**Національна академія наук України**

**ІНСТИТУТ ФІЗИКИ КОНДЕНСОВАНИХ СИСТЕМ**

ICMP–98–35E

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STATISTICAL THEORY FOR DIFFUSION OF RADIONUCLIDES IN GROUND AND SUBTERRANEAN WATER

ЛЬВІВ

**УДК:** УДК 537.311.32:532:541.135.1.

**PACS:** 05.60.+w; 73.40; 66.10.Cb; 82.65.Fr

**Статистична теорія процесів дифузії радіонуклідів у грунтових та підземних водах**

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**Анотація.** В об'єкті "Укриття" Чорнобильської АЕС міститься багато радіоактивних матеріалів. Водне оточення підвищує їхню рухливість, так що радіоактивні елементи постійно проникають у довколишнє середовище з грунтовими водами. Це становище обговорюється в світлі недавніх вимірювань. Вважається, що вихід радіоактивних частинок має дифузійний характер. Запропоновано модель для дифузійного опису міграції радіоактивних елементів у грунтових та підземних водах. Методом нерівноважної статистичної механіки отримано коефіцієнти дифузії. Вони виражаються через унарну та парну функції розподілу. Їх розраховано для радіоактивних частинок біля діелектричної стінки, яка моделює грунт, при різних концентраціях.

**Statistical theory for diffusion of radionuclides in ground and subterranean water**

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**Abstract.** The object "Shelter" at Chernobyl nuclear power plant contains a lot of radioactive materials. Aqueous medium increases their mobility so that dangerous elements are persistently penetrating into environment with ground water. This situation and recent measurements are discussed. The egress is supposed to have a diffusion nature. A model for the diffusion treatment of active elements migration with ground and subterranean water is proposed. The diffusion coefficients are derived from nonequilibrium statistical mechanics. They are expressed through singlet and pair equilibrium distribution functions to be evaluated for radioactive particles near a dielectric wall modeling ground at various concentrations.

**Подається в** журнал "Радіаційна фізика та хімія" (Голандія)
**Submitted to** Radiation Physics and Chemistry

## 1. Introduction

One of main ecological problems in the exclusion zone, that is connected with consequences of the accident at the Chernobyl nuclear power plant, consists in transport (migration) of radionuclides with ground and subterranean water in the system "water pumping of near-by Chernobyl plant zone – the river Prypjat' – cascade of Dnieper storage lakes". Spreading radionuclides in atmosphere, ground, lakes, rivers after Chernobyl disaster is studied in many works [Accident...(1986), Arutyunyan R.V. et al (1993), Asmolov V.G. et al (1988), Avramenko V.I. et al (1992), Baryakhtar V.G., Honchar V.Yu. and Yanovskij V.V. (1993), Bogatov S.A. et al (1990), Borzilov V.A. (1991), Garcher E.K. et al (1992), Information...(1986), Lebedyev I.A. et al (1992), Makhonko K.P. et al (1992), Orlov M.Yu. et al (1992), Silantjev A.N. et al (1989), Sobotovich E.V. and Chabanenko S.I. (1990), Sobotovich E.V., Olkhovik Yu.A. et al (1991)]. Actual sources of radioactive elements entering into ground and subterranean water are: the very object"Shelter" carrying ecological and nuclear hazard and industrial territory surrounding it; points for temporary location of active wastes (temporary burials) and points for burial of radioactive wastes (permanent burials).

Every year, a great amount of water penetrates into the object "Shelter" with rain, snow, by way of indoor condensation and sprinkling of radioactive air by water solutions. At low points of block "B" and machine hall water is concentrated with a volume of 3000 - 3500 $m^3$ after Chernobyl plant data [Main Scientific and Technical Results...(1994), I.R.Yukhnovskii, A.E.Kobryn et al (1997)]. Availability of water in the object causes several kinds of danger:

- interaction with fuel containing masses (FCM) brings about the increase of neutron multiplication factor in the system;
- transport of solved salts of enriched uranium $^{235}$U also increases the object's nuclear hazard;
- disruption of FCM by water gives rise to uncontrollable indoor transport of radioactivity and radionuclide egress beyond the object since FCM disruption results in nuclear dust;
- parcels of "block" water penetrate into soil surrounding the object and migrate afterwards with ground and subterranean water of the exclusion zone.

The test of "block" water from "Shelter" for elements revealed that aqueous solutions are unique and hold isotopes of uranium $^{234}$U, $^{235}$U, $^{236}$U, $^{238}$U, plutonium $^{239}$Pu, $^{240}$Pu, americium $^{241}$Am, curium $^{242}$Cm, $^{244}$Cm, cesium $^{137,134}$Cs, strontium $^{90}$Sr and other elements. The rise of uranium to be leached out of FCM, is observed from 10 mg to 0.1 - 0.7 g per 1 liter of water. As a result, there is a jeopardy of progressive penetration of "block" radioactive water into ground and subterranean parcels outside the object as well as their pollution by not only cesium and strontium but also plutonium, americium that constitutes an ecological hazard for the environment.



Temporary burials are an active sources of radioactive elements (cesium, strontium, plutonium, americium). There are burial places for parts of buildings, radioactive soil, forest, parts of metal constructions and others. Temporary burials were erected under the condition of intensive radioactive background during a short time without proper technical design (the lack of waterproof layers) when deactivating the territory around the Chernobyl plant. At present, the list of temporary burials comprises information on 265 burial places in the exclusion zone among more then 800 existing ones. Data on the temporary burial "Oil storage" containing active wastes with 1672 Cu activity, are represented most entirely. A special attention is attracted by temporary burials near the station "Yaniv" and "Oil storage" [Ledenev A.I. et al (1995), Mishunina I.B. et al (1995)], "Red wood" [Dzhepo S.P. et al (1995), Kopeikin V.A. (1995)]. A peculiarity of the burial place "Oil storage" is its positioning near the river Prypjat' to become the most dangerous in view of radioactive pollution outflow into water parcels. Examination of the temporary burial "Oil storage" has discovered 14 constantly flooded trenches with radioactive element volume 6900 $m^3$. 25 trenches are filled with water during river floods. The total volume of radioactive wastes in flooded temporary burials trenches is 23000 $m^3$ [Zhylinskii V.V. et al (1997)]. In fact, all temporary burials in addition to elements $^{137,134}$Cs, $^{90}$Sr, $^{106}$Ru, $^{125}$Sb and $^{154}$Eu contain plutonium isotopes $^{239}$Pu, $^{240}$Pu. Especial anxiety is felt because the "cocktail" of the above isotopes can find their way through Prypjat' into Dnieper! In [Bogatov S.A. et al (1990)] the problem of americium, plutonium solved in ground water of "Red wood" was emphasized. In particular, the Table 1 exhibits isotope abundance (probes were taken on February 1, 1991) in ground water of "Red wood".

