MATHEMATICAL MODEL OF ADSORPTION OF HIGH MOLECULAR WEIGHT COMPOUNDS IN A COLUMN-TYPE APPARATUS

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Abstract. This article is devoted to researching the adsorption of high-molecular compounds in a column-type apparatus, which allows for taking into account the specific properties of adsorbed substances and the design features of the equipment. The purpose of the study was to create a mathematical model that describes the process of adsorption of high-molecular compounds in a column-type apparatus, taking into account the specific properties of the adsorbed substances and the design features of the equipment. Equations that consider the kinetic aspects of adsorption and desorption are used to describe the dynamics of the process. It was noted that the large size of molecules, their complex structure, and environmental conditions can significantly affect the efficiency of the adsorption process. The model considers phenomena such as diffusion in a porous medium, the influence of competition between different components of the mixture, and possible changes in the structure of adsorbed molecules. The obtained data made it possible to determine the optimal conditions for achieving maximum adsorption efficiency and assess the effect of changing the process parameters on the initial products. Comparison of experimental and theoretical data indicates the adequacy of the obtained model and high convergence of results. The developed model can be used for forecasting and optimizing industrial processes, where the adsorption of high-molecular compounds is a crucial stage, Including biotech manufacturing, the pharmaceutical industry, water treatment, and other industries. Thanks to the possibility of predicting the system's behaviour when conditions change, the model can be a tool for improving existing technological processes, reducing costs and improving the quality of the final product. The developed mathematical model is essential to a deeper understanding of the adsorption processes of high-molecular compounds in column-type devices. It not only allows for the analysis of the current state of the system but also provides for the possibility of its adaptation to new production conditions and needs. This opens up new opportunities for the development of technologies in various industries.

Keywords: wastewater, macromolecular compounds, adsorption dynamics, albumin, activated carbon, column-type devices.

1. Introduction

Extraction of high molecular weight organic compounds from wastewater is a critical task for environmental protection and human health. High molecular weight organic compounds such as proteins, synthetic polymers, humic substances, dyes and other macromolecules can enter wastewater from various sources, including industrial production, agriculture and domestic waste. Untimely and insufficient purification of wastewater from these substances can lead to serious environmental and health problems. High molecular weight organic compounds can have a negative impact on aquatic ecosystems because they are able to absorb light, reducing the penetration of sunlight into water. This can lead to a decrease in the photosynthetic activity of algae and a decrease in the level of oxygen in the water, which negatively affects

fish and other aquatic organisms. Some compounds can be toxic to aquatic life or promote the development of pathogenic microorganisms. High molecular weight organic compounds can penetrate into aquifers and sources of drinking water. They can not only deteriorate the organoleptic properties of water (e.g. smell, taste) but also pose a threat to human health, especially if these compounds are potential carcinogens, mutagens or toxins. Also, the presence of high molecular weight organic compounds can complicate traditional water treatment methods, such as coagulation, filtration or biological treatment. They can form stable colloidal systems or protect smaller molecules, making their removal difficult. This can lead to higher water treatment costs and the need for additional or more efficient treatment technologies. One of the key challenges in these industries is the efficient separation and purification of such compounds. Adsorption is one of the most widely used methods for this purpose, as it allows the efficient removal of unwanted impurities or the concentration of target molecules. In modern conditions, the development of mathematical models that allow not only to understand the processes of adsorption but also to predict the behaviour of the system under different conditions is becoming more and more relevant. However, high molecular compounds often have a complex structure and specific properties that can significantly affect the adsorption process. In addition, column-type devices have their own design features that can affect the hydrodynamics of the system and mass transfer processes. It is not enough to consider only the static characteristics of adsorption; it is necessary to develop dynamic models that will allow evaluation of the behaviour of the system in actual operating conditions. Thus, there is a need to create an adequate mathematical model that takes into account all the above-mentioned factors and allows for the effective prediction of the process of adsorption of high-molecular compounds in column-type devices. Mathematical modelling of adsorption processes of high-molecular compounds in column-type devices is an extremely important direction of modern science and technology, particularly in the fields of chemical, pharmaceutical, food industry and environmental protection. High-molecular compounds, such as proteins, polymers, ligands and other macromolecules, have unique physicochemical properties that determine their specific behavior during adsorption. Mathematical models make it possible to predict the behaviour of the system, which makes it possible to optimize adsorption processes from the point of view of energy efficiency, consumption of raw materials and time. This is especially important in the conditions of industrial production, where efficiency processes directly affect economic indicators. This contributes to increasing the efficiency of technological processes, reduction ecological impact and ensuring high quality of final products.

