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PREDICTING THE DURABILITY OF CONCRETE STRUCTURES WITH REGARD TO MASS TRANSFER AND PORE COLMATATIONS DURING CORROSION

Abstract. *The object of research is the corrosion process of destruction of concrete and reinforced concrete structures in aggressive media. The purpose of the investigation is to establish a set of relationships and ratio between the characteristics of this process and various external and internal factors that affect the growth of corrosion. To solve the tasks set on the basis of the obtained experimental data, the method of mathematical modeling of corrosion processes was applied. The theories of physical and chemical transformations and heat and mass transfer laws suggest that different stages of corrosion can be simulated by differential equations of interrelated heat and mass transfer with boundary conditions. They are defined by arbitrary functions of initial transfer potentials and nonlinear boundary conditions of all known types. The possibilities of developed physical and mathematical models of mass transfer in the processes of studying corrosion of various types are demonstrated. They allow to evaluate the concentration of the transported component throughout the thickness of the concrete structure and in the biofilm itself at any time, as well as the concentration of "free" calcium hydroxide in the liquid phase. As a result, it is succeeded to predict the durability and reliability of concrete structures with a minimum error. The proposed author's model of the destruction of cement concretes, taking into account the colmatation of pores and capillaries, ensures the adequacy of judgments about the kinetics of mass transfer processes during corrosion of the II type.*

Keywords: *corrosion, mathematical model, porous and non-porous materials, colmatation.*

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ПРОГНОЗИРОВАНИЕ ДОЛГОВЕЧНОСТИ БЕТОННЫХ КОНСТРУКЦИЙ С УЧЕТОМ МАССОПЕРЕНОСА И КОЛЬМАТАЦИИ ПОР ПРИ КОРРОЗИИ

Аннотация. *Объектом исследования является коррозионный процесс разрушения бетонных и железобетонных конструкций в агрессивных средах. Цель исследования – установление совокупности соотношений и взаимосвязи характеристик указанного процесса с различными внешними и внутренними факторами, влияющими на развитие коррозии. Для решения поставленных задач на основании полученных экспериментальных данных применялся метод математического моделирования коррозионных процессов. С позиции теорий физико-химических превращений и тепломассообменных закономерностей, различные стадии коррозии могут моделироваться дифференциальными уравнениями взаимосвязанного тепломассопереноса с краевыми условиями. Их характеризуют произвольные функции начальных потенциалов переноса и нелинейные граничные условия всех известных типов. Продемонстрированы возможности разработанных физико-математических моделей* *массопереноса в процессах изучения коррозии различных видов. Они позволяют оценить концентрации переносимого компонента по толщине бетонной конструкции и в самой биопленке в любой момент времени, а также концентрации «свободного» гидроксида кальция в жидкой фазе. В результате удается прогнозировать долговечность и надежность бетонных конструкций с минимальной погрешностью. Предлагаемая авторская модель разрушения цементных бетонов с учетом кольматации пор и капилляров обеспечивает адекватность суждений о кинетике массообменных процессов при коррозии второго вида.*

Ключевые слова: *коррозия, математическая модель, пористые и непористые материалы, кольматация.*

Introduction.

The concept of "corrosion" came from the depths of time and by the 20th century was finally defined as "the process of spontaneous destruction of metals and alloys under the influence of aggressive environmental factors" [1, 2]. Historically, the term came into use with the wide spread of metals and their use for peaceful purposes (kitchenware, jewelry) and military purposes (weapons).

The acquisition and comprehension of the results of worldwide studies on metal corrosion helped the scientists identify the causes and driving forces behind this destructive phenomenon. In addition, various classification attributes of corrosion processes have been defined [3]:

– according to the type of aggressive media: gas; atmospheric; underground; corrosion in non-electrolytes (in liquid media); corrosion in electrolytes; biological corrosion; corrosion by stray currents;

– by the conditions of the corrosion process: contact corrosion; crevice corrosion; corrosion by incomplete immersion; corrosion at full immersion; friction corrosion; intergranular corrosion; stress corrosion;

– by the nature of the destruction: solid corrosion; uniform corrosion; irregular corrosion; selective, local (spots, ulcers), pitting, through; intergranular (delaminating – in deformed blanks, stabbing - in welded joints).

