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CORRELATION OF HIGHLY DISPERSED AEROSOL PARTICLES AND AEROIONS, FORMED BY SYLVINITE SURFACES AND MATERIALS

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КОРРЕЛЯЦИЯ ВЫСОКОДИСПЕРСНЫХ АЭРОЗОЛЬНЫХ ЧАСТИЦ И АЭРОИОНОВ, ФОРМИРУЕМЫХ СИЛЬВИНИТОВЫМИ ПОВЕРХНОСТЯМИ И МАТЕРИАЛАМИ

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The use of materials based on natural potassium salts is a known way to create a high-quality, up to healing, indoor air which is modified due to the effect of sylvinite, carnallite and halite aerosol particles. Facing or decorating protective surfaces of walls, floor or ceiling in special ground facilities – speleoclimatic chambers – can enrich the indoor air with a highly dispersed salt aerosol and aeroions of light mobility group.

It is proposed to look over the interrelation between distribution of aerosol particles and concentration of light aeroions in sylvinite speleoclimatic chambers, considering the ionization and recombination equation of formation and disappearance of light aeroions. By extrapolation the main parameters of the highly dispersed salt aerosol were determined for size less than 0.3 microns based on the experimentally determined parameters of aerosol particles distribution by size (greater than 0.3 microns) taking into account possible solutions of the aeroionic balance equation and applying the superposition model of several logarithmically normal distributions.

On example of Verkhnekamskoie potash deposit the article shows the main parameters of aerosol particles` size distribution in sylvinite speleoclimatic chambers with surfaces of different constructions made of sawn natural sylvinite blocks, panels and molded salt tiles with a high content of potassium chloride, a component of potash salts.

Study results confirm high efficiency of sylvinite building materials application to create a high-quality medical or wellness air environment saturated with highly dispersed salt aerosol, and allow to optimally select special constructive and decorative materials on the basis of sylvinite, depending on required parameters of the aerosol distribution in order to create a high-quality indoor air.

Ключевые слова:
сильвинит, спелеоклиматическая камера, распределение аэрозольных частиц по размерам, высокодисперсный аэрозоль, логнормальное распределение, экстраполяция, диффузионная зарядка аэрозоля, легкие аэроионы.

Применение материалов на основе природных калийных солей является известным способом создания качественной, вплоть до лечебной, воздушной среды помещений, которая модифицируется ввиду влияния аэрозольных частиц сильвинита, карналлита и галита. Облицовка или декорирование ограждающих поверхностей стен, пола или потолка специальных наземных комплексов – спелеоклиматических камер – позволяют обогатить воздух помещения высокодисперсным соляным аэрозолем и аэроионами легкой группы подвижности.

Взаимосвязь распределения аэрозольных частиц с концентрацией легких аэроионов в сильвинитовых спелеоклиматических камерах предлагается проследить, рассматривая ионизационно-рекомбинационное уравнение образования и исчезновения легких аэроионов. Путем экстраполяции определены основные параметры высокодисперсного соляного аэрозоля размером менее 0,3 мкм исходя из экспериментально определенных параметров распределения аэрозольных частиц по размерам (более 0,3 мкм), принимая во внимание возможные решения уравнения аэроионного баланса и применив модель суперпозиции нескольких логарифмически нормальных распределений.

На примере горных пород Верхнекамского калийного месторождения представлены основные параметры размерного распределения аэрозольных частиц в сильвинитовых спелеоклиматических камерах с поверхностями разных конструкций, выполненных из пиленых сильвинитовых блоков природного сильвинита и из панелей и прессованной соляной плитки с высоким содержанием хлористого калия, входящего в состав калийных солей.

Полученные результаты подтверждают высокую эффективность применения сильвинитовых строительных материалов для создания высококачественной лечебной или оздоровительной воздушной среды, насыщенной высокодисперсным соляным аэрозолем, и позволяют оптимально выбирать специальные строительные отделочные и декоративные материалы на основе сильвинита в зависимости от требуемых параметров аэрозольного распределения с целью формирования качественного воздуха помещений.

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Introduction

It is known that one of the main rocks' biologically active factors and certain, natural or artificially passed, underground cavities (such as salt mining workings [1-3]) is a highly dispersed aerosol [4-6]. To create the unique microclimate of the closed facilities special facilities [7, 8] – speleoclimatic chamber are increasingly used, where at facing material [9] and/or materials of the load-bearing structures sylvinite ore is used. Due to the natural salt effect air environment in such complexes possess unique properties [10] such as the air has high volume concentrations of highly dispersed salt spray and light ions.

