UDC 628.16: 517.95
DOI: 10.15587/1729-4061.2019.177537

PREDICTION OF THE PROCESS OF BIOLOGICAL DEFERRIZATION OF UNDERGROUND WATER IN A BIOREACTOR

A. Kvartenko
PhD, Associate Professor
Department of Water Supply, Water Sewerage and Drilling
National University of Water and Environmental Engineering
Soborna str., 11, Rivne, Ukraine, 33028
E-mail: o.m.kvartenko@nuwm.edu.ua

I. Prysiazhniuk
PhD, Associate Professor
Department of Higher Mathematics
Rivne State University of Humanities
S. Bandery str., 12, Rivne, Ukraine, 33000
E-mail: igorpri79@gmail.com

Дослідження в галузі очищення підземних вод вказують на перспективність розвитку їх комплексної очистки за участю різних морфологічних типів мікроорганізмів, закріплених на інертних контактних матеріалах. Вказано, що при певних параметрах якості води (pH 6–7; Eh 50…200 мВ, в присутності розчиненого диоксиду вуглецю та величинах перманганатної окисності до 5 мг O_2/дм^3) в підземних водах превалює розвиток бактерій роду Gallionella, а при значеннях pH 6,5–7,5; Eh=–200…300 мВ та ПО>5 мг O_2/дм^3 – розвиток бактерій родів Lepthothrix, Crenothrix. Це надає ряд переваг при застосуванні біохімічного методу перед традиційними фізико-хімічними, зокрема прискорення процесу очищення води від сполук феруму.

Показано, що моделюванню кінетики процесів очищення підземних вод в біореакторах приділялося значно менше уваги ніж традиційним фізико-хімічним методам, для яких були розроблені сучасні математичні моделі. Тому розвиток напрямку моделювання біохімічного процесу очищення води від сполук заліза є актуальним завданням. Математична модель представлена задачею Коші для нелінійної системи диференціальних рівнянь в частинних похідних першого порядку. Система задачі Коші складається з п'яти рівнянь з п'ятьма невідомими функціями, які описують розподіл концентрацій катіонів феруму, бактерій а також матриксних структур в двох фазах (рухомій та іммобілізованій) як у просторі, так і у часі. При побудові моделі були використані як технологічні (максимальна брудомісткість (2,6– кг/м^3), гранична величина біомаси бактерій в матриксних структурах (9,5 г/м^3), максимальна питома швидкість їх росту (0,17–0,18 год$^{-1}$), коефіцієнт насичення (0,65–0,7 г/м^3), швидкість потоку в діапазоні 5–20 м/год), так і конструктивні параметри (висота контактного завантаження біореактра 1,3 м). В розглянутій моделі час ефективної роботи біореактора залежить від концентрацій катіонів Fe^{2+}, які в природних водах можуть знаходитися в межах 0,5–20 мг/дм^3, кількості феробактерій (10^2–10^4 кл/дм^3), а також швидкості потоку води. Враховано зворотний вплив характеристик процесу, зокрема концентрації матриксних структур в міжпоровому просторі, а також характеристик середовища за допомогою коефіцієнтів масообміну та пористості. Модель дозволяє визначати оптимальний час роботи біореактора між промивками

Ключові слова: біохімічні процеси, кінетична модель біологічного знезалізнення, матриксні структури, метод характеристик

Received date 23.04.2019
Accepted date 27.08.2019
Published date 31.10.2019

1. Introduction

In the practice of water treatment, the methods of removing iron compounds from water are represented by three groups: non-reagent, reagent, and biological [1]. When treating weakly acidic (5.5–6.5) and near-neutral (6.5–7.5) waters with low (<2.0 mmol/dm^3) and medium (2–4 mmol/dm^3) alkaline reserve, the use of the biological method has advantages over conventional physical and chemical methods [2, 3], in particular, acceleration by several times of the rate of the process of iron compounds oxidation in both mineral form and in the form of organic complexes [2, 4, 5]. That is why the relevant task of today is to switch over the existing stations, which operate by the method of simplified aeration – filtration, to the operation with the use of the method of biochemical oxidation [2]. Over the past few decades, the biochemical method has become quite widespread in many countries of the world [3]. Papers [5, 6] emphasized the dominance of the biological method over the physical and chemical method in the treatment of iron-containing weakly acidic and near-neutral groundwater with low and medium alkaline reserve. The empirical data were mainly obtained as a result of the implementation of the technology of biological treatment of underground water [2, 5]. That is why the research into the operation of bioreactors under different operating conditions with the subsequent development of a mathematical model of the process is currently a relevant task.

2. Literature review and problem statement

Existing mathematical models of the removal of ferrous compounds are divided into the models describing the processes of physical-chemical and biological treatment of groundwater. According to the degree of consideration of various factors and processes, the models, which describe the processes of physical-chemical treatment, can be divided into several groups. The first group includes the models that describe only kinetics of the process of oxidation of Fe^{2+}. In paper [7], the authors presented a kinetic model, which describes the processes of removal of Fe^{2+} and $Fe(OH)_3$ ions by the depth of a multilayer filtering load. The model considers the parameters of water quality, in particular, Ph magnitude, concentration of Fe^{2+}, Mn^{2+}, homogenous oxidation in the over-filter space and heterogeneous oxidation in the thickness of the filtering load. The model does not take into account the processes of convective transfer of the studied elements in the depth of contact load during a filter cycle, as well as the change of porosity of the interpore space.

The most complex type includes the multicomponent models that describe different processes of physical-chemical deferrization of groundwater. The most modern of them are the models developed, in particular, in papers [8, 9]. The model, presented in [8], describes the dynamics of the process of water deferrization on fast filters at constant values of filtering rates. The model considers the influence of the processes of mass transfer, kinetics of exchange and various transformations that occur both in liquid and in the solid phases of the system in relation to Fe^{2+}, oxygen, various forms of $Fe(OH)_3$. Paper [9] contains theoretical research into dynamics of accumulation of iron compounds during water purification through two-layer filters. The mathematical model consists of a hydrodynamic (filtration) unit and a unit of dynamics of glandular compounds in the filtration medium. The hydrodynamic unit, in turn, consists of the equations of filtration and continuity of the filtration flow under conditions of changing hydraulic characteristics in the filter layers. The unit of dynamics of iron compounds consists of the equations of material balance, written down relative to concentrations of Fe^{2+} and iron hydroxide $Fe(OH)_3$ in the solution and in solid (fixed) phases.

Article [10] presents the modern kinetic model of physical-chemical deferrization of groundwater through filters, which takes into account the mass transfer for two forms of iron. The model also reflects the intensification of iron removal under the influence of resulting sediment, provides continuous filtration, and takes into consideration the limitation of sorption resource.

