

Research Article

Simulation of the Industrial Oil Adsorption Purification Process for Automation Tasks

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Objective reasons associated with an increase of transport quantity and the volume of industrial goods and energy service production lead to the accumulation of used oils and greases. These substances can be reused in the case of their purification. Such resource saving is facilitated by the development of technologies, in particular, adsorption purification and efficient systems for their automation. The article carried out a detailed analysis of the continuous adsorption purification technological system and indicated its significant differences from the point of view of control systems with special preparation of raw materials. The conclusion about the presence of nonstationarity and stochasticity sources of continuous adsorption as a control object is substantiated, which made it possible to impose requirements on the properties of the control model. The existing methods of adsorption mathematical description are analyzed within their use for continuous control. Structures of dynamic models based on a combination of analytical and experimental methods are proposed. Analytical models are based on mass balances of substances, known forms of approximations for describing equilibrium conditions and adsorption kinetics are studied, and experimental and statistical modeling is carried out to determine possible structures of the connection between technological variables during control. A structural-parametric scheme of the model and a scheme for its adaptation in control systems have been formed. An example of an adsorption simulation model formation using *MATLAB+Simulink* is shown. The results can be used to develop software for control systems of continuous adsorption and to test control algorithms.

1. Introduction

Waste industrial oils and greases (OG) are substances that cannot decompose under normal environmental conditions and are hazardous environmental pollutants. At the same time, they contain a significant number of substances that can be used in various spheres of human life. The creation and operation of purification facilities for the waste oil and grease (WOG) recovery is now a perspective direction for the industrial development, which is aimed at improving the Earth's ecosystem state through resource conservation and biosphere protection. Continuous adsorption technology of WOG is the most effective way of their purification. The creation of software for control systems of such indus-

tries is an urgent task due to the complexity of processes, raw material instability, and poor information support.

The efficiency of automatic control systems (ACS) largely depends on the type and parameters of the control algorithm used. Modern technical means of automation (TMA) make it possible to implement various control algorithms, so there is a need for preliminary testing of competing algorithms on mathematical models of the technological control object (TCO).

Equally important is the task of maintaining parameters of the ACS controllers in accordance with the adsorption process properties as the control object. This task is related to the necessity to adapt the software of control systems.

Justification for the choice and adaptation of ACS algorithms to a greater extent relate to complex technological objects of large capacity, especially those that are at the implementation stage [1]. Such objects can be considered continuous adsorbers designed for WOG recovery in large volumes.

The above problems are primarily related to the properties of the TCO mathematical models. For use in control systems, they must satisfy conditions for the reproduction accuracy of the objects properties and be convenient for adaptation. To study various control algorithms, models should be able to simulate the dynamic properties of input material flows and external and internal disturbances.

These conditions require a reasonable choice of the type for the adsorption process mathematical model and the mathematical processor for its creation. Important tasks are the formation of criteria for selecting models and the formation of plans for their testing.

Given the processes' nonstationarity occurring in industrial adsorbers during the industrial oil's continuous purification, as well as the significant difficulties in organizing and conducting experiments in production conditions, the importance and relevance of modeling these TCOs should be noted.

The aim of the study is to create a model of the adsorption process, designed both to compare alternative control algorithms for industrial adsorbers and for use in control systems for these adsorbers. The objectives of the study are as follows:

- (i) Determine a modeling method that is convenient for correcting the model based on information from a real adsorber
- (ii) To carry out structural identification of the adsorption model based on experimental studies
- (iii) Create a structural-parametric scheme of the model
- (iv) Implement the model using *MATLAB+Simulink*

2. Literature Review

A lot of attention is devoted to the modeling of technological processes. Models are performed in different ways, depending on the TCO complexity, the information support of the automation system, and the purpose of the model. In [2, 3], mathematical expressions for modeling adsorption processes as nanosystems and analysis of these expressions from the point of view of practical application are presented. Methods based on the heat and mass transfer theory of the adsorption process are given in [4–11]. These works show that the use of analytical models is possible only under assumptions about the properties of substances and simplifications regarding the behaviour of these substances in apparatuses. An example of modeling for a one-component adsorbate (water vapor) is given in [12]. The authors of this work investigated mass transfer in a fixed layer of a solid phase using silica gel. In [5], mathematical dependences are given for describing

single-component adsorption of copper on different adsorbents. In [6], various aspects of the analytical modeling of adsorption processes are considered in search of the model that adequately describes the experimental data.

Consideration of multicomponent mixtures is found in a few works [13–15], which use complex analytical dependences to describe heat and mass transfer in heterogeneous systems. The work [16] describes adsorption processes of several components from the standpoint of determining the highest selectivity degree of the adsorbent, i.e., what quantity of the studied mixture components will be most adsorbed on the proposed adsorbents. The studies carried out in [17] generally describe multicomponent adsorption, but from the position of a mixture of components that must be removed, without taking into account adsorption properties of each separately. In [18], the authors developed the mathematical model of the dynamic mode of adsorption of inclusions on the interface. Such a model takes into account the physicochemical properties of media in contact with each other but contains many assumptions that can introduce significant errors into the model, depending on the properties of the adsorbent and the raw material.

There are works devoted to computer simulation of adsorption processes in specialized mathematical packages. In particular, in [19], the authors use the *MATLAB+Simulink* environment to simulate the dynamic mode of the catalytic cracking process, and in [20], the creation of a 3D model of the adsorption process in the *ANSYS CFX* software product is described. In [21], the mathematical model was developed to solve the problem of pollutant transport in groundwater based on the stationary incompressible two-dimensional Navier-Stokes equation for the fluid flow and the two-dimensional convection-diffusion equation for the pollutant (one type of pollutant was used). This model was implemented using *FreeFEM* and *MATLAB* environments. Since the adsorption process of multicomponent mixtures is difficult for its behaviour description and adequate reproduction in models that will be used in control systems, it is necessary to provide such a mathematical apparatus that will have the functionality to respond to changes in the properties of raw materials, adsorbent, and technological parameters (such as temperature and pressure directly inside the installation) of the process as a whole.

An analysis of the studies described above shows that the use of exclusively analytical methods is limited for the separation of multicomponent mixtures with an unstable composition. The methods require knowledge of many variables that characterize the properties of material flows and the state of technological equipment, which is impossible in production conditions. Sophisticated computer models are effective in scientific research to determine the form of relationships between adsorption process variables. Since control tasks require models that can be satisfied only by the capabilities of the information subsystem of the production automation system, it is useful to work with the above laboratory studies and computer modeling ideas.

3. Materials and Methods

We consider a general technological system (TS) for the WOG property recovery and outline the boundaries of the part that should be taken into account when modeling the adsorption process.

Contaminated raw materials (WOG) and a circulating adsorbent enter the input of the common TS (in one part of the TS, it becomes contaminated, adsorbing substances from the raw materials, and in the other part, it is regenerated). The general technology of WOG recovery involves several stages: preparation of WOG (raw materials), adsorption, and adsorbent recovery (desorption, drying, regeneration, and cooling). Figure 1 shows a structural scheme of part of the TS that contains the adsorber and others most closely associated with its apparatuses.

