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ВЛИЯНИЕ ВХОДНОЙ ТЕМПЕРАТУРЫ НА ИЗОТЕРМИЧЕСКУЮ ОЧИСТКУ ГАЗА ОТ МОНОПРИМЕСИ НЕПОДВИЖНЫМ СЛОЕМ АДСОРБЕНТА

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Проведена оценка влияния температуры входного газа, содержащего монопримесь, на процесс физической адсорбционной очистки в пористом неподвижном слое гранулированного адсорбента. В основу теоретического анализа положена классическая математическая модель динамики изотермической адсорбции в пористой стационарной среде, структурно состоящей из дисперсной фазы твердых частиц, при принятых допущениях: очищаемый газ содержит малоконцентрированную монопримесь, его движение в адсорбере является однонаправленным, а выравнивающий эффект профиля скорости в поперечном сечении пористой среды позволил принять гидродинамический режим идеального вытеснения; аксиальное перемешивание в потоке газа незначимо; теплота адсорбции пренебрежимо мала; пористость слоя однородна; скорость адсорбции определяется уравнением кинетики сорбции с изотермой, подчиняющейся закону Генри. Сформулирована начально-краевая задача для системы дифференциальных уравнений в частных производных первого порядка, решение которой относительно концентраций моно-примеси в потоке газа и адсорбенте получено в явном аналитическом виде с помощью одностороннего интегрального преобразования Лапласа. Сравнительный анализ результатов вычислительного эксперимента с известными опытными данными показал, что предложенная модель при системе сделанных допущений вполне адекватно качественно и количественно описывает процесс адсорбционной сепарации. На примере функционирования промышленного адсорбера в блоке комплексной очистки ЦБ-120/120 в системе мобильной газодобывающей станции показано, что увеличение температуры входного осушенного атмосферного воздуха после компримирования, содержащего диоксид углерода, на 10 К сокращает время отработки в стадии адсорбции на 45%. Установлено, что изменение температуры входного потока газа оказывает существенное влияние на эффективность работы адсорбера и должно быть учтено при идентификации габаритных характеристик блока комплексной очистки.

Ключевые слова: адсорбция, изотермичность, входная температура, неподвижный слой, диоксид углерода, математическая модель, цеолит

INLET TEMPERATURE INFLUENCE ON ISOTHERMAL GAS CLEANING FROM MONO-IMPURITIES BY FIXED LAYER OF ADSORBENT

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The influence of the temperature of the inlet gas containing mono-impurity was evaluated for the process of physical adsorption purification in a porous fixed bed of granular adsorbent. The theoretical analysis is based on the classical mathematical model of the isothermal adsorption dynamics in a porous stationary medium structurally made-up of the dispersed phase of solid particles, under the assumptions made: the gas to be purified contains a low-concentration mono-impurity, its motion in the adsorber is unidirectional, and the levelling effect of the velocity pro-

file in the porous cross section environment allowed to adopt the hydrodynamic regime of plug flow; axial mixing in the gas flow is negligible; the adsorption heat is negligible; the layer porosity is uniform; adsorption rate is determined by the sorption kinetics equation with an isotherm, obeying Henry's law. An initial-boundary-value problem is formulated for a system of first-order partial differential equations, which solution with respect to mono-impurity concentrations in the gas stream and adsorbent is obtained in an explicit analytical form using the one-sided integral Laplace transform. A comparative analysis of the computational experiment results with known experimental data showed that the proposed model with an assumptions system is quite adequate qualitatively and quantitatively describes the adsorption separation process. Using the example of an industrial adsorber functioning in the ZB-120/120 complex purification unit in a mobile gas production system, it is shown that the temperature increase by 10 K of inlet dried air after compression containing carbon dioxide reduces the working time in the adsorption stage by 45%. It has been established that the temperature change in the gas inlet flow has a significant effect on the adsorber efficiency and should be taken into account when identifying the overall characteristics of the complex cleaning unit.

Key words: adsorption, isothermality, inlet temperature, fixed bed, carbon dioxide, mathematical model, zeolite

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INTRODUCTION

Mobile air separation units (ASU), for example TOPS-100V (transportable oxygen and nitrogen production station), are designed to produce high purity oxygen and nitrogen and can be operated in harsh temperature conditions [1]. In this regard, for ASU reliable and safe operation is necessary to remove associated impurities from atmospheric air (water vapor, carbon dioxide, carbon-hydrogen and others) after compression and pre-cooling, which is carried out in the complex air drying and purification unit (CCU) [2], which works on base of the principles of physical adsorption in a fixed granular layer of zeolite adsorbents [3-5]. The adsorbent operating volumetric average temperature in the CCU is approximately 281 K, however, the standard deviation during the cleaning process can reach 10 K [6]. However, it is known that with temperature increasing the adsorption activity of adsorbents decreases [7, 8]. Therefore, an undesirable penetration of impurities into the ASU elements occurs with the formation of their crystalline phase, which violates the functioning mode and creates explosive situations [9, 10]. The lack of technical ability to monitor the drift and localization of solidified impurities does not allow operation of the ASU for a long time in safe mode [11]. For this reason, the continuous ASU operation is determined empirically by a chromatographic analysis method based on the results of the purge gas heating. But at the same time, to ensure representativeness of the statistical sample and identify the causes of the excessive presence of impurities, a large amount of experimental material is required, which is not always possible [12, 13].