Our analytical review [7–10] shows that during the theoretical solution of the problem of removal of Fe^{2+} cations from neutral waters, the classical method based on studying the regularities of their removal by the physical and chemical mechanism in the process of filtration was mainly used. In theoretical studies, the role of the biological factor, which exists in the treatment of weakly acidic and near-neutral groundwater was not taken into consideration. Papers [11–14] address the mentioned factor. Thus, [11] shows the results of research into the removal of Fe^{2+} and Mn^{2+} cations from underground water in the continuously operating reactor. The authors use the modernite mineral as contact load. Physical activation of the mineral was carried out by means of its heating up to 400–600 °C. However, the work shows only the results of experimental data without constructing a mathematical model and corresponding results of numerical calculations, which makes it impossible for the authors to model the studied process.

It is this approach that was used in research [12] whose authors, based on the experimental research, developed a model that takes into account a number of parameters of water quality, namely: concentrations of Fe^{2+}, Mn^{2+}, magnitude of pH. The model is sensible to experimentally determined adsorption parameters and allows predicting the kinetics of oxidation of Fe^{2+} compounds in the filtering load. However, it should be noted that when developing the model, the authors did not take into account the development of bacterial populations, formation of matrix structures with their further influence on the deferrization process. The reason for this can be objective difficulties associated with determining the biomass concentration by the depth of contact load of a bioreactor. The mutual influence of the main characteristics of the process was not taken into consideration either, which does not make it possible to predict more accurately the time of effective operation of a bioreactor, as well as to obtain the distribution of concentrations of the core components of the process by layers of contact load.

In paper [13], the process of deferrization of underground shaft waters with the help of the fixed thionic bacteria of the genus *Thiobacillus ferrooxidans*, with nutrition of bacteria in a bioreactor was modeled. As a result of the research carried out by the authors, it was found that the effectiveness of operation of bioreactors with fixed microflora is from 7 to 20 times better at deferrization of mine waters than at their passive treatment in horizontal sumps. It should be noted that when developing the model, the authors did not take into account the influence of fixed biomass and matrix structures on cleaning processes. In addition, the factors of fixing-separation of bacteria and sediment under the influence of the hydrodynamics forces with their transfer to the lower layers of contact load were not taken into consideration, which does not give a complete idea of operation of a bioreactor between washings.

This problem was partially solved in paper [14]. The authors presented the results of comparative studies of physical-chemical and biological removal of ferrum compounds from drinking water. The concentration of ferrum ions accounted for 1, 2, 3 and 4 mg/dm^3, the dissolved oxygen up to 7 and 8 mg/dm^3, the water temperature was 14 °C, the magnitude of Ph was 7.5. The paper provides the model with a series of assumptions for combined removal of ferrum ions. In particular, the authors accepted that microorganisms were uniformly immobilized on the surface of the material of contact load as a monolayer of constant thickness. Biological oxidation occurred only in the biofilm, and only physical-chemical oxidation occurred in the liquid volume.

The model is represented by the equation of mass balance, which includes the transport and iron oxidation in the result of aeration and bacterial activity. However, in this model, the authors did not take into account the influence on the processes of oxidation of ferrobacteria, which come to a bioreactor with outlet water, as well as the bacteria located on the matrix structures in the interpore space. In addition, they did not take into account the processes of adhesion and separation of bacteria from the matrix structures, the processes of formation of matrix itself with the passage of the processes of fixation-separation with their subsequent transportation into the lower layers of contact load. The model does not allow determining the optimal operating time of a bioreactor between washings.

All this allows us to argue that at the present time, the problems of modeling the processes of biological purification of groundwater from iron compounds in contact load of bioreactors have not been solved completely. In particular, not only determining the optimal operating time of a bioreactor between washings, but also determining the efficiency of purification by the depth of contact load under the influence of a number of accompanying factors (bacterial component, matrix structures, additional source of carbon dioxide, etc.) remain unresolved.

3. The aim and objectives of the study

The aim of this study is to develop a mathematical model of kinetics of the process of treatment of underground water-containing waters in bioreactors. This will enable obtaining the distribution of concentrations of bacteria, bivalent and trivalent iron in the middle of a bioreactor to set its optimum physical dimensions, predict the time of its effective operation between washings.

To accomplish the aim, the following tasks have been set:

– to identify the necessary components of the process to solve the model, to construct a physical model of the process;

– to substantiate the structure of the model, to set its parameters, to determine the initial and boundary conditions;

– to construct the algorithm for solving the set problem, using the method of characteristics of solving the Cauchy problem for differential equations in partial derivatives of the first order and numerical methods;

– to conduct numerical experiments of the process of water purification from ferrite compounds in contact load of a bioreactor.

4. Materials and methods to model water purification process in a bioreactor

The model is based on modern theoretical and microbiological research into the biochemical deferrization process presented in papers [2, 15–17]. The proposed model mainly focuses on developing the concept of kinetics of biochemical treatment of groundwater. Based on these concepts, the kinetic model, which should take into account the main phenomena and mechanisms in biochemical purification of groundwater from ferrite compounds, is being developed.

4. 1. Substantiating the selection of stage in the bioreactor operation for studying

According to the results of years of research conducted in the town of Berezne (2010–2018), the settlement of Rokytne (2011–2013), Ukraine, it was possible to state that setting a bioreactor in a working state is divided into two stages. Stage 1 is the initial stage of «charging» the contact load. During this period, the water from the well, which contains iron compounds, carbon dioxide, and ferrobacteria, passes through the load of a bioreactor. Gradually, the active catalytic surface from cells and matrix structures is formed at the surface of the granules of contact load. The process takes place as a result of adhesion of bacteria with the negative charge on the primary catalytic film, according to the known mechanism [18]. The duration of «a charge» depends on the parameters of water quality: pH magnitude, hydrocarbonic alkalinity, concentration of Fe(II) ions and the presence of ferrobacteria. During this period, the initial matrix structure is formed. New concentrations of bacteria come to a bioreactor with each portion of coming water, increasing their total quantity. New bacterial structures begin to form their matrices on previous sediments.

The second stage (described by the kinetic model) is the working stage during the operation of a bioreactor between washings. As a result of the research at the pilot and production plants carried out by the authors (the town of Berezne), it was found that after washing, the number of bacteria in the load of the bioreactor amounted to 10^3–10^5 kl/cm^3. This is explained by the fact that not only bacteria from the previous filter cycle, but also bacteria that came along with washing water from the well and were fixed on the surface of its contact load remain in the bioreactor. Thus, a certain amount of biomass which remained in the load (0.1 g/m^3), as well as the amount of biomass that came with the original water (0.001 g/m^3), were accepted during the statement of the boundary conditions. In addition, based on the results of own experimental research, we established: maximal specific rate of growth of microorganisms (μ_{max} 0.17–0.18 h^{-1}) and constant of saturation (K_F 0.65–0.7 g/m^3).

