IRSTI 61.13.15

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https://doi.org/10.55956/XWHV9151

CALCULATION OF ADSORPTION APPARATUSES WITH A LAYER OF EXPANDED SUBSTANCES

Abstract. This work is a part of the project for investigations of hydrodynamics and mass transfer in an adsorption apparatus with ion-exchange resins during the extraction of stevioside from stevia extract. The issues of improving the structural-sorption characteristics while increasing selectivity to certain substances, expanding the range, simplifying the manufacturing technology and reducing the cost of production of industrial adsorbents are extremely relevant. The work is devoted to mathematical modeling and a theoretical description of adsorption in the layer of expanded ion-exchange resins. The main aspects of adsorption in a layer of porous material have been considered, and a model of the adsorption layer structure has been developed. The contribution of the work and its scientific novelty lie in the fact that a mathematical model of the evolution of a two-component dispersed phase has been developed and qualitatively analyzed taking into account competitive adsorption and the non-equilibrium nature of mass transfer between the adsorption surface of the porous material and the dispersed phase. The results and conclusions are recommended to be used in planning and conducting research work for intensification of heat and mass transfer processes and for engineering design of adsorption chemical apparatuses and adsorption columns.

Keywords: adsorption, adsorption layer, adsorption wave, fractal dimension, ion-exchange resins multicomponent systems, stevia, porosity.

Bakhram Z.S., Bayisbay O.P., Azimov A.M., Seitkhanov N.T., Brener A.M., Pecherskiy V.N. Calculation of adsorption apparatuses with a layer of expanded substances // Mechanics and Technology / Scientific journal. – 2024. – No.2(84). – P.272-283. https://doi.org/10.55956/XWHV9151

Introduction. Adsorption apparatuses based on layers of expanded substances, ion-exchange resins in particular [1], have been widely used currently [2] both to purify multicomponent mixtures from impurities [3] and as an intermediate step for extracting useful components by adsorption [4]. During the porous adsorbents producing [5], the main problem is to create high porosity [6]. Besides it, the porous layer hold be also characterized by a uniform distribution of pores throughout the volume of the layer [7]. The rheological characteristics of the binder in the production of porous materials with an even cellular structure from the viscous-flowing compositions by swelling depend on the consumption of the binder and the duration of the mixing and molding cycles [8]. Ensuring optimal modes of swelling process [9] with a developed porous internal structure of the layer, and achieving the necessary geometric characteristics of the adsorption layer [10] require both an understanding of the features of the formation of the layer structure and a theoretical description of this complex process [11]. These requirements determine the method and parameters of the molding process [12], as well as the type of porous structure and properties of the products [13].

The rheological properties when producing highly porous adsorbents can be controlled by using a set of various techniques [14]: external influences on structured mixtures, in particular vibrations that cause a thixotropic effect, or the introduction of surfactants, temperature changes, etc. [15]. The need for such a combination of different techniques is due to the fact that none of the known techniques is without drawbacks [16].

Water, highly volatile liquids, and burnable solid additives are used as blow-forming agents. When using burnable additives, as a rule, a cellular porous structure is formed; when using evaporating liquids, a porous-capillary structure is formed [17]. If the mass concentration of the pore-forming agent in the material and the mass concentration of the structure-forming material are known, then the porosity of the resulting material can be calculated in advance.

One of the most widely used methods is swelling [18]. This method is based on the release or introduction of a gas phase into a plastic-viscous mass in the form of hydrogen, oxygen, carbon dioxide, water vapor, air, isopentenyl, Freon, or other substances.

As a result of saturation of the mass with the gas phase, its volume increases – swelling occurs (foaming). During swelling, a cellular porous structure is formed, the total volume of porosity of which depends on the amount of the gaseous component introduced and retained by the mass. Common to all types of swelling is the plastic-viscous state of the porous masses during the period of their porosity, i.e., the porous masses must have the ability to irreversibly deform (flow) without breaking the continuity. However, the process of swelling of the ion-exchanged resins has some peculiarities [19].

Namely, in this system, the so-called lamellar porosity is observed due to the sliding apart of the grains of the dispersed system by inter-packet water, which turns into a vapor state when the grains are heated. In the case of using a porous binder in the form of a foam mass, the material is obtained with cellular porosity, consisting of the porosity of the grains and the porosity of the binder [20].

The results of studies of the internal structure of adsorption layers using adsorption isotherm methods leads to the conclusion that the internal surface of the porous layer is characterized by an extremely complex developed shape [21]. This problem can be solved by using fractal geometry methods [22].