2. Experimental part

To study the kinetics of adsorption of high-molecular compounds on activated carbon, a column apparatus with a polyethene column with a diameter of 1 cm and a variable height of the sorbent layer from 1 to 30 cm was used (Sabadash, 2023). The column apparatus provided a constant flow of the solution through the sorbent layer, which made it possible to simulate adsorption processes in dynamic conditions. 0.2 dm³ of an aqueous solution of high molecular weight compounds with an initial concentration of 0.5 mg/dm³ was introduced into the studied system. A model solution of albumin was used as a high-molecular compound. The albumin solution was prepared in distilled water to ensure the absence of impurities that could affect the adsorption results. Activated carbon was loaded into the column, previously washed and dried to a constant mass. The height of the sorbent layer varied from 1 to 30 cm to study the influence of the layer thickness on the adsorption efficiency. The solution was injected into the upper part of the column at a constant rate, ensuring a uniform flow through the sorbent layer. An important parameter was controlling the flow rate to avoid breakthrough phenomena or insufficient interaction between the solution and the sorbent. The flow rate was adjusted using a peristaltic pump, establishing optimal conditions for maximum contact of the compounds with the sorbent. Samples of the solution were taken at the exit from the column after particular time intervals. The selected samples were analyzed for albumin content using a photometric and appropriate analysis methods (Hyvlud et al., 2019). The adsorption efficiency was determined as a change in the albumin concentration in the solution after passing through the sorbent layer. In parallel, control experiments were conducted without a sorbent to determine possible adsorbate losses due to other processes not related to adsorption. The obtained data made it possible to construct adsorption kinetic curves, determine the influence of the sorbent layer's thickness on the process's speed and efficiency, and also evaluate the potential of activated carbon as an adsorbent for albumin removal.

3. Results and Discussion

To adapt the mathematical model for the adsorption of high-molecular compounds in a layer of activated carbon, the specific properties of such compounds and the adsorbent material can be taken into account (Sabadash, Gumnitsky, 2019). The main points of adaptation include correcting diffusion parameters and mass transfer coefficients and considering specific conditions of adsorption of high-molecular-weight compounds.

Diffusion flow model taking into account internal and external diffusion:

1. Equation for the mobile phase:

$$\frac{\partial C}{\partial t} = D_Z \frac{\partial^2 C}{\partial z^2} - V_i \frac{\partial C}{\partial Z} - \frac{3k_f(1-\varepsilon)}{R\varepsilon}\left[C - C_p(r=R)\right], \quad (1)$$

where C is the concentration in the mobile phase; D_Z is the diffusion coefficient along the axis of the column; V_i is the transfer rate in the mobile phase; k_f is the mass transfer coefficient; ε is the layer porosity; R is the radius of the activated carbon granule; *Cp* is the concentration in the pore space on the surface of the granule.

2. Equations for the stationary phase (pore and surface transport):

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left[r^2 D_s \rho_a \frac{\partial q}{\partial r} + r^2 D_p \varepsilon_p \frac{\partial C_p}{\partial r}\right] = \frac{\partial}{\partial t}\left[\rho_a q + \varepsilon_p C_p\right], (2)$$

where *r* is the radial coordinate inside the granule; *D*s is the surface diffusion coefficient; ρ_a is the bulk density of the adsorbent; q is the adsorbed concentration; *D*p is the pore diffusion coefficient; ε_p is the sorbent porosity.