The bacteria of the genus *Gallionella* create helical structures as a result of their vital activity [19]. Gradually, from separate covers of these bacteria, the porous structure from bio-minerals γ-FeOOH begins to be formed in the interpore space of contact load, which eventually fills the whole interpore spaces, through which the flow passes convectively. An increase in the volume of the matrix structures of biominerals in the interpore space led to their gradual migration with the descending flow of water to the lower layers with their gradual clogging, which was proved by the piezometric filming of pressure losses by the height of contact load, as well as sampling to determine the concentrations of matrix structures and bacteria by the height of contact load. It should be noted that when a certain magnitude of contamination is reached at the corresponding height (maximum contamination content), the rate of gain in pressure losses in this area decreased, while the rate of their gain on the lower layers increased. A gradual increase in pressure losses in height resulted in an increase of the water level over contact load and maintained the rates averaged in time in the calculated limits. The local rates in the interpore space may change in time between washings, but for simplification, we accept that the integral rate is equal to the averaged rate.

4. 2. Mathematical modeling of the process of water purification from ferrum compounds

We will model the considered process of water deferrization in a bioreactor with the height of contact load ℓ by the following problem:

$$V=-\theta(M)\,\mathrm{grad}\,P, \tag{1}$$

$$\sigma(M)\frac{\partial B}{\partial t}=W(x,t)B-\upsilon(x)\frac{\partial B}{\partial x}-\gamma(x,t)H(x,t)-\chi B, \tag{2}$$

$$\frac{\partial U}{\partial t}=W(x,t)U+\gamma(x,t)\cdot H(x,t)-\chi U, \tag{3}$$

$$\sigma(M)\frac{\partial F}{\partial t}=-\upsilon(x)\frac{\partial F}{\partial x}-\beta_1 W(x,t)B-\beta_2 W(x,t)U-R_{chem}, \tag{4}$$

$$\sigma(M)\frac{\partial S}{\partial t}=-\upsilon(x)\frac{\partial S}{\partial x}-k_1 S+H^*(x,t)+\eta_1 W(x,t)B, \tag{5}$$

$$\frac{\partial M}{\partial t}=k_1 S-H^*(x,t)+\eta_2 W(x,t)U. \tag{6}$$

Write down the initial and boundary conditions for equations (2) to (6):

$$B(x,t)\big|_{t=0} = B_0^0(x); \quad F(x,t)\big|_{t=0} = F_0^0(x);$$

$$S(x,t)\big|_{t=0} = S_0^0(x); \tag{7}$$

$$B(x,t)\big|_{x=0} = B_*(t); \quad F(x,t)\big|_{x=0} = F_*(t); \quad S(x,t)\big|_{x=0} = S_*(t);$$

$$U(x,t)\big|_{t=0} = U_0^0(x), \quad M(x,t)\big|_{t=0} = M_0^0(x),$$

where x is $(0, \ell)$, ℓ is the height of the working part of a bioreactor, m; t is $(0, T)$, where T is the time of effective operation of the filter, determined in the process of solving the problem and equal to time within which the maximum contamination saturation of a bioreactor is reached.

M_*^* is the maximum concentration of matrix in a bioreactor, g/m^3 (maximum contamination capacity).

M is the total concentration of matrix in a bioreactor at the explored moment, g/m^3.

B is the mean magnitude of the biomass of ferrobacteria in the unit of volume of original water that goes through a bioreactor, g/m^3.

U is the mean magnitude of biomass of ferrobacteria immobilized on the surface of the primary stationary shell of contact load and matrix structures in the interpore space, g/m^3.

$F(x, t)$ is the concentration of divalent iron in original water, g/m^3.

B^{**} is the boundary magnitude of the biomass of bacteria in the matrix structures of a bioreactor, g/m^3.

γ is the function of the rate of bacteria fixing on the surfaces of matrix structures and grains of load per unit of time, h^{-1}, depends on the ionic strength of solution and pH of original water (for pH 5.0–7.2, it is in the range of 0.72–0.66 [4]).

β is the coefficient of mass fraction of Fe^{2+} ions, necessary for formation of 1 g of cell biomass of ferrobacteria and ensuring their respiration process is 279 g of Fe^{2+} ions.

η is the coefficient of mass fraction of matrix structures of biominerals g-FeOOH, it is 530 g per 1 g of the biomass.

v is the water flow rate, m/h.

χ is the coefficient of bacteria dying rate, h^{-1}.

$v(x)S_x$ is the component that characterizes the dynamics of convective transfer of matrix structures formed in the flow by the depth of contact load.

$(-k_1S)$ is the component that describes the adhesion of matrices formed in dynamics of flow by non-fixed bacteria on the pre-formed stationary matrix in the interpore space.

$\eta_1 W(x, t)B$ та $\eta_2 W(x, t)U$ are the components that characterize the formation of new matrix structures in the interpore space with the help of bacteria.

When the water flow passes through the interpore contact load of a bioreactor, which is gradually filled with matrix structures, there is their partial separation, which is described by function $k_2 \cdot M(x, t)$ when condition $M(x,t) < M_*^*$ is satisfied. When matrix structures are accumulated in the interpore space in the concentration, which exceeds the maximum contamination capacity, i. e. when the condition $M(x,t) \geq M_*^*$ is reached, matrix structures are separated and transfer into the water flow $k_3\left(M(x,t) - M_*^*\right)$.

k_3 is the coefficient which characterizes the fraction of detached excess matrix structures per unit of time, h^{-1}.

The first component of equation (2), W is the rate of growth of microorganisms according to the Mono equation:

$$W(x,t) = \frac{\mu_{\max} F(x,t)}{\left(F(x,t) + K_F\right)}, \tag{8}$$

where $\mu_{\max}$ is the maximum specific rate of growth of microorganisms, h^{-1}; K_F is the constant of saturation, g/m^3.

