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DEVELOPMENT OF MATHEMATICAL MODEL OF INFILTRATION OF IRON SULFATE ACID SOLUTION

The chemical industry is the leader, which is characterized by significant volumes of production and multiplicity of waste. The accumulation of a large amount of waste negatively affects the environment. The most obvious example is the state of the soils, where there are enterprises producing pigmentary titanium dioxide. The purpose of the work is to study the processes of penetration and accumulation of acidic solutions from chemical industry wastes into the soil and development of mathematical model of infiltration of iron sulfate acid solution. The study of the infiltration of solutions of sulfuric acid was carried out on the example of gray soil, since this type dominates in the area of the inkstone dump. Soil infiltration is estimated by the coefficient of filtration, which is determined by Darcy's law. The diffusion density was determined experimentally – $1.63 \cdot 10^{-10}$ kg/m^2, and the diffusion coefficient was calculated – $1.51 \cdot 10^{-8}$ m^2/s. On the basis of experimental research, an effective mathematical model for the propagation of acid solutions of iron sulfate in soil was developmented. By numerical modeling of the developed model, the acidity distribution in the 3D projection is obtained. At the initial stage, there is no water movement in dry soil. However, in the presence of slight convection, an increase in the acidity of the soil occurs. Soil oxidation occurs faster and its leveling is observed at different depths. Comparison of the results of numerical simulation and statistical data obtained experimentally shows the relationship between the hydrolytic acidity of the soil environment and the depth of impregnation of acid solutions of ferrous sulfate. It is established that between the values obtained by the experimental method and the results of the solution of the mathematical model, there is a close correlation connection. The correlation coefficient was 0.89.

Keywords: iron sulfate; sulfuric acid; water permeability; acidification; diffusion.

1. Problem statement.

The main ecological problem of today is the storage of industrial solid waste in open territories, resulting in environmental pollution. Since the real conditions of the functioning of natural processes in the natural environment are characterized by the influence of a complex set of negative factors, the assessment of their result should be based on the existing dynamic models of environmental feedback under the influence of certain factors [1].

The danger of storing iron sulfate is that it consists of free sulfuric acid, which is 24 %. Free sulfuric acid in this case is a pollutant of the fertile soil layer, which also makes it impossible to diagnose its accumulation in the lower layers of soil ecosystems, which ultimately causes a slow pollution of underground water basins which can be located at a rather insignificant depth from the soil surface (from 3 to 30 m).

In view of this definition of the level of danger of the place of storage of iron sulfate is to create a mathematical model of infiltration of its acid solutions to the soil environment.

2. Analysis of the recent researches and publications.

Waste from production and consumption are sources of anthropogenic pollution of the environment on a global scale and arise as the inevitable result of the consumer attitude and low resource utilization rate [2]. Getting into the environment, the components of the waste have a negative effect on the ecosystem. In addition, the waste composition is often not investigated or studied only by the main components. The biggest problem in the ecosystem is acid, alkaline and water soluble waste, taking into account the aggressiveness of the environment, high chemical activity and the ability to diffusion and infiltration processes. As a result of chemical interaction of pollutants it is possible and their transformation with the formation of new chemical compounds – xenobiotics, which are often more toxic than the source pollutants. The probability of such a transformation is greatly increased in the lithosphere and soil ecosystems [3].

The terrible case of environmental pollution is the large-scale waste with the acidic reaction of the production environment of pigment titanium dioxide by sulfate acid technology. In the worked out literary sources, no information was found on the direct influence of acid solutions of iron sulfate of different concentrations on soils [4]. Due to the dissolution of some masses of waste containing residual sulfuric acid, acid rain solutions whose pH can reach 1. In the periods of rain and snow falling into the lithosphere may occur acidic solutions can be made. It can be assumed that the effect of leakage on the soil ecosystem is predicted to be similar to the effects of acid rain that contain SO_3. Strong changes in the acidity of the soil may cause the acid, containing sulfur and nitrogen compounds, to enter on its surface. Hydrogen ions that enter the soil can be replaced by cations that are in the ground, resulting in either the removal of calcium, magnesium and potassium, or their deposition in dehydrated form. The mobility of toxic heavy metals, such as: manganese, copper, cadmium, is increasing. The solubility of heavy metals strongly depends on pH. Dissolved by these heavy metals, which are easily digested by plants, is a poison for plants and can lead to death [4]. Transferring soluble pollutants in the environment depends on a large number of environmental factors and processes occurring in it. The degree of influence of one or another process may be different for different environments, pollutants, considered moments of time and space [5, 6].