However, to date research, generalizing from common physical positions theoretical and experimental results of the effect of these special decorative materials on generated indoor air aerosol and aeroion compositions is not conducted. As a rule, research of aeroionic and aerosol compositions is limited to in natural measurements without regard to interconnection between them. In particular, in the speleoclimatic chambers' air environment erosol particles with a size greater than 0.5-1.0 microns are most studied, because of the possibility selecting by filters for subsequent microscopic and chemical analysis. However, the currently accepted the gravimetric approach to aerosol pollution's harmful effects evaluation method does not satisfy modern trends, since without definition of aerosol particles' dispersion and their physical and chemical properties the design concept and speleological chambers' operation mode can not be objectively choosen. Indeed, their size is the most important parameter in determining the characteristic effect of respirable aerosol particles on human [11-14]. At high dispersion aerosol has high chemical activity because of a large total surface of the particles. Highly dispersed aerosol particles have unique physical and chemical properties, as practically they do not settle for a long time and are in a suspended state.

It should be noted that the measurement of the highly dispersed aerosol particles' concentration with a diameter less than 0.5 microns is technically very difficult [15]. According to this receiving the information about the highly dispersed aerosol particles instrumentally by extrapolating certain size distribution of particles larger than about 0.5 microns or smaller fraction is the relevant task. For it to carry out simulation airborne particle with nanometers and hundreds of nanometers diameters, considering a system of "aerosol particles–airoions" is proposed

Theoretical and empirical foundations

Interconnection of aerosol particles distribution and the concentration of light ions can be traced by considering the formation's ionization-recombination equation and light ions disappearance, that establishes the interconnection of light ions' volumetric enumeration concentrations with the ionization level and aerosol distribution, taking into account the recombination of ions between itself and deposited on aerosol particles:

$$\begin{aligned} \frac{\partial n_i}{\partial t} &= \nu - \alpha n_+ n_- - \\ &- n_i \sum_{\chi=-\infty}^{\chi=+\infty} \int_{D_{\min}}^{D_{\max}} dD \beta_\chi^i(D) N(\chi,D,t), \end{aligned} \qquad (1)$$

where n_i – the light aeroions concentration with mobility μ_i = 0.5-2 $cm^2B^{-1}s^{-1}$; i – ionic charge characteristics, if the charge is positive, then i is denoted by a "+" if the charge is negative then i is denoted by "–"; ν – ionization intensity; α – recombination factor of oppositely charged light aeroions with each other, according to calculations by attachment theory as a result of triple collisions for normal conditions $\alpha = 1.4 \cdot 10^{-6}$ cm^3/s, which is close to the experimentally observed values $\alpha \cong 1.6 \cdot 10^{-6}$ cm^3/s [16]; $\beta_\chi^i(D)$ – combination factor of light positive (or negative) aeroions and aerosol particles with a diameter D and charge χe (or $-\chi e$); $N(\chi,D,t)$ – concentration of the aerosol particle with diameter D and charge χe at the time t; e – elementary charge; t – time.

Suppose that the system "aerosol particles – airoions" is in thermodynamic and electric equilibrium (ie, the distribution of aerosol particle by size is quasi-stationary and by charges is symmetrically). Then the equation (1) can be written as

$$\begin{aligned} \frac{\partial n_+}{\partial t} &= \nu - \alpha n_+ n_- - S_p^+ n_+, \\ \frac{\partial n_-}{\partial t} &= \nu - \alpha n_- n_+ - S_p^- n_-, \end{aligned} \qquad (2)$$

where S_p^i – factor reflecting the value of the decrease aeroions on the aerosol particles, $S_p^i = \sum_{\chi=-\infty}^{\chi=+\infty} \int_{D_{\min}}^{D_{\max}} dD \beta_\chi^i(D) N(\chi,D)$.