Function $H(x, t)$ establishes the relation between the biomass of bacteria in the unit of volume of original water $B(x, t)$, average biomass of bacteria $U(x, t)$ on the surface of grains of contact load and matrix structures in the interpore space [21, 22]:

$$H(x,t) = \begin{cases} B(x,t), & \text{if } B(x,t) < B^{**} - U(x,t), \\ B^{**} - U(x,t), & \text{if } B(x,t) \geq B(x,t) \geq B^{**} - U(x,t). \end{cases}$$

Function $H^*(x, t)$ ensures the transition of matrix from the stationary state to the dynamic state with taking into account the boundary saturation of matrix:

$$H^*(x,t) = \begin{cases} k_2 \cdot M(x,t), & \text{if } \quad (x,t) < M_*^*, \\ k_3 \cdot (M(x,t) - M_*^*), & \text{if } M(x,t) \geq M_*^*. \end{cases}$$

The choice of the initial conditions was determined by the fact that at the initial moment of time we know the distribution of concentration of cations of Fe^{2+} g/m^3, matrix structures and biomass, respectively, in the moving (S, B, g/m^3), and unmoving (M, U g/m^3) phases in a bioreactor. The choice of boundary conditions is determined by the fact that at the inlet of a bioreactor we know: distribution of concentrations of bacteria (B), Fe^{2+} cations, moving phase of matrix (S).

Equation (1) is the equation of filtration, equations (2) to (4) describe the process of biologically abiotic oxidation of ferrum compounds with the help of consortia of ferrobacteria. Note that bacteria are fixed both on the surface of the stationary catalytic shell of grains and on the matrix structures of bio-minerals in the interpore space of contact load. In addition, bacteria that move freely in the interpore space together with the water flow that must be cleaned are considered. Equations (5), (6) describe the processes of accumulation-transfer of the matrix structures formed in the process of bioreactor operation by the height of contact load of a bioreactor. In considering this process, we take into account the reverse influence of the characteristics of the process [21, 22], in particular, the concentration of the matrix that settled down in the load of a bioreactor and characteristics of the medium through the coefficients of mass exchange, porosity, etc. We believe that the average rate is maintained at the expense of self-regulation of flow hydrodynamics and biochemical processes.

4. 3. Construction of the algorithm to solve a model problem

Reduce the above set problem (2) to (6) to solving n problems respectively on time intervals $n\Delta t \leq t \leq (n+1)\Delta t$ [22]:

$$\sigma(M_n)\frac{\partial B_n}{\partial t} = W_n(x,t)B_n - \upsilon(x)\frac{\partial B_n}{\partial x} - \gamma(x,t)H_n(x,t) - \chi B_n, \tag{9}$$

$$\frac{\partial U_n}{\partial t} = W_n(x,t)U_n + \gamma(x,t)\cdot H_n(x,t) - \chi U_n, \tag{10}$$

$$\sigma(M_n)\frac{\partial F_n}{\partial t}=-\upsilon(x)\frac{\partial F_n}{\partial x}-\beta_1 W_n(x,t)B_n-$$
$$-\beta_2 W_n(x,t)U_n, \tag{11}$$

$$\sigma(M_n)\frac{\partial S_n}{\partial t}=-\upsilon(x)\frac{\partial S_n}{\partial x}-k_1 S_n+H_n^*(x,t)+$$
$$+\eta_1 W_n(x,t)B_n, \tag{12}$$

$$\frac{\partial M_n}{\partial t}=k_1 S_n-H_n^*(x,t)+\eta_2 W_n(x,t)U_n, \tag{13}$$

$$W_n(x,t)=\frac{\mu_{max}F_n(x,t)}{\left(F_n(x,t)+K_F\right)}. \tag{14}$$

Boundary conditions for the process:

$$B_n\Big|_{x=0}=B_*(t),$$

$$F_n\Big|_{x=0}=F_*(t),$$

$$S_n\Big|_{x=0}=S_*(t).$$

Initial conditions on time interval $n\Delta t\le t\le(n+1)\Delta t$ will take the form:

$$B_n\Big|_{t=n\Delta t}=B^{**}(x)=\begin{bmatrix}B_0^0(x), n=0,\\ B_{n-1}(x,n\Delta t), n>0,\end{bmatrix}$$

$$U_*^*(x,(n-1)\Delta t)=\begin{bmatrix}U_0^0(x), & \text{if } n=0,\\ U_{n-1}(x,(n-1)\Delta t), & \text{if } n>0,\end{bmatrix}$$

$$F_n\big|_{t=n\Delta t}=F_*^*=\begin{bmatrix}F_0^0(x), n=0,\\ F_{n-1}(x,n\Delta t), n>0,\end{bmatrix}$$

$$M_*^*(x,(n-1)\Delta t)=\begin{bmatrix}M_0^0(x), & \text{if } n=0,\\ M_{n-1}(x,(n-1)\Delta t), & \text{if } n>0,\end{bmatrix}$$

$$S_n\big|_{t=n\Delta t}=S_*^*=\begin{bmatrix}S_0^0(x), n=0,\\ S_{n-1}(x,n\Delta t), n>0.\end{bmatrix}$$

Function H_n^* describes the process of adhesion of matrix structures in the interpore space of contact load taking into consideration its boundary saturation:

$$H_n^*(x,t)=\begin{cases}k_2\cdot M_n(x,t), & \text{if } M_n(x,t)<M_*^*,\\ k_3\left(M_n(x,t)-M_*^*\right), & \text{if } M_n(x,t)\ge M_*^*.\end{cases}$$

Function H_n describes the similar process for ferrobacteria taking into consideration their boundary saturation on each of time intervals $n\Delta t\le t\le(n+1)\Delta t$:

$$H_n(x,t)=\ge\begin{bmatrix}B_n(x,t), & \text{if } B_n(x,t)<B^{**}-U_n(x,t),\\ B^{**}-U_{n(x,t)}, & \text{if } B_n(x,t)\ge B^{**}-U_n(x,t).\end{bmatrix}$$

Taking into account the inverse effect of the concentration of adsorbed matrix on the porosity of the medium, as well as the inverse effect of the concentration of Fe^{2+}, bacteria and matrices on mass exchange coefficients, we obtain the following somewhat simplified dependences:

$$\sigma\left(M_{n-1}(x,t-\Delta t)\right)\frac{\partial B_n}{\partial t}=W_{n-1}(x,t)B_{n-1}(x,t-\Delta t)-$$
$$-\upsilon(x)\frac{\partial B_n}{\partial x}-\gamma(x,t)H_{n-1}(x,t)-\chi B_{n-1}(x,t-\Delta t), \tag{15}$$

$$\frac{\partial U_n}{\partial t}=W_{n-1}(x,t)U_{n-1}(x,t-\Delta t)+$$
$$+\gamma(x,t)\cdot H_{n-1}(x,t)-\chi U_{n-1}(x,t-\Delta t), \tag{16}$$