Preparation of raw materials: the diaphragm mixer *DM* receives contaminated raw material *RM* and solvent *SOL1* used to reduce the viscosity of *RM*. The resulting mixture *MS1* is cooled in the refrigerator *R1* to the adsorption temperature with water *W1* fed through the coils.

Adsorption: the adsorbent *A* (from the intermediate hopper *B1*) and the cooled mixture of raw material and solvent *MS2* are simultaneously fed countercurrently into the adsorber *Ad*. The adsorbent saturated with aromatic hydrocarbons and resins leaves the adsorber in the suspension form. Solvent *SOL2* is fed into the adsorber water seal to prevent the removal of raw materials with the suspension. The purified product—the raffinate solution *RS1*—exits the top of the adsorber.

Contaminated adsorbent *CA1* from the adsorber enters the technological recovery units. It can be considered regenerated (adsorbent *A1*) after the stages of contaminant desorption, drying and burning resins out of it.

Cooling of the regenerated adsorbent: the regenerated hot adsorbent stream *A1* is cooled with water in a fluidized bed refrigerator *R2* to a predetermined adsorption temperature. The cooled adsorbent *A2* enters the hopper *B1* and from it to the adsorber *Ad* (adsorbent *A*). Desorption, drying, and burning are not considered, since the regenerated adsorbent properties are finally determined after it has been cooled in the refrigerator *R2*.

Each of the material flows shown in Figure 1 has several properties that form the properties of the reduced raw material *RS1* at the adsorber outlet. Determining the model type requires the analysis of these properties as information flows. Figure 2 shows a technological-parametric scheme for the contaminated oil and grease recovery.

The designation of material flow properties that are indicated in Figure 2 is revealed.

- (i) Raw material (*RM*): F_{rm} : consumption; Θ_{rm} : temperature; V_{rm} : viscosity, C_{ah0} : aromatic hydrocarbon (AH) content; C_{s0} : sulphur content; C_{r0} : resin content; C_{c0} : content of other impurities
- (ii) Solvent (*SOL1*): F_{sol1} : consumption; Θ_{sol1} : temperature.
- (iii) Mixture of raw material and solvent (*MS1*): F_{ms1} : consumption; Θ_{ms1} : temperature; V_{ms1} : viscosity

- (iv) Water (*W1*) at the refrigerator *R1* inlet: F_{w1} : consumption; Θ_{w1} : temperature
- (v) Water (*W2*) at the refrigerator *R1* outlet: F_{w2} : consumption; Θ_{w2} : temperature
- (vi) Solvent for the water seal (*SOL2*): F_{sol2} : consumption; Θ_{sol2} : temperature
- (vii) Cooled mixture of raw material and solvent (*MS2*): F_{ms} : consumption; Θ_{ms} : temperature; V_{ms} : viscosity; $C_{ms,ah}$: aromatic hydrocarbon content; $C_{ms,s}$: sulphur content; $C_{ms,r}$: resin content; $C_{ms,c}$: content of other impurities
- (viii) Regenerated adsorbent hot (*A1*): F_{a1} : consumption; Θ_{a1} : temperature; M_{a1} : moisture content; C_{ah1} : aromatic hydrocarbon content; C_{s1} : sulphur content; C_{r1} : resin content
- (ix) Inlet water flows (*W3*, *W5*, *W7*, *W9*) in the refrigerator *R2*: F_{w3} , F_{w5} , F_{w7} , F_{w9} : consumption; Θ_{w3} , Θ_{w5} , Θ_{w7} , Θ_{w9} : temperature
- (x) Outlet water flows (*W2*, *W4*, *W6*, *W8*, *W10*) from the refrigerator *R2*: F_{w4} , F_{w6} , F_{w8} , F_{w10} : consumption; Θ_{w4} , Θ_{w6} , Θ_{w8} , Θ_{w10} : temperature

For the simulation model, we will take into account only the flows *W3*, *W4*.

- (i) Regenerated adsorbent cooled (*A2*): F_{a2} : consumption; Θ_{a2} : temperature; M_{a2} : moisture content, C_{ah2} : aromatic hydrocarbon content; C_{s2} : sulphur content; C_{r2} : resin content
- (ii) Adsorbent (*A*): F_a : consumption; Θ_a : temperature; M_a : moisture content, $C_{a,ah}$: aromatic hydrocarbon content; $C_{a,s}$: sulphur content; $C_{a,r}$: resin content
- (iii) Adsorber (*Ad*): P : pressure in the apparatus (or the pressure difference between the upper and lower parts of the adsorption column); Θ : adsorption temperature; L : solvent level (*SOL2*) in the water seal
- (iv) Raffinate solution I (*RS1*): F_{rs} : consumption; Θ_{rs} : temperature; V_{rs} : viscosity; $C_{rs,ah}$: aromatic hydrocarbon content; $C_{rs,s}$: sulphur content; $C_{rs,r}$: resin content; $C_{rs,c}$: content of other impurities
- (v) Contaminated adsorbent (*CA1*): F_{ca} : consumption; Θ_{ca} : temperature; M_{ca} : moisture content, $C_{ca,ah}$: aromatic hydrocarbon content; $C_{ca,s}$: sulphur content; $C_{ca,r}$: resin content

The control action [1] ACS adsorber can be the consumption of the adsorbent (F_a), the mixture of raw material and solvent (F_{ms2}), and water (F_{w1} , F_{w3}) at the stages of cooling the input streams and the solvent (F_{sol1}). The most important possible controlled variables are the concentrations of contaminants in the product ($C_{rs,ah}$, $C_{rs,s}$, $C_{rs,r}$), pressure (P) and adsorption temperature (Θ), and level (L); controlled values are the concentrations of aromatic

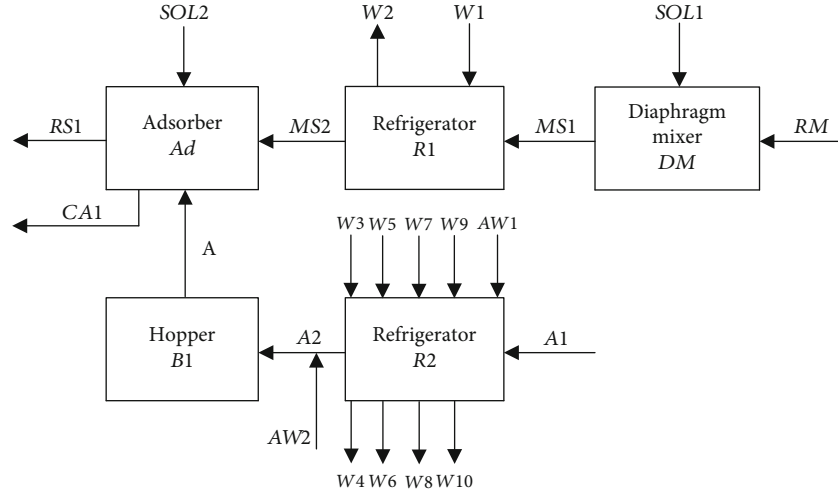


FIGURE 1: Structural scheme of the TS for the preparation processes of raw material and adsorption and cooling of the adsorbent. RM: contaminated raw material; SOL1: solvent at the diaphragm mixer inlet; MS1: mixture of raw material and solvent; W1, W2: water at the refrigerator R1 inlet and outlet, respectively; SOL2: solvent for the water seal; MS2: cooled mixture of raw material and solvent; A1, A2: regenerated adsorbent hot and cooled, respectively; W3, W5, W7, W9: water at the coil inlet of the refrigerator R2; W4, W6, W8, W10: water at the coil outlet of the refrigerator R2; AW1: air to create a fluidized bed in the refrigerator R2; AW2: air for the pneumatic transport system; A: the adsorbent at the adsorber inlet (hereinafter, referred to as the adsorbent); RS1: purified product; CA1: contaminated adsorbent.

