Fluid Mass-Exchange Process in the Near-Surface Zone of Massif Aeration

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Abstract: - Based on a series of laboratory experiments on a unique installation for upward filtration lifting of solutions of water-soluble salts of nonferrous metals, the regularities of mass transfer and surface deposition at the evaporative barrier have been obtained. It was found that the intensity of evaporation has a linear tendency to decrease with time, which is associated with changes in the porosity of the massif in the aeration zone. Redistribution of vertical zonality of salt deposition concentration in the surface zone of the array is possible by controlling the evaporation intensity. Regularities of salt concentration distribution in different levels of the massif have been established, which forms the basis of the mathematical model of the fluid-mass transfer process.

Key-Words: - massif, aeration zone, evaporation barrier, aqueous solution of salts, upward mass transfer, capillary filtration, crystallization concentration of salts.

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1 Introduction

Evaporation of water from the surface of the massif is a natural process. The rise of fluids from the depths of the massif to the surface and the evaporation of water into the atmosphere can form the basis for the formation of artificial concentration zones in the near-surface region of the massif. Studying the regularities of the process of "sliding" mass transfer will allow using its mechanism in a number of technological solutions in the development of minerals of both geogenic and technogenic origin. It can be used, for example, in the storage of enrichment waste or substandard ores of deposits. In rock massifs of the subsoil, the mechanisms of fluid mass transfer processes are the driving force for the transformation and transfer of ore matter, [1]. Migration of useful components in massifs of waste from the processing of mineral raw materials and the occurrence of in situ concentration processes with the formation of zones of both increased and decreased contents of various elements. These processes are inherent in both the original geological structures of the massifs and newly created technogenic mineral formations, [2]. However, these processes are not formalized enough to date for possible application in the artificial formation of highly concentrated zones in situ. Studies of the transformation of substances during mass transfer at technogenic sites and model experiments have shown that geological hypergene transformations lead to changes in the technological properties of both ore and non-ore minerals, [3], [4]. For example, enrichment waste, which usually has a finely dispersed composition, represents both the basis for secondary mineral formation and a potential environmental hazard, [5]. In nature, geological processes are not aimed at creating highly concentrated zones of useful components in massifs. They do not have such a goal. The appearance of deposits in the subsoil, including rich ores or large reserves in massifs, is associated with the patterns of substance transformation mechanisms in geological processes. Most deposits out of the total number are caused by fluid mass transfer, [6]. Filtration movement of fluids in the subsoil and the presence of geochemical barriers predetermine mobilization, immobilization, and local accumulation of useful components up to deposits. Taking into account the hypothetical direction of filtration movement of fluids in a certain area of the subsoil, we can assume the natural potential for the creation of deposits and ore occurrences of certain useful minerals. If the direction of fluid movement is controlled or even directed, then the formation of deposits can be localized in the most suitable places for technological development. Knowledge of the mechanism of the directed process of material-structural transformation of the massif will allow the formation to be initiated in the future. The essence of initiation and control of processes determines the creation of a nonequilibrium state in the selected area of the subsoil, in which the potential energy of transformation of substances sets in motion the homeostatic mechanisms of the geological environment, [7]. Of course, not all geological processes are suitable for modeling and subsequent applicability on a geological scale. The most productive and technologically acceptable, in our opinion, is the process of mass transfer of fluids in the direction of the surface of the massif. Many researchers have devoted their studies to the processes of fluid filtration in natural and man-made rocks. [8], [9], [10]. As a rule, concentrations of useful component compounds are confined to zones of geochemical and physical barriers in the subsoil. If there are no physical or chemical barrier conditions in the massif, then filtration mass transfer of concentrated zones of useful components is created. One such physical and effective barrier is evaporation. It is associated with the surface of the massif, with its contact with the air atmosphere. A large number of rich and even unique (in terms of useful component content) deposits are known, confined to surface weathering crusts, [11]. From a technological point of view, this type of barrier reprecipitation is especially attractive for several reasons. Mainly, this process is controllable, relatively dynamic, and scalable. An important advantage is that useful components are concentrated in the near-surface aeration zone of the massif, which is economically and technologically attractive during development. In this case, the direction