Despite the large amount of accumulated waste containing residual sulfuric acid, today there are virtually no methods for assessing environmental pollution and modeling impregnation processes. Unfortunately, there is no information on the relationship between sour solutions of iron sulfate and ecosystems [7]. Studies of the infiltration of substances into the soil and the development of mathematical models of infiltration are solved by scientists [8], but for managing catchment basins and forecasting floods, without taking into account the penetration of toxic substances. Also scientists are currently studying the hydrodynamic models of infiltration, which are characterized by the movement of fluid in a hollow environment by the theory of filtration [9] and the harm of acid solutions of iron sulfate for living organisms. Unidentified mechanisms of impregnation of acid solutions of iron sulfate in soil ecosystems remained.

3. Statement of the problem and its solution.

Taking into account the above and analyzing the current situation, accumulation of waste and penetration of its acidic solutions to the soil, is an urgent problem. The purpose of the work is to study the processes of penetration and accumulation of acidic solutions from chemical industry wastes into the soil and development of a mathematical model of infiltration of iron sulfate acid solution. Therefore, in this paper we solve such problems:

– finding the density and coefficient of soil filtration;

– finding the coefficient and diffusion density;

– development of mathematical model of infiltration of iron sulfate acid solution.

The distribution of acidic solutions of iron cumulus penetrating into the depth of soils depends on many factors, first of all, on climatic conditions of soil layout, physical and chemical properties of soils, nature and sources of their input into ecosystems, chemical and granulometric composition of breeding rocks [4, 5]. Infiltration of acid solutions takes place due to rain fall and melting of snow cover. The process of infiltration occurs periodically, depending on favorable meteorological conditions at certain intervals of time. This process occurs to a certain depth, because the flow of water from the surface is absorbed in its path by particles of soil ecosystems or falls into the flow of groundwater, which determines the depth of its penetration.

The intensity of rainfall is the main factor in the formation of moisture in the environment that transports pollutants. After rainfall rain falls, water is quickly absorbed by the medium, and then the moisture flow stabilizes. This initial stage of rapid penetration of water to an unsaturated moist environment is called absorption or infiltration. Further, due to the saturation of the entire threshold environment of the medium, the moisture flow is stabilized. There comes the stage of motion of water in a saturated environment – filtration.

3.1. Materials and methods of investigation of sulfuric acid solution infiltration.

Investigation of sulfuric acid solution infiltration was carried out on the example of gray soil, since this type dominates in the area of the dump of iron sulfate. In the study of the penetration of acid solutions to the soil, the dependence of the coefficient of filtration on the density of the soil was established: with increasing density (with depth) the coefficient of filtration decreased. On the basis of experimental studies, the following factors, such as the diffusion coefficient ($1.51 \cdot 10^{-8}$ m^2/s) and the density of diffusion flux ($1.63 \cdot 10^{-10}$ kg/m^2), were calculated. The conducted study of the phenomenon of soil infiltration and its basic characteristics allows to construct an effective model of accumulation and distribution of acid solutions of iron sulfate at different depths to the soil environment.

For scientific substantiation and adequate numerical study of patterns of distribution in soil ecosystems of soluble substances it is expedient to use methods of mathematical modeling. To determine the dependency type when constructing a mathematical model for the distribution of sulfuric acid content in the isolated elementary phase of the landscape, the Wolfram Matematika 8.0.0 program was used. With a wide range of spatial-temporal scales, hydrological and climatic factors, these methods do not lose their credibility.