In this case, the inner surface of the layer is characterized by a fractal dimension higher than the “usual” geometric dimension of the surface [23].

The specific value of porosity and internal surface depends on the initial geometric characteristics of the layer, i.e. the size of the granules and the method of their laying [24].

This paper is devoted both to mathematical modeling of the geometric structure of porous adsorbents with account of the peculiarities of substances [20] exposed to swelling and to building a govern model for describing mass transfer of two-phase liquids flowing through the adsorbent porous layer [25].

Materials and methods. This section of paper is devoted to description of the main concepts for calculating the height of the adsorbent layer in the adsorption apparatuses. The process of removing a contaminant from a carrier gas in an adsorbent layer can be described using such a concept as the "adsorption wave" shown in Figure 1.

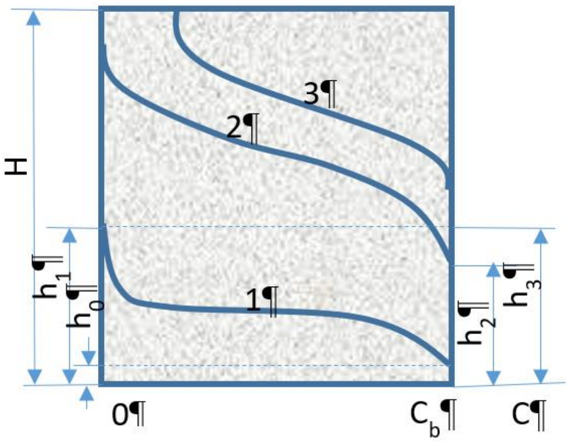


Fig. 1. Movement of an adsorption wave along a stationary layer

Curve 1 characterizes the dependence of the pollutant concentration on the position of the layer for fresh adsorbent, and the dashed horizontal line C_0 characterizes the maximum permissible concentration of the pollutant in gas emissions after adsorption treatment. With proper adsorption apparatus design, the output concentrations are much lower than this value. During operation, saturation occurs at the entrance to the adsorption layer and pollutants begin to penetrate further into the depth of the layer, where their effective adsorption occurs (curve 2). Finally, in cases where the maximum permissible concentration at the outlet of the layer is reached (curve 3), the adsorbent must be regenerated.

The adsorption process involves staged or continuous contacting, as in the case of liquid absorption, in which preferably only one component of the gas phase is absorbed. When more than one component is adsorbed, the process is more similar to fractionation, in particular extraction.

For dilute solutions, neglecting thermal effects, it is possible to obtain formulas corresponding to an isothermal process. Regardless of the nature of the adsorption forces, the magnitude of adsorption is influenced by the following factors: the nature of the adsorbed substance; temperature; pressure; impurities in the phase from which the substance is adsorbed [26]. As for the nature of the adsorbed substance that it is believed that the equilibrium concentration x^* is higher, the higher the molecular weight of the adsorbed gas, and in the case of solutions, the lower the solubility of the adsorbed substance in the liquid [23]. With increasing temperature, other things being equal, the equilibrium concentration x^* decreases. With increasing pressure in the vapor-gas phase, the equilibrium concentration increases.

As for the impurities in the phase from which the substance is absorbed that if there is a competing (displacing) substance B in the phase from which the adsorbent adsorbs substance A, i.e. of a substance that is also capable of being adsorbed by this adsorbent, the equilibrium concentration x* of substance A decreases. In this case, substance B either partially or completely displaces or replaces substance A in the adsorbent. Adsorbents are characterized by static and dynamic activity. After a certain period of operation, the adsorbent ceases to completely absorb the extracted component and a “breakthrough” of the component through the adsorbent layer is observed. From this moment, the concentration of the component in the exhaust gas-vapor mixture increases until equilibrium is reached.

The amount of substance absorbed per unit mass (or volume) of the adsorbent during the time from the beginning of adsorption to the beginning of the “breakthrough” determines the dynamic activity of the adsorbent. The amount of a substance absorbed by the same amount of adsorbent during the time from the beginning of adsorption to the establishment of equilibrium characterizes static activity (equilibrium activity).

As a result of the occurrence of some complex processes, such as, for example, wetting of the adsorbent or changes in the shape of the capillary openings and pores of the solid phase, the parameters characterizing the desorption equilibrium differ from the values determined for the adsorption equilibrium. The adsorption process is characterized by the presence of hysteresis, and the desorption pressure is always lower than the value obtained during adsorption.