of fluid movement to the surface is natural and can be controlled. Natural conditions for the occurrence of an evaporation barrier in nature are the value of air humidity and the connection with the "mirror" (level of the table water) of groundwater occurrence. If the connection exists, then the fluids are guaranteed to rise from the "mirror" of the aquifer to the surface through the aeration zone. The mechanism of fluid ascent in the massif occurs due to the pressure gradient and surface tension forces in the capillaries of the massif. In the aeration zone of the massif, the aqueous part of the fluid in the capillaries begins to evaporate, increasing the concentration of dissolved salts, and evaporation from the surface "pulls" like a pump from the aeration zone, creating a pressure gradient. This is the basis for the upward rise of fluids to the surface of the aquifer. And this mechanism operates continuously, [12]. Continuous evaporation at such barriers is capable of forming and usually does form, zones of concentration in the near-surface areas of the massif. Under natural conditions, fluids constantly move in an ascending flow to the surface. In essence, this mechanism is a powerful natural mechanism that sets fluids in motion through the pores of the massif to the surface. Potentially, this mechanism is capable of passing large volumes of fluids from the subsurface to the surface with the evaporation of water into the atmosphere. This is a natural process, [13]. In addition, the evaporation of water from the surface itself creates a pressure gradient in the aeration zone. These mechanisms serve as criteria for controlling the process of ascending fluid flow, [14], [15], [16]. Evaporation of water from the fluid complex increases the concentration of dissolved salts. When the concentration exceeds the solubility limits of a particular compound in the fluid, these useful compounds will precipitate where the excess occurs, including in the capillaries of the massif in the aeration zone or directly on the surface. Different mineral compounds have different concentration limits of aqueous solutions, therefore different compounds can be selectively precipitated at different levels of the massif aeration zone depending on their maximum concentration values. The aeration zone is a zone of natural evaporation barrier. This property of the aeration zone should be used in technological solutions during the creation and preparation of mineral deposits. Selective precipitation and accumulation of heterogeneous salt solutions at different levels of the massif aeration zone can be achieved by a controlled capillary rise at sedimentation levels. Information about the evaporation barrier is also known in agriculture. The negative impact of the salinization process of agricultural soils is an important aspect of plant growth. The main patterns of natural capillary rise and its effect on changes in soil properties have been studied in the literature, [17]. It is reflected that the structure, size of particles, and capillaries are the main criteria for the nature of filtration, [18], [19]. To formalize the process of filtration mass transfer in a controlled process solution in the field of extraction of useful components by the method of in situ conversion and sedimentation in the aeration zone on the evaporation barrier, it is important to study a number of parameters, the main ones of which are the capillary flow rate, the value of the surface area of contact of the solution with the solid material and the parameters of the capillary system, [20]. To use this phenomenon in the technological process, it is necessary to optimize the main parameters that ensure complete completion of extraction and control of the process of capillary movement of liquids. Based on laboratory experiments, estimates of a number of certain parameters of the ascending filtration flow were obtained. Based on the obtained patterns, the basic principles of mathematical modeling of the process can be substantiated.

2 Description of the Experiment and Discussion of the Results

The study was conducted on water-soluble salts of non-ferrous metals. It is known that copper, nickel, and cobalt sulfates are formed during the oxidation of sulfides of the corresponding metals, for example, in oxidation zones of massifs under hypergenesis conditions. Sulfates are water-soluble salts and, as a rule, are part of the fluids in the subsoil. The study of the process of filtration movement and sedimentation of complex salts of non-ferrous metals was carried out on the evaporation barrier. An experimental study of the vertical distribution in the aeration zone and in the massif (by zones) was carried out. To study the process, two laboratory mock-up installations were specially created (Figure 1, Appendix), where the imitation of the massif material was performed by quartz sand with a size of -0.5+0.2 mm, which is very typical for the granulometric composition of the weathering crust massif material. The column-type installation (Figure 1, Appendix) was made for the condition of vertical ascending filtration movement of liquids. A sulfate solution (complex) with a concentration of 20% to 80% was fed into the lower region (the concentration value was calculated based on the maximum solubility in water at a temperature of 20°C). A constant liquid feed level was maintained at the base of the column according to the Mariotte vessel principle (the liquid "mirror" level was at the bottom of the column). The containers for feeding solutions into the column with the material were isolated from daylight (Figure 1(b), Appendix) to prevent the development of microorganisms.