$$\sigma\left(M_{n-1}(x,t-\Delta t)\right)\frac{\partial F_n}{\partial t}=$$
$$=-\upsilon(x)\frac{\partial F_n}{\partial x}-\beta_1 W_{n-1}(x,t)B_{n-1}(x,t-\Delta t)-$$
$$-\beta_2 W_{n-1}(x,t)U_{n-1}(x,t-\Delta t), \tag{17}$$

$$\sigma\left(M_{n-1}(x,t-\Delta t)\right)\frac{\partial S_n}{\partial t}=-\upsilon(x)\frac{\partial S_n}{\partial x}-k_1 S_{n-1}(x,t-\Delta t)+$$
$$+H_{n-1}^*(x,t)+\eta_1 W_{n-1}(x,t)B_{n-1}(x,t-\Delta t), \tag{18}$$

$$\frac{\partial M_n}{\partial t}=k_1 S_{n-1}(x,t-\Delta t)-H_{n-1}^*(x,t)+$$
$$+\eta_2 W_{n-1}(x,t)U_{n-1}(x,t-\Delta t), \tag{19}$$

$$W_{n-1}(x,t)=\frac{\mu_{max}F_{n-1}(x,t)}{(F_{n-1}(x,t-\Delta t)+K_F)}, \tag{20}$$

$$H_{n-1}(x,t)=$$
$$=\begin{bmatrix}B_{n-1}(x,t-\Delta t), & \text{if } B_{n-1}(x,t-\Delta t)<B^{**}-U_{n-1}(x,t-\Delta t),\\ B^{**}-U_{n-1}(x,t-\Delta t), & \text{if } B_{n-1}(x,t-\Delta t)\ge B^{**}-U_{n-1}(x,t-\Delta t),\end{bmatrix}$$

$$H_{n-1}^*(x,t)=$$
$$=\begin{bmatrix}k_2 M_{n-1}(x,t-\Delta t), & \text{if } M_{n-1}(x,t-\Delta t)<M_*^*,\\ k_3(M_{n-1}(x,t-\Delta t)-M_*^*), & \text{if } M_{n-1}(x,t-\Delta t)\ge M_*^*.\end{bmatrix}$$

The boundary and initial conditions remain the same (7). Note that in this case, the mass exchange components are the known functions for each of the problems on time intervals $n\Delta t\le t\le(n+1)\Delta t$. Porosity coefficient is a constant magnitude on each next stage and each section of a bioreactor.

Equations (15), (17), (18) are the linear heterogeneous differential equations in fractal derivatives of first order, which are solved by the method of characteristics similarly to [21, 22].

$$B_n(x,t)=$$
$$=\begin{cases}\int_0^x \frac{g_n(\tilde{x},\sigma_n f(\tilde{x})-\sigma_n f(x)+t)}{\upsilon(\tilde{x})}\mathrm{d}\tilde{x}+B_*(t-\sigma_n f(x)), t\geq\sigma_n f(x),\\ \frac{1}{\sigma_n}\int_{n\Delta t}^{t} g_n\left(f^{-1}\left(\frac{\sigma_n f(x)-t+\tilde{t}}{\sigma_n}\right),\tilde{t}\right)\mathrm{d}\tilde{t}+B_*^*\left(f^{-1}\left(\frac{\sigma_n f(x)-t}{\sigma_n}\right)\right), t<\sigma_n f(x),\end{cases} \quad (21)$$

$$F_n(x,t)=$$
$$=\begin{cases}\int_0^x \frac{Fg_n(\tilde{x},\sigma_n f(\tilde{x})-\sigma_n f(x)+t)}{\upsilon(\tilde{x})}\mathrm{d}\tilde{x}+F_*(t-\sigma_n f(x)), t\geq\sigma_n f(x),\\ \frac{1}{\sigma_n}\int_{n\Delta t}^{t} Fg_n\left(f^{-1}\left(\frac{\sigma_n f(x)-t+\tilde{t}}{\sigma_n}\right),\tilde{t}\right)\mathrm{d}\tilde{t}+F_*^*\left(f^{-1}\left(\frac{\sigma_n f(x)-t}{\sigma_n}\right)\right), t<\sigma_n f(x),\end{cases} \quad (22)$$

$$S_n(x,t)=$$
$$=\begin{cases}\int_0^x \frac{Sg_n(\tilde{x},\sigma_n f(\tilde{x})-\sigma_n f(x)+t)}{\upsilon(\tilde{x})}\mathrm{d}\tilde{x}+S_*(t-\sigma_n f(x)), t\geq\sigma_n f(x),\\ \frac{1}{\sigma_n}\int_{n\Delta t}^{t} Sg_n\left(f^{-1}\left(\frac{\sigma_n f(x)-t+\tilde{t}}{\sigma_n}\right),\tilde{t}\right)\mathrm{d}\tilde{t}+S_*^*\left(f^{-1}\left(\frac{\sigma_n f(x)-t}{\sigma_n}\right)\right), t<\sigma_n f(x),\end{cases} \quad (23)$$

where

$$Fg_n(x,t)=$$
$$=-\beta_n W_{n-1}(x-f^{-1}(\Delta t),t)\mathrm{B}_{n-1}(x-f^{-1}(\Delta t),t-\Delta t)-$$
$$-\beta_2 W_{n-1}(x-f^{-1}(\Delta t),t)U_{n-1}(x-f^{-1}(\Delta t),t-\Delta t),$$

$$Sg_n(x,t)=$$
$$=\eta_1 W_{n-1}(x-f^{-1}(\tau),t)\mathrm{B}_{n-1}(x-f^{-1}(\tau),t-\Delta t)+$$
$$+H_{n-1}^*(x-f^{-1}(\tau),t)-k_1 S_{n-1}(x-f^{-1}(\tau),t-\Delta t).$$

Equations (16) and (19) are conventional differential equations with separated variables, their solutions taking into account respective initial conditions are as follows:

$$U_n(x,t)=$$
$$=\int_{(n-1)\Delta t}^{t} W_{n-1}(x-f^{-1}(\Delta t),s-\Delta t)\cdot U_{n-1}(x-f^{-1}(\Delta t),s-\Delta t)-$$
$$-\chi U_{n-1}(x-f^{-1}(\Delta t),s-\Delta t)+\gamma(x,t)H_{n-1}(x,s)\mathrm{d}s+$$
$$+U_*^*(x,(n-1)\Delta t), \quad (24)$$

$$M_n(x,t)=$$
$$=\int_{(n-1)\Delta t}^{t} \eta_2 W_{n-1}(x-f^{-1}(\Delta t),s-\Delta t)U_{n-1}(x-f^{-1}(\Delta t),s-\Delta t)+$$
$$+k_1 S_n(x-f^{-1}(\Delta t),s-\Delta t)-H_{n-1}^*(x-f^{-1}(\Delta t),s)\mathrm{d}s+$$
$$+M_*^*(x,(n-1)\Delta t), \quad (25)$$

where Δt is the time within which the basic characteristics of the medium change little. Its choice depends on pre-assigned accuracy.