Americium $^{241}$Am is the result of $^{241}$Pu $\beta$-decay with 14.3 years period, which had been present in the 4-th power block by the accident. It had activity 60 times as large as $^{238-240}$Pu. By the accident there had been almost 700 kg of plutonium ($^{239}$Pu – 420 kg, $^{240}$Pu – 175 kg,$^{241}$Pu – 50 kg, $^{242}$Pu – 15 kg) in reactor's active core. Remembering that nuclear fuel was utilized during 3 years before the disaster and $^{241}$Pu $\beta$-decay period one can conclude that the amount of dangerous americium $^{241}$Am

| Radioactive element | Probe Cu/l | Soluble forms after electrodialysis |
|---|---|---|
| $^{90}$Sr | $(860\pm 260)\cdot 10^{-10}$ | $(180\pm 50)\cdot 10^{-10}$ |
| $^{125}$Sb | $< 24\cdot 10^{-10}$ | $< 17\cdot 10^{-10}$ |
| $^{106}$Ru | $< 55\cdot 10^{-10}$ | $< 35\cdot 10^{-10}$ |
| $^{134}$Cs | $(33\pm 5)\cdot 10^{-10}$ | $(24\pm 3)\cdot 10^{-10}$ |
| $^{137}$Cs | $(394\pm 12)\cdot 10^{-10}$ | $(330\pm 8)\cdot 10^{-10}$ |
| $^{144}$Ce | $(55\pm 26)\cdot 10^{-10}$ | $\cdot 10^{-10}$ |
| $^{238}$Pu | $(280\pm 30)\cdot 10^{-12}$ | $(52\pm 8)\cdot 10^{-12}$ |
| $^{239}$Pu,$^{240}$Pu | $(580\pm 60)\cdot 10^{-12}$ | $(101\pm 11)\cdot 10^{-12}$ |
| $^{241}$Am | $(410\pm 90)\cdot 10^{-12}$ | $(87\pm 20)\cdot 10^{-12}$ |
| $^{244}$Cm | $(17\pm 5)\cdot 10^{-12}$ | $(5.4\pm 1.5)\cdot 10^{-12}$ |

Table 1. Isotope abundance in ground water of the "Red wood" on February 1, 1991.

tends to increase this year and in the future. $^{241}$Am is an intensive source of $\alpha$-particles creating neptunium $^{237}$Np. Its high level into water may not exceed $1.9\cdot 10^{-9}$Cu/l, in air - $1.0\cdot 10^{-16}$ Cu/l.

Less intensive sources of radioactive elements in ground water of the zone are permanent burials – "Kompleksnyi", "Pidlisnyi", "Buryakivka", with total activity about 120 thousands Cu . The first two of them are not isolated from the environment absolutely because of violating nature protection rules at their design. In general the amount of active wastes in permanent and temporary burials is estimated to be 1 million m$^3$ with total activity 380 thousands Cu [Baryakhtar V.G. (1995)].

To reduce ecological hazard of permanent burials, temporary burials and radioactive water egress from the object "Shelter" it is necessary first of all to list radioactive objects and estimate their hazard level. In so doing main ways to minimize permanent and temporary burials danger are conservation, termination of water flooding and reburial. To work out the strategy for the number of measures one needs scientific researches on radioactive elements migration in ground water as well as thorough GIS for both permanent and temporary burials.

There is a certain nonconformity between huge quantity of experimental information about the content, distribution and transport of radionuclides in ground, on one hand, and insufficient level of generalization of the information on the basis of ideas on the migration mechanism of ions and molecules carrying radionuclides in ground, on the other



hand. The lack of sufficient analytical information about complex processes of radionuclide transport in ground does not enable to solve a lot of important problems related to choice and taking measures radionuclides not to spread out of shelters. We deal with natural shelters for radionuclides, so the main problems are studying the change of capacity of radiation dose above the surface of ground at migration of radionuclides from the surface inside ground; studying adsorption features of ground (exchange adsorption of cations) and environment (polydispersivity, availability of organic combinations to be humic acids) in which radionuclide migration takes place as well as influence of these features on the interaction of radionuclides with ground; description of complex processes of radionuclide penetration in grounds with known properties during a fixed time.

At present experimental measurements in 30-km zone show the vertical radionuclide migration in ground occurs [Silantjev A.N. et al (1989)], due to molecular diffusion and convection flows of moisture and about 95% radionuclides are located in upper 2-5 cm of ground. It means that radionuclide spreading primarily takes place in fertile layer of ground. Thus another dangerous way of transport through the root system of plants is activated. There is a number of basic moving forces causing active migration of radionuclides in grounds, namely water movement along the ground surface, filtration of atmospheric precipitations inside ground, thermal transfer of moisture under the action of temperature gradients, diffusion of free and adsorbed ions, transfer on colloid particles and through the root system of plants as well as practical activity of people. In particular, the vertical migration of radionuclides $^{90}$Sr, $^{137}$Cs, $^{239,240}$Pu in upper ground layers at the Chernobyl nuclear power plant was considered experimentally and by mathematic modeling in recent papers [Ivanov Yu.A. et al I, II, III (1996), Avramenko V.I. et al (1994)]. It was proposed a mathematic model for radionuclide migration in upper ground layer to be treated as a continuous medium. Another actual aspect of investigations is connected with modeling of radionuclide migration for $^{90}$Sr, $^{137}$Cs from the "Shelter" object into a geologic medium [Kivva S.L. (1997)]. For this purpose a combined model of moisture and radionuclide transport adapted by MSTS code [White M.D., and Nichols W.E. (1992), White M.D., and Nichols W.E. (1993)] in saturated - nonsaturated porous medium was used. The lack of suitable experimental and field studies excluded the possibility of complete validating the model. Mathematic simulation of radionuclide transport by surface water from the vicinity of the Chernobyl nuclear power plant is presented in [Zheleznjak M.J. (1997)] using macroscopic equations of

radionuclide transport hydrodynamics [Onishi Y. et al (1981), Cunge J.A. et al (1986), Hofer H. and Bayer A. (1993)]. The results of modeling have been used to substantiate countermeasures in the Chernobyl zone and to predict seasonal and annual radionuclide dynamics in the cascade of Dnieper storage lakes.

Experimental and theoretical investigations of radionuclide migration in grounds [Eisenbud M. (1966), Prokhorov V.M. (1981)], that had been of active interest before Chernobyl disaster too, show that the main transport way consists in radionuclide diffusion complicated by the ground features such as heterogeneity, porosity, property to adsorb ions and so on. To describe radionuclide diffusion theoretically in grounds, mainly a phenomenological approach [Prokhorov V.M. (1981)] is used on the basis of modifying the diffusion Fick's equations by parameters characterizing the features of ground underconsideration as a continuous medium with constant diffusion coefficient for radionuclides, obtained from experiment. Perhaps, such an approach can not describe complicated migration processes of radionuclides including specific interactions with ions, molecules, colloid particles in ground. The mathematic modeling of radionuclide transport in upper ground layers in papers [Avramenko V.I. et (1994), Ivanov Yu.A. et III. (1996), Kivva S.L. (1997), Zheleznjak M.J. (1997)] is performed on the basis of macroscopic diffusion and hydrodynamic equations in which transport coefficients, namely, diffusion coefficient, adsorption - desorption constants are problem's parameters to be primarily determined experimentally. These parameters, i.e. diffusion coefficients, constants of adsorption, desorption involve mechanisms of radionuclide interaction with water subsystem and the very ground particles. They depend on the temperature, particle concentration, pressure, medium pH and contain more important information on mechanisms of radionuclide interaction with particles of aqueous solutions and ground than diffusion or hydrodynamics equations. In inhomogeneous mediums these parameters depend on spatial coordinates essentially. Hence, to study radionuclide migration statistical approaches are needed. They should be based on equivalent taking into account both of radioactive particles and ions, water molecules, colloid particles whose availability can strongly affect the radionuclide movement. We start a number of papers on the problem of such a clayey barrier that would allow water molecules to diffuse but filtrate radioactive elements.