The massif temperature was controlled and regulated by blowing hot air through a column unit. The air temperature and humidity parameters were controlled by a hygrometer and an atmospheric thermometer. It is known that to assess the ascending kinetics of fluid filtration in the massif, it is necessary to know the physical and filtration-capacitive properties of the rock. The principles of filtration are characterized by the following known dependencies, [21]. The critical parameter of the massif for assessing filtration is the porosity of the medium, which is characterized by the effective diameter of the particles that make up the porous medium of the massif:

$$\frac{1}{d_e} = \sum_1^n \frac{g_i}{d_i}, \tag{1}$$

where:

$\boldsymbol{g_i}$ - is the specific content of the i-th fraction, in fractions of a unit from the total content;

$\boldsymbol{n}$ - is the number of fractions; di is the average diameter of the i-th fraction.

The permeability of rocks is characterized by:

$$k = m\frac{R^2}{8} \tag{2}$$

where $\boldsymbol{m}$ - is the porosity of rocks (for the material presented with the grain size distribution m= 0.259 – 0.476);

$\boldsymbol{R}$ - is the hydraulic radius of the pores of the model of the presented material (µm).

$$R = d_i\sqrt[3]{\frac{\rho}{8\rho_n}} \tag{3}$$

where $\boldsymbol{d_i}$ - is the particle diameter, $\boldsymbol{\rho}$ - is the soil density; $\boldsymbol{\rho_n}$ - is the bulk density of the soil.

According to the conditions of the experiment of the ascending filtration movement of fluids, there is a water-saturated capillary layer above the level of the mirror. The height of the capillary rise of the fluid is determined by the formula depending on the size of the capillary.

$$h = \frac{2\alpha cos\theta}{\rho gr} \tag{4}$$

where $\boldsymbol{\alpha}$ - is the surface tension, J/m2; $\boldsymbol{\theta}$ - is the contact angle; $\boldsymbol{\rho}$ - is the density of the liquid;
$\boldsymbol{r}$ - is the radius of the capillary, m.

The calculations of the filtration properties of the experimental massif are compared with the models of the massif in the installation. The calculated filtration-capacitive properties of sand are presented in Table 1 (Appendix).

In accordance with the calculated data on the characteristics of the experimental material, the parameters of the laboratory setup were taken into account, in particular, the minimum diameter and height of the columns.

The experiment was long-term, lasting 2000 hours with control of the main parameters listed above. At the end of the experiment, the massif material was removed from the setup in vertical zones, in each of which a sample was taken for chemical analysis.

The results of the precipitation of non-ferrous metal salts were compared with the theoretical principles of crystallization precipitation of salts in the mouths of capillaries, [21]. The basis of crystallization can be represented as follows:

$$Proportional \propto \leq \frac{D}{L} Ln\left(\frac{C_f}{C_0}\right) \qquad (5)$$

where $\boldsymbol{\propto}$ - is the evaporation rate, cm/s; $\boldsymbol{D}$ - is the diffusion coefficient of the salt in the solution, cm^2/s; $\boldsymbol{L}$ - is the length of the capillary, cm; $\boldsymbol{C_f}$ -is the concentration of the saturated solution, g/cm^3; $\boldsymbol{C_0}$ - is the initial concentration of the solution, g/cm^3.