In this connection, the estimation of the impurities amount entering the ASU after CCU at an undetermined integral thermal regime is of theoretical and practical interest.

METHODOLOGY OF THEORETICAL ANALYSIS

The following assumptions are made (their detailed justification can be found, for example, in [14]): the gas being purified contains a low-concentration mono-impurity, its movement in the adsorber is unidirectional, and the equalizing velocity effect in the porous medium cross section allows one to adopt the ideal displacement mode; axial mixing in the gas stream is negligible; the adsorption heat is negligible; the layer porosity is constant; adsorption rate is determined by the kinetics sorption equation; the adsorption isotherm obeys Henry's law, which generalized form was used in [15]. Then, as the basic mathematical model of isothermal adsorption in a porous stationary medium structurally consisting of the dispersed phase of solid particles, we selected the classical model of the dynamics of gas purification in a granular adsorbent layer [16, 17], which, taking into account the accepted assumptions, is reduced to the following form:

$$-u\frac{\partial c(z,\tau)}{\partial z}-\frac{\partial a(z,\tau)}{\partial \tau}=\varepsilon\frac{\partial c(z,\tau)}{\partial \tau}; \qquad (1)$$

$$\frac{\partial a(z,\tau)}{\partial \tau} = \beta[c(z,\tau) - c^*(z,\tau)]; \quad (2)$$

$$a(z,\tau) = Hc^*(z,\tau), \quad (3)$$

where z is the axial Cartesian coordinate, m; τ is the time, s; u is the average gas velocity over the adsorber cross section, m/s; ε is the porosity; $c(z, \tau)$, $a(z, \tau)$ impurity concentration in the gas and adsorbent, kg/m^3; β kinetic coefficient of mass transfer at the gas-granule adsorbent interface, s^{-1}; $c^*(z, \tau)$ is the equilibrium impurity concentration in the gas, kg/m^3; $H = a_0/c_0$ is Henry's constant [18], where a_0 is the adsorbent capacity by impurity, kg/m^3; c_0 is the concentration of impurities in the gas at the inlet to the adsorber, kg/m^3. System (1)-(3) is supplemented with initial

$$c(z,0) = a(z,0) = 0 \quad (4)$$

with boundary conditions

$$c(0,\tau) = c_0; \;\; a(0,\tau) = Hc_0\left[1 - \exp\left(-\frac{\beta\tau}{H}\right)\right], \quad (5)$$

Please note that the form of the boundary condition for $a(0, \tau)$ is obtained from consideration of the kinetics of the adsorbent frontal layer filling.

Let h be the characteristic size of the cross section of the adsorbent layer, m; then system (1)-(5) using the relative variables $Z = z/h$, $\theta = \tau u/h$, $C(Z, \theta) = c(z,\tau)/c_0$, $A(Z, \theta) = a(z, \tau)/a_0$, Pe = $\beta h/u$ is the Peclet mass transfer number, takes a dimensionless form:

$$-\frac{\partial C(Z,\theta)}{\partial Z} - H\frac{\partial A(Z,\theta)}{\partial \theta} = \varepsilon\frac{\partial C(Z,\theta)}{\partial \theta}; \quad (6)$$

$$\frac{\partial A(Z,\theta)}{\partial \theta} = (\mathrm{Pe}/H)[C(Z,\theta) - A(Z,\theta)]; \quad (7)$$

$$C(Z,0) = A(Z,0) = 0; \quad (8)$$

$$C(0,\theta) = 1, \;\; A(0,\theta) = 1 - \exp\left(-\frac{\mathrm{Pe}\theta}{H}\right). \quad (9)$$

System (6)-(9) is translated using the one-sided integral Laplace transform to image format:

$$-\frac{dC_L(Z,s)}{dZ} - HsA_L(Z,s) = \varepsilon sC(Z,s); \quad (10)$$

$$sA(Z,s) = (\mathrm{Pe}\cdot H)[C_L(Z,s) - A_L(Z,s)]; \quad (11)$$

$$C_L(0,s) = \frac{1}{s}, A_L(0,s) = \left(\frac{\mathrm{Pe}}{H}\right)/\left[s\left(s + \frac{\mathrm{Pe}}{H}\right)\right], \quad (12)$$

where $C_L(0, s)$, $A_L(0, s)$ is the image $C(Z, \theta) = A(Z, \theta)$; s is a parameter.

