

Nonlinear inverse problem for a model of ion-exchange filter: numerical recovery of parameters

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Abstract

In the article considers the problem of calculating the impurities concentration for an ion-exchange filter model. The model consists of the material balance equation, the equation of the kinetics of ion exchange for the nonequilibrium case and of the ion exchange isotherm. In the material balance equation there is a nonlinear term depending on the kinetics of ion exchange. A numerical solution of the direct problem of calculating the impurities concentration at the outlet of the filter is provided. The inverse problem is formulated as refers to finding the parameters of a mathematical model of an ion-exchange process in non-equilibrium conditions. A method for determining the effective values of that parameters is developed based on measurements of the impurities concentration at the outlet of the filter. The method is used to solve the inverse problem of recovering one and two unknown parameters of the ion-exchange filter model.

Keywords: ion-exchange filter, non-linear problem, exchange isotherm, non-linearity parameters, non-linear inverse problem.

1. Introduction

The basic source of heat and electrical energy are thermal power plants (TPP) burning fuel: gas and coal [1].

Power-generating units of TPP operate under severe corrosive conditions: high temperature (515 - 530°C) and pressure (15 MPa) of hot steam [2]. Some units are made of cheap corrosion and heat-resistant steel which is resistant to fuel combustion products, so the main reason for failures of power equipment is failure of the most thermally loaded parts thereof, namely pipes in which water, the coolant, flows[2].

Water is a good solvent of various salts featuring negative solubility coefficient, i.e., when the water temperature rises, the amount of dissolved salts is reduced, and the excess amounts of these salts are precipitated in heated tubes, forming scale with a very low coefficient of thermal conductivity [1].

Scale restricting specific flow of heat energy to the coolant - water - does not allow to cool pipes surface, heating surface metal temperature increases reaching critical melting point resulting in pipe burnout leading to emergency stop of the boiler [1].

The rate of corrosion of the metal under scale on the surface of the tubes sharply increases, i.e. scale is a corrosion catalyst [2-5].

Reducing metal corrosion not only requires using materials with high corrosion resistance, but also taking all measures to reduce aggressiveness of the environment - water [6-8].

The primary source of harmful impurities is make-up water from the water treatment plant designed to compensate for loss of water coolant in the power plant cycle.

One of the key technologies for the preparation of water at thermal power plants and nuclear power plants is the use of ion-exchange materials, which require periodic regeneration [9-12].

Water treatment using methods of ion exchange is based on passing of source water or partially treated water through a filter bed of ion-exchange material, substantially insoluble in water but capable of ion exchange with ions contained in the treated water. Materials having the properties of exchanging cations are called cation exchangers, which are after water softening periodically regenerated with NaCl brine solution and sulfuric acid regardless of whether the cation exchanger is in the Na-form or H-form.

Regeneration of ion-exchange filters results in highly mineralized, acidic and alkaline waste water [9]. With the continuous circulation of water through the production, when water is taken from the surface sources for the needs of production and is returned as a high-mineralized aqueous solution, in rivers and lakes an increase in salinity can be observed, and consequently, deterioration of water quality, which is a very negative impact on the environment.

When operating the water treatment plant equipment, switching from filtering (water treatment) to backwash, regeneration and washing modes, is carried out either time-wise or based on the volume of passed water and solution, which leads to an increase in errors of the equipment and accordingly, a sharp increase in water and reagents consumption for own needs.

About 25-30% of the water purifier output is for own needs of water treatment plants. For example, given the power of a thermal power plant of 1200 MW, water treatment plant capacity shall be about 250 m³/hour, i.e. an average of about 62,5 m³/hour or 547500 m³/year of water is spent for own needs

The problem of water treatment for thermal power plants using ion exchange method is a multiparameter task. The existing techniques to design the operation of either a separate ion-exchange filter or the whole plant for water purification from the dissolved impurities exclude the possibility of calculating the optimal mode of operation [12-14]. It makes relevant the issue of determining the resource of the filter and planning of technological process of its operation on the basis of analyzes of water quality at the outlet of the filter. That is, it is necessary to solve the inverse problem for finding the optimal process parameters.

In this article we propose our method of determining the optimal process parameters of the technological process, the use of which will allow to determine the optimal mode of operation of the water treatment plant, which will, in turn, increase the operating cycle of the ion-exchange filter and the whole water

treatment plant, decrease the amount of water consumed for own needs and reduce the amount of waste water. The method is rather simple and can be used to simulate the operation of water treatment equipment either on the existing TPP or at the stage of designing the water treatment plants equipment.