3 Experimental Results

Evaporation from the surface usually leads to the precipitation of salts with the formation of crystals at the mouth of the capillaries. The speed of fluid movement depends on the temperature of the massif material and the combination of temperature + atmospheric humidity. This determines the kinetics of the process of water evaporation from the surface. The evaporation rate, in turn, determines the dynamics of the concentration of salts in the fluid near the meniscus. When the concentration of a particular salt in the capillaries in the aeration zone can exceed the maximum concentration Cf, this will lead to precipitation and crystallization on the surface of the capillaries. At low evaporation rates, the critical concentration of the solution remains constant and is formed directly on the surface with the formation of a surface salt crust, [22], [23].

$$\frac{C_f}{C_0} = 1 + \left(\frac{\pi\beta}{D}\right)^{\frac{1}{2}} \qquad (6)$$

where $\boldsymbol{\beta}$ - is the evaporation coefficient, cm^2/s

The composition of the fluid fed to the columns in the experiment is given in Table 2 (Appendix).

During the experiment, the evaporation rate was monitored in order to determine the dependencies for the mathematical model of the mass transfer and sedimentation process in the aeration zone and elsewhere. The obtained dependencies are shown in Appendix in Figure 2, Figure 3 and Figure 4.

The most significant factors were the duration of the process, humidity, and air temperature. It is characteristic that over time, the pores in the aeration zone calcify when salts are deposited from the fluid. As a result, the cross-sectional area of the capillaries decreases and the porosity in the aeration zone decreases. In addition to temperature, air humidity has a significant effect. In the experiment, this parameter was maintained within 20 - 49%. It is known that the moisture capacity of the atmosphere depends significantly on temperature, so an increase in air temperature by 1 degree Celsius increases moisture absorption by 7-11% in the temperature range from 15 to 24 degrees Celsius, [24], [25].

The results of the experiment confirmed the theoretical assumptions [26], according to which, during evaporation, an increase in the concentration of the solution is compensated by the diffusion removal of the crystallization point deep into the capillary. A decrease in the rate of fluid flow into the aeration zone will accordingly reduce the evaporation rate at constant parameters of temperature and humidity of the atmospheric air. Experimental studies conducted with different solutions and materials also indicate a decrease in the evaporation rate due to the transfer of salt to the surface layers of the massif, [27]. For more intensive precipitation with the formation of a crystalline crust on the surface, according to the regularity (5), it is necessary to change the evaporation rate and, accordingly, the speed of movement of solutions in the capillaries of the massif. Experimentally, this was achieved by gradually increasing the temperature of the massif to 40 °C and additionally using warm air blowing on the surface of the massif at a speed of 5-7 m / s. Such conditions ensured an increase in the speed of solution movement to the surface to values of 3 - 5 mm / h, which is 7 - 9 times higher than the speed of movement under normal experimental conditions (20 ° C and without blowing on the surface). During cavitation with precipitation and its crystallization in the capillaries, the evaporation rate decreases somewhat, [28]. During the experiment, the first appearance of crystals on the surface of the massif is observed already in the second week. After 120 days from the start of the experiment, the main part (up to 80%) of the salt entering the column is in a crystalline state in the aeration zone. It was experimentally established that the concentration of the solution corresponds to the concentration of the maximally saturated solution and the precipitation of crystals does not begin, and the evaporation rate decreases by 2 times. In the variant with low evaporation rates (t ~ 20°), crystallization on the surface began to be visually observed only in the second month from the start of the experiment. After the start of crystallization, the evaporation rate begins to decrease linearly (Figure 5, Appendix).