From (10)-(12) it follows:

$$C_L(Z,s) = F_1(Z,s)F_2(Z,s); \quad (13)$$

$$A_L(Z,s) = \left(\frac{\mathrm{Pe}}{H}\right)C_L(Z,s)/\left(s + \frac{\mathrm{Pe}}{H}\right); \quad (14)$$

$$F_1(Z,s) = \left(\frac{1}{s}\right)/\left[-\mathrm{Pe}Zs/\left(s + \frac{\mathrm{Pe}}{H}\right)\right]; \quad (15)$$

$$F_2(Z,s) = \exp(-\varepsilon Zs). \quad (16)$$

The original from (15) was found by analogy with [19], that is, first the image is arranged in a series in s, and then its original is identified in the form of a converging nested series

$$f_1(Z,\theta) =$$

$$= 1 + \exp\left(-\frac{\mathrm{H}\theta}{\mathrm{Pe}}\right)\sum_{n=0}^{\infty}\sum_{k=0}^{n}\frac{(-ZH)^{n+1}\left(-\frac{H\theta}{\mathrm{Pe}}\right)^{\mathrm{k}}}{(n+1)[(n-k)!](k!)^2}. \quad (17)$$

The original (17) is equal [20]

$$f_2(Z,\theta) = \delta(\theta - \varepsilon Z), \quad (18)$$

where $\delta(\ldots)$ is the Dirac function [21]. By convolution theorem [20]

$$C(Z,\theta) = \int_0^{\theta} f_1(Z,\xi)f_2(Z,\theta-\xi)d\xi. \quad (19)$$

Taking into account the integral property of the Dirac function and its parity, it follows from (19)

$$C(Z,\theta) = 1 +$$

$$\exp\left[-\frac{\mathrm{H}(\theta-\varepsilon Z)}{\mathrm{Pe}}\right]\sum_{n=0}^{\infty}\sum_{k=1}^{n}\frac{(-ZH)^{n+1}\left[-\frac{H(\theta-\varepsilon Z)}{\mathrm{Pe}}\right]^{\mathrm{k}}}{(n+1)[(n-k)!](k!)^2}. \quad (20)$$

Original (14)

$$A(Z,\theta) = \frac{\mathrm{Pe}}{H}\int_0^{\theta} C(Z,\xi)\exp\left[-\frac{\mathrm{Pe}}{\mathrm{H}}(\theta-\xi)\right]d\xi. \quad (21)$$

The comparative analysis of the computational experiment results with the data from [22] (Fig. 1) showed that the proposed model, under the assumptions made, quite adequately describes the gas adsorptive separation process from a mono-impurity in a fixed adsorbent layer and, as an instrument, can be correctly used to assess the influence of the inlet temperature medium for the purification degree.

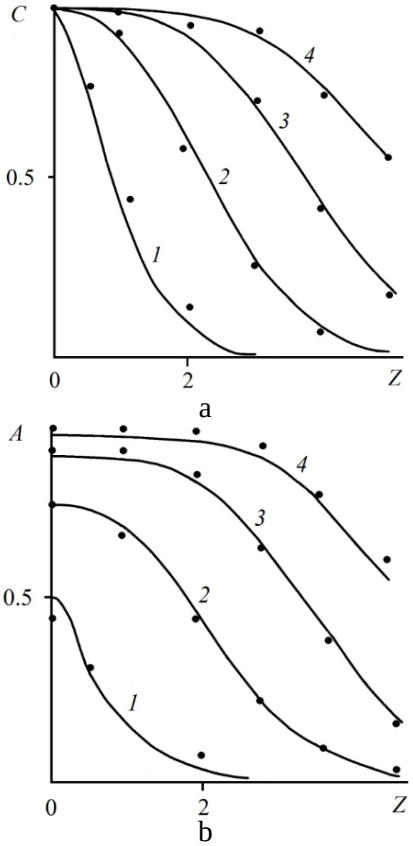

Fig. 1. The adsorption dynamics in a fixed bed at H=2, Pe=1, ε=0.4 (a – concentration of mono-impurity in the gas, b – concentration of mono-impurity in adsorbent) and various values of dimensionless time θ: 1 – 1.0; 2 – 3.0; 3 – 5.0; 4 – 7.0; - calculation using formulas (20) and (21); ● – data from [22]