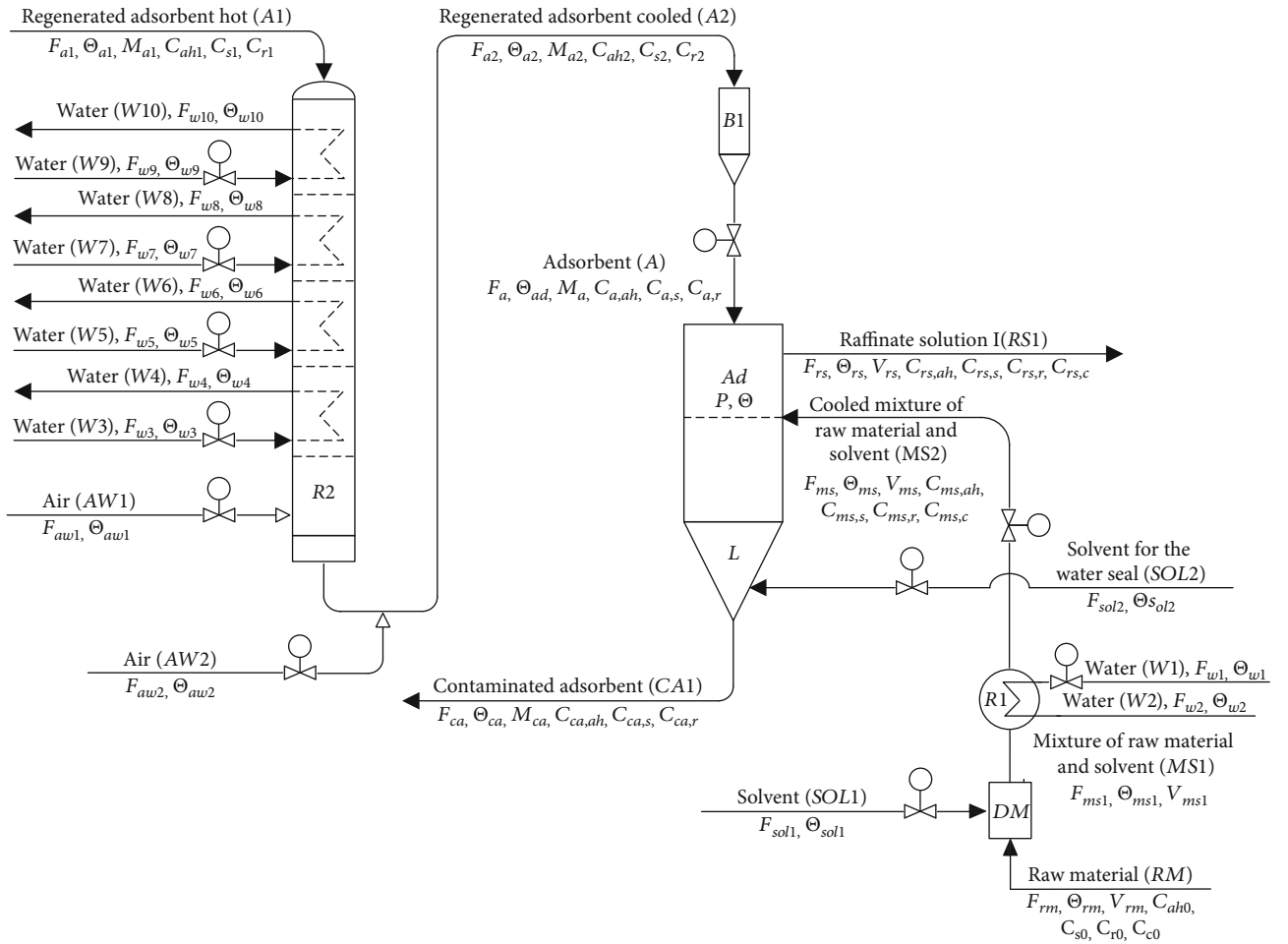


FIGURE 2: Technological and parametric scheme of the processes of raw material preparation, adsorption, and adsorbent recovery with information flow designation.

hydrocarbons, sulphur, and resins in the contaminated adsorbent ($C_{ca,ah}$, $C_{ca,s}$, $C_{ca,r}$). Disturbances are the concentrations of pollutants, i. e. AH, sulphur, resins, in the raw material ($C_{ms,ah}$, $C_{ms,s}$, $C_{ms,r}$) and adsorbent ($C_{a,ah}$, $C_{a,s}$, $C_{a,r}$), temperatures of the raw material (Θ_{ms}) and adsorbent (Θ_a). The final structure of the automation system depends on many factors.

Figure 2 shows that the preparation processes and adsorption itself are complex multiparameter control objects with a very small number of control actions. Therefore, the model concept is as follows: when applying the same control actions and basic disturbances to the model and the adsorber, the model must be adequate to the object and satisfy the given accuracy indicator. There are no requirements for the application of specific mathematical formalisms to parts of the model or the model as a whole.

First, explore the possibilities of analytical modeling for the processes occurring in the adsorber with a continuous operating principle. Consider the general patterns of transfer of the energy amount, mass, and momentum between different parts of the technological system [22]:

$$\begin{aligned}\frac{\partial \Theta}{\partial t} &= -\left(w_x \frac{\partial \Theta}{\partial x} + w_y \frac{\partial \Theta}{\partial y} + w_z \frac{\partial \Theta}{\partial z}\right) + k_\Theta \nabla^2 \Theta + \frac{q_r}{\rho c_p}, \\ \frac{\partial c_i}{\partial t} &= r_{iv} - \left(w_x \frac{\partial c_i}{\partial x} + w_y \frac{\partial c_i}{\partial y} + w_z \frac{\partial c_i}{\partial z}\right) + D_i \nabla^2 c_i, \\ \frac{\partial w_x}{\partial t} &= -\left(w_x \frac{\partial w_x}{\partial x} + w_y \frac{\partial w_x}{\partial y} + w_z \frac{\partial w_x}{\partial z}\right) + \nu \nabla^2 w_x + \left(X - \frac{1}{\rho} \frac{\partial p}{\partial x}\right),\end{aligned}\quad (1)$$

where Θ is the temperature; t is the time; x , y , and z are the designation of Cartesian coordinates; w_j is the fluid velocity along the j -th coordinate; $k_\Theta \nabla^2 \Theta + q_r / \rho c_p$ is the Fourier equation describing the pattern of heat propagation in the system due to heat conduction; k_Θ is the thermal diffusivity; $\nabla^2 = \partial^2 / \partial x^2 + \partial^2 / \partial y^2 + \partial^2 / \partial z^2$, e.g., $\nabla^2 \Theta - \partial^2 \Theta_i / \partial x^2 + \partial^2 \Theta_i / \partial y^2 + \partial^2 \Theta_i / \partial z^2$; q_r , ρ , and c_p are power of the heat source per unit volume, density, and heat capacity, respectively; i , c_i , r_{iv} , and D_i are the component number, concentration, rate of formation per unit volume, and diffusion coefficient of the i -th component, respectively; ν is the volume; p is the pressure; and X is the projection of the mass force on the x coordinate.