Equilibrium during adsorption can be determined using various equations [23]. The sorption isotherm according to the Dubinin [22] equation is considered the most universal today:

$$C_p = \frac{k_0}{v_k} \exp\left[-b \left(\frac{T_{cp}}{z} \lg \frac{C^*}{C_0} \right) \right] \qquad (1)$$

where C is the equilibrium concentration, (kmol pollutant/kg adsorbent); k is a constant characterizing the maximum adsorption volume of micropores; , b is a constant characterizing the size of micropores; T – average process temperature, K; is the contaminant breakthrough concentration at the average process temperature and is the initial contaminant concentration.

For a non-stationary adsorption apparatus with a fixed layer, it is necessary also to determine the breakthrough moment. A slip occurs when the changing concentration (1) of a pollutant in the effluent stream reaches a certain set point, which may be, for example, the value allowed by standards for a given release. The time required to achieve breakthrough is determined from the mass transfer equations and equilibrium conditions; this, in turn, allows you to determine the required amount of adsorbent. Adsorption diameter layers are calculated, as in the case of absorption, based on the permissible value of hydraulic resistance [24].

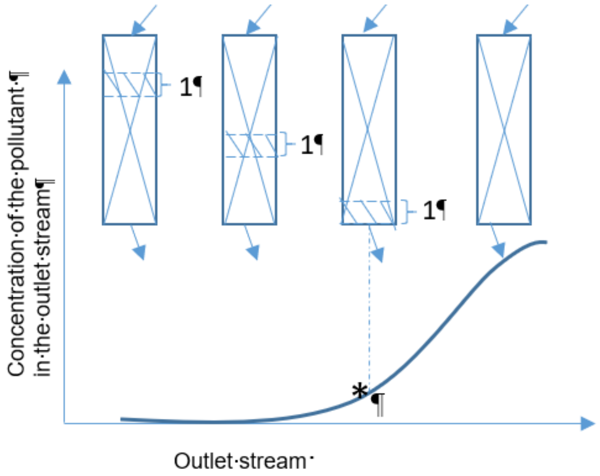
First, the adsorbent comes into contact with the incoming concentrated solution. As a result of passing through the entire adsorbent layer, almost all of the solute is removed from the solution. Figure 2 illustrates the process when the flow is supplied from top to bottom and corresponds to the case where the exit stream contains practically no solute. The uppermost part of the layer becomes saturated, and the main adsorption occurs in a relatively narrow part of the sorbent layer, where the concentration changes rapidly.

This narrow adsorption zone moves down the layer, forming a concentration wave; the displacement velocity is much less than the linear velocity of the gas passing through the sorbent layer. Over time, the solute concentration in the effluent increases. When it reaches a predetermined limit value, which, for example, may be determined by environmental regulations, it is considered that a breakthrough has occurred. After this, the concentration of the dissolved substance increases rapidly, since the adsorption zone extends beyond the sorbent layer; eventually the solute concentration at the outlet becomes the same as that in the original solution. The dependence of the concentration on the volume of the outlet flow in this region is called the breakthrough curve.

Using Figure 2, you can determine the required height of the adsorbent layer in the apparatus for a given degree of purification.

If the rate of filtration of the mixture through the layer is known, and the speed of the breakthrough wave is known also, then the height of the layer is determined by the formula:

$$H = \tau \lambda W \tag{2}$$



* – breakthrough point; 1 – adsorption zone;

Fig. 2. Non-stationary adsorption in a fixed sorbent layer

where τ is the residence time of the mixture in the apparatus.

Calculation procedure for periodic adsorption apparatus can be written as follows:

The most common adsorption process is cleaning in devices with a fixed layer of solid phase. Processes of this kind are non-stationary and periodic. In this case, the concentrations in the solid material and gas located inside the apparatus change over time.

In the fixed adsorbent layer, the adsorption zone moves along the column.

Calculations of periodic adsorption apparatuses with a stationary adsorbent using the Dubinin equation (1) and relation (2) can be performed in the following order [22].

1. Determine the equilibrium concentration of the pollutant in the solid phase. The concentrations of the pollutant in the adsorbent, which are in equilibrium at a given temperature with its concentrations in the gas phase, are expressed as a sorption isotherm. Using the known sorption isotherm, the amount of pollutant that can be absorbed by the adsorbent at a given temperature is determined if the process continues to an equilibrium state.

The sorption isotherm of the pollutant in question is calculated using the special thermodynamic graphs [22].