Correction of diffusion coefficients is possible in the case of adsorption of high-molecular compounds. Due to their large size and mass, high-molecular-weight compounds have much less diffusion. This may require reducing the values of *D*Z, *D*s, and *D*p.

It is necessary to take into account possible steric effects and limited access to micropores in the adsorbent. This can change the volumes available for diffusion and adsorption, as well as affect mass transfer coefficients (Garba, 2019).

High-molecular compounds can have different kinetic characteristics of adsorption, particularly a slower saturation rate, which needs to be taken into account in the model.

It is also necessary to consider the possible influence of temperature and other conditions on the stability of high-molecular compounds and their interaction with the surface of activated carbon.

If necessary, additional parameters or submodels can be added to consider specific interactions between high-molecular compounds and the surface of activated carbon, for example, taking into account specific sorption centres or chemical adsorption (Sabadash, 2023).

The adsorption equilibrium isotherm (AEI) describes the relationship between the adsorbate concentration in the solution and the amount adsorbed on the adsorbent surface. The Langmuir adsorption model is one of the most common models used for the mathematical description of adsorption equilibrium in aqueous systems. This model provides a basic understanding of the adsorption process, assuming that adsorption occurs on specific homogeneous areas of the adsorbent surface, where each can hold only one adsorbate molecule (Sabadash & Omelianova, 2021). In addition, the model assumes that all areas of the adsorbent have the same adsorption energy and that there is no interaction between adsorbed molecules.

According to the Langmuir model, the equation for describing adsorption can be written in the form:

$$a^* = a_\infty \frac{KC}{1+KC}$$

where a* is the amount of adsorbed substance per unit mass of adsorbent (mg/g); a $_\infty$ is the maximum adsorption capacity, which corresponds to the complete coverage of the surface of the adsorbent (mg/g); K is the Langmuir adsorption equilibrium constant, which is related to the adsorption energy (dm³/mg); C is the equilibrium concentration of the adsorbate in the solution (mg/dm³).

The Langmuir model is widely used due to its simplicity and effectiveness in modelling various systems where adsorption occurs on the surface of a solid material. However, it has certain limitations, in particular, it does not consider possible interactions between adsorbate molecules on the surface and the variety of adsorbent sites. For more complex systems, other models may be needed, such as the Freundlich or Dubinin-Radushkevich model, BET, which considers the heterogeneity of the adsorbent surface and other factors. The complexity of solving the nonlinear partial differential equation for the mass balance of the stationary phase (Equation (2)) led to a simplification based on the assumption of a linear nature of the equilibrium isotherm for highly diluted solutions. This simplification made it possible to

assume that at the initial stages of adsorption, the achievement of dynamic equilibrium on the surface of the sorbent occurs faster. As a result, pore and surface diffusion were combined into a single intraparticle diffusion process, denoted as Di. This assumption simplifies the modelling of adsorption, reducing the number of unknown parameters and facilitating the solution of equations that describe the processes of transport of substances in the system (Sabadash & Omelianova, 2021). The combination of diffusion processes makes it possible to more effectively describe the mass transfer inside the pores of the adsorbent, taking into account their joint contribution to the adsorption dynamics.

$$D_i = \frac{D_p \varepsilon_p + D_s \rho_a K}{\varepsilon_p + \rho_a K}. \tag{3}$$

Accordingly, equation (2) will take the form:

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 D_i \frac{\partial C_p}{\partial r} \right] = \frac{\partial C_p}{\partial t}. \tag{4}$$

At the same time, equation (1) remains unchanged. This combination is known as the dispersed flow model with an intradiffusive component. However, if we consider most of the physical transport mechanisms but ignore pore diffusion and axial dispersion, we obtain a homogeneous surface diffusion model. When axial dispersion is neglected, the equation of the mobile phase (equation (1)) is simplified to:

$$\frac{\partial C}{\partial t} = -V_i \frac{\partial C}{\partial z} - \frac{3k_f(1-\varepsilon)}{R\varepsilon} \left[C - C_p \right]. \tag{5}$$

To find the solution of these two partial differential equations, we will use the methods of separation of variables and the Laplace transform. Let us start with the first equation:

$$\frac{\partial C}{\partial t} = D_Z \frac{\partial^2 C}{\partial z^2} - V_i \frac{\partial C}{\partial z} - \frac{3k_f(1-\varepsilon)}{R\varepsilon} \left[C - C_p(r = R) \right]. \tag{6}$$

Initial and boundary conditions:

Initial condition: $C(z,t) = 0$ for $0 \le z \le L$, $t = 0$.