Multicomponent adsorption in the case of WOG (n -component) will require consideration of a system of $n-1$ equations of System 1.

For the mathematical formalization of adsorption, equations relating to the concept of phase equilibrium and adsorption kinetics are also used. Equilibrium conditions are equations whose factors are the temperature Θ and the concentration c of the substance adsorbed by the adsorbent in the balanced moving phase, and the initial variable is the adsorption capacity a (the amount of substance adsorbed by a unit mass or volume of the adsorbent):

$$a = f(c, \Theta_i). \quad (2)$$

The adsorption theory considers the external (to the adsorbent surface) and internal (inside adsorbent particles) transfer of an adsorbed substance. The rate of this process characterizes the kinetics of external adsorption. It is given using the following expression:

$$\frac{da}{dt} = \beta(c - c^*), \quad (3)$$

where β is the coefficient of external mass transfer per unit volume of the adsorbent and c and c^* are the adsorbed substance content in the volume of the mixture and on the surface, respectively.

Adsorption inside particles is the most complex process in terms of modeling. The equations that can be applied to describe it should contain the parameters of the geometry of the adsorbent internal chambers and the physicochemical properties of the adsorbent, adsorbate, and adsorptive as nanoparticles. For multicomponent mixtures, theoretical equations with such exponents are highly general [23–25].

The analysis of the above mathematical expressions shows that even models of the type as in Equations (1)–(3) require significant amount of information and can be adequate to TCO only if all the assumptions regarding the physicochemical properties and methods of mutual movement of the interacting substances are performed. Existing models of molecular adsorption can only be considered as possible ways of mathematical description of the substance's behaviour under certain conditions. Therefore, the use of complex analytical models also requires adaptation to production conditions when changing the properties of the WOG. However, due to the bulkiness, multiparametric nature, and lack of the necessary TMA, the process of model adaptation will be very difficult.

For these reasons, the model is based on a system of simplified dynamic equations for pollutants [26, 27]:

$$\begin{aligned}F_m y_{ah0} - F_m y_{ah1} - SK_{m,ah} \frac{(y_{ah0} - y_m(x_{ah1})) + (y_{ah1} - y_m(x_{ah0}))}{2} \\ = V_m \rho_m \frac{d}{dt} \left(\frac{y_{ah0} + y_{ah1}}{2} \right) + V_a \rho_a \frac{d}{dt} \left(\frac{x_{ah0} + x_{ah1}}{2} \right), \\ F_a x_{ah0} - F_a x_{ah1} + SK_{a,ah} \frac{(x_a(y_{ah1}) - x_{ah0}) + (x_a(y_{ah0}) - x_{ah1})}{2} \\ = V_a \rho_a \frac{d}{dt} \left(\frac{x_{ah0} + x_{ah1}}{2} \right) + V_m \rho_m \frac{d}{dt} \left(\frac{y_{ah0} + y_{ah1}}{2} \right), \\ F_m y_{s0} - F_m y_{s1} - SK_{m,s} \frac{(y_{s0} - y_m(x_{s1})) + (y_{s1} - y_m(x_{s0}))}{2} \\ = V_m \rho_m \frac{d}{dt} \left(\frac{y_{s0} + y_{s1}}{2} \right) + V_a \rho_a \frac{d}{dt} \left(\frac{x_{s0} + x_{s1}}{2} \right), \\ F_a x_{s0} - F_a x_{s1} + SK_{a,s} \frac{(x_a(y_{s1}) - x_{s0}) + (x_a(y_{s0}) - x_{s1})}{2} \\ = V_a \rho_a \frac{d}{dt} \left(\frac{x_{s0} + x_{s1}}{2} \right) + V_m \rho_m \frac{d}{dt} \left(\frac{y_{s0} + y_{s1}}{2} \right),\end{aligned}$$

$$\begin{aligned}
& F_m y_{r0} - F_m y_{r1} - SK_{m,r} \frac{(y_{r0} - y_m(x_{r1})) + (y_{r1} - y_m(x_{r0}))}{2} \\
& = V_m \rho_m \frac{d}{dt} \left(\frac{y_{r0} + y_{r1}}{2} \right) + V_a \rho_a \frac{d}{dt} \left(\frac{x_{r0} + x_{r1}}{2} \right), \\
& F_a x_{r0} - F_a x_{r1} + SK_{a,r} \frac{(x_a(y_{r1}) - x_{r0}) + (x_a(y_{r0}) - x_{r1})}{2} \\
& = V_a \rho_a \frac{d}{dt} \left(\frac{x_{r0} + x_{r1}}{2} \right) + V_m \rho_m \frac{d}{dt} \left(\frac{y_{r0} + y_{r1}}{2} \right), \quad (4)
\end{aligned}$$

where F_m and F_a are the flow rates of the mixture of raw material and solvent and adsorbent, respectively; y_{i0} and x_{i0} are the input concentrations in the mixture of raw material and solvent and adsorbent, respectively; y_{i1} and x_{i1} are the output concentrations in the mixture of raw material and solvent and adsorbent, respectively; $i = ah, s, r$; ah is the aromatic hydrocarbons; s is the sulphur; r is the resins; $y_m(x_{i0})$, $y_m(x_{i1})$, $x_a(y_{i0})$, and $x_a(y_{i1})$ are the equilibrium concentrations of aromatic hydrocarbons, sulphur, and resins in the mixture of raw material and solvent and adsorbent, respectively; $K_{m,i}$ and $K_{a,i}$ are the mass transfer coefficients expressed in terms of the component molar particles in the mixture of raw materials and solvent and adsorbent, respectively; V_m and V_a are the column volume filled with the mixture of raw material and solvent and adsorbent, respectively; and ρ_m and ρ_a are the densities of the mixture of raw material and solvent and adsorbent, respectively.

The simplification of this system consists, in particular, in the following assumptions: the adsorber is a control object with lumped parameters; mass transfer coefficients are constant values; the motive force for the transfer of pollutants from adsorbate to adsorbent is directly proportional to the average value of the differences in the equilibrium concentrations of these substances in adsorbate and adsorbent; mass accumulation is proportional to the average concentrations: $(x_{ah0} + x_{ah1})/2$, $(y_{ah0} + y_{ah1})/2$, $(x_{s0} + x_{s1})/2$, $(y_s + y_{s1})/2$, $(x_{r0} + x_{r1})/2$, and $(y_{r0} + y_{r1})/2$.

These simplifications will reduce the number of unknown parameters and, thus, the duration of the model adaptation in production conditions.