Section 2 of the article contains a description of the mathematical model of the ion-exchange filtration process. Section 3 describes the main idea of solving the inverse problem. In Section 4 a description of the method to solve the direct problem is provided. In Section 5 the method considered carefully for a simple inverse problem, when the two parameters of the model, namely, k and γ are shown. In Section 6 is provided a description of the results of the numerical solution of the inverse problem with one and two unknown parameters of the model of the ion-exchange filter. The final Section contains a brief summary of the research conducted.

2 The mathematical model of the ion-exchange filtration process

Mathematical modeling and calculation of various ion exchange processes focused of the dynamics of ion exchange have been investigated in articles [15-18]. In general, the ion exchange dynamics can be described by a system of following differential equations [12]:

- balance equation;
- kinetic equation;
- isotherm equation.

On this basis, we draw up a mathematical model of an ion-exchange filter operating in non-equilibrium conditions.

2.1 Balance equation

Sorption of ions mixture by ion exchange material is subject to the law of conservation of mass, so the ion-exchange filter mathematical model is based on the mass balance equations.

For all cases the balance relation at ion-exchange water treatment is written in the following form [12]:

$$w_{ap} \frac{dC}{dx} + \varepsilon \frac{dC}{dt} + \frac{dq}{dt} = 0 \quad (1)$$

where w_{ap} is the flow velocity in the free section of the filter, m/sec

ε is porosity of ion exchange material, m^3/m^3 ;

$\frac{dC}{dx}$ is the rate of the concentration change of ions in the solution (C) along the filter axis (x) (meq/kg)/m;

$\frac{dC}{dt}$ is the rate of concentration change of ions in the solution (C) with respect to time (t), (meq/kg)/sec;

$\frac{dq}{dt}$ is the rate of concentration change of ions in the ion exchange material (q) with respect to time (t), (meq/kg)/sec.

2.2 The kinetic equations

Processes of anion-exchange and cationization of fresh natural waters for which ($C_0 \leq 10^{-2} \text{H}$), as well as processes of ion-exchange of condensate type water run in accordance with the mechanism of the external diffusion kinetics [12].

One of the equations which is customarily used to describe the rate of ions exchange in the external diffusion kinetics is the Nernst equation kinetics [12]. But the Nernst theory is not always convenient to describe the diffusion kinetics, because of the large number of experimental variables in the equation. So, in the study [12] a simple equation of the general theory of mass transfer is used. This equation expresses the hypothesis that the rate of ions exchange is proportional to the deviation of concentration of ions in the sorbent from the corresponding equilibrium value. The equation is mathematically written in the following form:

$$\frac{dq}{dt} = \beta(C - C^*) \quad (2)$$

where β is mass transfer coefficient (proportionality), m^3/m^3 ;

q is the concentration of exchanging ions in the sorbent, mEq/kg ;

C is concentration of the exchanging ions in the solution, mEq/kg ;

C^* is equilibrium concentration of the ion in solution, which borders with the surface of the ion exchange material, mEq/kg , in which case $C > C^*$.

2.3 The equation of exchange isotherm

The adsorption isotherm equation is used to determine the capacity of each sorbent for various sorbates. In the ion exchange theory isotherms of Langmuir [19], Freundlich [20] and Redlich-Peterson [21] are commonly used. These studies have shown that for a mathematical description of the ion-exchange process in nonequilibrium conditions, the Langmuir isotherm equation described in [12] may be used. The corresponding mathematical expression can be written as follows:

$$\varphi^* = \frac{k(1-\varphi_a)\theta}{1-(1-k)\theta} \quad (3)$$

Here φ^* is relative concentration of exchanging ions in the solution phase, $\varphi^* = C^*/C_0$ where C_0 is the total concentration of exchanging ions in the original solution, mEq/kg ; k is the constant of exchange isotherm, $k > 1$; φ_a is the relative concentration of additional ions in the ion exchange material. These ions form a weakly dissociated compound, mEq/kg , $\varphi_a = C_a/C_0$ where C_a is concentration of ions in the original solution that form a weakly dissociated compound, mEq/kg ; θ is relative concentration of exchanging ions in the ion exchanger phase, mEq/kg , $\theta = q/q_0$ where q_0 is the total exchange capacity of the ion exchange material, mEq/kg .