In section 2 we obtain inhomogeneous diffusion equations for radionuclides in the system "water radioactive solution–ground" by means of Zubarev nonequilibrium statistical operator method [Zubarev D.N. (1971), Zubarev D., Morozov V., Röpke G. (1996)] as well as perform



an approximate analytical calculation of inhomogeneous diffusion coefficients for particles, expressed via interaction potentials, singlet and many-body distribution functions. In section 3 approximate analytical and numerical calculations of singlet and pair distribution functions are carried out for ions $UO_2^{2+}$, $Sr^{2+}$, $Cs^+$ in the system "water radioactive solution–clayey ground", water subsystem being considered as a dielectric medium. In section 4 within the framework of such an ionic approach, numerical computations of inhomogeneous diffusion coefficients for ions $UO_2^{2+}$, $Sr^{2+}$, $Cs^+$ near the interface "water radioactive solution–clayey ground" are done.

## 2. Inhomogeneous diffusion equations for the system "water radioactive solution–clayey ground".

Specific feature of water solutions containing radioactive elements such as uranium ($^{238}$U, $^{235}$U, $^{234}$U) plutonium, strontium, cesium and so on is that the radioactive elements can make up various types of hydrated ions, molecules, double and shifted complexes, mononuclear and polynuclear hydrolysis products, colloid particles [Davydov Yu.P. (1978), Hoekstra H.R. and Kutz J.J. (1954), Milyukov M.S. et al. (1965)]. Experimental investigations for studying radionuclide concentrations in grounds in the 30-km zone [Sobotovich E.V. and Chabanenko S.I. (1990)] point to the availability of a specific type of technogeneous uranium with high content of $^{235}$U isotope which demonstrates anion properties in salt acidic solutions. Obviously, when describing the transfer of radionuclides that are active in forming complexes, hydrolysis and radiolysis in solutions, the concept of ion exchange chromatography [Rachinskij V.V. (1964), Belitskij A.S. and Orlova E.I. (1968), Prokhorov V.M. (1981)] is not correct and it requires a statistical modification taking into account processes and character of interparticle interactions. The formation of complex organic mineral and metal organic combinations in cation, anion, neutral forms in the presence of radionuclides was searched in many works. In [Straub C.P. (1965)] the existence of anion and neutral complexes of radioactive cesium (23% and 9% of general content respectively) was found on the basis of electrodialysis results of river radioactive water. The formation of molecular complexes is a characteristic for radioactive ruthenium (till 50% of general content in solution). All three types of strontium (cation - 66%, anion - 26%, neutral - 8%) were discovered in ground water gathered in regions of $^{90}$Sr precipitation on the territory of Ural (during 1961-1966) [Dibobes M.K. et al (1967)]. The same forms of radionuclide migration are found in the Chernobyl zone [Avramenko

V.I. et (1994), Ivanov Yu.A. et III. (1996), Kivva S.L. (1997), Zheleznjak M.J. (1997), see Baryakhtar V.G. (1995)].

It follows from this rather short review about forms, in which radionuclides are available in water or other solutions, that radionuclides form mainly complicated complexes of three types, namely cation, anion and neutral (colloid particles, polymers). The existence of these forms should be taken into account when describing the radionuclide transport in ground, mixture separation (extraction of radioactive elements) by ultrafiltration.

In ground a part of ions is adsorbed while another one is solved. So the diffusion of every isolated ion consists of consequent processes. These are slow diffusion in adsorbed state, transition into a solution as the result of ionic exchange, faster diffusion in the solution and returning in the adsorbed state. At the same time cation diffusion differs from anion one in grounds. In particular, grounds usually (except phosphate-ions) adsorb anions slightly. That is why the latter diffuse easy in a liquid phase. Solid phase of ground will affect anion transport as a geometrical barrier as well as by double electric layer at the interface if exists. On the contrary, cations are involved into ionic exchange with those adsorbed by solid phase surface.

Here we do not treat slow diffusion in the adsorbed state or ionic exchange processes which could be described as the chemical reactions

$$\mathrm{A} + \mathrm{B} \rightleftarrows \mathrm{B'},$$

where B - adsorption centres on ground's solid phase, A-particles (ions, molecules) of a water solution. Formulating the problem of flow of a water solutions with radioactive elements through clayey ground we consider the statistical model in which the solution is a mixture of water molecules, molecular complexes containing radioactive elements as well as positively ($UO_2^{2+}$, $Sr^{2+}$, $Cs^{+}$, $PuO_2^{2+}$ and their hydrated groups) and negatively $(OH)_p^-$ charged ions. We model clayey ground with dielectric constant $\varepsilon_t$ as a porous medium consisting of certain molecules of t species and water molecules, concentration of the latter being considerably lower in comparison with that in solution. Under the situation diffusion flow of solution particles into clayey ground phase appears due to different concentrations of water molecules in solution and clayey ground. The question arises at what characteristic features of clayey ground water molecules will diffuse through it but ions and molecular complexes containing radioactive elements will be detained and till what depth ions will penetrate into ground due to interaction unless dynamic equilibrium of solutions in two phases comes.

Let us assume water solution as the first phase, volume $V_1$, clayey



ground the second one, volume $V_2$ . We represent the Hamiltonian of the system in the form:

$$H = \sum_{\alpha}\sum_{j=1}^{N_\alpha}\frac{p_j^2}{2m_\alpha} + \frac{1}{2}\sum_{\alpha,\beta}\sum_{j\neq l}\Phi_{\alpha\beta}(\boldsymbol{r}_{jl}) + \sum_{\alpha}\sum_{j=1}^{N_\alpha}\sum_{t=1}^{M}\Phi_{\alpha s}(\boldsymbol{r}_{jt}) + \frac{1}{2}\sum_{s\neq s'}^{M}\Phi(\boldsymbol{r}_{ss'}) + \sum_{s=1}^{M}W_s(z), \qquad (2.1)$$

where $\boldsymbol{p}_j$ - momentum of j-th particle of $\alpha$-species in solution, $m_\alpha$, $m_\alpha$- mass and amount of $\alpha$-species particles. $\alpha$, $\beta$ may denote water molecules, positively, negatively charged ions, neutral molecules with radionuclides. $\Phi_{\alpha\beta}(\boldsymbol{r}_{jl})$- interaction potential for particles of $\alpha$- and $\beta$-species in a solution. $\Phi_{\alpha s}(\boldsymbol{r}_{jt})$- interaction potential between solution particles and clayey ground ones, $\Phi(\boldsymbol{r}_{ss'})$- potential of interaction between ground molecules,

$$W_s(z) = \infty, \quad z_i < 0$$
$$W_s(z) = 0, \quad z_i \geq 0 \qquad (2.2)$$

– Gibbs potential that prevents penetration of s-species molecules into the first phase. Information about the interaction energy between water molecules, ions and molecules containing radionuclides is very important for such a formulation of the problem but at the same time it is a complicated question of the interaction theory. In this paper we use ionic model of water solutions in spatially inhomogeneous case [Kurylyak I.I. and Yukhnovskii I.R. (1982), Kurylyak I.I. (1979), Yukhnovskii I.R., Holovko M.F. and Sovyak E.N. (1982), Kurylyak I.I. (1983)]. We will present a concrete form of interaction potentials in the next chapter when searching for equilibrium distribution functions of molecules and ions in solution in the presence of clayey ground.