In equilibrium, the derivatives of equations (2) are equal to zero and $S_p^+ n_+ = S_p^- n_-$. Solution of the equations (2) relative to n_+ and n_- gives

$$n_+ = \frac{\sqrt{\left(S_p^-\right)^2 + 4\frac{S_p^-}{S_p^+}\alpha\nu} - S_p^-}{2\alpha},$$
$$n_- = \frac{\sqrt{\left(S_p^+\right)^2 + 4\frac{S_p^+}{S_p^-}\alpha\nu} - S_p^+}{2\alpha}. \qquad (3)$$

Assuming that the both charges light aeroions' concentration are equal ($n_+ = n_- = n$), the initial equations of aeroionic balance (2), dropping the index *i*, would be written as

$$\frac{\partial n}{\partial t} = \nu - \alpha n^2 - S_p n\,. \qquad (4)$$

In the ionic balance equation, aeroions decrease S_{pn} due to aerosol particles sedimentation on the assumption that the total number of aerosol particles per unit volume $N = N_0(D) + 2\sum_{\chi=1}^{\infty} N_\chi(D)$ and $N_+ = N_-$, can be written as

$$S_p n = n \sum_{\chi=-\infty}^{\chi=+\infty} \int_{D_{\min}}^{D_{\max}} dD \beta_\chi(D) N(\chi, D) =$$
$$= n \sum_{D=D_{\min}}^{D=D_{\max}} \left(\beta_0(D) N_0(D) + \sum_{\chi=-\infty}^{\chi=+\infty} N_\chi(D)\beta_\chi(D) \right), \qquad (5)$$

where $\beta_0(D)$ – recombination factor between the aeroions and neutral aerosol particles; $\beta_\chi(D)$ – recombination factor between the aeroions and charged particles to the number of elementary charges χ = 1, 2, 3, ... (or $-\chi$ = 1, 2, 3, ...); $N_0(D)$ – the concentration of chargeless particles; $N_\chi(D)$ – concentration of particles, charged to a value χ (or $-\chi$).

In the system equilibrium state applied to the condition of symmetrical (ie when $n_+ = n_-$) diffusion charging of aerosol particles by light aeroions combination of aeroions and aerosol particles (5) over the entire range of particle sizes can be described by the empirical formula [17]

$$S_p n \approx n \int_{D_{\min}}^{D_{\max}} N(D) \left[25 \sqrt{\frac{D - 0{,}001}{D + 0{,}005}} D \cdot 10^{-6} \right] dD, \qquad (6)$$

whre D – diameter of the aerosol particles, micron; $N(D)$ – distribution of aerosol particle by size.

Taking into account the accepted designations for stationary conditions equation (4) takes the form

$$\nu = \alpha n^2 + S_p n. \qquad (7)$$

Solving the obtained quadratic equation and excluding negative solution, we have

$$n = \frac{\sqrt{S_p^2 + 4\alpha\nu} - S_p}{2\alpha}. \qquad (8)$$

Note that for the solution of equation (8) linking the ionization intensity, calculating the light aeroions concentration and the distribution of aerosol particle by size the information about the aerosol dispersed composition in a wide range of particle sizes from $D_{\min}$ (at least on the order of 0.05 microns) to $D_{\max}$ is necessary.

In practical studies often the distribution of aerosol particles greater than 0.1 microns (usually in the range of a diameter of 0.3-0.5 microns) is defined. Then, if there is no data distribution of aerosol particles in a wide range of $D_{\min}$ to $D_{\max}$ applying equation (8) becomes incorrect.

Information about highly dispersed aerosol particles, having data about a particle distribution with size more or about 0.1 microns, receives considering empirical models that describe characteristic distribution of the aerosol particle by size.

Logarithmically normal distributionis considered to be the most reasonable for the analytical description of the disperse composition of aerosol pollution, especially in the highly dispersed fractions. It is known [18, 19], that in most cases, the distribution of aerosol particles can be reduced to the superposition of multiple logarith micallynormal distributions

$$\frac{\partial N(D)}{\partial (D)} =$$
$$= \sum_{j=1}^{p} \frac{N_{0j}}{\sqrt{2\pi} \ln\sigma_j D} \exp\left(-\frac{(\ln D - \ln D_{0j})^2}{2(\ln\sigma_j)^2} \right), \qquad (9)$$

where N_{0j} – the total number of particles *j-th* mode; σ_j – the standard geometric deviation of the *j*-th mode; D_{0j} – the geometric mean of mode diameter.

In general, the number of modes *p* in the distribution (9) is taken equal to four [20]. Wherein one of the modes (j = IV) lies in the geometric particles having an average diameter D_{04} more than 1 micron, and in particle sizes less than 1 micron are considered possible remaining three modes I, II, III (Fig. 1).