The mass of each of the pollutants that have passed from the adsorbate to the adsorbent per unit time, in Equation (4), is represented by expressions that are identical in structure. For example, an expression for aromatic hydrocarbons

$$SK_{m,ah} \frac{(y_{ah0} - y_m(x_{ah1})) + (y_{ah1} - y_m(x_{ah0}))}{2}. \quad (5)$$

In the case of WOG, the input properties of the raw material are usually unstable, so even Equation (5) cannot provide the necessary information in real time with the required accuracy.

A significant list of factors and disturbances even in such a simplified model complicates its practical application in ACS. Based on this conclusion and recognizing the convenience of the current model adaptation as a very important requirement, based on System 4 and taking into account the transport delay in the TCO, it is proposed to present

the models of the main control channels in the form

$$W_n(s, t) = \frac{k_n(t)}{T_{2n}(t)s^2 + T_{1n}(t)s + 1} e^{-\tau_n s}, \quad (6)$$

where n is the control channel number, $n = 1, 2, 3$; (ah, S, r); T_{1n} and T_{2n} are the adsorber time constants; s is the Laplace transform symbol; t is the time; k_n is the transmission coefficient; and τ_n is the transport delay time of the control object.

Since the above simplifications distort actual relationships between the variables of heat and mass transfer processes in the adsorber, it is proposed to represent each of the parameters of Equation (6) by the function φ_{wk} , which will be adjusted when comparing the values of the selected variable at the outputs of the adsorber and its model.

To select the form of the functions φ_{wk} nonlinearity consider experimentally confirmed mathematical expressions that characterize various aspects of adsorption processes [1, 22], in particular, formulas for calculating the index a . Since there are no recommendations for calculating a , for the adsorption WGO purification, several formulas are used that can be taken as a basis. Among them are the formulas of Langmuir, Freundlich, Redlich-Peterson, BET (the theory of polymolecular (multilayer) adsorption by Brunauer, Emmett, and Teller), Dubinin-Radushkevich, Boyd, and Lagergren [22, 27, 28]. Each of the formulas is based on a certain set of various assumptions related both to the properties of material flows and to the process conditions.

So, in the Dubinin-Radushkevich formula [22], the adsorption index a depends nonlinearly on the properties of material flows and regime parameters as follows:

$$a = \frac{a_m BP}{1 + BP}, \quad (7)$$

where a_m is the capacity of a monolayer of adsorbed substance in adsorbent, P is the partial pressure, and B is the parameter, which is proposed to be calculated by the formula

$$B = \frac{\alpha_m \exp(q_m/kT)}{k_0 \sqrt{(2\pi mkT)}}, \quad (8)$$

where m is the mass of the molecule, k is Boltzmann's constant, T is the absolute temperature, k_0 is the entropy coefficient, and q_m is the adsorption heat of one molecule.

The Freundlich formula (for energetically inhomogeneous surfaces) has the following form:

$$\ln a = \ln K_F + \frac{1}{n} \ln C, \quad (9)$$

where K_F and n are the Freundlich adsorption constants characterizing the adsorption capacity and the interaction intensity of substances "adsorbent-adsorbate," respectively.

The structure of these expressions has common features—they are nonlinear with exponential, logarithmic, and power components of the adsorbate concentration,

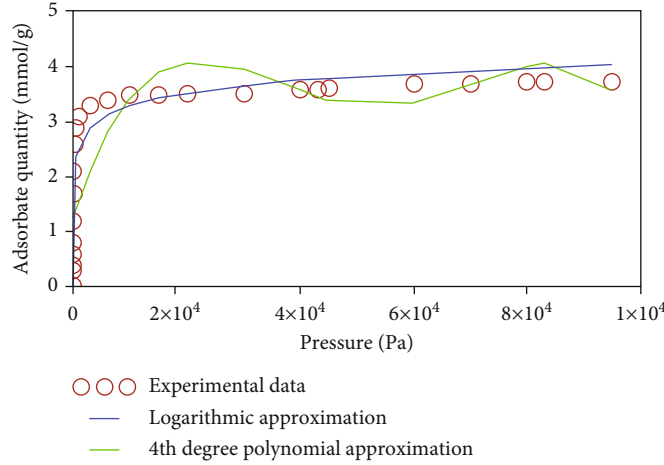


FIGURE 3: Approximation of the dependence of the adsorbed substance quantity on pressure during the adsorption of tetrafluoroethane on NaY zeolite.

temperature, and pressure in the adsorber. Based on this, it is necessary to substantiate which structures of the functions φ_{wk} should be introduced into the model designed to control and study algorithms.

The definition of the form φ_{wk} was made using the results of studies given in [6, 14, 15].

Thus, the adsorption of tetrafluoroethane isomers on NaY zeolite was studied in [6]. Figure 3 shows experimental data displaying the dependence of the adsorbed substance quantity on pressure and its logarithmic and polynomial approximations.

The work [14] considers the separation of Krypton (Kr) from Xenon (Xe) on different adsorbents. Figure 4 shows the dependence of Kr quantity on relative pressure using FAU LSX K—faujasite zeolite with Si/Al = 1—and, accordingly, its approximation by exponential and quadratic parabolas.

The authors in [15] considered the problem of used fluorine-containing compound utilization by extracting from them and separating fluorocarbons by adsorption methods. Figure 5 shows the dependence of adsorbed difluoromethane R32 (adsorbate) quantity on pressure during adsorption on 5A zeolite with modified pore sizes and the approximation of the obtained dependence of adsorbate amount on pressure by an exponent, quadratic, and cubic polynomials.

Figures 6 and 7 show dependencies of fluorocarbon adsorbate quantity on temperature at various pressures [15]: dependence of R32, R22 (chlorodifluoromethane), and R125 (pentafluoroethane) quantity on temperature during adsorption on zeolite 5A and a pressure of 1.5 bar (Figure 6); dependence of R134a (1,1,1,2-tetrafluoroethane), R125, and R143a (1,1,1-trifluoroethane) quantity on temperature during adsorption on zeolite 13X and a pressure of 1 bar (Figure 7).

Figure 8 shows results of fluorocarbon components R134a, R125, and R143a adsorption on zeolite 13X, obtained from the two-level Langmuir-Freundlich equation at temperatures of 293 K, 313 K, and 333 K.

Figure 9 shows the dependencies of the fluorocarbon adsorbate R32, R22, and R125 quantity on temperature at

a pressure of 1 bar on HKUST-1 zeolite, which has a three-dimensional porous structure with three types of cages formed by the connection of dimeric copper clusters coordinated with four carboxylate groups of trimesic acid [15].

Based on the studies on structural identification, which are illustrated in Figures 3–9, for the control channel transmission coefficient, it is proposed to choose two competing structures $\varphi_{n,kn}$ —exponential and polynomial—of the following types:

$$\begin{aligned} \varphi_{n,kn}(t) &= a_0 + a_1 e^{a_2 \Theta(t)} + a_3 e^{a_4 P(t)}, \\ \varphi_{n,kn}(t) &= \varphi_{n,kn,\Theta}(\Theta(t)) + \varphi_{n,kn,P}(P(t)) + \varphi_{n,kn,c}(c(t)), \end{aligned} \quad (10)$$

where a_i is the exponential model parameters; $\varphi_{n,kn,\Theta}(\Theta(t))$, $\varphi_{n,kn,P}(P(t))$, and $\varphi_{n,kn,c}(c(t))$ are polynomials of the 2nd or 3rd degree for temperature, pressure, and concentration.