The filtration process slows down almost linearly over a fairly long period (up to 6 months), as well as the crystal growth, which is very slow and stable. This can be explained by the fact that the sediment reduces the capillary cross-section and reduces the evaporation rate with a decrease in the real evaporation surface. According to theory (5), the concentration of salt at the meniscus should be constant, and this should lead to the dissolution of part of the formed crystals and the transfer of the substance along the capillaries into the sediment closer to the contact with the atmosphere. This, in turn, partially increases the evaporation rate and promotes an increase in concentration. This characterizes the architectural growth of crystals on the surface, which, as a whole, then again reduces the evaporation surface. In this mode of change in the volume of crystals, precipitation with the growth of the salt crust can occur over a long period. In the course of our experiment, over a two-year period of experiments, the speed of movement of solutions stabilizes, decreasing after 2 months with a stable tendency of linear decrease within 7-12% per month. It should be noted that during precipitation, the salt crust is formed on the surface unevenly (Figure 1(b), Appendix). In our opinion, this is primarily due to the heterogeneity of the structure of the salt crust and capillaries between the crystals, which in turn is determined by the growth points of crystallization. The study of the salt crust showed that the pores in the salt crust itself are distributed chaotically. In the aeration zone, near the surface, as in the massif, dead-end pores are formed without an outlet to the surface, which inhibits the exit of solutions to the surface areas. In areas where there are through pores, a relatively rapid exit of solutions to the surface and their crystallization occurs. Crystals are formed primarily in the area of the exit of solutions to the surface. It was also experimentally noted that in all cases, including intense evaporation, salt crystallization gravitates toward the periphery of the surface of the massif or elevations on the surface (closer to the walls of the column). In our opinion, this is due to a higher level of ventilation of the outskirts and, accordingly, a higher evaporation rate. Under the experimental conditions, a threefold excess of the content was recorded in the peripheral zones, along the walls of the columns, compared to the central part of the experimental array. As for the specific conditions of our experiment, the increased crystallization rate along the perimeter of the columns is due to the higher speed of movement of solutions along the smooth surfaces of the glass columns. This effect is also consistent with the studies cited in [29]. During evaporation without increasing the temperature, the evaporation flow is much more uniform over the surface, and the deposition of salts is comparatively uniform.

4 Conclusion

Sedimentation on evaporation barriers in the massif aeration zone serves as a combined concentration zone for complex salts of non-ferrous metals while maintaining the water table level of the massif humidity level and directing the upward movement of fluids to the surface. The intensity of water evaporation from the surface causes an increase in the concentration of useful components as a technological layer for preliminary enrichment in the near-surface aeration zone of the massif in situ. The nature of the precipitation of water-soluble sulfates of non-ferrous metals (copper, nickel, and cobalt) depends on the conditions of fluid passage through the evaporation barrier. The speed of solution movement (depending on the evaporation intensity) predetermines the nature of the distribution and concentration of components in the aeration zone of the massif. At a low speed of movement, the evaporation zone spreads inside the massif itself, moving towards the central part of the massif (from the surface), due to which the filtration rate of the solution decreases, since the decrease in humidity from the middle part of the massif is compensated by its inflow from the lower, more saturated layer. This leads to an increase in precipitation and, accordingly, the concentration of the solution in the lower and central parts of the aeration zone of the massif. The low velocity of the solution movement at low air temperature and high humidity does not provide pronounced zonality of salt crystallization in the massif. In this case, crystallization precipitation spreads throughout the aeration zone with a decrease in content in the direction from the lower layers to the surface. Such a distribution is typical for both the initial highly concentrated solutions and relatively lean fluids. The scale and concentration of crystallization of sediments in the surface zone increase with the growth of the filtration rate of fluids. It can be assumed that additional phenomena of heat and moisture conductivity of the evaporation barrier occur. It was found that an increase in the evaporation intensity due to heating of the massif material and blowing over the surface, crystallization begins in the pores of the massif, starting from 7-10% (in the aeration zone) from the surface. With the onset of crystallization on the surface, the rate of filtration rises of the solution and, accordingly, the evaporation rate begins to fall linearly, decreasing to 12% per month. Based on the experiments, it was found that the intensity of the solution movement and the formation of a salt crust during crystallization can be controlled by changing the filtration rate before the evaporation barrier and directly inside it.

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Contribution of Individual Authors to the Creation of a Scientific Article (Ghostwriting Policy)

The authors equally contributed in the present research, at all stages from the formulation of the problem to the final findings and solution.