The mechanisms of corrosion processes course and development are theoretically possible: in non-porous materials; in porous materials.

In practice, corrosion failure of materials occurs everywhere (figure 1).

Figure 1 – Corrosion destruction of building structures.

The substance (solid phase) is a dense homogeneous non-porous medium. Its typical representatives are metals. It is assumed that the metal structure is free of cracks and ulcers and that the metal product itself is in a continuous medium (gas or liquid). At the initial moment of time ($\tau = 0$), in this environment, a component A appears. It is aggressive in relation to the metal. Let us denote the Concentration of this component as C_A^*. In general, the numerical value of this quantity is a function of time (figure 2). Free-convective flows of a continuous medium always exist near the surface of a solid body [4, 5]. This creates a boundary layer, the magnitude of which is represented by Δ in figure 2.

There is an interaction [6] at the interfacial boundary (1):

$$4Fe + 3O_2 = 2Fe_2O_3 \quad (1)$$

The primary task of mathematical analysis of corrosion processes is to take into account the kinetic features and dynamics of mass transfer of an aggressive component through a boundary layer of a continuous medium. As the thickness of the reaction product (rust) layer increases, the influence of diffusion of the aggressive reagent through it becomes increasingly important. This process is shown schematically in figure 2, *a* and *b*.

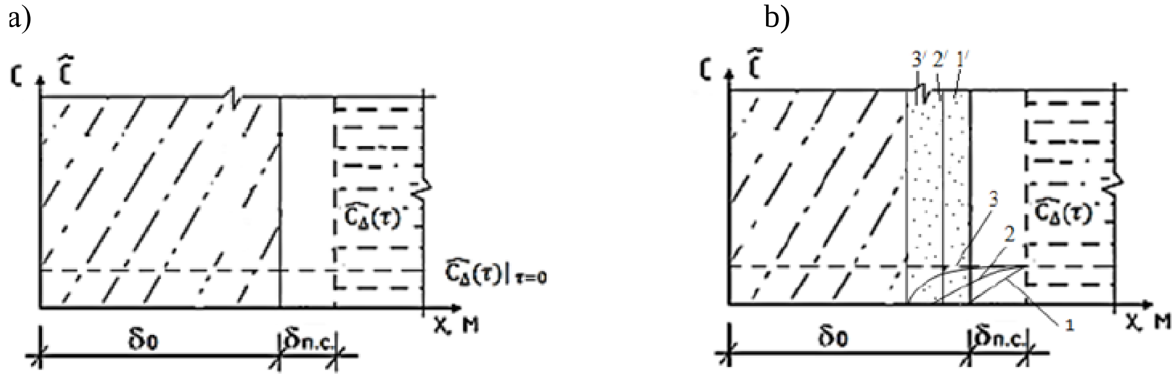

Figure 2 - Corrosion processes in a non-porous substance:
1' - a chemical reaction begins to take place on the surface; 1', 2' - corrosion product areas at time τ_1;
2', 3' - corrosion product areas at time τ_2; 1-3 - concentration curves.

The issues of corrosion destruction of concretes were considered in details by Prof. V.M. Moskvin and co-authors, as shown in [6]. As a result of the research, the corrosion processes occurring in concretes were divided into three main types: corrosion of the washoutcorrosion (I type); carbon dioxide, salt corrosion (II type); sulfate corrosion, etc. (III type).

Taking into account how broad the classification is, it is advisable to use methods of mathematical modeling for a comprehensive study of corrosion processes taking place. In our opinion, they can be used at each stage of changing the state of the environment leading to material failure.

The theoretical basis for mathematical modeling of the kinetic regularities of chemical reactions proceeding during corrosion damage are the fundamental physical and chemical laws.