The diffusion of particles from solution phase into clayey ground is a process of straight osmosis. To investigate such diffusion processes, nonequilibrium average particle densities $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$ of each species $\alpha$ in two phases are accepted to be parameters of abbreviated description, where

$$\hat{n}^\alpha(\boldsymbol{r}_l) = \sum_{j=1}^{N_\alpha}\delta(\boldsymbol{r}_l - \boldsymbol{r}_j) \qquad (2.3)$$

-microscopic particle density for $\alpha$-species in a phase $l$. $\langle\cdots\rangle^t = \int\frac{(dx)^N}{N!}\cdots$

$\cdots \rho(x^N;t)$ is the definition of mean value with total nonequilibrium distribution function for ions and molecules of solution and clayey ground particles, which satisfies the Liouville equation:

$$\frac{\partial}{\partial t}\rho(x^N;t) + iL_N\rho(x^N;t) = 0, \tag{2.4}$$

$iL_N$ is the Liouville operator appropriate to the Hamiltonian (2.1) of the system:

$$\begin{aligned} iL_N &= \sum_{\alpha,j=1}^{N_\alpha} \frac{\boldsymbol{p}_j}{m_\alpha}\frac{\partial}{\partial \boldsymbol{r}_j} - \sum_{\alpha\beta}\sum_{j\neq k}^{N_\alpha,N_\beta}\left\{\frac{\partial}{\partial \boldsymbol{r}_j}\overline{\Phi}_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k)\frac{\partial}{\partial \boldsymbol{p}_j}\right. \\ &\left. + \frac{\partial}{\partial \boldsymbol{r}_k}\overline{\Phi}_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k)\frac{\partial}{\partial \boldsymbol{p}_k}\right\} - \sum_{\alpha}\sum_{j=1}^{N_\alpha}\frac{\partial}{\partial \boldsymbol{r}_j}\overline{\Phi}_{\alpha s}(\boldsymbol{r}_j,\boldsymbol{r}_t)\frac{\partial}{\partial \boldsymbol{p}_j} \end{aligned}$$

with

$$\overline{\Phi}_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k) = \Phi_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k) + \varphi_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k),$$

$\Phi_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k)$ being a long-range part whereas $\varphi_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k)$ is a short-range part of interaction. They are given in the next chapter. The structure of the Liouville operator reflects the fact that the subsystem "clayey ground" is considered as an equilibrium one. $x_j = \{\boldsymbol{r}_j,\boldsymbol{p}_j\}$ is the coordinate and momentum of $j$-th particle.

In accordance to the abbreviated description of diffusion processes in two-phase system "water solution – clayey ground", the nonequilibrium distribution function will be sought as a functional of nonequilibrium mean particle densities $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$ of each species in two phases

$$\rho(x^N;t) = \rho(\ldots\{\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t\}\ldots) \tag{2.5}$$

It is important to note that the change of quantity $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$ in space and time occurs due to diffusion, interdiffusion caused by intrinsic interactions in the system. Moreover, $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$ can vary for radionuclides such as U, Pu, Am, Sr, Cs and others because of $\alpha$-, $\beta$-, $\gamma$-radiation, processes of nuclear capture and fission when interacting with neutrons in the system. Hence, a more consistent description requires the processes related to $\alpha$-, $\beta$-, $\gamma$-radiation and radionuclide interaction with neutrons to be incorporated in Hamoltonian (2.1). In so doing, besides radionuclide diffusion it is necessary to study in equal terms the diffusion of heat neutrons in the system. It makes the statistical description of particle diffusion in the two-phase system "water solution – clayey ground" much more complicated. So we note in advance that variations



of $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$ related to $\alpha$-, $\beta$-, $\gamma$-radiation and nuclear processes of interaction with neutrons will be taken into account correctly by suitable terms in final diffusion equations and they will not be treated explicitly.

To find a nonequilibrium distribution function $\rho(x^N;t)$ of all particles and, hence, transport equations for $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$ one has to solve Liouville equation (2.4) with appropriate boundary conditions. For this purpose we apply the Zubarev nonequilibrium statistical operator method [Zubarev D.N. (1971), Zubarev D., Morozov V., Röpke G. (1996)] and represent a general solution to the Liouville equation on the basis of ideas of an abbreviated description in the form:

$$\begin{aligned} \rho(x^N;t) &= \rho_q(x^N;t) - \\ &- \int_{-\infty}^{t} e^{\varepsilon(t'-t)}T(t,t')(1-\mathcal{P}_q(t'))iL_N\rho_q(x^N;t')dt' \end{aligned} \tag{2.6}$$

wherein $\varepsilon \to +0$ after the thermodynamic limiting transition, $T(t,t')$ is time evolution operator taking into account projection

$$T(t,t') = \exp_+\left\{\int_{t'}^{t}(1-\mathcal{P}_q(t''))iL_N dt''\right\},$$

$\mathcal{P}_q(t)$ is the Kawasaki-Gunton projection operator, its structure being determined by the quasiequilibrium distribution function $\rho_q(x^N;t)$ for particles in the system.

For the defined set of abbreviated description parameters $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$, quasiequilibrium distribution function, which serves as an auxiliary one to find $\rho_q(x^N;t)$, is derived from extremum of informational entropy [Zubarev D.N. (1971)]:

$$\rho_q(x^N;t) = \exp\left\{-\Phi(t) - \beta\left(H - \sum_\alpha\sum_l\int_{V_l} d\boldsymbol{r}_l\mu_\alpha(\boldsymbol{r}_l;t)\hat{n}^\alpha(\boldsymbol{r}_l)\right)\right\}, \tag{2.7}$$

in which $\Phi(t)$ is found from the normalization condition $\int d\Gamma_N\rho_q(x^N;t) = 1$:

$$\Phi(t) = \ln\int d\Gamma_N \exp\left\{-\Phi(t) - \beta\left(H - \sum_\alpha\sum_l\int_{V_l} d\boldsymbol{r}_l\mu_\alpha(\boldsymbol{r}_l;t)\hat{n}^\alpha(\boldsymbol{r}_l)\right)\right\},$$