Initial value of Δt is assigned intuitively and is finally determined in the process of solving the problem by numerical methods and ensures convergence according to the algorithm [21, 22].

5. Results of numerical calculation

Based on the obtained algorithms for solving the model problem, software in the Mathcad environment was developed for computer implementation and appropriate calculations. This takes into account the changes in the concentration of bacteria, matrix structures of biominerals, divalent iron, on the basis of which it is possible to predict the time of bioreactor operation between washings of contact load.

Numerical calculations were performed at $V=10$ m/h, $B^{**}=9.5$ g/m^3, $\mu_{max}=0.17$ h^{-1}, $K_F=0.7$ g/m^3, $\lambda=10^{-6}$ h^{-1}, $F_0^0(x)=1.6$ g/m^3, $F_*(t)=1.6$ g/m^3, $B_0^0(x)=0.001$ g/m^3, $B_*(t)=$ $=0.001$ g/m^3, $\beta_1=\beta_2=279$, $\eta_1=\eta_2=530$, $K_1=0.78$ h^{-1}, $K_2=$ $=0.1$ h^{-1}, $K_3=0.9$ h^{-1}, $U_0^0(x)=0.1$ g/m^3, $S_*(t)=0$ g/m^3, $S_0^0(x)=$ $=0$ g/m^3, $M_0^0(x)=10$ g/m^3, $\gamma=0.66$ h^{-1}, $M_*^*(x)=2.65$ g/m^3.

Fig. 1 shows the results of theoretical and experimental studies of distribution: *a* – Fe^{2+} ions, *b* – ferrobacteria; *c, d* – matrix structures in the interpore space. It also presents the distribution of the concentration of movable matrix (Fig. 2) by the depth of contact load at the averaged rate of 10 m/h for different intervals of time after washing.

The results of numerical calculations are represented by continuous lines. The given ranges of the studied parameters were obtained as a result of the conducted experimental research under production conditions on the setup, mounted in the premises of the filtration hall of the station of underground water treatment of the town of Berezne of Rivne oblast (Ukraine). The height of the setup was 3,500 mm, the diameter of the bioreactor section was 219 mm. The first layer (200 mm in thickness, dimensions of fractions of 6–12 mm) was intended for water distribution around the area of the bioreactor. Polystyrene foam with granules of 2–6 mm and the height of 1,100 mm was used as working load. A change in the kinetics of contamination by the height of the layers of contact load was studied using the system of samplers, uniformly located throughout the height of the load.

The physical-chemical parameters were determined according to the standard procedures at the certified hydrochemical laboratory of the Department of water supply, drainage and drilling business at the National University of Water and Environmental Management (Ukraine). The microbiological research was conducted at the bacteriological laboratory of SI «Rivne regional laboratory center of the Ministry of Health of Ukraine», accredited in the ISO 17025:2006 system.

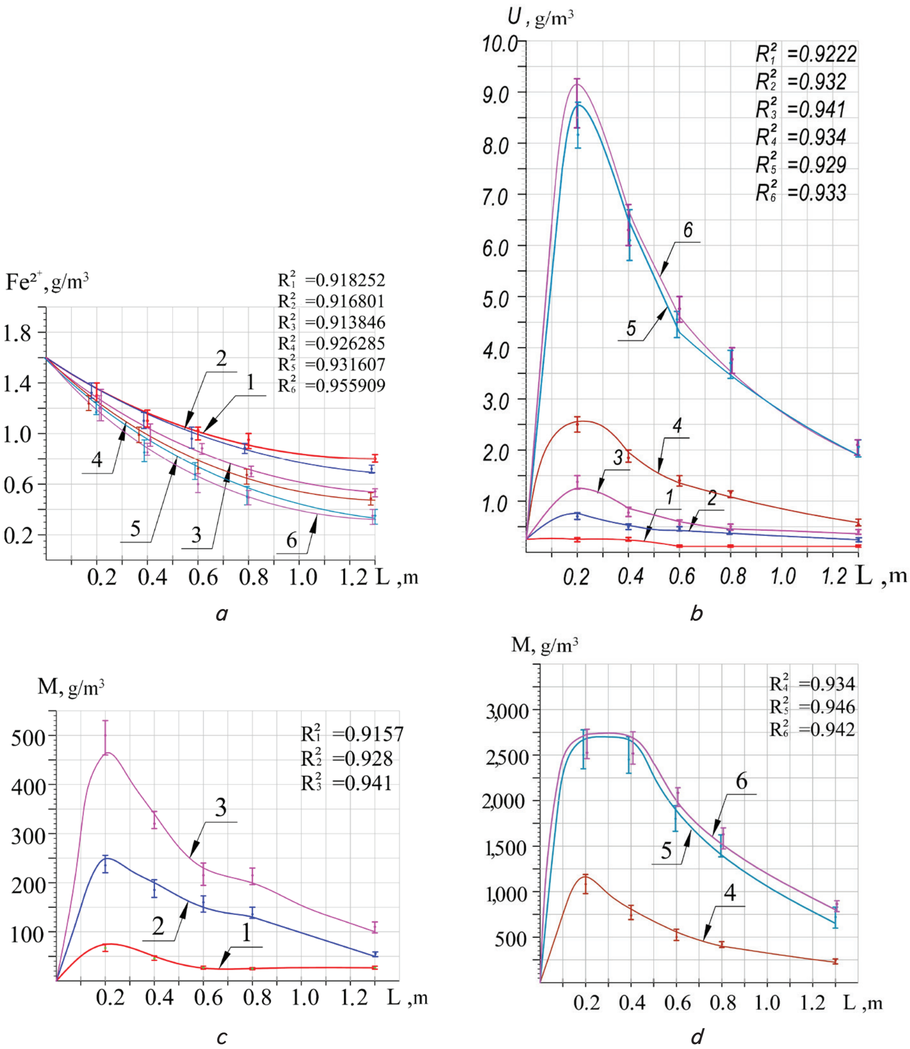

Fig. 1. Distribution of desired components by the depth of contact load, obtained as a result of theoretical and experimental studies within the time of bioreactor operation between washings: *a* — Fe^{2+} concentrations: 1 — 4 hours; 2 — 16 hours; 3 — 24 hours; 4 — 48 hours; 5 — 143 hours; 6 — 150 hours; *b* — ferrobacteria: 1 — 4 hours; 2 — 16 hours; 3 — 24 hours; 4 — 48 hours; 5 — 143 hours; 6 — 150 hours; *c, d* — matrix structures in interpore space: 1 — 4 hours; 2 — 16 hours; 3 — 24 hours; 3 — 24 hours; 4 — 48 hours; 5 — 143 hours; 6 — 150 hours