Various methods of mathematical modeling based on mass transfer equations are used to describe the nature of corrosion. That said:

– simple mathematical approxi-mations of experimental results.

– complex models are represented as elementary processes, each of which is described by differential equations.

An essential contribution to development of theory and construction of mathematical models of processes of corrosion was made by Corresponding Member of the Russian Academy of Sciences B.V. Gusev and his colleagues [6].

The most well-known approach is the mathematical modeling of corrosion damage by representing the process investigated as a complex of separate stages occurring in space and time, taking into ac-count the kinetics of chemical reactions, mass transfer processes and phase transfers. The rate of each stage is expressed by phenomenological equations. Modeling based on these equations allows simple calculations of the required coefficients.

The application of the theory of mathematical modeling in solving such problems makes it possible to obtain dimensionless complexes that most accurately express the chemical and physical essence of the phenomena under study. In this case, the corrosion process is considered to be quasi-stationary, i.e. along the coordinate and time intervals, the process is considered as an equilibrium one [7].

Any mathematical model must satisfy the principle of universality and adequacy, since only in this case it can be applied to solve practical problems. A universal mathematical model should be based on generalized equations describing processes in the most general case.

In this regard, the aim of the study is to establish a set of relations linking the properties of the corrosion process with various external and internal factors affecting the development of corrosion. To achieve this goal, it is necessary to solve the following tasks:

– create mathematical models of various types of corrosion damage in order to compare and determine the parameters of mass transfer to predict the durability of materials in aggressive opeaing environments;

– determine the time to reach the critical value of $Ca(OH)_2$ concentration, at which the decomposi-tion of highly basic compounds will begin;

– to propose an engineering method for predicting corrosion damage in various industries.

Methods.

In this study, we used a method of creating a mathematical model of corrosion mass transfer based on a system of differential equations including the molecular diffusion equation, which is supplemented by initial and boundary conditions. When using the zone calculation method, the entire process is divided into N elementary microprocesses. Each of them implies constant mass conductivity factors. Thus, the nonlinear mass transfer problem is reduced to a set of N linear problems. The mass conductivity factors and external mass yield were calculated based on the obtained data. The solution of the system made it possible to complete the picture of the substance distribution over time and to present an analysis of the kinetics and dynamics of the processes. In order to verify the described mathematical models of various corrosion processes, a number of experimental studies were carried out using the following method. The objects of corrosion resistance study were cubes with a facet of 0.03 m, made of Portland cement of the CEM I 42.5 N class with a water-cement ratio W/C = 0.3. The system under study was composed of 1x3x3 cm plates tightly fitted to each other (figure 3).

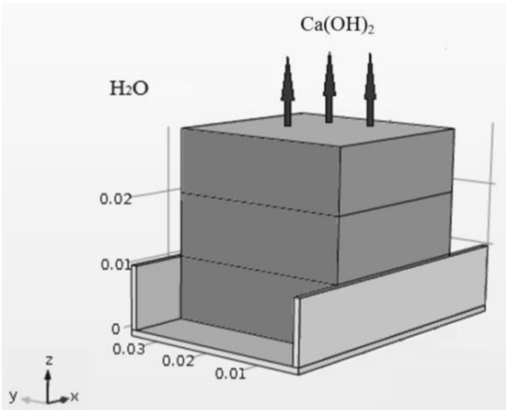

Figure 3 – Experimental sample for corrosion resistance.

After 28-day preliminary hardening, the samples were immersed in an aqueous medium. Five surfaces of the system under study were isolated; only one surface remained open for interaction with an aggressive environment. After certain time intervals, the samples were taken out of the container and the content of Ca^{2+} ions was determined in each of the plate elements, as a result, the concentration distribution curves along the coordinate were plotted.

Results and Discussion.