The initial approach three modes are considered in a range of particle sizes less than 1.0 microns. Particles' average geometric diameter, standard geometric deviation and concentration of the particles are assumed to be the values shown in Fig. 1. Further the values of D_{0j}, σ_j, N_{0j} are chosen method of the least squares so as to ensure that the known experimental data distribution of aerosol particles and the equation's convergence (5) connecting the distribution and concentration of particulate aeroions. For this in the modes the values are fitted consecutively: а) of average geometric diameter D_{0j}; б) of standard geometric deviation σ_j in the range 1.2-2.1; в) of the total particles number N_{0j}.

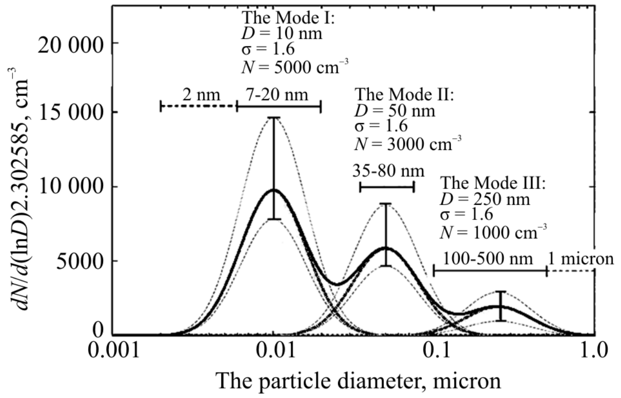

Fig. 1. The iterations' initial conditions in modeling the distribution of the aerosol particles (according to [20])

Since in some cases the distribution in particle sizes less than 1 micron can be reduced to a bimodal or even to unimodal as the further step of approximation the possible to reduce the number of modes in the particle size range of less than 1 micron from three to two (if needed accuracy is achieved) is considered. There are two variants: a) in the particle size range below 0.1 microns two modes are observed, and the particle size range of 0.1-1.0 microns mode is no; b) at the range of particles less than 0.1 microns only one mode is observed (particle range of 0.003-0.100 m), and the second mode is in the size range of 0.1-1.0 microns. Further, for condition of achievement desired accuracy, the possibility of reducing the number of events from two to one is taken into account. In the case of single-mode distribution similar iteration are carried out for the full range of 0.003-1.000 micron particle sizes.

Lognormal distribution's parameters of highly dispersed particles are determined by method of the least squares that the concentrations of heavy aeroions, calculated on the basis of the charge Boltzmann distribution on the aerosol particles

$$\frac{N_\chi(D)}{N} = \left[\sqrt{\frac{4\pi^2\varepsilon_0 DkT}{e^2}}\left(1+\frac{e^2}{48\pi\varepsilon_0 DkT}\right)\right]^{-1} \times \exp\left(-\frac{\chi^2 e^2}{4\pi\varepsilon_0 DkT}\right), \qquad (10)$$

as well as the empirical relation linking the value of the aerosol particles' diameter and mobility [21]

$$\mu = \left(\frac{9{,}44\chi}{D}\right) \times \left[1+\frac{165}{D}\left(1+0{,}336\exp\left(-\frac{D}{153}\right)\right)\right]10^{-3}, \qquad (11)$$

are coincided with the experimentally observed.

In equation (10) the following designations are accepted: ε_0 – electric constant; k – Boltzmann constant; T – temperature.

Thus, the use of the several logarithmically normal distributions' superposition to describe the dispersed composition of aerosol particles allows to conduct the approximation of the experimental data distribution up to the range of diameters D ~ 0.003 m and thus to evaluate the quantitative characteristics of highly dispersed aerosol fraction.

Methods and research equipment, the results of modeling

The above theoretical and empirical apparatus was experimentally tested on real special rooms, lined with sawn blocks of natural sylvinite or sylvinite panels of pressed salt tiles with a high content of potassium chloride, a component of potash.

The distribution of aerosol particle by size in an air environment was investigated using aerosol counter PKZV-1 (range of investigated aerosol particles sizes – 0.3-100 microns) and aerosol counter AZ-6 (particle size range – 0.3-1.0 microns).

The study aeroions concentration was carried out using the integral spectrometer aeroions UT-8401, which allows register the airoions of positive and negative polarities in the mobility range from 0.00032 up to 2.0 cm^{2}B^{-1}s^{-1} and more.

All studies were performed without people in the rooms (except the researcher).