Thus, taking into account equations (1)-(3), the mathematical model of the ion-exchange filtration process for nonequilibrium conditions can be written as follows:

$$\begin{cases} \varphi_t + c\varphi_x + \gamma\beta(\varphi - f(\theta)) = 0, \\ \theta_t = \beta(\varphi - f(\theta)), t \in (0, T], x \in (0, L) \\ f(\theta) = m\theta / (k + (1 - k)\theta) \end{cases} \quad (4)$$

The corresponding initial-boundary conditions are the following:

$$\begin{aligned} \varphi(x, 0) = 0, \theta(x, 0) = 0, \\ \varphi(0, t) = \varphi_0(t), \quad 0 \leq \varphi_0(t) < 1 \end{aligned} \quad (5)$$

Here $\theta(x, t)$, $\varphi(x, t)$ are relative concentrations of ion absorbed from the solution in the ion-exchange material and the solution respectively, normalized to q_0 , C_0 ; L - thickness of the ion-exchange material layer; the function $f(\theta)$ determines the value of the equilibrium concentration of exchanging ion in the solution at a given concentration θ of the same ion in the ion-exchange material; m is the constant determining possible chemical reactions with other impurities in the solution, $0 < m < 1$.

The numerical parameters of the model (4) are determined using the following formulas:

$$c = w_{ap}/\varepsilon, \beta = q_0/(\varepsilon C_0), \gamma = \eta \cdot C_0/q_0$$

here η is a proportionality factor that determine the rate of release of ions from the ion-exchange material. This ratio is called intradiffusion mass transfer coefficient and is determined by chemical kinetics of ion exchange. It depends on the area of free surface of the ion-exchange material pellets, on the rate of exchanging ions valences. Often the coefficient η is determined by some empirical relationships, for example, according to [12] $\eta \approx 0.009\alpha w_{ap}^{0.53}/d^{1.47}$ where α is an empirical coefficient, d is the diameter of the granules, m . At this stage of the study, we neglect the change of porosity ε of the material.

3 Formulation of the inverse problem

During the operation process the measurement of impurities concentration at the outlet of the filter are available:

$$\varphi(l, t) = \varphi_1(t), \quad 0 < t < T \quad (5)$$

Here T is the observation time. Due to the complexity of the sorption process and heterogeneity of the mixture to filter, parameters γ in the model (4), k and m in the equation

$$f(\theta; k, m) = \frac{m\theta}{k+(1-k)\theta} \quad (6)$$

cannot be determined accurately. In practice, we can only operate with some effective values of k , m and γ .

In practice we can assume the constant k to be known. Then we still have two uncertain model parameters: exchange isotherm parameter m , and parameter γ .

Having measurements (5), the problem is actually reduced to an inverse problem which can be stated as follows:

Find the model parameters m , γ of the problem (4)-(5a) via boundary measurements (5).

Although in the literature there are examples of formulations and solution of inverse problems for filtration processes [22], this statement of the inverse problem is new.

The standard method to solve inverse problems is the method of quasisolution [23], according to which the unknown parameters of the model are determined by minimizing the residual functional.

Earlier, in [24], we investigated the behavior of the residual functional of the following form

$$J(k, m) = \frac{1}{2} \int_0^T (\varphi(L, t; k, m) - \varphi(L, t; k_0, m_0))^2 dt \quad (7)$$

with the fixed value of the mass transfer coefficient γ .

On the base of the direct numerical simulation described in [24], we have shown that the functional (7) is not convex. Additionally, low sensitivity of the functional (7) was established with respect to the model parameters k, m . This means that the routine use of gradient methods is non efficient.

In this paper, we propose an alternative approach to the solution of the formulated inverse problem.

The main idea of the method is the following: as we have two unknown parameters of the model - m and γ , it is required to find at least two integral characteristics of the measured data (3), which clearly define these parameters.

Below, in Section 5 and 6 we provide examples of such characteristics, calculate the table of their values for the grid of the required parameters and use these tables to approximate the solution of the inverse problem.

4 The numerical solution of the direct problem

A necessary step in solving the inverse problem is the study and solution of direct problems. At the first stage to develop algorithms solving inverse problems one needs to study the behavior of the residual functional. Then other observable quantities should be determine to find the range of their sensitivity to changes in the sough-for parameters. In addition, the algorithm for solving the inverse problem should be tested primarily on the so-called synthetic measured data.