As an example of corrosion in porous material, consider the case of biodegradation of concrete structures in liquid media. In general, biocorrosion refers to the violation of the functional properties of materials and structures by natural objects [7, 8]. One of the most common types of biocorrosion is microbiological corrosion resulting from contact of concrete with sea and fresh water. Fungal corrosion has the most aggressive and destructive effect on materials.

According to other researchers [9-12], vertical surfaces in areas with low fluid pressures (in areas of flow failure) experience the greatest accumulation of biomass. Based on this, in mathematical modeling, it was assumed that in the considered system "concrete – liquid", accumulation of biomass will occur at the interface, i.e., on the right border.

The model of mass transfer in an unbounded two-layer plate can be represented as a system of differential equations of parabolic type in partial derivatives with boundary conditions of the second (concrete – liquid boundary) and the fourth kind (concrete – biofilm boundary) [9] (2), (3):

$$\frac{\partial C_1(x,\tau)}{\partial \tau} = k_1 \cdot \frac{\partial^2 C_1(x,\tau)}{\partial x^2}, \quad \tau > 0, \ -\delta_1 \le x \le 0, \tag{2}$$

$$\frac{\partial C_2(x,\tau)}{\partial \tau} = k_2 \cdot \frac{\partial^2 C_2(x,\tau)}{\partial x^2}, \quad \tau > 0, \ 0 \le x \le \delta_2, \tag{3}$$

Here, $C_1(x,\tau) -$ is the concentration of "free" calcium hydroxide converted to CaO in concrete at time τ at an arbitrary point with coordinate x (kg CaO · kg^{-1} concrete); $C_2(x,\tau) -$ concentration of "free" calcium hydroxide converted to CaO in biofilm at time τ at anarbitrary point with coordinate x (kg CaO · kg^{-1} biomass); k_1 – is the mass conductivity coefficient in the solid phase, m^2·s^{-1}; k_2 – coefficient of mass conductivity in biofilm, m^2·s^{-1}; δ_1 — thickness of the concrete structure, m; δ_2 – biofilm thickness, m.

Initial conditions (4), (5):

$$C_1(x,\tau)\big|_{\tau=0} = C_1(x,0) = C_{1,0}. \tag{4}$$

$$C_2(x,\tau)\big|_{\tau=0} = C_2(x,0) = C_{2,0}. \tag{5}$$

Boundary conditions. On the left border (6):

$$\left.\frac{\partial C_1(x,\tau)}{\partial x}\right|_{x=-\delta_1} = 0. \tag{6}$$

At the point of contact between the concrete and the biofilm (7), (8):

$$C_1(x,\tau)\big|_{x=0} = m \cdot C_2(x,\tau)\big|_{x=0}, \tag{7}$$

$$-\rho_{бет} \cdot k_1 \cdot \left.\frac{\partial C_1(x,\tau)}{\partial x}\right|_{x=0} = -\rho_{биом} \cdot k_2 \cdot \left.\frac{\partial C_2(x,\tau)}{\partial x}\right|_{x=0}. \tag{8}$$

On the right border (9):

$$-k_2 \cdot \left.\frac{\partial C_2(x,\tau)}{\partial x}\right|_{x=\frac{\delta_2}{\delta_1}} = q_н(\tau). \tag{9}$$

Here, $C_{1,0} -$ is the initial concentration of "free" CaO, kg CaO · kg^{-1} of concrete; $C_{2,0} -$ is the initial concentration of "free" CaO, kg CaO · kg^{-1} biomass; m – is Henry's equilibrium constant (kg biofilm · kg^{-1} concrete); $\rho_{бет}, \rho_{биом} -$ are densities of concrete and biomass, kg·m^{-3}; $q_н(\tau) -$ is the mass flux density leaving the biofilm into the liquid stream.

Then the original system of equations was mapped into the domain of complex numbers, where the solution was obtained. Then we translated the solution into the original domain.