$\beta = \frac{1}{k_B T}$, $k_B$ is the Boltzman constant, $T$ is the equilibrium temperature. Parameters $\mu_\alpha(\boldsymbol{r}_l; t)$ are determined from the selfconsistency conditions

$$\langle \hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t = \langle \hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t_q \tag{2.8}$$

to be local chemical potentials for particles of $\alpha$-species in the phase $l$. $\langle\cdots\rangle^t_q = \int \frac{(dx)^N}{N!}\cdots\rho_q(x^N;t)$. In the case of quasiequilibrium distribution function (2.7), the projection operator $\mathcal{P}_q(t)$ in expression (2.6) for total distribution function $\rho(x^N;t)$ has the following form

$$\mathcal{P}_{\rm q}(t)\rho' = \left(\rho_{\rm q}(x^N;t) - \sum_\alpha\sum_l \int\limits_{V_l} d\boldsymbol{r}_l \frac{\delta\rho_{\rm q}(x^N;t)}{\delta\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t}\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t\right)\times\int {\rm d}\Gamma_N\,\rho'$$
$$+\sum_\alpha\sum_l\int\limits_{V_l} d\boldsymbol{r}_l\frac{\delta\rho_{\rm q}(x^N;t)}{\delta\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t}\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t\int d\Gamma_N\hat{n}^\alpha(\boldsymbol{r}_l)\rho'$$

and properties

$$\mathcal{P}_{\rm q}(t)\,\mathcal{P}_{\rm q}(t') = \mathcal{P}_{\rm q}(t), \qquad \mathcal{P}_{\rm q}(t)\rho_{\rm q}(x^N;t) = \rho_{\rm q}(x^N;t),$$
$$\mathcal{P}_{\rm q}(t)(1-\mathcal{P}_{\rm q}(t)) = 0, \qquad \mathcal{P}_{\rm q}(t)\rho(x^N;t) = \rho_{\rm q}(x^N;t).$$

With the help of nonequilibrium distribution function $\rho(x^N;t)$ and relationship $\frac{\partial}{\partial t}\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t = \langle iL_N\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$, we obtain the nonlinear diffusion equation for $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$:

$$\frac{\partial}{\partial t}\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t = \sum_\beta\sum_f\int\limits_{V_f} d\boldsymbol{r}_f\int\limits_{-\infty}^{t} e^{\varepsilon(t'-t)}\frac{\partial}{\partial\boldsymbol{r}_l}D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_f;t,t')$$
$$\times\frac{\partial}{\partial\boldsymbol{r}_f}\mu_\beta(\boldsymbol{r}_f;t')dt', \tag{2.9}$$

where

$$D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_f;t,t') = \int d\Gamma_N(1-\mathcal{P}(t))\hat{\boldsymbol{J}}^\alpha(\boldsymbol{r}_l)$$
$$\times T(t,t')(1-\mathcal{P}(t'))\hat{\boldsymbol{J}}^\beta(\boldsymbol{r}_f)\rho_{\rm q}(x^N;t') \tag{2.10}$$

are generalized diffusion coefficients. $\hat{\boldsymbol{J}}^\alpha(\boldsymbol{r}_l)$ is a vector of flux density for $\alpha$-species particles coupled with $\hat{n}^\alpha(\boldsymbol{r}_l)$ by the discontinuity equation

$$\frac{\partial}{\partial t}\hat{n}^\alpha(\boldsymbol{r}_l) = -\frac{\partial}{\partial\boldsymbol{r}_l}\hat{\boldsymbol{J}}^\alpha(\boldsymbol{r}_l),$$



$\mathcal{P}(t)$ is the time dependent Mori projection operator that acts on dynamic variables,

$$\mathcal{P}(t)\hat{a}(\boldsymbol{r}) = \langle\hat{a}(\boldsymbol{r})\rangle^t_{\rm q} + \sum_\alpha\sum_l\int\limits_{V_l} d\boldsymbol{r}_l\frac{\delta\langle\hat{a}(\boldsymbol{r})\rangle^t_{\rm q}}{\delta\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t}\left(\hat{n}^\alpha(\boldsymbol{r}_l) - \langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t\right),$$

with the properties $\mathcal{P}(t)\mathcal{P}(t') = \mathcal{P}(t)$, $\mathcal{P}(t)(1-\mathcal{P}(t)) = 0$, $\mathcal{P}(t)\hat{n}^\alpha(\boldsymbol{r}_l) = \hat{n}^\alpha(\boldsymbol{r}_l)$.

We have obtained the system of nonlinear equations (2.9) describing nonlinear nonstationary diffusion processes in which moving forces are gradients of chemical potentials $\frac{\partial}{\partial\boldsymbol{r}_l}\mu_\alpha(\boldsymbol{r}_l;t)$ of particles in solution in the phase $l$.

We consider below linear stationary diffusion processes in the system "water radionuclide solution – ground". They can be governed by diffusion equations in linear approximation on gradients of local chemical potentials $\mu_\alpha(\boldsymbol{r}_l;t)$. Then eliminating with the help of selfconsistency conditions (2.8) and (2.7) local chemical potentials from (2.9) in linear approximation on $\delta\mu_\alpha(\boldsymbol{r}_l;t)$ ($\delta\mu_\alpha(\boldsymbol{r}_l;t) = \mu_\alpha(\boldsymbol{r}_l;t) - \mu_\alpha(\boldsymbol{r}_l)$) we arrive at the system of diffusion equations which after a correct consideration of terms connected with the variation of $\langle\hat{n}^\alpha(\boldsymbol{r}_l)\rangle^t$ due to nuclear transformations is modified to yield:

1. Water solution region, volume $V_1$:

$$\frac{\partial}{\partial t}\delta n^\alpha(\boldsymbol{r}_1;t) = -\sum_\beta\int\limits_{V_1} d\boldsymbol{r}'_1\int\limits_{-\infty}^{t} e^{\varepsilon(t'-t)}\frac{\partial}{\partial\boldsymbol{r}_1}D^{\alpha\beta}(\boldsymbol{r}_1,\boldsymbol{r}'_1;t,t')$$
$$\times\frac{\partial}{\partial\boldsymbol{r}'_1}\delta n^\beta(\boldsymbol{r}'_1;t')dt'$$
$$-\sum_\beta\int\limits_{V_2} d\boldsymbol{r}'_2\int\limits_{-\infty}^{t} e^{\varepsilon(t'-t)}\frac{\partial}{\partial\boldsymbol{r}_1}D^{\alpha\beta}(\boldsymbol{r}_1,\boldsymbol{r}'_2;t,t')$$
$$\times\frac{\partial}{\partial\boldsymbol{r}'_2}\delta n^\beta(\boldsymbol{r}'_2;t')dt'$$
$$-\sum_\beta A_1^{\alpha\beta}(\boldsymbol{r}_1,t)\delta n^\beta(\boldsymbol{r}_1;t) - \sum_\beta A_1^\alpha(\boldsymbol{r}_1,t)\delta n^\beta(\boldsymbol{r}_1;t) \quad , \tag{2.11}$$

where $D^{\alpha\beta}(\boldsymbol{r}_1,\boldsymbol{r}'_1;t,t')$ are generalized diffusion coefficients for ions and molecules, describing transport in the volume $\rm V_1$ of water solution. $D^{\alpha\beta}(\boldsymbol{r}_1,\boldsymbol{r}'_2;t,t')$ describe diffusion of ions and molecules from the volume $\rm V_1$ into the volume $\rm V_2$ – clayey ground.