The direct problem (4) with known constants k and m is nonlinear, but, however, the nonlinear function $f(\theta)$ satisfies the Lipschitz condition [25], so it can be shown that the solution of the problem (4) - (5) exists and is unique. For the numerical solution of the problem (4) - (5) we apply the method of Finite Difference Scheme. The direct problem is approximated by implicit differential scheme, because it was found that the explicit scheme is very time consuming due to stability conditions. Due to the nonlinear nature of the problem, it turned out that for the stability of the calculation, even for the point-to-point computation scheme a very small time step is required.

To investigate the influence of variables k , m on the measurement data (6) the grids of parameters k and m is given at their characteristic changes intervals. Then for each pair of discrete values of k , m the concentration function $\varphi_1(t)$ is calculated at the output of the filter on a grid of values of time.

Calculations are made for values $w_{ap} = 5.55 \times 10^{-3}$ m / sec, $\varepsilon = 0.4$ m³ / m³, $\gamma = 2500$, $L = 1.8$ m, $C_0/q_0 = 5.75 \times 10^{-3}$, which are typical for hydrogenation exchange filters, used at the thermal power plants at the first stage of water treatment.

5 Solution of the inverse problem with one unknown parameter m of the model.

Assume first that the variable k , γ in model (4) are known and state the problem of determining the m -value.

The natural measured value is the total amount of impurity Q passed through during the ions observation period:

$$Q(T, k, m, \gamma) = \int_0^T \varphi_1(t) dt \quad (8)$$

We guess that this is one of the values which is sensitive to the model parameters, and examine its dependence on the k , m values. Due to the extremely large scatter $Q(T)$ it proved to be more convenient to use its logarithm $I_1 = \log Q$.

It was found that the value I_1 depends on the ratio $s = k/m$, and practically does not depend on m . This fact indicates that using the values of variable I_1 it is possible to recover the relationship $s = k/m$, and it is impossible to recover the variable m .

It follows that the models with the same value of the function $s(k, m) = k/m$ are equivalent with respect to the observed variable $\log Q$. On figures 1 and 2 we show the characteristic graphs of $\log Q$ depending on s and m .

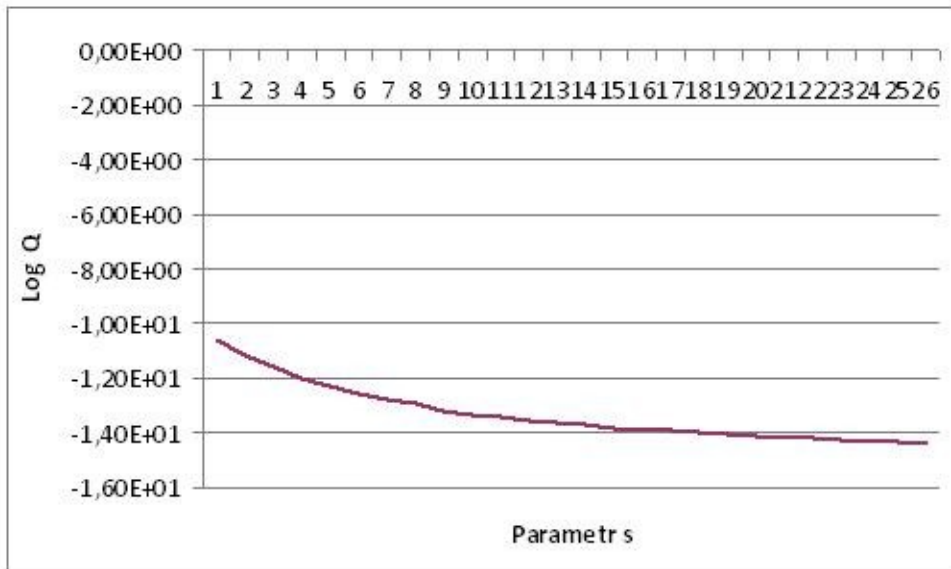


Figure 1: The dependence of $\log Q$ on the parameter s .