The general solution of the mass-conductivity problem is Eqs. (10), (11), (12):

$$Z_1(\bar{x}, Fo_m) = \frac{1}{1+NK_k K_\delta}\left\{1 - NK_\delta + NKi_H^*\left[Fo_m + \frac{(1-\bar{x})^2}{2} + \varphi(K_k, N, K_\delta)\right]\right\} +$$
$$+2\sum_{n=1}^{\infty}\frac{1}{\mu_n^2 \psi_1^{/}(\mu_n)}(\mu_n \sin\mu_n \left[\cos(\mu_n \bar{x})\cos(\mu_n\sqrt{K_k}K_\delta) - \sqrt{K_k}K_\delta \sin(\mu_n \bar{x})\sin(\mu_n\sqrt{K_k}K_\delta)\right] - \tag{10}$$
$$-\frac{N}{\sqrt{K_k}}\cos(\mu_n(1+\bar{x}))\exp(-\mu_n^2 Fo_m).$$

$$Z_2(\bar{x}, Fo_m) = \frac{1}{1+NK_kK_\delta}(1 - NK_\delta + Ki_H^*\left[\bar{x} - Fo_m K_k K_\delta\right] + NKi_H^*(\varphi(K_k, N, K_\delta) -$$

$$-\frac{1+K_k\bar{x}^2}{2}) - 2\sum_{m=1}^{\infty}\frac{J}{\mu_m^2\psi_1^{/}(\mu_m)}(\mu_m \sin\mu_m \cos\left[\mu_m\sqrt{K_k}(K_\delta - \bar{x})\right] -$$

$$-\frac{\mu_m}{\sqrt{K_k}}\sin(\mu_m\sqrt{K_k}K_\delta)\left[N\cos\mu_m\cos(\mu_m\sqrt{K_k}\,\bar{x}) + \frac{1}{\sqrt{K_k}}\sin\mu_m\sin(\mu_m\sqrt{K_k}\,\bar{x})\right] + \quad (11)$$

$$+Ki_H^*\left[N\cos\mu_m\cos\left(\mu_m\sqrt{K_k}\,\bar{x}\right) + \frac{1}{\sqrt{K_k}}\sin\mu_m\sin\left(\mu_m\sqrt{K_k}\,\bar{x}\right)\right]\exp(-\mu_m^2K_kFo_m).$$

where: μ_m — roots of the characteristic equation;

$$\varphi(K_{k,}N, K_\delta) = \frac{1+K_kK_\delta(3K_\delta + 3N + NK_kK_\delta^2)}{6\,(1+NK_kK_\delta)},\ J = \int_0^1 Z_{1,0}(\xi)cos\left[\mu_m(1-\xi)\right]d\xi,$$

$$tg\mu_m = N\sqrt{K_k}tg(\mu_m\sqrt{K_k}K_\delta). \quad (12)$$

Here: $Z_1(\bar{x}, Fo_m)$ is the dimensionless concentration of the transferable component over the thickness of the concrete; $Z_2(\bar{x}, Fo_m)$ is the dimensionless concentration of the transferred component over the biofilm thickness; $\bar{x} = x/\delta_1$ is the dimensionless coordinate; $K_k = k_2/k_1$; $K_\delta = \delta_2/\delta_1$; q_H are mass flux density leaving the biofilm into the liquid stream; m is Henry's constant (kg biofilm · kg^{-1} concrete); $N = (\rho_{биом}\cdot k_2)/(\rho_{бет}\cdot k_1 \cdot m)$ is a coefficient that takes into account the phases properties; $Fo_m = (k_1\cdot\tau)/\delta_1^2$ is the Fourier criterion; $Ki_H^* = \frac{q_H\cdot\rho_{бет}\cdot m\cdot K_\delta}{\delta_2\cdot\rho_{биоп}\cdot k_2\cdot C_0}$ is the Kirpichev's mass exchange criterion.

