The greatest velocity difference is achieved in the first second after the valves are opened, when the velocity of the incoming flow in the adsorber is maximum. The velocity distribution over the cross section of the granular layer is consistent with the data obtained in [9–11].

Figure 5 shows the dependences of the velocity distribution in the central part of the adsorber and at its walls at the inlet to the granular layer at various $G_{in}/V_a$ ratios.

The analysis of the graphs in figure 5 shows that at the inlet moment into the granular layer at the adsorber wall there is a slight decrease in the gas-air flow velocity, which is especially noticeable in figure 5b (until time $t = 2$ s). Despite the apparent significant discrepancy between the velocities (for example, curves 1, 2, figure 5b), it disappears in the first 2 seconds of the process due to the redistribution of the incoming flow inside the adsorber. It was found that the redistribution of the flow inside the adsorber and the production of a flow, which is almost uniform over the cross section (with
a deviation of no more than 5%), occur in the first 6-20 granular layers, which is no more than 4-6% of the height of the adsorbent layer ($L/2L_1$).

Figure 5. Velocity distribution in the central part of the adsorber (curves 1, 3, 5, 7) and at the walls (curves 2, 4, 6, 8) at the inlet to the granular layer at $G_{in}/V_a$: 0.250 – curves 1, 2; 0.510 – curves 3, 4; 1.010 – curves 5, 6; 2.020 – curves 7, 8: (a) $L_i/L = 0.042$; (b) $L_i/L = 0.034$.

The analysis of the dependences of the velocity distribution in the central part of the adsorber and at its walls at the inlet to the granular layer at various ratios $L_i/L$ and $\alpha$ shows that an increase in the distance $L_1$ between the granular layer and the adsorber inlet point of the gas flow contributes to a more uniform distribution of the gas flow over its cross section – the velocity difference at the wall and in the center of the vertical cylindrical adsorber is less than 1%. In addition, a quick levelling of the gas flow velocity profile over the cross section is ensured: in the first 4 layers at $L_1/L = 0.083$, against 16 layers of adsorbent particles at $L_1/L = 0.021$. An increase in $G_{in}/V_a$ from 0.250 to 0.510 leads to a decrease (~2.5 times) in the levelling time of the gas flow velocities in the adsorber with a greater (by ~40%) discrepancy between the velocities at the wall and in the center of the adsorber at the initial time (up to $t = 3$ s).

The most uniform velocity field at the initial time is provided at the diffuser angle $\alpha = 30^\circ$. At $\alpha = 45^\circ$, the gas-air flow tends to the central part of the adsorber. With an increase in $G_{in}/V_a$ from 0.250 to 0.510, the nature of the dependences does not change.

In the course of the research it was found that changing the fitting diameter affects the formation of stagnant zones in an empty adsorber. In an adsorber filled with a granular adsorbent, the flow is quickly redistributed in the initial section. A much larger effect on the velocity field in the adsorber is exerted by the distance from the flow inlet to the adsorber to the adsorbent frontal layer (determined by $L_1/L$), which is also mentioned in [8].

Figure 6 shows the dependence of the velocity distribution in the central part of the adsorber and at its walls at the inlet to the granular layer at various $d_p$, and, accordingly, the ratio $d_p/D_a$.

The analysis of the results presented in figure 6 shows that an increase in the adsorbent particle size $d_p$ negatively affects the uniformity of the flow distribution over the adsorber cross section. The gas-air flow rushes to the place where the aerodynamic drag is less, and, accordingly, the equivalent diameter of the pore channels is larger. The analysis of the gas-air flow distribution in the adsorber showed that the gas-air flow tends to slip through the central part of the adsorber at $d_p$ more than 2 mm, which leads to an increasingly significant difference in velocities in the center of the adsorber and at its walls (figure 6).
4. Conclusion
In the course of computational experiments, it was found that the following structural parameters have the greatest influence on the velocity distribution in the vertical cylindrical adsorber: the ratio of the free volume (not occupied by the adsorbent layer) to the height of the adsorber, the diameter of the adsorbent particles, and the angle of the diffuser part of the adsorber. The granular adsorbent layer in the adsorber operates as a distribution grid, which ensures the uniformity of the gas-air flow over the cross section (in the first 6-20 layers); the smaller the diameter of the adsorbent granules, the faster the uniformity of the velocity field over the adsorber cross section is reached.

It was found that when simulating the aerodynamic situation in a vertical cylindrical adsorber filled with a granular adsorbent with particle diameter less than 2 mm, the use of a two-dimensional model is impractical, since it provides an increase in the accuracy of calculations by an average of only ~1-2% with a significant increase in the requirements for computational resources.

The mass and heat transfer equations used in the mathematical description of cyclic adsorption-desorption processes include components describing convective mass and heat transfer in the gas phase, as well as mass and heat transfer coefficients from the gas phase to the adsorbent and vice versa, which determine the intensity of the mass and heat transfer. These components depend on the values of the gas-air flow velocity, while the absence of a significant difference in the velocity fields when using one-dimensional and two-dimensional models for the adsorbent layer with particles of less than 2 mm in diameter suggests the inexpediency of constructing a two-dimensional mathematical model of cyclic adsorption-desorption processes in the adsorber of the PSA unit.

However, when using granular adsorbent with a particle diameter of more than 2 mm in the PSA unit, the use of two-dimensional (three-dimensional) mathematical models for calculations is promising, especially at gas flow velocities greater than 0.3 m/s, due to increasing divergence of velocities in the center of the vertical cylindrical adsorber and at its walls.

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