2. Select the type of adsorption apparatus and its design parameters. Take a fictitious speed of cm6e in the adsorption apparatus w = 0.05...0.1 m/s, calculate the diameter of the apparatus at a given flow rate and select the closest standard size of the adsorption apparatus of the selected design. Based on the design characteristics of the apparatus and the speed of the breakthrough wave, an acceptable height of the adsorbent layer is selected.

3. Determine the mass transfer coefficient.

During adsorption on the expanded substance, the mass transfer coefficient in the continuous phase can be found from the equation [8]:

$$Nu = 0{,}355\mathrm{Re}^{0{,}64}\frac{\mathrm{Pr}^{0{,}333}}{\varepsilon} \tag{3}$$

The mass transfer coefficient from the continuous phase to the surface of the adsorbent is found by the formula:

$$\beta_1 = Nu\frac{D_a}{d_p} \tag{4}$$

To estimate the value of the internal diffusion coefficient, you can use the equation [16]:

$$D_a = \frac{\varepsilon D}{2}\left[1-\exp\left(-\frac{2d_n}{3D}\sqrt{\frac{8RT}{m}}\right)\right] \tag{5}$$

Mass transfer coefficient:

$$K = \frac{1}{1/\beta_1 + 1/\beta_2} \tag{6}$$

Deviation from the ideal displacement regime due to longitudinal mixing can be taken into account by introducing additional diffusion resistance, for which the longitudinal mixing coefficient is determined [8]:

$$\Lambda = 0{,}057\frac{W\,\mathrm{Re}^{0{,}22}}{1-\varepsilon} \tag{7}$$

The mass transfer coefficient (m/s) adjusted for longitudinal mixing is found from the relationship:

$$K' = \frac{1}{1/K + 1/\Lambda} \tag{8}$$

Set of formulas (3) - (8) can be considered as methods for calculation the main control parameters for the mass transfer description.

It is convenient to present the results of calculations of the output curve graphically in the form of a dependence of the dimensionless concentrations of C/C_b on the dimensionless time of the process T. From this graph, for a given output (final) concentration, it is not difficult to find the corresponding dimensionless calculated time and the estimated duration of the adsorption stage.

Check the convergence of the found parameters, drawing up a material balance. To do this, the amount of pollutant entering the adsorption apparatus is calculated and compared with the amounts of pollutant absorbed by the adsorbent, remaining in the gas phase of the adsorbent layer and leaving the apparatus in accordance with a given breakthrough.

Determine the duration of auxiliary stages of the mixture processing process.

When designing an adsorption scheme, the duration of each of the auxiliary stages – desorption, drying and cooling of the adsorbent – is assumed to be the same as the calculated adsorption duration.

The duration of the remaining phases of the cycle (desorption, drying and cooling of the adsorbent) is calculated, as a rule, on the basis of experimental data or empirical equations (due to the complexity of the mathematical description of the corresponding processes, due mainly to the internal porosity of the adsorbent).

Research results and discussions. An assessment of the porosity of a layer of a plate-like adsorbent, in particular, vermiculite, taking into account two characteristic scales, can be made based on the following relationship [9]:

$$\varepsilon \approx \left(l_1/l_2\right)^{3-D} \tag{9}$$

From here an estimation for the fractal dimension of the swollen adsorbent layer can be obtained as follows [2, 8]:

$$D \approx 3 - \left[\ln\left(\frac{\varepsilon_1}{1.4} + \varepsilon_0\right) \Big/ \ln\left(l_1/l_2\right)\right] \tag{10}$$

where the indices 1 and 0 for porosity ε refer to the final and initial states of the layer, respectively. The derivation of Eqs (9) and (10) is based on the ideas proposed in the work [8].

Typically, the main source of data on disperse systems is the results of rheological experiments. At the same time, it is known that during the flow of multicomponent multiphase liquids in porous structures, nonlinear effects are observed, which manifest themselves both in the form of adsorption effects, which affects the dynamics of adsorption efficiency indicators, and at the level of specific rheological effects, for example, in the form of a blocking effect flow [22]. Therefore, the dynamic properties of complex disperse systems are not determined only by rheological characteristics.

Unusual flow regimes of complex multicomponent liquids in porous media are manifested in the nonuniform distribution of the adsorbed substance in the adsorption layer. This is explained by the interaction of the components with each other and their competition when filling free adsorption centers on a solid surface, since many multicomponent mixtures are colloidal systems in which surfactant components form a dispersed phase.