The most promising way to solve this problem is to:

– conducting mathematical calculations;

– construction of an effective mathematical model for the accumulation of acid solutions of iron sulfite at different depths of the soil to the parent rock;

– filtration processes in the soil environment, which establish the functional dependence of acidification in the soil on spatial coordinates and time.

3.2. Development of mathematical model of infiltration of iron sulfate acid solution

The concept of concentration of solutions of iron sulfate was based on the mathematical model of accumulation and distribution of acid solutions of iron sulfate at different depths to the soil environment. Under the concept of dissolved substance per unit volume of solution. In this case, water containing a certain amount of acid solutions of iron sulfate fills the empty medium, with the concentration in it can vary both in length and in time. In this case, the set of concentrations over time has the form of a concentration field. The analytical field of concentration is described by the functional dependence (1):

$$C = f(x, y, z, t), \qquad (1)$$

where C – concentration of acid solutions of iron sulfate (acidity, mg equiv); x, y, z – spatial coordinates; t – time, s.

From function (1) it is observed that the analytic field of concentration has the form of non-stationary one. A stationary analytic field of concentration is only a coordinate function and has the form (2):

$$C = f(x, y, z),\ \text{or}\ \frac{\partial C}{\partial t} = 0. \quad (2)$$

On a plane, the distribution of concentration has the form of isoconcentration line. The change in concentration during the transition from one isoconcentration surface to another is characterized by a derivative of the normal to the surfaces, and the vector directed towards the growth of concentration serves as a concentration gradient, and numerically, it is equal to the module of the value of the function (2). Then the diffusion flow can be considered as the rate of passage of matter. That is, there is a certain amount of matter passing through the isoconcentration surface per unit time. If we deduce the diffusion flow to unit area of the isoconcentration surface, then we obtain the density of the diffusion stream. Analytically it looks like (3):

$$q = \frac{dG}{Fdt}, \quad (3)$$

where G – the weight of the diffused substance, kg; F – area of isoconcentration surface, m^2; t – time, s.

In accordance with Fick's law, the flow of matter is described by the relation (4):

$$q = -D\frac{dC}{dn}, \quad (4)$$

where n – the derivative of the normal (the thickness of the soil), m; D – diffusion coefficient, m^2/s; C – concentration (water acidity of the soil).

The "–" sign indicates that the flow is directed toward a decrease in concentration. The density of the diffusion stream is directly proportional to the gradient of concentration.

Soils can be neutral to diffusion. The object of the study should be an inhomogeneous medium consisting of at least two components (an empty medium and a liquid that fills it). Each component takes an active part in the diffusion process. If the medium is statistically homogeneous, then the mathematical model of the displacement of acid solutions of iron sulfate is described by the following equation (5):

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - \vartheta\frac{\partial C}{\partial x}, \quad (5)$$

where – speed, m/s; x – vertical coordinate; D – diffusion coefficient, m/s^2.

According to equation (5), the diffusion coefficient and the velocity of motion are empirical in nature and their values are ambiguous in the account of a number of transfer characteristics in the sorption environment.

To solve equation (5) it is necessary to supplement it with the boundary and initial conditions, in the case when the field is unsteady and boundary, if it is stationary. Boundary conditions reflect the situation on the border where infiltration occurs, and the initial – in the middle of the area before the course of the infiltration process. In the initial concentration state of the medium the whole of its previous concentration situation is reflected and for the subsequent concentration changes no matter how the given concentration field was formed. Without convective member of equation (5) will have the following form (6):

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}. \quad (6)$$

According to the obtained laboratory data, the initial acidity of Ng entering the soil environment is c_1 = 1.68 mg-eq. The diffusion coefficient D = $1.51\cdot10^{-8}$ m^2/s, and the initial concentration in the soil is taken with c_0 = 0. Using Mathematica 8, the solution of equation (5) is obtained without a convective term:

$$C(x,t) = \frac{1{,}68x}{2\sqrt{\pi D}}\int_0^t \frac{e^{-\frac{x^2}{4D(t-\tau)}}}{(t-\tau)^{\frac{3}{2}}}dt = \quad (7)$$
$$= 0{,}4326\left(3{,}883 - 3{,}883\cdot E\cdot r\cdot f\cdot\left[\frac{0{,}4564x}{\sqrt{t}}\right] + 0{,}1326\right).$$

The developed mathematical dependencies allow us to proceed directly to the mathematical modeling of the processes of penetration of iron sulfate into the soil environment.