Figure 4 shows the results of calculations according to the above proposed mathematical model of biocorrosive failure of concrete, confirming the adequacy of the previously adopted provisions [9, 13-15]. Such a mathematical model makes it possible to determine the value of concentrations of the transferred component along the thickness of the concrete structure and in the biofilm itself at any time, as well as to calculate the concentrations of "free" calcium hydroxide in the liquid phase. The end result is the ability to predict the durability and reliability of concrete structures with a minimum of error.

a) b)

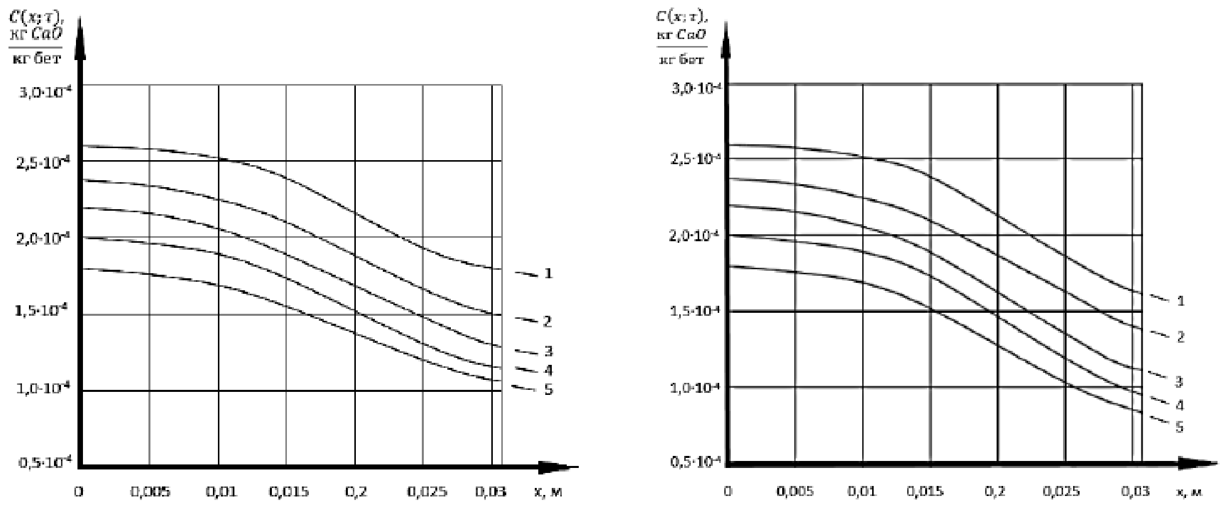

Figure 4 – The profiles of "free" calcium hydroxide concentrations along the concrete thickness and biofilm in bacterial (a) and fungal (b) corrosion at the time point:
1 – 14 days; 2 – 28 days; 3 – 42 days; 4 – 56 days; 5 – 70 days.

Mathematical model of cement concrete corrosion taking into account colmatation of pores and capillaries is presented below. The differential equation for determining the dimensionless speed of the clogging front has the form (13):

$$\frac{dL(Fo_m)}{dFo_m} = K_{pc} \cdot \frac{\partial \theta_B(\overline{R}_x, Fo_m)}{\partial \overline{x}} \cdot \frac{\nu_{AD}}{(1-\varepsilon_{oc})}, \qquad (13)$$

where: ε_{oc} – is the porosity of sediment layer, characterizing the volume of voids in the sediment layer; $K_{pc} = K_p \cdot C_{v.o.}$; $K_p = \frac{\rho_{бет}}{\rho_{oc}}$ – density coefficient; R_k – surface of the structure (interface) through which the transfer of the target component from the solid phase to the liquid phase takes place; $Fo_m = \frac{k_B \cdot \tau}{\delta^2_{бет}}$ – Fourier mass transfer criterion; k_v is the mass conductivity coefficient of the component B_{tv} (calcium hydroxide in concrete), $m^2 \cdot s^{-1}$; $\rho_{бет}$ – concrete density, $kg \cdot m^{-3}$; $\overline{x} = \frac{x}{\delta_{бет}}$ – dimensionless coordinate; τ – time, s; ν_{AD} is the stoichiometric coefficient; x – coordinate, m.