2. Clayey ground, volume $V_2$,

$$\frac{\partial}{\partial t}\delta n^{\alpha}(\boldsymbol{r}_2;t) = -\sum_{\beta}\int\limits_{V_1} d\boldsymbol{r}_1' \int\limits_{-\infty}^{t} e^{\varepsilon(t'-t)}\frac{\partial}{\partial \boldsymbol{r}_2}D^{\alpha\beta}(\boldsymbol{r}_2,\boldsymbol{r}_1';t,t') \times\frac{\partial}{\partial \boldsymbol{r}_1'}\delta n^{\beta}(\boldsymbol{r}_1';t')dt' - \sum_{\beta}\int\limits_{V_2} d\boldsymbol{r}_2' \int\limits_{-\infty}^{t} e^{\varepsilon(t'-t)}\frac{\partial}{\partial \boldsymbol{r}_2}D^{\alpha\beta}(\boldsymbol{r}_2,\boldsymbol{r}_2';t,t') \times\frac{\partial}{\partial \boldsymbol{r}_2'}\delta n^{\beta}(\boldsymbol{r}_2';t')dt' - \sum_{\beta}A_2^{\alpha\beta}(\boldsymbol{r}_2,t)\delta n_2^{\beta}(\boldsymbol{r}_2;t) - \sum_{\beta}A_2^{\alpha}(\boldsymbol{r}_2,t)\delta n_2^{\beta}(\boldsymbol{r}_2;t) \quad , \quad (2.12)$$

Correlation functions $D^{\alpha\beta}(\boldsymbol{r}_2,\boldsymbol{r}_1';t,t')$ describe diffusion of ions and molecules from clayey ground phase into solution and $D^{\alpha\beta}(\boldsymbol{r}_2,\boldsymbol{r}_2';t,t')$ – diffusion of ions and molecules in clayey ground. In the system of transport equations (2.11), (2.12) $\delta n^{\alpha}(\boldsymbol{r}_l;t)$ are nonequilibrium averages of particle density fluctuations for $\alpha$-species particles in a phase $l$:

$$\delta n^{\alpha}(\boldsymbol{r}_l;t) = \langle n^{\alpha}(\boldsymbol{r}_l)\rangle^{t} - \langle n^{\alpha}(\boldsymbol{r}_l)\rangle_0, \quad (2.13)$$

$\rho_0(x^N;P)$ – equilibrium distribution function for the two phase system:

$$\rho_0(x^N) = \Xi^{-1}\exp\Big\{-\beta(H-\sum_{\alpha}\sum_{l=1}^{2}\int\limits_{V_l} d\boldsymbol{r}_l\,\mu_{\alpha}(\boldsymbol{r}_l)\,\hat{n}_l^{\alpha}(\boldsymbol{r}_l)\Big\}\,, \quad (2.14)$$

$$\Xi = \int\frac{(dx)^N}{N!}\exp\Big\{-\beta(H-\sum_{\alpha}\sum_{l=1}^{2}\int\limits_{V_l} d\boldsymbol{r}_l\,\mu_{\alpha}(\boldsymbol{r}_l)\,\hat{n}_l^{\alpha}(\boldsymbol{r}_l)\Big\}, \quad (2.15)$$

$\Xi$ – partition function in which inhomogeneity of the system "water solution-clayey ground"is taken into account by the term

$$-\sum_{\alpha}\sum_{l=1}^{2}\int\limits_{V_l} d\boldsymbol{r}_l\,\mu_{\alpha}(\boldsymbol{r}_l)\,\hat{n}_l^{\alpha}(\boldsymbol{r}_l)$$

with $\mu_{\alpha}(\boldsymbol{r}_l)$ as chemical potential of $\alpha$-species particles in a phase $l$. $H$ - the system's Hamiltonian, its potential part will be defined in the next chapter.



The last two terms in (2.11) and (2.12) describe a radionuclide density variation in time because of nuclear transformations promoted by neutrons and spontaneous decay. The first one describes of $\alpha$-species radionuclide from all the other nuclei $\beta$ as a result of (n,$\gamma$), $\alpha$-, $\beta$-, $\gamma$- decays as well as (n,f) fission if the relevant $\alpha$-species radionuclide belongs to fission products. The second term describes a decay (disappearance) of $\alpha$-species radionuclide under the action of neutrons and natural radioactive decay. Functions $A^{\alpha\beta}(\boldsymbol{r},t)$, $A^{\alpha}(\boldsymbol{r},t)$ are rates for reactions to be represented

$$A^{\alpha\beta}(\boldsymbol{r},t) = \int\limits_0^{\infty} J(\boldsymbol{r};E;t)\,\sigma_{\alpha\beta}(E)\,dE + L_{\alpha\beta}\,\lambda_{\alpha\beta} \quad , \quad (2.16)$$

$$A^{\alpha}(\boldsymbol{r},t) = \int\limits_0^{\infty} J(\boldsymbol{r};E;t)\,\sigma_{\alpha}(E)\,dE + \lambda_{\alpha\beta} \quad , \quad (2.17)$$

Here $J(\boldsymbol{r};E;t)$ is the spectrum of neutron density fluxes in a point $\boldsymbol{r}$ at the moment $t$; $\sigma_{\alpha\beta}(E)$ is the microscopic section of $\alpha$-species radionuclide formation at catching neutron with energy $E$ by $\beta$-species nucleus. $\sigma_{\alpha}(E)$ is the microscopic section of neutron capture with energy $E$ by $\alpha$-species nucleus. $L_{\alpha\beta}$ is the probability for creation of $\alpha$-species radionuclide at radioactive decay of $\beta$-species nucleus. $\lambda_{\alpha}$,$\lambda_{\beta}$ are decay constants of $\alpha$-, $\beta$-species nuclei. The time dependence of reaction rates $A^{\alpha\beta}(\boldsymbol{r},t)$,$A^{\alpha}(\boldsymbol{r},t)$ is determined by that of spectrum of neutron density fluxes $J(\boldsymbol{r};E;t)$. Neutron spatial energetic distribution in its turndepends on spatial distribution and concentration of radionuclides in the system so actually neutron field and time variation of density of actinides or fission products are closely related. One has to note that the equation system for diffusion of solution's ions and molecules (2.11), (2.12) does not take into account the transport processes, related to rotational degrees of freedom of water molecules and molecular complexes with radionuclides but it will be considered through equilibrium distribution functions for ions and molecules at calculation of generalized diffusion coefficients.