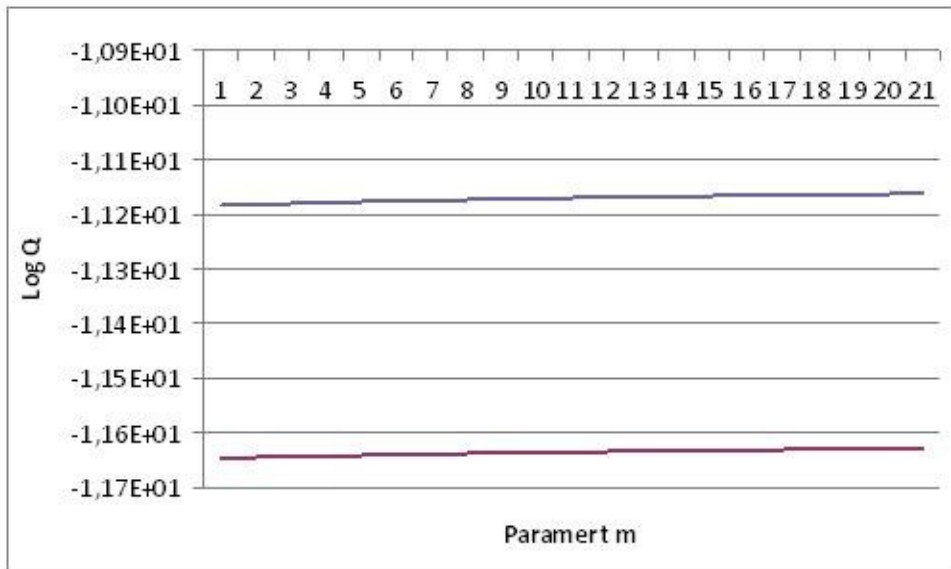


Figure 2: The dependence of $\log Q$ on the parameter m .

Thus, the following equivalence principle is obtained for the problem of filtration (4)-(5): *mathematical models with different m, k and the same $s = k/m$ are equivalent with respect to the observed value I_1 .*

Note that similar principles of equivalence occur also in geophysical problems [26] and are a mathematical expression of ill-posedness of the most of inverse problems.

Calculations show monotonic dependence $I_1(s)$ of s in practically acceptable range of k, m parameters. This implies that if we know the value k , then on the basis of the observed value I_1 the value $s = k/m$ is uniquely determined, and through it, in turn, the value $m = k/s$ can be recovered.

Thus, we have two uncertain parameters of the model: $s = m/k$, that is associated with a set of parameters of exchange isotherm and the parameter γ .

For practical applications of this result, we have generated a table of values $I_1 = \log Q$ for a range of values of s in the interval [1,5]. It is enough to interpolate the data of the generated table to restore the desired value of s_0 using the measured value of $\log Q_0$.

Table 1 shows the values of the experimental (s_{exp}) and restored (s_{rest}) parameters s for hydrogen-cation exchange filters used at the first stage of make-up water treatment.

Table 1: The values of the experimental (s_{exp}) and restored (s_{rest}) parameters s .

Parameters	Parameter values								
s_{exp}	1.45	1.7	2.0	2.45	3.1	3.5	4.0	4.45	4.7
s_{rest}	1.453	1.71	2.14	2.453	3.12	3.52	4.14	4.451	4.71

The test results showed satisfactory accuracy of m variable restoration.

6 Restoring parameters m and γ

Suppose now that we do not know the two parameters - m and γ . To calculate the range of change of γ we can use the following formula [12]:

$$\gamma = \eta \cdot C_0 / q_0 \quad (9)$$

Here η is the mass transfer coefficient; C_0 is the total concentration of exchanging ions in the original solution, mEq/kg; q_0 is the total exchange capacity of the ion exchange material, mEq/kg.

Parameter η are calculated according to [12], using the empirical formula:

$$\eta \approx 0.009 \alpha w_{ap}^{0.53} / d^{1.47} \quad (10)$$

where w_{ap} is the rate of flow of fluid in the unit, m/s; d is the ion exchange grain size, m; α is an empirical coefficient.

For the calculation we use the parameter values that are typical for hydrogen-cation exchange unit of the first stage of water treatment: $w_{ap} = 0,0056$ m/s, the value of d for the ion exchange material KU-2-8 varies from 0,315 to 1,250 mm. The coefficient α for the exchange of H^+ ions for Na^+ is 1.

Based on these data, the values of γ in the processes of interest are in the range of $1,523 \leq \gamma \leq 3,165$.