4. Results and Discussion.

The results of the solving equations (7) obtained using the Mathematica 8 package are presented in figures 1 and 2.

The solution of equation (5) is considered, taking into account the convective term. The boundary conditions have been set that were. Then the solution of the equation will have the following form:

$$C(x,t) = \int_0^\infty c_0 G(x,\varepsilon,t)d\varepsilon + D\int_0^t c^1\Delta(x,t-\tau)d\tau, \quad (8)$$

where $G(x,\varepsilon,t)$ – Green's function:

$$G(x,\varepsilon,t) = \frac{1}{2\sqrt{\pi Dt}}\exp\left[\frac{\vartheta(\varepsilon - x)}{2D} - \frac{\vartheta^2 t}{4D}\right]\times$$
$$\times\left[\exp\left(-\frac{(x-\varepsilon)^2}{4Dt}\right) - \exp\left(-\frac{(x+\varepsilon)^2}{4Dt}\right)\right],$$

and

$$\Delta(x,t) = D\int_0^t c^1\Delta(x,t-\tau)d\tau. \quad (9)$$

The results of the research on the solution of the equation (9) obtained in Mathematica 8 are presented in figures 3 and 4.

At the initial stage, in the dry soil there is practically no movement of moisture. It makes sense to use equation (6). However, in the presence of insignificant convection there is an increase in the acidity of the soil. The hardening of the soil occurs faster and there is a leveling at different depths.