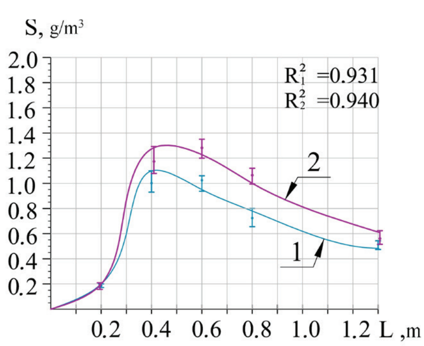

Fig. 2. Distribution of movable matrix by the depth of contact load within the time of bioreactor operation: 1 — 143 hours; 2 — 150 hours

Statistical treatment of data was carried out with the use of the rank determination coefficient. Deviation between the theoretical and experimental research accounted for not more than 6–9 %.

6. Discussion of results of studying water treatment within a contact load of bioreactor

Consideration of the results of theoretical and experimental studies (Fig. 1, *a*, *b*) reveals that as bacteria are gradually accumulated in the upper layers of contact load, the intensity of oxidation of iron compounds with the formation of matrix structures increases. Thus, a change in the concentration of Fe^{2+} depends on the concentration of bacteria. In particular, at the initial depth of contact load (0.2 m) it is possible to observe over time a gradual increase in the concentration of bacteria, which at the same time led to the gradual decrease in Fe^{2+} ions and accumulation of the respective amount of immovable matrix in the interpore space. The largest concentration of bacteria fixed on the matrix structures was noted in the depth range from 0.2 to 0.4 m, which corresponded to the occurrence of intensive processes for oxidation of Fe^{2+} compounds and the formation of «stationary» matrix structures. Over time, the concentration of active bacteria gradually decreased by the depth of contact load, which is explained by several factors. First, it is a decrease in the concentration of Fe^{2+} ions, which are known [16, 17] to be electron suppliers to the respiratory chain of *Gallionella* bacteria. Second, it is a decrease in concentration of dissolved oxygen and inorganic carbon required for the construction of cell biomass [16, 17].

This resulted into a gradual inhibition of the processes of biological oxidation of Fe^{2+} ions with the practical coming out onto the plateau at the depths of 0.8–1.3 m (Fig. 1, *a*). The concentration of active bacteria on these sections is several times lower in comparison with the upper layers (Fig. 1, *b*).

At the same time, during the whole period of bioreactor operation, there was gradual filling of its interpore space with matrix structures.

Moreover, the concentrations of matrix structures in each of the studied areas increased over time (Fig. 1, *c*). In addition, one should note the influence of the movable part of the matrix on the operation of bioreactors mostly during the hours which precede its washing (Fig. 2). In particular, it was traced in its gradual movement by the depth of contact load. The maximum magnitude of the movable matrix in the studied example was at the depth of 0.4 m, that is, at the depth, where we observe the accumulation by the «stationary» matrix of the concentration, which is approximately equal to the maximum possible contamination capacity (Fig. 1, *c*). When this contamination capacity is reached, the matrix is separated and subsequently transported to the lower layers.

It should be noted that the use of above-mentioned fractions of polystyrene foam makes it possible to increase the depth of accumulation of the matrix structures up to 60–80 cm with gradual filling the interpore space.

It is important to note that the research into the process of purification of groundwater from Fe^{2+} compounds takes into account the mutual influence of both fixed and non-fixed ferrobacteria, the matrix structures created by them, as well as a number of characteristics of the medium, in particular contamination capacity, flow rate, porosity, maximum specific rate of ferrobacteria growth, and the height of bioreactor load.

It was found that operation efficiency (from 8 to 10 days) is influenced by water quality parameters, in particular, the concentration of Fe^{2+} cations, magnitudes of hydrocarbonate alkalinity, pH, as well as the number of bacteria both in groundwater and in the interpore space of contact load. The influence of pulse (sudden) filtering rate, associated with a sharp change of hydraulic load, should be studied subsequently.

The considered bioreactor in technological circuits usually acts as the first degree in purification of groundwater. The filters with different types of inert loads (polystyrene foam, gravel sand) are used as the second degree for additional water treatment. The electrolysis plant «Polumia-2» is used for water disinfection. According to this technology, the first ground water treatment system with the capacity of 2,000 m^3/day was put in operation in 2010 at the Berezne water supply system of Rivne oblast. After putting the water treatment station in operation, the concentration of Fe_{con} in the water supply network of the town of Berezne was within the limits from <0.05 to 0.2 mg/dm^3.

7. Conclusions

1. The coefficients of maximum specific growth rate of microorganisms (μ_{max} 0.17–0.18 h^{-1}) and constant of saturation ($K_{S(Fe)}$ 0.65–0.7 g/m^3) were determined. The number of bacteria in aquifers under consideration (10^3–10^4 kl/cm^3), as well as concentrations of bacteria in contact load of a bioreactor, which changed during the whole period of its operations between washings from 10^5 kl/cm^3 to 10^9 kl/cm^3, was determined. The maximal contamination capacity was found, which in the accepted calculations amounted to 3–4 kg/m^3.

2. The multicomponent mathematical model of the process of biological purification of underground waters from iron compounds was developed. It was established that the largest increase in biomass was observed in the depth range from 0.2 to 0.4 m, which corresponded to the flow of intensive processes of Fe^{2+} oxidation and formation of «stationary» matrix structures. It was noted that over time, the concentration of active microorganisms gradually decreases by the length of the contact load, which was explained by a decrease in the concentration of Fe^{2+} ions, which are electron suppliers to the respiratory chain of *Gallionella* bacteria, dissolved oxygen and inorganic carbon needed for the formation of cell biomass. As a result, there was a gradual inhibition of the processes of oxidation of Fe^{2+} ions with the practical coming out to the plateau at depths of 0.8–1.3 m.

3. The algorithm for solving the model problem was constructed based on a combination of the method of characteristics and numerical methods.

4. As a result of computer realization of the model and conducted numerical experiments, we established the time of effective operation of a bioreactor between washings (up to 9 days), as well as the optimum height of contact load (1.2 m), distribution of concentrations of the components of the process in the middle of contact load of a bioreactor. Based on this, the main load was found to fall on its upper layers (0.2–0.6 m).