Boundary condition: $C(z,t) = C_0$ for $z = 0$, $t > 0$.

Boundary condition: $\frac{\partial C(z,t)}{\partial z} = 0$ for $z = L$, $t > 0$.

Solution of the first equation:
Method of separation of variables.
Suppose that the solution can be given in the form: $C(z,t) = Z(z)T(t)$. Then:

$$\frac{\partial C}{\partial t} = Z(z)T'(t)$$

$$\frac{\partial^2 C}{\partial z^2} = Z''(z)T(t)$$

$$\frac{\partial C}{\partial z} = Z'(z)T(t).$$

Substituting these expressions into the original equation, we get:

$$Z(z)T'(t) = D_Z Z''(z)T(t) -$$
$$- V_i Z'(z)T(t) - \frac{3k_f(1-\varepsilon)}{R\varepsilon} Z(z)T(t). \tag{7}$$

Dividing both parts by $Z(z)T(t)$, we get:

$$\frac{T'(t)}{T(t)} = D_Z \frac{Z''(z)}{Z(z)} - V_i \frac{Z'(z)}{Z(z)} - \frac{3k_f(1-\varepsilon)}{R\varepsilon}. \tag{8}$$

This equation is divided into two independent differential equations:

For the function $T(t)$:

$$\frac{T'(t)}{T(t)} = -\lambda. \tag{9}$$

For the function $Z(z)$:

$$D_Z Z''(z) - V_i Z'(z) - \left(\frac{3k_f(1-\varepsilon)}{R\varepsilon} + \lambda \right) Z(z) = 0. \tag{10}$$

Solution for $T(t)$:

$$T(t) = T_0 e^{-\lambda t}. \tag{11}$$

Solution for $Z(z)$:

It depends on the solution of the characteristic equation. Depending on the roots, we get complex, real, or multiple roots.

We present equation (2) in the following form:

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 D_s \rho_a \frac{\partial q}{\partial r} + r^2 D_p \varepsilon_p \frac{\partial C_p}{\partial r} \right] =$$
$$= \frac{\partial}{\partial t} \left[\rho_a q + \varepsilon_p C_p \right]. \tag{12}$$

Initial and boundary conditions:
Initial condition: $q = 0$ for $0 \le r \le R$, $t = 0$.
Boundary condition: $\frac{\partial q}{\partial r} = 0$ for $r = 0$.

Boundary condition: $D_s \rho_a \frac{\partial q}{\partial r} = k_f \left[C(t) - C_p(r = R) \right]$ for $r = R$.

We solve the second equation using the method of separation of variables

Suppose that the solution has the form $q(r,t) = R(r)T(t)$. After substitution and separation of variables, we get:

For the function $T(t)$:

$$\frac{T'(t)}{T(t)} = -\lambda. \tag{13}$$

For the function $R(r)$:

$$\frac{1}{r^2} \left[r^2 D_s \rho_a R'(r) \right] + \frac{1}{r^2} \left[r^2 D_p \varepsilon_p R'(r) \right] =$$
$$= -\lambda R(r). \tag{14}$$

For a complete solution of these equations, it is necessary to determine the integration constants taking into account the boundary conditions and possib-

le physical parameters of the system. This usually involves numerical methods or special functions corresponding to physical conditions.

To derive the analytical expression that gives the ratio between the concentration in the liquid phase and the initial concentration C_0 (relative concentration) for the considered model, we will use the analytical method to solve the equations of adsorption dynamics. The basis for derivation is the solution of a differential equation system that describes mass transfer in the liquid phase and the processes occurring in the adsorbent (Kindi et al., 2023).