Correction of functions 10 and 11 when adapting the model should not be difficult.

Consider the main control channel $\ll F_a \rightarrow C_{rs,ah} \gg$. Its transfer function, for example, is as follows:

$$W_1(t) = W_{F_a \rightarrow C_{ah}}(t) = \frac{k_{11}(t)}{T_{21}(t)s^2 + T_{11}(t)s + 1} e^{-\tau_1 s}, \quad (11)$$

$$k_{11}(t) = a_{10} + a_{11} e^{a_{12} \Theta(t)} + a_{13} P(t)^{a_{14}}, \quad (12)$$

$$T_{11}(t) = b_{11}, \quad T_{21}(t) = b_{21}. \quad (13)$$

It is appropriate to choose the values T_{11} , T_{21} , and k_{11} , obtained from simplified material balance 4 or from the available experimental data as the starting values of the parameters for corrective functions.

Figure 10 shows a generalized structural scheme of the system for adsorption model adapting through several control channels and channels of disturbances.

It allows to create a model adaptation algorithm.

According to the algorithm, vectors of input signals \vec{U}

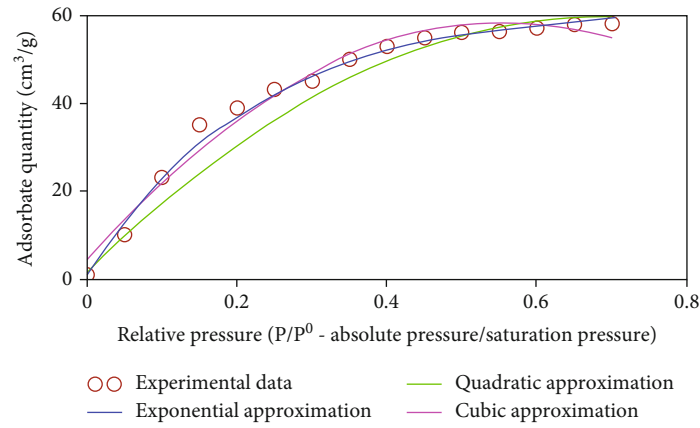


FIGURE 4: Approximation of the dependence of Kr quantity on relative pressure during the adsorption on FAU LSX K zeolite.

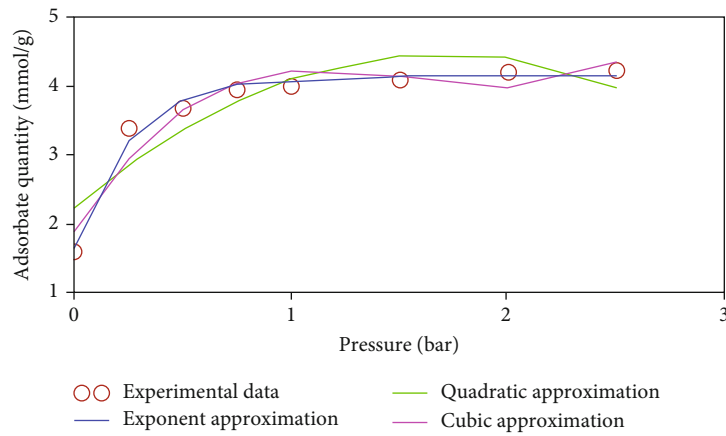


FIGURE 5: Approximation of the dependence of difluoromethane R32 quantity on pressure during adsorption on 5A zeolite.

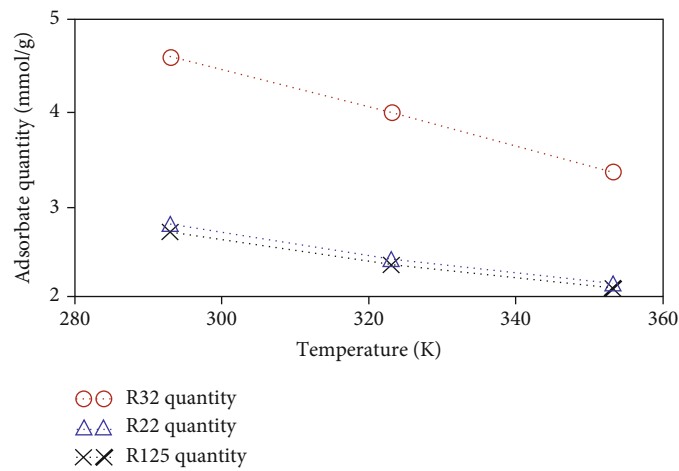


FIGURE 6: Dependence of the adsorbate quantity on temperature using zeolite 5A (pressure 1.5 bar).

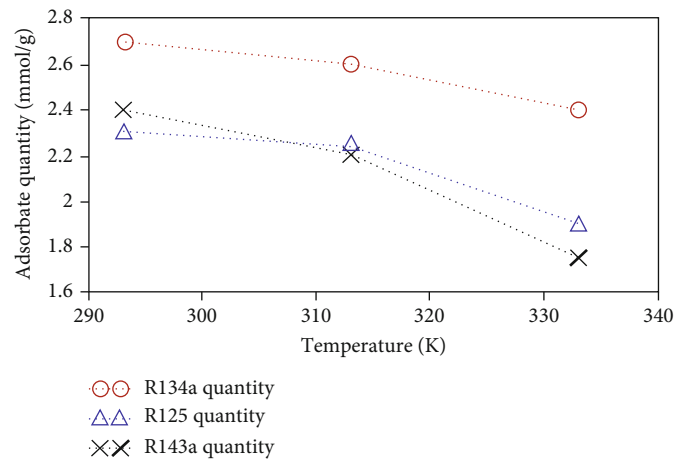


FIGURE 7: Dependence of the adsorbate quantity on temperature using zeolite 13X (pressure 1 bar).

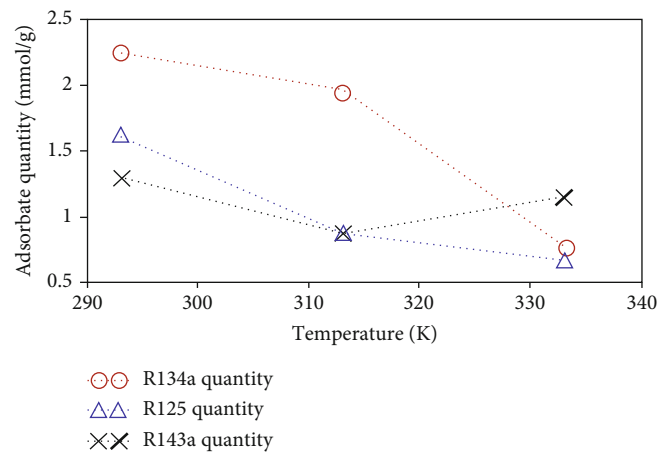


FIGURE 8: Dependence of the adsorbate quantity on temperature during adsorption on zeolite 13X.