References

1. Zhurba, M. G., Govorova, Zh. M. (2008). Vodosnabzhenie. Uluchshenie kachestva vody. Vol. 2. Moscow: Izdatel'stvo ASV, 544.
2. Mouchet, P. (1995). Biological Filtration for Iron and Manganese Removal: Some Case Studies. WQTC 95 (AWWA) New Orleans LA, 12–16.
3. Kvartenko, A. N. (2016). Using biochemical methods in modern treatment technologies of underground water. Voda i vodoochysni tekhnolohiyi. Naukovo-tekhnichni visti, 2 (19), 51–65.
4. Scholl, M. A., Harvey, R. W. (1992). Laboratory investigations on the role of sediment surface and groundwater chemistry in transport of bacteria through a contaminated sandy aquifer. Environmental Science & Technology, 26 (7), 1410–1417. doi: https://doi.org/10.1021/es00031a020
5. Sharma, S. K., Petrusevski, B., Schippers, J. C. (2005). Biological iron removal from groundwater: a review. Journal of Water Supply: Research and Technology-Aqua, 54 (4), 239–247. doi: https://doi.org/10.2166/aqua.2005.0022
6. Van Beek, C. G. E. M., Dusseldorp, J., Joris, K., Huysman, K., Leijssen, H., Schoonenberg Kegel, F. et. al. (2015). Contributions of homogeneous, heterogeneous and biological iron(II) oxidation in aeration and rapid sand filtration (RSF) in field sites. Journal of Water Supply: Research and Technology-Aqua, 65 (3), 195–207. doi: https://doi.org/10.2166/aqua.2015.059
7. Vries, D., Bertelkamp, C., Schoonenberg Kegel, F., Hofs, B., Dusseldorp, J., Bruins, J. H. et. al. (2017). Iron and manganese removal: Recent advances in modelling treatment efficiency by rapid sand filtration. Water Research, 109, 35–45. doi: https://doi.org/10.1016/j.watres.2016.11.032
8. Oleynik, A. Ya., Semenko, G. I. (1997). Matematicheskoe modelirovanie protsessa udaleniya zheleza iz prirodnyh vod fil'trovaniem. Himiya i tehnologiya vody, 19 (5), 451–457.
9. Oliynyk, O. Ya., Sadchykov, O. O. (2013). Teoretychni doslidzhennia znezaliznennia vody na dvosharovykh filtrakh. Problemy vodopostachannia, vodovidvedennia ta hidravliky, 21, 14–22.

10. Poliakov, V. L., Martynov, S. Yu. (2017). Do teoriyi fizyko-khimichnoho znezaliznennia pidzemnykh vod ta yii informatsiinoho zabezpechennia. Chysta voda. Fundamentalni, praktychni ta promyslovi aspekty. Materialy V Mizhnarodnoi naukovo-praktychnoi konferentsiyi. Kyiv, 178–181.
11. Zevi, Y., Dewita, S., Aghasa, A., Dwinandha, D. (2018). Removal of Iron and Manganese from Natural Groundwater by Continuous Reactor Using Activated and Natural Mordenite Mineral Adsorption. IOP Conference Series: Earth and Environmental Science, 111, 012016. doi: https://doi.org/10.1088/1755-1315/111/1/012016
12. Vries, D., Bertelkamp, C., Schoonenberg Kegel, F., Hofs, B., Dusseldorp, J., Bruins, J. H. et. al. (2017). Iron and manganese removal: Recent advances in modelling treatment efficiency by rapid sand filtration. Water Research, 109, 35–45. doi: https://doi.org/10.1016/j.watres.2016.11.032
13. Sheng, Y., Kaley, B., Bibby, K., Grettenberger, C., Macalady, J. L., Wang, G., Burgos, W. D. (2017). Bioreactors for low-pH iron(II) oxidation remove considerable amounts of total iron. RSC Advances, 7 (57), 35962–35972. doi: https://doi.org/10.1039/c7ra03717a
14. Tekerlekopoulou, A. G., Vasiliadou, I. A., Vayenas, D. V. (2006). Physico-chemical and biological iron removal from potable water. Biochemical Engineering Journal, 31 (1), 74–83. doi: https://doi.org/10.1016/j.bej.2006.05.020
15. Chan, C. S., Fakra, S. C., Edwards, D. C., Emerson, D., Banfield, J. F. (2009). Iron oxyhydroxide mineralization on microbial extracellular polysaccharides. Geochimica et Cosmochimica Acta, 73 (13), 3807–3818. doi: https://doi.org/10.1016/j.gca.2009.02.036
16. Emerson, D., Field, E. K., Chertkov, O., Davenport, K. W., Goodwin, L., Munk, C. et. al. (2013). Comparative genomics of freshwater Fe-oxidizing bacteria: implications for physiology, ecology, and systematics. Frontiers in Microbiology, 4. doi: https://doi.org/10.3389/fmicb.2013.00254
17. Hallbeck, L., Pedersen, K. (1991). Autotrophic and mixotrophic growth of Gallionella ferruginea. Journal of General Microbiology, 137 (11), 2657–2661. doi: https://doi.org/10.1099/00221287-137-11-2657
18. Bukreeva, V. Yu., Grabovich, M. Yu., Eprintcev, A. T., Dubinina, G. A. (2009). Sorption of colloidal iron and manganese oxides by iron bacteria on the sand filter of water-lifting facilities. Sorbtsionnye i Khromatograficheskie Protsessy, 9 (4), 506–514.
19. Sakai, T., Miyazaki, Y., Murakami, A., Sakamoto, N., Ema, T., Hashimoto, H. et. al. (2010). Chemical modification of biogenous iron oxide to create an excellent enzyme scaffold. Org. Biomol. Chem., 8 (2), 336–338. doi: https://doi.org/10.1039/b919497e
20. Kvartenko, A., Prysiazhniuk, I. (2017). Modelling the kinetics of ferrum compouunds removal in a bioreactor. Technical sciences and technologies, 4 (10), 247–254. doi: https://doi.org/10.25140/2411-5363-2017-4(10)-247-254
21. Sivak, V. M., Bomba, A. Ya., Prysiazhniuk, I. M. (2005). Kompiuterne modeliuvannia protsesiv ochyshchennia stichnoi vody na karkasno-zasypnykh filtrakh. Visnyk NUVHP, 4 (32), 164–169.
22. Bomba, A. Ya., Baranovskyi, S. V., Prysiazhniuk, I. M. (2008). Neliniyni synhuliarno-zbureni zadachi typu «konvektsiya-dyfuziya». Rivne: NUVHP, 254.