After the Laplace transform and using the boundary conditions, we will obtain the solution for the concentration in Laplace space and then perform the inverse Laplace transform to obtain the solution in time-space.

The resulting solution looks like this:

$$\frac{C}{C_0} = \frac{1}{2}\left[1 + \mathrm{erf}\left(\sqrt{\tau} - \sqrt{\xi} + \frac{1}{8\sqrt{\tau}} + \frac{1}{8\sqrt{\xi}}\right)\right], \quad (15)$$

where $\mathrm{erf}(x)$ is the error function arising from the integration of the normal distribution, τ is a dimensionless parameter characterizing mass transfer in the liquid phase, ξ is a dimensionless parameter that considers the time and speed of the adsorption front.

3. Definition of parameters:

To obtain an expression in terms of dimensionless parameters:

$$\tau = \frac{k_f K_o Z}{V_i}\left(\frac{1-\varepsilon}{\varepsilon}\right), \quad (16)$$

$$\xi = k_f\left(t - \frac{Z}{V_i}\right). \quad (17)$$

The obtained expression $\frac{C}{C_0}$ as a function of time and spatial coordinates is the result of solving the equation of convective-dispersive transport taking into account intradiffusive mass transfer. It makes it possible to estimate the concentration of the dissolved substance at any point of the liquid phase as a function of time, which is essential for predicting the efficiency of the adsorption process.

An alternative solution for this model can also be presented as:

$$\frac{C(L,T)}{C_0} = \frac{1}{2}\mathrm{erfc}\left[\frac{Pe^{1/2}}{2}\frac{(1-T)}{T^{1/2}}\right] + \frac{1}{\pi^{1/2}}\left[\frac{T}{Pe}\right]^{1/2}, \quad (18)$$

where T is the throughput of the column, and Pe is the Peclet number.

Taking into account the Freundlich isotherm, the system of equations (1) and (2) takes the following form:

$$C = \left(\frac{C_0^{n-1}}{1 + A \cdot \exp(-rt)}\right), \quad (19)$$

where r and A are constants of the logistic function. A defined as:

$$A = \left((C_0^{n-1}/C_b) - 1\right)/\exp(-rt_b), \quad (20)$$

where $C_к$ is the adsorbate concentration after $t_к$ the service time, as during breakthrough.

Fig. 1 compares theoretical and experimental data on adsorption by activated carbon.

y 2 = 0.1143x y 5 = 0.025x - 0.0146 y 7 = 0.0187x - 0.1216 y 20 = 0.0232x - 0.9523
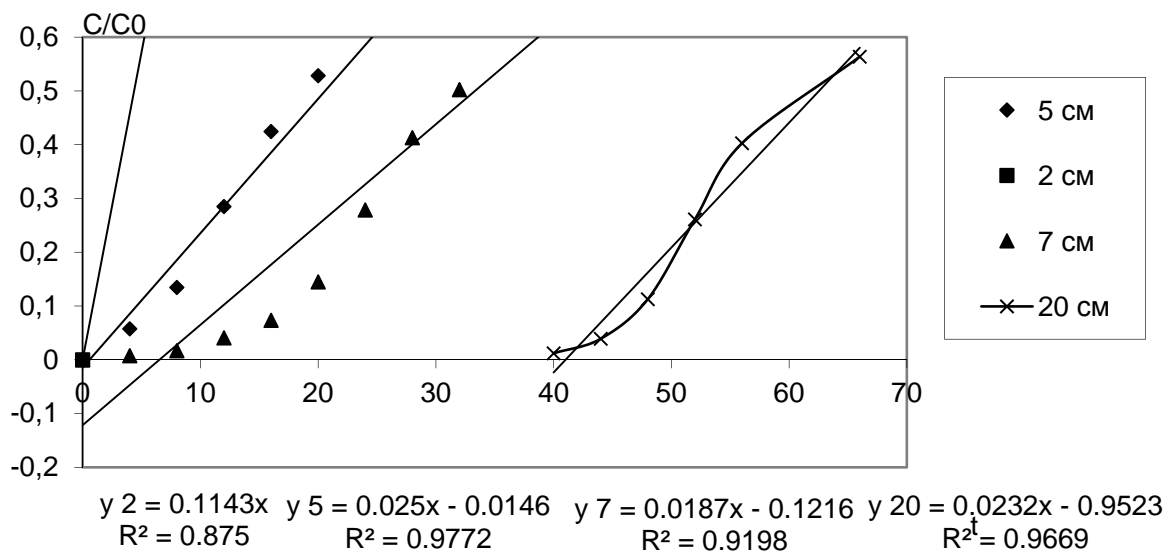R² = 0.875   R² = 0.9772   R² = 0.9198   R²t = 0.9669