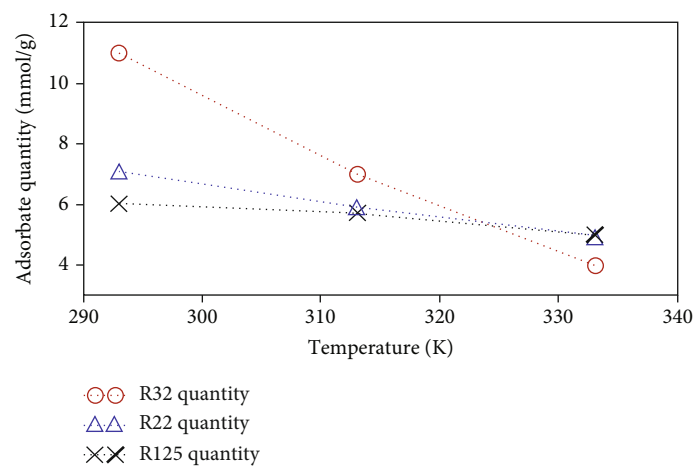


FIGURE 9: Dependence of the adsorbate quantity on temperature using zeolite HKUST-1 at a pressure of 1 bar.

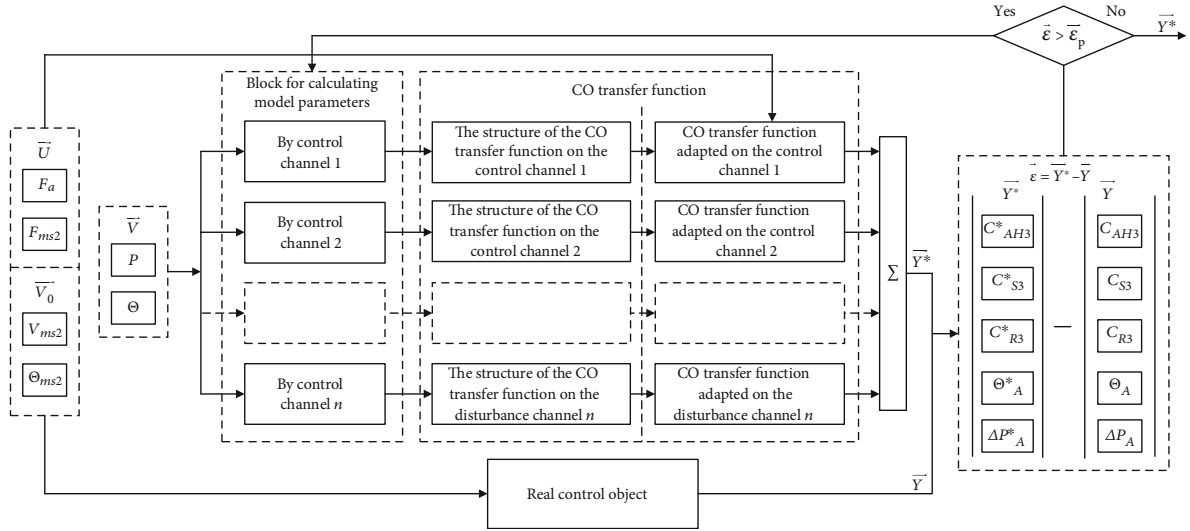


FIGURE 10: Structural scheme of the adsorber model adaptation system.

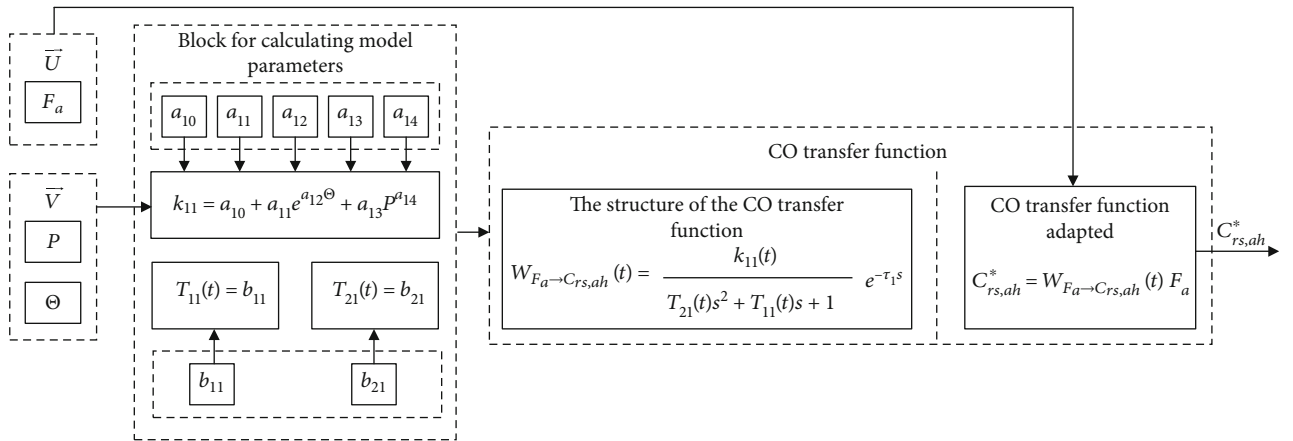


FIGURE 11: Structural-parametric scheme of the model.

(control actions) and \vec{V}_0 (mode parameters) are fed to the inputs of each control channel and the real TCO. The adaptive model implies the presence of an adaptation block to determine the transfer function coefficients of the model, the structure of which is known. The vector of mode parameters \vec{V} is fed to the input of the same block. The vector of the adaptive model \vec{Y}^* is compared with the vector of the output signals of the adsorber \vec{Y} . If the error value $\vec{\epsilon}$ is not acceptable, then in the adaptation block, the transfer function coefficients are tuned until the condition $\vec{\epsilon} \leq \vec{\epsilon}_p$.

Figure 11 shows an example of a structural-parametric scheme of the model with the proposed corrective functions for the control channel " $F_a \rightarrow C_{rs,ah}$ ".