$$D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_f';t,t') = \sum_{\xi}\sum_{\gamma}\int\limits_{V_{\xi}} d\boldsymbol{r}_{\xi}''\,\langle(1-P_0)\hat{\boldsymbol{J}}_l^{\alpha}(\boldsymbol{r}_l) \times T(t,t')(1-P_0)\hat{\boldsymbol{J}}_{\xi}^{\gamma}(\boldsymbol{r}_{\xi}'')\rangle_0\Big(\tilde{F}^{-1}(\boldsymbol{r}'',\boldsymbol{r}')\Big)_{\alpha\beta}^{\xi f}\,, \quad (2.18)$$

where

$$T_0(t,t') = \exp\{(1-P_0)(t'-t)\,iL_N\}$$

is the time evolution operator taking into account projection; $P_0$ is linear Mori projection operator that acts on dynamic variables:

$$P_0 A = \langle A \rangle_0 + \sum_{\alpha\beta} \sum_{lf} \int d\boldsymbol{r}_l d\boldsymbol{r}'_f \, \langle A \hat{n}_l^\alpha(\boldsymbol{r}_l) \rangle_0 \left( \tilde{F}^{-1}(\boldsymbol{r}, \boldsymbol{r}') \right)_{lf}^{\alpha\beta} \hat{n}_f^\beta(\boldsymbol{r}'_f) \, .$$

$\left( \tilde{F}^{-1}(\boldsymbol{r}, \boldsymbol{r}') \right)_{\alpha\beta}^{lf}$ are elements of the matrix $\tilde{F}^{-1}(\boldsymbol{r}, \boldsymbol{r}')$, inverted to $\tilde{F}(\boldsymbol{r}, \boldsymbol{r}')$, consisting of elements $f_{\alpha\beta}^{lf}(\boldsymbol{r}, \boldsymbol{r}')$:

$$f_{\alpha\beta}^{lf}(\boldsymbol{r}, \boldsymbol{r}') \;=\; \langle \hat{n}^\alpha(\boldsymbol{r}_l) \, \hat{n}^\beta(\boldsymbol{r}'_f) \rangle_0 \qquad (2.19)$$

to be pair equilibrium distribution functions for ions and molecules in two phases, that may be calculated from partition function (2.15).

To analyze the system of diffusion equations for ions and molecules (2.11), (2.12) in the system "water solution - clayey ground" one has to calculate generalized coefficients (2.18) for two phase system. In the paper [Tokarchuk M.V. et al (1993)] their analytical calculation in Gauss approximation of time dependence was carried out:

$$D^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}_f; t, t') = \lambda_0^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}_f) \, \exp \left\{ - \frac{\overline{\lambda}_2^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}_f)}{2!} \, (t - t')^2 \right\}, \qquad (2.20)$$

$$\overline{\lambda}_2^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}_f) = \frac{\lambda_2^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}_f)}{\lambda_0^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}_f)}, \qquad (2.21)$$

where the zeroth moment and the second one are expressed in terms of equilibrium distribution functions and interaction potentials for ions and molecules:

$$\lambda_0^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}'_f) \;=\; \sum_\xi \sum_\gamma \int\limits_{V_\xi} d\boldsymbol{r}''_\xi \, \langle (1 - P_0) \hat{\boldsymbol{J}}_l^{\,\alpha}(\boldsymbol{r}_l) (1 - P_0) \hat{\boldsymbol{J}}_\xi^{\,\gamma}(\boldsymbol{r}''_\xi) \rangle_0$$

$$\times \left( \tilde{F}^{-1}(\boldsymbol{r}'', \boldsymbol{r}') \right)_{\xi f}^{\gamma\beta} = \frac{1}{m_\alpha} K_B T f_l^\alpha(\boldsymbol{r}_l) \left( \tilde{F}^{-1}(\boldsymbol{r}, \boldsymbol{r}') \right)_{lf}^{\alpha\beta} \, , \qquad (2.22)$$

$$\lambda_2^{\alpha\beta}(\boldsymbol{r}_l, \boldsymbol{r}'_f) \;=\; \sum_\xi \sum_\gamma \int\limits_{V_\xi} d\boldsymbol{r}''_\xi \, \langle (1 - P_0) \hat{\boldsymbol{J}}_l^{\,\alpha}(\boldsymbol{r}_l) \, [(1 - P_0) i L_N]^2$$

$$\times (1 - P_0) \hat{\boldsymbol{J}}_\xi^{\,\gamma}(\boldsymbol{r}''_\xi) \rangle_0 \left( \tilde{F}^{-1}(\boldsymbol{r}'', \boldsymbol{r}') \right)_{\xi f}^{\gamma\beta} \, . \qquad (2.23)$$

After some tedious algebra we obtain

$\overline{\lambda}_2^{\alpha\alpha}(\boldsymbol{r}_l, \boldsymbol{r}_f) =$



$$= \frac{1}{3m_\alpha} \Big( \frac{\partial^2}{\partial \boldsymbol{r}_l^2} U_\alpha(\boldsymbol{r}_l) + \sum_\gamma \rho_\gamma \int \frac{\partial^2}{\partial \boldsymbol{r}_l^2} \Phi_{\alpha\gamma}(\boldsymbol{r}_l, \boldsymbol{r}_f) \, \frac{F_2^{\alpha\gamma}(\boldsymbol{r}_l, \boldsymbol{r}_f)}{f_1^\alpha(\boldsymbol{r}_l)} \, d\boldsymbol{r}_f \Big) +$$

$$+ \frac{5}{3m_\alpha} \Big( \frac{\partial^2}{\partial \boldsymbol{r}_l^2} U_\alpha(\boldsymbol{r}_l) + \sum_\gamma \rho_\gamma \int \Big( \frac{\partial^2}{\partial \boldsymbol{r}_l^2} \Phi_{\alpha\gamma}(\boldsymbol{r}_l, \boldsymbol{r}_f) \, \frac{F_2^{\alpha\gamma}(\boldsymbol{r}_l, \boldsymbol{r}_f)}{f_1^\alpha(\boldsymbol{r}_l)} +$$

$$+ \frac{\partial}{\partial \boldsymbol{r}_l} \Phi_{\alpha\gamma}(\boldsymbol{r}_l, \boldsymbol{r}_f) \, \frac{\partial}{\partial \boldsymbol{r}_l} \frac{F_2^{\alpha\gamma}(\boldsymbol{r}_l, \boldsymbol{r}_f)}{f_1^\alpha(\boldsymbol{r}_l)} \Big) \, d\boldsymbol{r}_f \Big). \qquad (2.24)$$

As it can be seen, the zeroth moment and the second one are expressed in terms of one- and two-body equilibrium distribution functions together with interaction potentials for solution's ions and molecules and clayey ground molecules. As a result from investigation of generalized diffusion coefficients for ions and molecules (2.20) the problem appears to calculate the distribution functions of ions and molecules for the two phase system "water solution - clayey ground".

## 3. Approximate calculation of equilibrium distribution functions in a two phase system

Active elements are supposed to be present in water in small amounts. Within this model, the active elements particles ($UO_2^{2+}$, $Cs^+$, $Sr^{2+}$) are considered as charged hard spheres having the overall charge to be compensated according to the electroneutrality condition by negative $OH^-$-groups in continuous medium with the dielectric constant $\varepsilon_p = 81$ (water).