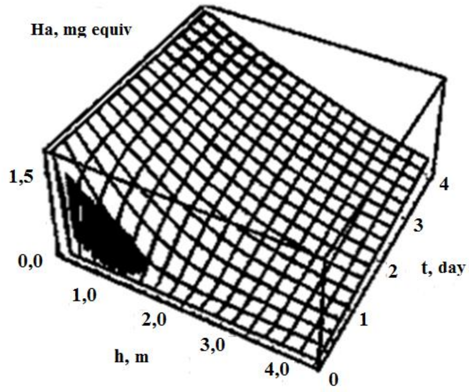

Figure 1 – Distribution of hydrolytic acidity in 3D projection

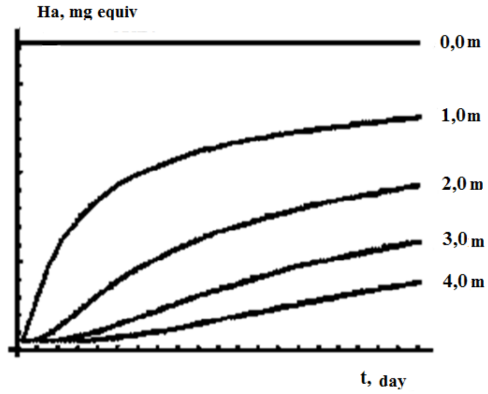

Figure 2 – Distribution of hydrolytic acidity deep into the soil

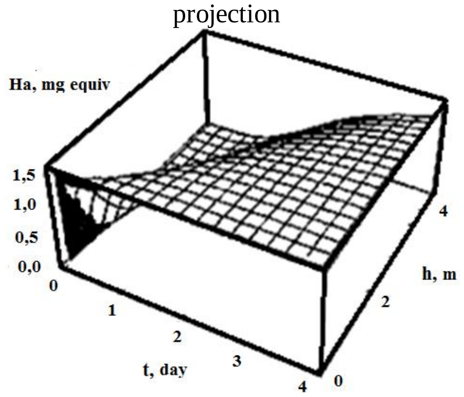

Figure 3 – Distribution of acidity in 3D projection

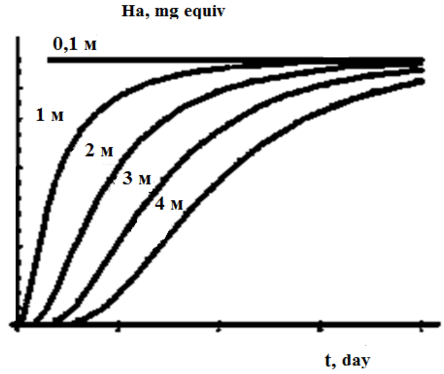

Figure 4 – Distribution of acidity deep into the soil

In the process of studying the accumulation and impregnation of acid solutions of iron sulfate on the model of soil samples, a connection was established between the values obtained by the experimental method and the results of solving the mathematical model (figure 5).

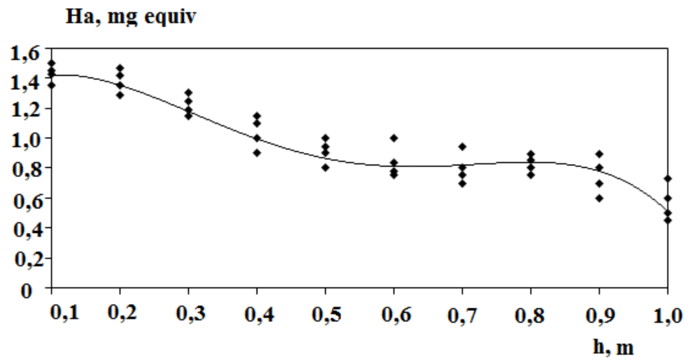

Figure 5 – The dependence of the distribution of acidity on the depth of soil

On the basis of the correlation analysis, a relationship was established between the hydrolytic acidity of the soil medium and the depth of impregnation of acid solutions of iron sulfate. The value of the received connection is characterized by the correlation coefficient, calculated on the basis of the experimental data obtained from the statistical sample of the values of hydrolytic acidity and the corresponding depth of soil.

The performed regression-correlation analysis confirms that there was a close correlation between the distribution of the content of acid solutions of iron sulfate on the profile of the soil, according to the calculation of the mathematical model and experimental analysis. The correlation coefficient was 0.89.

Conclusions.

Waste from production and consumption are sources of anthropogenic pollution of the environment on a global scale and arise as an inevitable result of a consumer attitude and a low utilization rate. Getting into the environment, the components of the waste have a negative effect on the ecosystem. In addition, the waste composition is often not investigated or studied only by the main components. The biggest problem in the ecosystem is acid, alkaline and water soluble waste, taking into account the aggressiveness of the environment, high chemical activity and the ability to diffusion and infiltration processes. As a result of chemical interaction of pollutants it is possible and their transformation with the formation of new chemical compounds – xenobiotics, which are often more toxic than the source pollutants. The probability of such a transformation is greatly increased in the lithosphere and soil ecosystems.

Creation of a mathematical model of the real process of penetration of acid solutions of iron sulfate in the full sense of this concept is a good way to mathematically describe the real process of propagation and accumulation of hydrolytic acidity in the soil environment. As a result of the research carried out:

1. A mathematical model of penetration into the soil of acidic solutions of iron sulfate, which can be formed at dissolution of some mass of iron sulfate of titanium production, is developed:

– when constructing a mathematical model, hydrodynamic models of geophylation were used and the main provisions of the theory of filtration;

– the mathematical model allows to calculate the hydrolytic acidity that characterizes the acidity of the soil.

2. According to the results of mathematical modeling, it is established that in the presence of convection in the soil, it is faster acidification than in its absence.

3. It is established that between the values obtained by the experimental method and the results of the solution of the mathematical model, there is a close correlation connection of 0.89.