This paper presents a mathematical model of the movement of a two-component dispersed phase, taking into account competitive adsorption and the non-equilibrium nature of mass transfer between the adsorption surface of the porous layer and the dispersed phase.

The basic system of model equations for a flow one-dimensional layer scheme is as follows [25]:

$$\varepsilon\frac{\partial c_i}{\partial t}+\frac{\partial a_i}{\partial t}=D_i\frac{\partial^2 c_i}{\partial x^2}-u\frac{\partial c_i}{\partial x}, \tag{11}$$

$$\frac{\partial a_i}{\partial t}=\beta_i\left(c_i-b_i a_i^2\left(1-\sum_k \lambda_k a_k\right)^{-2}+\sum_{j\neq i}k_{ij}a_i a_j\right), \tag{12}$$

The mathematical model (11), (12) is not closed and must be supplemented with a relation for the connection between equilibrium and non-equilibrium concentrations of components in the adsorbed state.

In the work by A. Brener [25] it was proposed to use the simplest form of such a connection:

$$\tau\frac{\partial a_i}{\partial t}+a_i=\tilde{a}_i \tag{13}$$

were $\tilde{a}_i$ is some pseudo-equilibrium value, and τ is the characteristic relaxation time.

System of equations (11), (12) is solved under the following initial and boundary conditions:

$$c_i(0,t)=c_i^0,\quad \left.\frac{\partial c_i}{\partial x}\right|_{x=L}=0,\quad t\geq 0 \tag{14}$$

$$a_i(x,0)=0,\quad c_i(x,0)=0,\quad 0<x<L \tag{15}$$

However, taking into account the relaxation time requires modification of the basic transport equation (1). Therefore, in this paper it is proposed to use the methodology of, based on the concept of relaxation transfer kernels [25]. In addition, this approach makes it possible to study the effect of non-isothermal process on the intensity of adsorption.

The system of equations (9) - (15) represents a development of methodology proposed in [25]. This methodology is based on the representation of transport equations in the form of integer-differential equations, which make it possible to take into account the phenomena of nonlocality and delay using a special form of kernels of integral operators in the equations.

The more detail analysis of the derived equations of adsorption kinetics showed the possibility of the existence of two types of solutions to the model, namely: solutions of the traveling concentration wave type for the case when the speed of adsorption or the degree of adsorption activity on the surface of a porous medium for one of the components clearly exceeds the others, as well as specific solutions describing oscillatory concentration processes of more complex dynamics [26]. At the same time, the detail analysis of the stability of the solutions obtained yet should be completed. Note that the resulting system of equations contains the entire variety of discovered solutions, and it describes the corresponding modes of adsorption processes. This gives grounds to propose the model outlined in the submitted paper for a detailed description of the flow of a two-phase liquid in a layer of a porous adsorbent of complex structure.

For a more detailed study of the system, a bifurcation diagram was constructed for a “nested” dynamic system of equations (11, 12) at a fixed point in space (x=0). As a result, it was revealed that in the various range of control parameters values the phase portrait can consist of unstable nodes, stable nodes and saddles. These states correspond to damped or unfading oscillations in the concentrations of adsorbed components. In all cases when the system exhibits oscillations in a nested “dynamic” system, a response to them appears in the form of a nonmonotonic distribution of concentrations of substances in the flow over space. Figure 3 shows the typical results of processing the data obtained on the penetration of various pollutants through a layer of expanded vermiculite. Both the case of fading (*A*) and unfading oscillations (*B*) is presented in Figure 4.

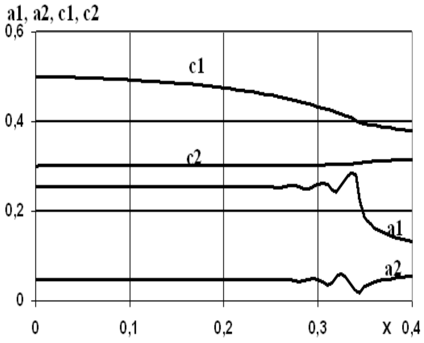


Fig. 3. Distribution of concentrations of substances the flow

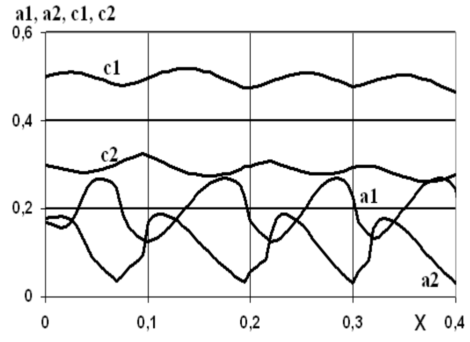


Fig. 4. Distribution of concentrations of substances the adsorbed state

A larger volume of computer research remains to be carried out, as well as a comparison of the results obtained with known experimental data. Such work will require separate specialized training to clearly define the verification concept.