Рис. 1. Динамика адсорбции в неподвижном слое при H=2, Pe=1, ε=0,4 (a – концентрация монопримеси в газе, b – концентрация монопримеси в адсорбенте) и различных значениях безразмерного времени θ: 1 – 1,0; 2 – 3,0; 3 – 5,0; 4 – 7,0; - расчёт по формулам (20) и (21); ● – данные из [22]

RESULTS AND DISCUSSION

Evaluation of the cleaning efficiency is considered on the example of the adsorber operation of the CCU ZB-120/120 (NPO «Kriogenmash») with the diameter and height of a non-moving layer of *NaX* zeolite particles (ε = 0.375, average granule diameter $d_p \approx 3\cdot10^{-3}$ m) respectively, h = 0.325 m and L = 1.380 m with a mass flow rate of dry air G = = 0.042 kg/s. The initial concentration of carbon dioxide (CO_2), which is a mono-impurity (the presence of acetylene and other impurities, is neglected due to their low concentration compared to the concentration of CO_2), is $4.6\cdot10^{-4}$ mass. According to the isobar of carbon dioxide adsorption on zeolite *NaX* at T = 281 K, there is a_r = 0.018 m^3/kg [6] (for dry atmospheric air). The values of molecular diffusion and density are calculated from the relations [23] at a working gas pressure p = 12 MPa

$$D = 2kT/(3\pi d_m^2\rho)\sqrt{\frac{RT}{\pi\mu}} = 1{,}06\cdot10^{-7}\ \text{м}^2/\text{с},$$

$$\rho = \frac{\mu\rho}{N_A kT} = 226{,}1\ \text{кг/м}^3,$$

where $k = 1.38\cdot10^{-23}$ J/K is the Boltzmann constant; $\mu = 44\cdot10^{-3}$ kg/mol – molar mass of CO_2; R = 8.31 J/(mol·K) – molar gas constant; $d_m = 2.9\cdot10^{-10}$ m is the effective diameter of the carbon dioxide molecule; $N_A = 6.022\cdot10^{23}$ mol^{-1} is the Avogadro number.

The adsorber cross-sectional area is $s = \pi h^2/4$ = = 0.083 m^2, then the gas velocity is $u = G/(\varepsilon\rho s)$ = = $6.0\cdot10^{-3}$ m/s. From the kinetics of filling granules of a spherical adsorbent granule using the «regular» mode [24] with averaging over the volume, the relation

$$\bar{A}(\theta) = \left\{1 - (6/\pi^2)\exp\left[-2\pi^2 Dh\theta/\left(d_p^2 u\right)\right]\right\},$$

which calculation (Fig. 2) indicates a uniform process, that is, $A(Z, \theta) = C(Z, \theta)$, then system (6)-(9) is simplified and transformed into an initial-boundary-value problem

$$-\frac{\partial C(Z,\theta)}{\partial Z} - H\frac{\partial C(Z,\theta)}{\partial \theta} = \varepsilon\frac{\partial C(Z,\theta)}{\partial \theta}; \quad (22)$$

$$C(Z,0) = 0,\ \ C(0,\theta) = 1. \quad (23)$$

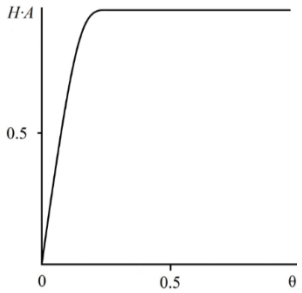

Fig. 2. Kinetics of adsorbent granule filling
Рис. 2. Кинетика заполнения гранулы адсорбента

Solutions (22) and (23):

$$C(0,\theta) = \mathbf{1}_+[\theta - Z(H+\varepsilon)], \quad (24)$$

where $\mathbf{1}_+(\ldots)$ is the one-way Heaviside function. Since $Z_L = L/h$ = 4.26, $a_0 = a\rho\rho_a$ = 2.442 kg/m^3 (zeolite density $\rho_a \approx$ 200 kg/m^3 [25], at an average volume pressure in the adsorber p = 4 MPa, c_0 is 0.026 kg/m^3 and, therefore, Henry's constant will be 108.053). From (24), the adsorber operating time $\tau = \frac{Z_L(H+\varepsilon)h}{3600u}$ = 6.93 h was obtained, which is consistent with the regulated operating time of the adsorbent before regeneration [6]. Recalculation at a_r = 0.015 m^3/kg, which corresponds to T = 291 K, gives τ = 3.83 h, that is, the working time of the adsorber is reduced by 45%.

CONCLUSION

The analysis using the proposed mathematical model showed that the gas inlet stream temperature change has a significant effect on the efficiency of the adsorber and should be taken into account when choosing its size characteristics.

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