The proposed model of colmatation makes it possible to predict the corrosion rate of cement concrete taking into account the movement of colmatation front and the thickness of colmatant layer [16]. Figure 5 shows particular cases of mass permeability coefficient changes during colmatation [17-19]. In turn, figure 6 illustrates the colmatation rate $\frac{dL(Fo_m)}{dFo_m}$ and colmatant layer thickness $L(Fo_m)$ dependence on Fourier mass-exchange criterion Fo_m at sediment layer porosity ε_{oc} = 0.5 [20]. Judging by the reason of the curves in figure 5, 6, the rate of colmatation of pores in concrete slows down in time due to the increase in thickness of the colmatant layer [21-24].

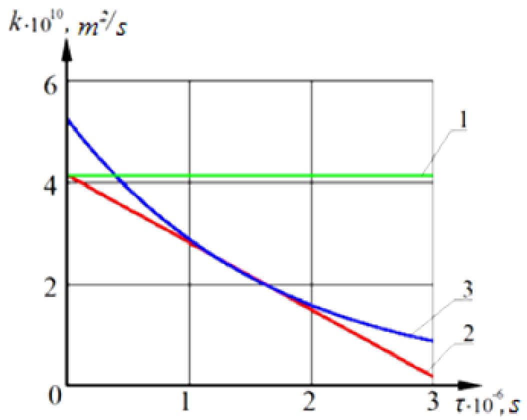

Figure 5 – Particular cases of changes in the mass permeability coefficient in colmatation: 1– no change in time; 2, 3 – change according to the linear (2) and exponential (3) dependence.

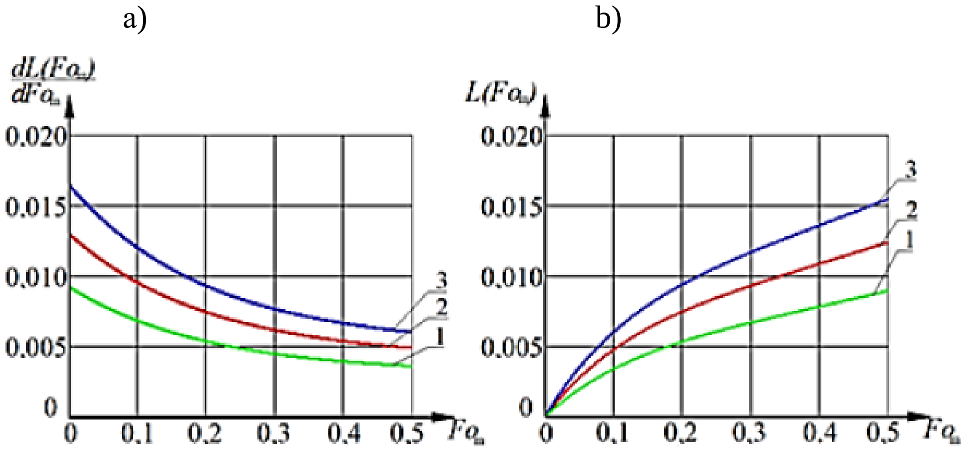

Figure 6 – Dependence of colmatation rate $\frac{dL(Fo_m)}{dFo_m}$ (a) and colmatant layer thickness $L(Fo_m)$ (b) on the Fourier mass transfer criteria Fo_m at sediment layer porosity ε_{oc} = 0.5 and mass transfer coefficient: 1 – k_{vo} = 4.11·10^{-10} m^2·s^{-1} (constant); 2, 3 – coefficient varying by a sloping linear (2) and exponential (3) dependence.

Let us consider the exposure of concrete to an aggressive salt solution. In this case, the wall of the concrete structure is modeled by an unrestricted plate. It is possible to trace the influence of Kirpitchev's mass transfer criterion on the dynamics of concentration fields of the target component "free" calcium hydroxide along the thickness of the concrete structure in contact with the fluid (figure 7). It can be seen (figure 7a), at the same value of Fourier mass transfer criterion, increasing of mass flow leads to acceleration of mass transfer process of target component in solid phase that agrees with data [25, 26].