Conclusion. The work devotes for carrying out the engineering model for describing mass transfer processes in non-equilibrium systems based on the methodology of relaxation transfer kernels applying to the dynamics of the adsorption process of a two-component liquid in the expanded adsorbents layer. The results of a qualitative and numerical study of the model are presented. At the same time, the possibility of the occurrence of wave modes of the adsorption process is shown. Possible types of emerging patterns have been identified. The fractal structure of the expanded adsorbent layers has been studied and corresponding estimates of the geometric parameters of the layer have been obtained. Further efforts will be aimed at identifying areas of different dynamics in the space of control parameters of the proposed novel model.

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Material received on 16.05.24.

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КЕҢЕЙГЕН СУБСТАНЦИЯЛАР ҚАБАТЫМЕН АДСОРБЦИЯЛЫҚ АППАРАТТАРДЫ ЕСЕПТЕУ

Аңдатпа. Жұмыс стевия сығындысынан стевиозидті алу кезінде ион алмастырғыш шайырлары бар адсорбциялық аппаратта гидродинамика мен масса алмасуды зерттеу жобасының бөлігі болып табылады. Жеке заттарға қатысты селективтілікті арттыру, ассортиментті кеңейту, өндіру технологиясын жеңілдету және өнеркәсіптік адсорбенттерді өндірудің өзіндік құнын төмендету кезінде құрылымдық және сорбциялық сипаттамаларын жақсарту мәселелері өте өзекті. Жұмыс кеңейтілген ион алмастырғыш шайырлар қабатындағы адсорбцияны математикалық модельдеуге және теориялық сипаттауға арналған. Кеуекті материал қабатындағы адсорбцияның негізгі аспектілері қарастырылып, адсорбциялық қабат құрылымының моделі жасалған. Жұмыстың қосқан үлесі және оның ғылыми жаңалығы бәсекелестік адсорбцияны және кеуекті материалдың беткі адсорбциясы мен дисперсті фаза арасындағы масса алмасудың тепе-теңдік сипатын ескере отырып, екі компонентті дисперсті фаза эволюциясының математикалық моделі әзірленіп, сапалы талданғандығында. Нәтижелер мен қорытындыларды жылу және масса алмасу процестерін интенсификациялау бойынша ғылыми-зерттеу жұмыстарын жоспарлау және жүргізу кезінде, сондай-ақ адсорбциялық химиялық аппараттар мен адсорбциялық колонналарды жобалау кезінде пайдалану ұсынылады.

Тірек сөздер: адсорбция, адсорбциялық қабат, адсорбция толқыны, фракталдық өлшем, ион алмастырғыш шайырлардың көпкомпонентті жүйелері, стевия, кеуектілік.

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РАСЧЕТ АДСОРБЦИОННЫХ АППАРАТОВ СО СЛОЕМ РАССЕЯННЫХ ВЕЩЕСТВ

Аннотация. Работа является частью проекта по исследованию гидродинамики и массопереноса в адсорбционном аппарате с ионообменными смолами при экстракции стевиозида из экстракта стевии. Крайне актуальны вопросы улучшения структурно-сорбционных характеристик при повышении селективности по отношению к отдельным веществам, расширении ассортимента, упрощении технологии изготовления и удешевлении производства промышленных адсорбентов. Работа посвящена математическому моделированию и теоретическому описанию адсорбции в слое вспученных ионообменных смол. Рассмотрены основные аспекты адсорбции в слое пористого материала и разработана модель строения адсорбционного слоя. Вклад работы и ее научная новизна заключаются в том, что разработана и качественно проанализирована математическая модель эволюции двухкомпонентной дисперсной фазы с учетом конкурентной адсорбции и неравновесного характера массопереноса между адсорбциями поверхности пористого материала и дисперсной фазы. Результаты и выводы рекомендуется использовать при планировании и проведении научно-исследовательских работ по интенсификации процессов тепло- и массообмена, а также при проектировании адсорбционных химических аппаратов и адсорбционных колонн.

Ключевые слова: адсорбция, адсорбционный слой, волна адсорбции, фрактальная размерность, многокомпонентные системы ионообменных смол, стевия, пористость.