Suppose that another observable quantity that is sensitive to the values of the unknown parameters of the model is the increment of the logarithm of the impurities concentration at the outlet of the filter:

$$I_2(T, k, m, \gamma) = \log (\varphi_1(T)) - \log ((\varphi_1(0)) \quad (11)$$

$$Q(T, k, m, \gamma) = \int_0^T \varphi_1(t)dt \quad (12)$$

By solving the direct problem in the range of variation of parameters s and γ we have generated a tables of values of variables $I_1 (T, s, \gamma)$ and $I_2 (T, s, \gamma)$.

Based on these two tables, we can calculate the appropriate values s_0, γ_0 from the measured values $I_{1,0}$ and $I_{2,0}$.

Below is described a numerical method for calculating s_0, γ_0 . Thus, suppose we have a table of values for the two measured variables at the filter outlet: $I_1 (s_i, \gamma_j)$ and $I_2 (s_i, \gamma_j)$ Suppose $I_{1,0}$ and $I_{2,0}$ are measured values. Using the tables we can determine a pair of values s_k, γ_l , for which the minimum deviation is reached.

$$\|I_1(s_k, \gamma_l) - I_{1,0}\|^2 + \|I_2(s_k, \gamma_l) - I_{2,0}\|^2 = \min_{i,j} \|I_1(s_i, \gamma_j) - I_{1,0}\|^2 + \|I_2(s_i, \gamma_j) - I_{2,0}\|^2 \quad (13)$$

Assuming that the quantities $I_1(s, \gamma)$ and $I_2(s, \gamma)$ are smooth functions of their arguments, we can write at a point of s_k, γ_l approximate equations:

$$\begin{aligned} I_{1,0} &= I_1(s_0, \gamma_0) = I_1(s_k, \gamma_l) + \frac{dI_1}{ds}(s_k, \gamma_l)(s_0 - s_k) + \frac{dI_1}{d\gamma}(s_k, \gamma_l)(\gamma_0 - \gamma_l), \\ I_{2,0} &= I_2(s_0, \gamma_0) = I_2(s_k, \gamma_l) + \frac{dI_2}{ds}(s_k, \gamma_l)(s_0 - s_k) + \frac{dI_2}{d\gamma}(s_k, \gamma_l)(\gamma_0 - \gamma_l) \end{aligned} \quad (14)$$

Partial derivatives $\frac{dI_1(s_k, \gamma_l)}{ds}$, $\frac{dI_1(s_k, \gamma_l)}{d\gamma}$, $\frac{dI_2(s_k, \gamma_l)}{ds}$, $\frac{dI_2(s_k, \gamma_l)}{d\gamma}$ are approximately calculated using the tables through formulas of finite differences:

The relations (14) represent a linear system of equations with indeterminates s_0, γ_0 . This system is solved numerically and determines approximate values of the unknown parameters. Table 2 shows the values of the experimental (γ_{exp}, m_{exp}) and restored (γ_{rest}, m_{rest}) parameters γ and m for hydrogen-cation exchange filters used at the first stage of make-up water treatment.

Table 2: The values of the experimental and restored parameters γ and m .

Parameters	Parameter values								
γ_{exp}	6.3	7.5	8.6	9.1	10.5	12.4	17.0	23.7	26.4
m_{exp}	0.55	0.6	0.66	0.7	0.73	0.78	0.82	0.85	0.89
γ_{rest}	6.35	7.53	8.61	9.21	10.54	12.43	17.15	23.72	26.43
m_{rest}	0.57	0.64	0.68	0.72	0.74	0.80	0.82	0.86	0.9

The test results showed satisfactory accuracy of m and γ variables restoration.

6. Conclusion and Discussions

The direct and inverse problems for a mathematical model of an ion-exchange filter are examined. The model is nonlinear and depends on a number of physical and chemical parameters. A series of numerical simulations for arrays of values of these parameters are carried out.

The new statement of the inverse problem for a nonlinear mathematical model of an ion exchange filter is provided. The equivalence principle of mathematical models for the set of parameters (k, m) is established for the first time.

A new method to recover the model parameters is developed based on the measurements at the outlet of the filter. The integral characteristics of the filtration process that uniquely determine the required coefficients of the inverse problem are found.

The use of the method is described for the case when one and two filtration model parameters are unknown.

The studies show satisfactory accuracy of the recovery of the parameters of the filtration process for ion exchange filters.

Conflict of Interests

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

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