To achieve this accuracy of modeling (correlation factor between the experimental data and the approximation in particle size bigger than 0.3 micrometers is not less than 0.99), the distribution of aerosol particles in an air environment space, special facing materials based on potassium salts, should be described at a superposition of two a logarithmically normal distributions (Fig. 2).

Presented in Fig. 2 the 2nd mode corresponds to mode with the index j = IV, which lies in the area of particles having an average geometric diameter greater than 1 micron [20]. The distribution of highly dispersed is characterized by modes with indexes j = I, II, III (Fig. 1), and it was reduced to single-mode – the 1st mode respectively (Fig. 2.).

The resulting measurements in rooms with salt decoration materials of logarithmically normal distributions' parameters' averaged values of for modes are shown in the table.

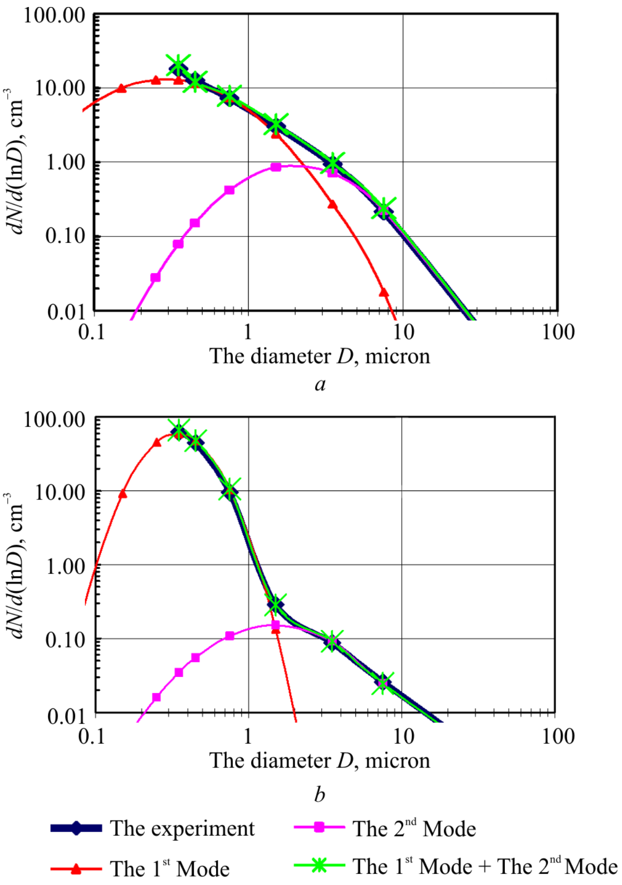

Fig. 2. Modeling the distribution of aerosol particles in rooms faced by: *a* – blocks of natural potassium salts; *b* – sylvinite panels and molded tiles

The averaged values of aeroions' volume concentrations and the results of aerosol particles' distribution by size approximation

Object of study	The concentration of aeroions, cm^{-3}		Mode 1			Mode 2		
	light	heavy	N_{01}	σ_1	D_{01}	N_{02}	σ_2	D_{02}
The rooms faced by blocs of natural potassium salts	2230 ± 390	2170 ± 520	29 ± 16	2.45 ± 0.68	0.29 ± 0.11	1.8 ± 0.2	2.2 ± 0.1	2.0 ± 0.3
The rooms faced by sylvinite panels and molded tiles	1340 ± 410	2810 ± 600	63 ± 29	1.53 ± 0.07	0.34 ± 0.01	0.32 ± 0.19	2.33 ± 0.11	1.5 ± 0.2

Conclusions

The received simulation results clearly demonstrate the difference in the salt spray dispersed composition according to the structural features of speleological chambers. Indeed, there is a pronounced dominant role of the 1st Mode compared with the 2nd Mode in rooms facing by sylvinite panels and molded tiles, compared with rooms, faced by blocs of natural potassium salt. Thus, the use of pressed tiles and panels increases the content of highly dispersed fraction of hydrochloric aerosols in the air. However, it should be noted that this increase inevitably leads to a relative decrease of the volume concentration of light aeroions.

The presented approach to the definition of the unique properties of these special facing and decorative materials based on potassium salts allows prove their use, identify options for use depending on the required parameters of the aerosol content in an air environment to form a high-quality indoor air. The presented results show that the panels and tiles made based on potassium salts by pressing are promising materials, because they allow generate more highly dispersed particulate salt fraction.

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