Fig. 1. Initial curves of albumin adsorption by activated carbon in a column-type apparatus

The data of Fig. 1 show the results of experiments on albumin adsorption, where the solution concentration (C, kg/m³), relative concentration (C/C₀), pH, solution volume (V, ml), time interval (min), absorbed mass were measured of albumin in each interval and the total mass of absorbed albumin. The data are presented for different experimental conditions, in particular, with different thicknesses of the adsorbent layer (5 cm, 7 cm, 20 cm) and different pH values. The data demonstrate the dynamics of albumin adsorption, which includes a decrease in the concentration of albumin in the solution and an increase in the mass of absorbed albumin until the adsorbent is saturated (Sabadash & Gumnitsky, 2019). The adsorption process depends on several parameters, including the adsorbent's thickness and the medium's pH, which can affect the absorption efficiency. Statistical evaluation of the data showed a satisfactory convergence of experimental and theoretical data. The coefficient of determination fluctuated within $R^2 = 0.875 \dots 0.977\ 2$, which indicates a satisfactory convergence of experimental and theoretical data.

The model made it possible to estimate the change in the albumin concentration in the solution during its passage through the adsorbent. It was found that a decrease in the concentration of albumin in the solution is accompanied by an increase in the mass of absorbed albumin, which indicates the efficiency of adsorption.

As can be seen from the experimental data, the adsorbent layer's thickness significantly affects the process's efficiency. Increasing the thickness of the layer promotes more efficient extraction of albumin from the solution, but this can lead to an increase in mass transfer resistance, which affects the overall performance of the process.

The results showed that the adsorption process has a phase character: in the initial stages, there is a rapid decrease in the albumin concentration, followed by a slowing down phase, where the system approaches the saturation state. This indicates the limitation of available adsorption centres on the surface of the adsorbent. One of the limitations of this model is the assumption of a uniform distribution of albumin concentration in the column, which only sometimes correspond to natural conditions, especially with large adsorbent layer thicknesses. Further research can be aimed at studying non-stationary conditions and the influence of hydrodynamic factors on the adsorption process.

It is also necessary to pay attention to the influence of temperature, ionic strength of the solution and other parameters that can affect the equilibrium and kinetic aspects of the adsorption process. Possible directions for further research include extending the model to account for these factors, as well as experimentally validating and refining the mathematical model.

4. Conclusions

The paper presents the results of studying the adsorption process of high-molecular compounds in a column-type apparatus based on the developed mathematical model. The conducted analysis allows us to conclude that the proposed model adequately describes the system's behaviour, considering the influence of critical parameters, such as diffusion and mass transfer coefficients, adsorbate concentration, and hydrodynamic conditions. The obtained data can be used to optimize the process and increase the efficiency and quality of liquid purification from high molecular weight compounds. The obtained results have important practical significance for designing and optimising column-type devices used for the purification and concentration of high-molecular compounds. This can be useful not only for the environmental industry but also in the biotechnological and pharmaceutical industries, where it is necessary to separate proteins and other high molecular weight compounds efficiently. The presented mathematical model of the adsorption of high molecular weight compounds in a column-type apparatus provides the possibility of optimizing the adsorption process. Despite some limitations, the model accurately predicts system behaviour and is the basis for further research. Future research will improve and adapt the model for a broader range of substances and conditions.

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