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Conflict of Interest

The authors have no conflicts of interest to declare that are relevant to the content of this article.

APPENDIX

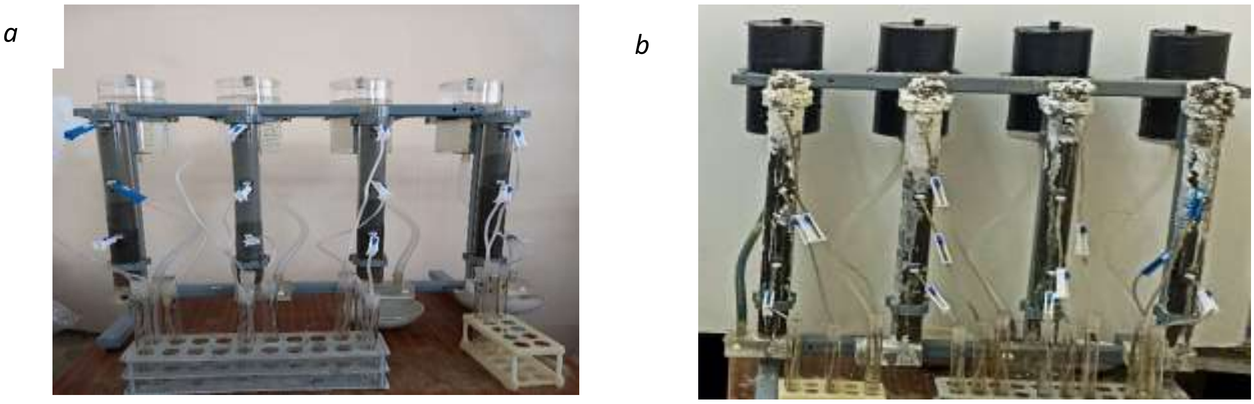

Fig. 1: Laboratory setup for ascending fluid flow, a - before the start of the experiment, b - after 3 months of the experiment