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РОЗРОБКА МАТЕМАТИЧНОЇ МОДЕЛІ ІНФІЛЬТРАЦІЇ КИСЛОГО РОЗЧИНУ СУЛЬФАТУ ЗАЛІЗА

Хімічна промисловість є лідером промисловості, яка характеризується значними обсягами виробництва і багатонажністю відходів. Накопичення великої кількості відходів негативно позначається на довкіллі. Найбільш очевидним прикладом є стан ґрунтів, де розташовані підприємства, що виробляють пігментний двоокис титану. Метою роботи є вивчення процесів проникнення і накопичення кислих розчинів з відходів хімічної промисловості в ґрунт і розробка математичної моделі інфільтрації розчину кислого лужного розчину. Вивчення інфільтрації розчинів сірчаної кислоти проводилося на прикладі сірої ґрунту, оскільки цей тип домінує в області скидання чорнильних каменів. Інфільтрація ґрунту оцінюється коефіцієнтом фільтрації, який визначається законом Дарсі. Щільність дифузії визначалася експериментально – $1,63 \cdot 10^{-10}$ кг/м^2, а коефіцієнт дифузії був розрахований – $1,51 \cdot 10^{-8}$ м^2/с. На основі експериментальних досліджень була розроблена математична модель поширення кислих розчинів сульфату заліза в ґрунті. При чисельному моделюванні із використанням розробленої моделі отримано розподіл кислотності в тривимірній проекції. На початковому етапі прийнято, що рух води в сухому ґрунті відсутній. Однак при невеликій конвекції відбувається підвищення кислотності ґрунту. Окислення ґрунтів відбувається швидше, і його вирівнювання спостерігається на різних глибинах. Порівняння результатів чисельного моделювання і статистичних даних, отриманих на експериментальній основі, показує взаємозв'язок між гідролізної кислотою ґрунтового середовища і глибиною просочення кислотних розчинів сульфату заліза. Встановлено, що між значеннями, отриманими експериментальним методом, і результатами чисельного моделювання з застосуванням розробленої математичної моделі, існує тісний кореляційний зв'язок. Коефіцієнт кореляції склав 0,89.

Ключові слова: залізний купорос; сірчана кислота; водопроникність; підкислення; дифузія.

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РАЗРАБОТКА МАТЕМАТИЧЕСКОЙ МОДЕЛИ ИНФИЛЬТРАЦИИ КИСЛОГО РАСТВОРА СУЛЬФАТА ЖЕЛЕЗА

Химическая промышленность является лидером промышленности, которая характеризуется значительными объемами производства и многотонажностью отходов. Накопление большого количества отходов отрицательно сказывается на окружающей среде. Наиболее очевидным примером является состояние почв, где расположены предприятия, производящие пигментный диоксид титана. Целью работы является изучение процессов инфильтрации и накопления кислых растворов из отходов химической промышленности в почву и разработка математической модели инфильтрации раствора кислого щелочного раствора. Изучение инфильтрации растворов серной кислоты проводилось на примере серой почвы, поскольку этот тип доминирует в области сброса медного купороса. Инфильтрация веществ в почву оценивается коэффициентом фильтрации, который определяется законом Дарси. Плотность диффузии определялась экспериментально – $1{,}63 \cdot 10^{-10}$ кг/м^2, а коэффициент диффузии был рассчитан – $1{,}51 \cdot 10^{-8}$ м^2/с. На основе экспериментальных исследований была разработана математическая модель распространения кислых растворов сульфата железа в почве. При численном моделировании с использованием разработанной модели получено распределение кислотности в трехмерной проекции. На начальном этапе принято, что движение воды в сухой почве отсутствует. Однако при небольшой конвекции происходит повышение кислотности почвы. Окисление почв происходит быстрее, и его выравнивание наблюдается на разных глубинах. Сравнение результатов численного моделирования и статистических данных, полученных на экспериментальной основе, показывает взаимосвязь между гидролизной кислотой почвенной среды и глубиной пропитки кислотных растворов сульфата железа. Установлено, что между значениями, полученными экспериментальным методом, и результатами численного моделирования с применением разработанной математической модели, существует тесная корреляционная связь. Коэффициент корреляции составил 0,89.

Ключевые слова: железный купорос; серная кислота; водопроницаемость; подкисление; диффузия.