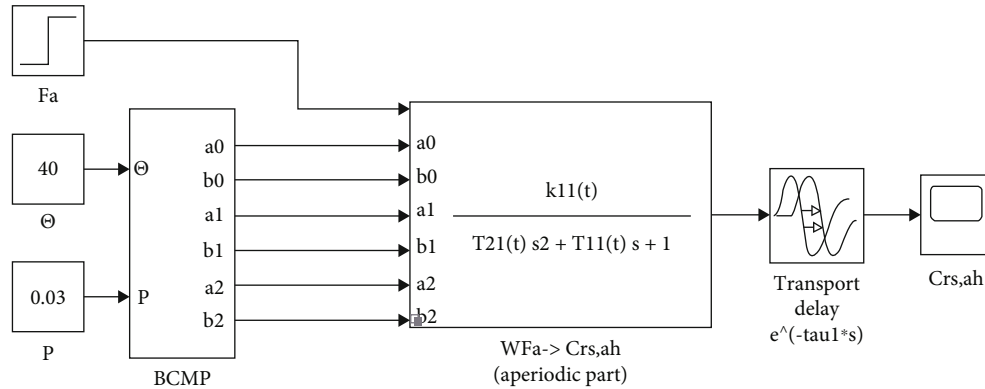
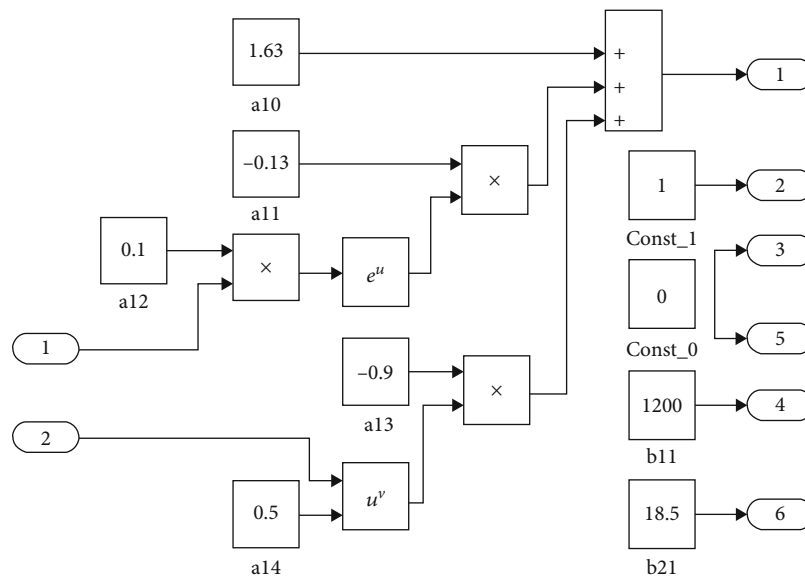
Figure 12 shows the structural scheme of the adsorption model by control channel " $F_a \rightarrow C_{rs,ah}$ " implemented by means of *MATLAB+Simulink*. The structure of the transfer function in Equation (12) is represented by two blocks: the

first has form of the aperiodic part $W F_a \rightarrow C_{rs,ah}$; the second is the transport delay $e^{-\tau_1 s}$, where τ_1 is the transport delay time, s. Control action F_a and the parameters calculated in the corresponding block for calculating model parameters (BCMP) are fed to the input of this function. Figure 13 shows the structure of the BCMP subsystem, which provides calculation of model parameters according to Equation (12).

Such a *Simulink* model can be used to compare adsorber control algorithms, supplementing it with disturbance blocks typical for production conditions.

4. Results and Discussion

An analysis of the technological system for purification (regeneration) of industrial (industrial and transport) oils and lubricating oils showed that it is characterized by two types of changes in the properties of substances (i.e., types of nonstationarity of processes in the FEC). The first of them

FIGURE 12: Structural *Simulink* model of adsorption by control channel " $F_a \rightarrow C_{rs,ah}$ ".FIGURE 13: Subsystem structure of the BCMP of *Simulink* adsorption model.

is systemic in nature, since it occurs every time when raw materials are received from another supplier. This type of nonstationarity can be more or less predictable (periodic) with one supplier of raw materials or unpredictable (random) due to its receipt from several suppliers.

The second type of change (nonstationarity) is purely random. It is related to the properties of the adsorbent (the impact of adsorbent regeneration processes) and raw materials (even from one supplier). The properties of both substances are random variables; we consider them as external perturbations. The indicators of the state of the internal elements of the design of the adsorber and other devices are considered internal perturbations [23].

The study of the chemical-technological system and the analysis of existing methods for modeling the adsorption process made it possible to determine the type of models and the algorithm for their use in computer systems for controlling the purification processes, namely, the PTO, taking into account the named nonstationarity.

Processing the results of open access studies made it possible to determine the structures of approximation equations

that can be applied in control systems for continuous adsorbers.

The obtained results allow software developers to use polynomial 1-3 orders and exponential models of control channels. Substantiation of the type of models designed to control the process of adsorption for PTO purification, based on theoretical provisions and experimental studies, can be considered a scientific novelty.

Taking into account the complexity of experimental work in production conditions and the inefficiency of research in laboratory conditions, such results are of practical importance.

5. Conclusions

As a result of the studies presented in the article, were obtained following:

- (i) It is substantiated that the adsorption process can be considered nonstationary due to the receipt of contaminated raw materials from different sources

and the lack of systems for stabilizing the properties of contaminated raw materials in each source

- (ii) It is determined that nonstationarity of processes has two causes and two types of nonstationarity of processes; this should be taken into account in process control algorithms
- (iii) The analysis of the structure and material flows of the technological purification system made it possible to determine the information flows
- (iv) It has been determined that the following should be taken into account when modeling: the adsorption purification process is a multiparameter TCO, the values of most of the process variables cannot be determined continuously, and under production conditions, they will not be measured even in a laboratory
- (v) Mathematical model of each control channel is proposed in the form of a second-order transfer function with a transfer coefficient that is a nonlinear function of temperature and pressure in the adsorber
- (vi) The study of data from open sources has identified the types of functions that can reflect the dependence of the transfer coefficients on the regime parameters; these include exponentials, polynomials of 2-3 orders, and logarithms
- (vii) A structural-parametric diagram of the model and a diagram of the system for adapting the adsorber model were created
- (viii) A simulation model of adsorption processes was created using *MATLAB+Simulink*, which corresponds to the concept of modeling

These results were obtained through the use of system analysis methods, in particular, the analysis of the technological system and its components and the analysis of methods for describing technological processes and adsorption, in particular, taking into account the requirements for the mathematical model of processes as control objects.

Such control-focused modeling research work related to continuous adsorption purification of used oils and greases based on the generalization of other adsorbents and adsorbates has not previously been reported in the literature. The achieved results can be considered as they have both scientific novelty and practical value. They can be used to create software for control systems for adsorption purification processes not only of WOG but also of other substances.

Proposed structures of the model allow solving the following problems related to the control of adsorption purification process, namely,

- (i) Determining the list of controlled variables necessary to control the adsorption process using the recommended model

- (ii) Determining the dynamic properties of controlled variables to synchronize their entry into the process model
- (iii) Choosing and investigating a computer method of the adsorption model adaptation to the conditions of this process in production conditions
- (iv) Creating and investigating the computer operation of an adaptive control system for the quality indicators of the contaminated industrial oil and grease purification both in the mode of unchanged raw materials and when the supplier of contaminated raw materials is changed
- (v) Coordinating functioning of separate control systems of the production system in order to save resources for implementation
- (vi) Developing plans for experimental studies of individual technological variables in production conditions to check the properties of controlled variables as random variables and to reconcile them when using models
- (vii) Developing plans for experimental studies of disturbance and control channels to check the model adequacy

The relevance of this project is in the fact that a process model was proposed to control the process of used oil and grease purification, the behaviour of which would correspond to the behaviour of this process in production conditions; it would be simple enough after the formula and in terms of the number and accuracy of the input variable measurement. The lack of mathematical models reduces effectiveness of control systems for complex technological apparatuses.

Also, the obtained model gives an opportunity to conduct simulation experiments to study various algorithms for controlling production processes, reducing time for designing and implementing control systems.

Data Availability

Data of this article is available upon request to the corresponding author.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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