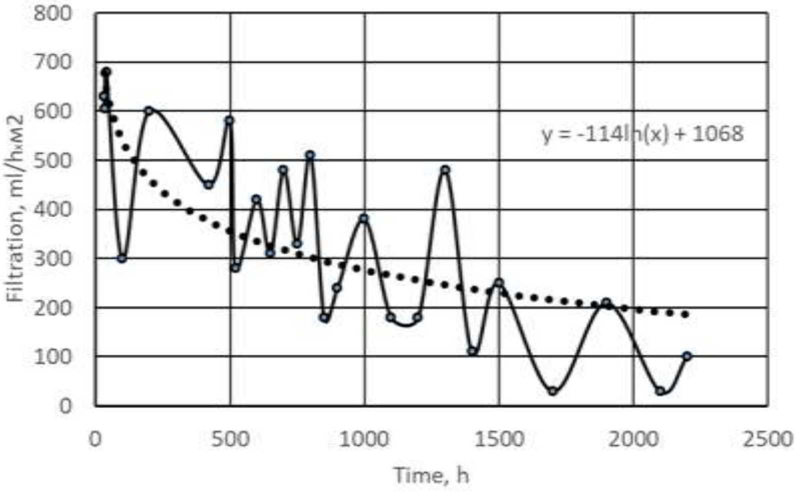

Fig. 2: Change in evaporation rate during the experiment

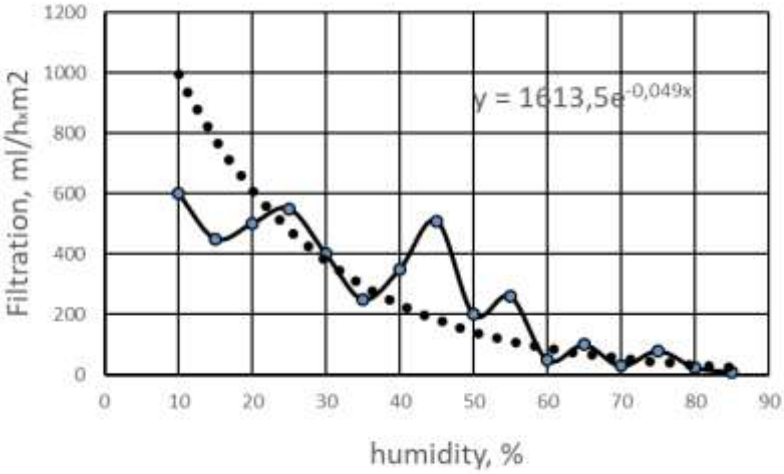

Fig. 3: Dependence of the evaporation rate on atmospheric humidity

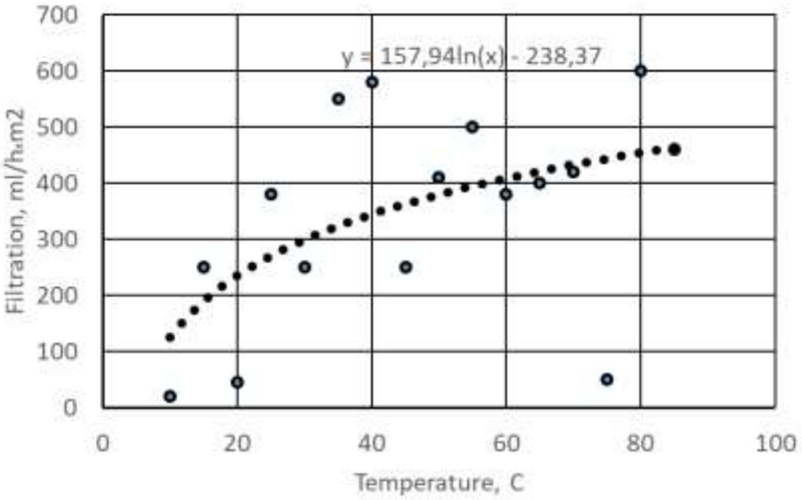

Fig. 4: Effect of air temperature on evaporation rate

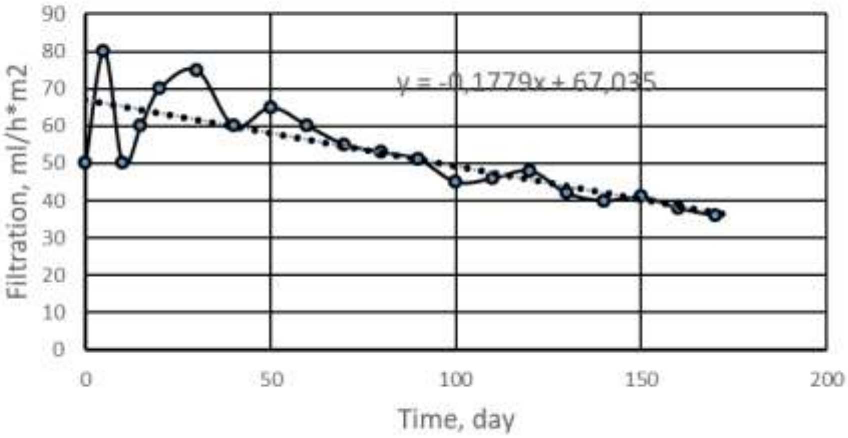

Fig. 5: Evaporation rate during surface crystallization of a complex salt of 80% concentration

Table 1. Characteristics of the experimental array material

Indicators	*Dimension*	*Value*
Average particle diameter	mm	0,242
Effective particle diameter	mm	0,020
Hydraulic radius	μm	3,210
Permeability	darcy	0,522
Capillary rise height	m	0,645

Table 2. Characteristics of the applied non-ferrous metal sulfate

Indicators	**Cu SO_4**	**$NiSO_4$**	**$ZnSO_4$**
Maximum solubility, g/l	349	220	383
Content at 80% solubility of maximum, g/l	280	176	306
Concentration of maximum solubility, %	26.7	26.7	26.7