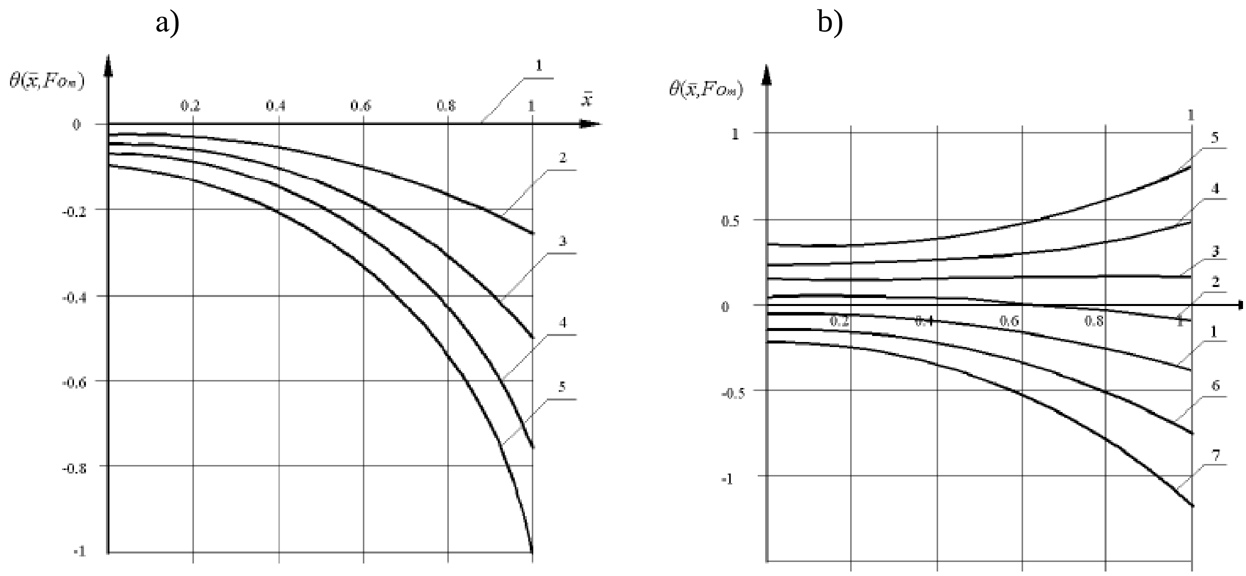

Figure 7 – Profiles of dimensionless concentrations of the transported component at Fo_m = 0.2 depending on: a – on the intensity of external mass transfer (1-5), Ki_m^* : 1 – 0; 2 – 0.5; 3 – 1.0; 4 – 1.5; 5 – 2.0; b – from the internal mass source (1-7), Po_m^*: 1 – 0; 2 – 0.5; 3 – 1.0; 4 – 1.5; 5 – 2.0; 6 – (-0.5); 7 – (-1.0), Ki_m^* =1

In turn, figure 7b illustrates the calculation results for various values of the modified Pomerantsev number (Po^*_m). This number shows the effect on the mass transfer process of a coordinate-distributed bulk source of mass due to reactions of release (curves 2-5) or absorption (curves 6, 7) of the substance at fixed Kirpitchev and Fourier criteria (Fo_m = 0.2; Ki^*_m = 1.0).

Based on such data, it can be judged that mass transfer processes during the destruction of cement concretes, taking into account the colmatation of pores and capillaries, proceed according to the kinetic patterns of corrosion of the II type.

Conclusions.

1. The presented mathematical models allow to calculate the kinetics of mass transfer processes of calcium hydroxide in the liquid and solid phases, to estimate the time to reach its concentration in free form at the interface corresponding to the beginning of decomposition of highly basic components.

2. It becomes possible to move to the subsequent stages of modeling of corrosion processes development using various materials for modern construction of industrial enterprises.

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