To investigate the diffusion of active particles it is necessary to know the structure of a solution near clayey wall. The problem reduces to a treatment of the model "aqueous radioactive solution-clayey wall", the second phase being a continuous medium with the dielectric constant $\varepsilon_c = 1 \div 15$ (clay). In this approach, profiles are affected by both structural ordering caused by own sizes and the presence of surface and electrostatic images to be most pronounced at small distances. However, their consecutive consideration is fairly intricate problem. To this end we used the first equation of BBGKY-chain modified to describe image charges. These latter are assumed to be fictitious charged particles inside the wall (clay, volume $V_2$), which have charges $\frac{\varepsilon_p - \varepsilon_c}{\varepsilon_p + \varepsilon_c} Z_i e$ and sizes $\sigma_i$, wherein $Z_i e$ , $\sigma_i$ are charges and sizes of particles in the solution (volume $V_1$).

Taking the above model into account we can write down explicitly

the potentials entering the Liouville operator:

$$\varphi_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k)=\varphi_{\alpha\beta}(|\boldsymbol{r}_j-\boldsymbol{r}_k|)=\varphi^{\rm hs}_{\alpha\beta}(r_{jk}), \tag{3.1}$$

$$\varphi^{\rm hs}_{\alpha\beta}(r_{jk}) \;=\; \infty, \quad r_{jk}<\frac{\sigma_j+\sigma_k}{2}, \tag{3.2}$$

$$\varphi^{\rm hs}_{\alpha\beta}(r_{jk}) \;=\; 0, \quad r_{jk}\geq\frac{\sigma_j+\sigma_k}{2} \tag{3.3}$$

– hard sphere potential; $\sigma_j$ stands for diameter of $j$-th particle.

$$\Phi_{\alpha\beta}(\boldsymbol{r}_j,\boldsymbol{r}_k)=\Phi^{\rm C}_{\alpha\beta}(r_{jk})=\frac{Z_je\;Z_ke}{r_{jk}} \tag{3.4}$$

–Coulomb interaction between ions; $r_{jk}$–distance between particles.

The potential $\overline{\Phi}_{\alpha s}(\boldsymbol{r}_j,\boldsymbol{r}_k)$ of a particle interaction with wall reduces in essence to that with electrostatic images. It has the obvious form

$$\overline{\Phi}_{\alpha s}(\boldsymbol{r}_j,\boldsymbol{r}_k)=\Phi^{\rm im}_{\alpha\beta}(r'_{jk})=\frac{\varepsilon_p-\varepsilon_c}{\varepsilon_p+\varepsilon_c}\frac{Z_je\;Z_ke}{r'_{jk}}, \tag{3.5}$$

where primed $r_{jk}$ means the $k$-th ion's image is in different phase 2 (ground) rather than in that of ionic solution. In the case of interaction with own image, the potential becomes singlet and dependent on the only distance from the wall $z_1$. We denote it as $U^\alpha_1(z_1)$, it is completely defined below by (3.8).

If to apply the method given in [Bogoliubov N.N. (1970)] and take into account symmetry along the wall we arrive at the following equation coupling singlet and pair distribution functions:

$$\begin{aligned}&\frac{\partial f^\alpha_1(z_1)}{\partial z_1}+\frac{\partial U^\alpha_1(z_1)}{\partial z_1}f^\alpha_1(z_1)+\\&+\sum_\beta\rho_\beta\int\limits_V\partial\boldsymbol{r}_{2\beta}\;\frac{\partial}{\partial z_1}(\varphi^{\rm hs}_{\alpha\beta}(r_{12})+\Phi^{\rm C}_{\alpha\beta}(r_{12}))F^{\alpha\beta}_2(z_{1\alpha},z_{2\beta},r_{2\beta})+\\&+\sum_\beta\rho'_\beta\int\limits_{V'}\partial\boldsymbol{r}'_{2\beta}\;\frac{\partial}{\partial z_1}\Phi^{\rm im}_{\alpha\beta}(r'_{12})F^{\alpha\beta}_2(z_{1\alpha},z'_{2\beta},r'_{2\beta})=0,\end{aligned} \tag{3.6}$$

wherein the interaction potentials have just been discussed, $f^\alpha_1$, $F^{\alpha\beta}_2$ are singlet and pair distribution functions entering also (2.24). $\rho_\beta$,$\rho'_\beta$ are the densities of $\beta$-species particles and their images. (Clearly $\rho_\beta=\rho'_\beta$). It is easily seen that the second integral term takes into account image effects on equal terms. But at small concentrations integral terms are minor (pair correlation is not essential) so ionic distribution near the



surface is determined by the potential $U^\alpha_1(z_1)$ that is advantageous to use as screened potential. The problem of point particles screened potential near a hard wall has a rigorous analytical solution [Blum L. (1975), Yukhnovskii I.R., Kurylyak I.I. and Sovyak E.N. (1979)]. In the case of ion-ion interaction the result is:

$$g(\boldsymbol{r}_1,\boldsymbol{r}_2)=\mp\frac{Z_1Z_2e^2}{\varepsilon_p}\left\{\frac{e^{-\kappa r_{12}}}{r_{12}}+\frac{\varepsilon_p-\varepsilon_c}{\varepsilon_p+\varepsilon_c}\,\frac{e^{-\kappa r'_{12}}}{r'_{12}}\right\}, \tag{3.7}$$

with $r'_{12}$–distance between the first particle and the image of the second

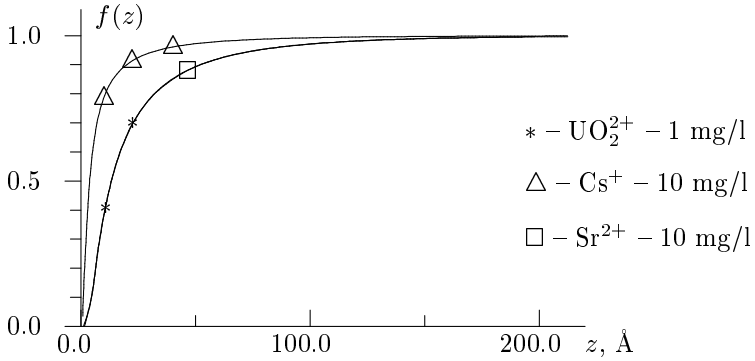


Figure 1. Qualitative distribution functions of active particles near the interface "aqueous solution–clayey wall".

one. It is evident that the potential consists of a bulk part dependent on $r_{12}$ and a surface one. The problem of bulk screened potential evaluation for arbitrary amount of ions having distinct sizes and valences is solved in [Blum L. (1975)]. We make use of the result for small concentrations in view of that the potential of an inhomogeneous system is expressed through bulk ones as is seen from (3.7). Thus

$$U^\alpha_1(z_1)=\frac{\varepsilon_p-\varepsilon_c}{\varepsilon_p+\varepsilon_c}\frac{(Z^\alpha_1e)^2}{\varepsilon_p}\frac{1}{2z_1}\left[\frac{2\Gamma}{\kappa}\right]^2\exp\{-2\Gamma(2z_1-\sigma_\alpha)\}, \tag{3.8}$$
$$z>\sigma_\alpha/2,$$

in which $\kappa$ is the standard Debye radius, $2\Gamma$ is new screening radius to

be specified by the following equation system

$$4\Gamma^2 = \frac{e^2}{\varepsilon_p}\sum_\beta \rho_\beta X_\beta^2, \quad X_\beta = \frac{Z_\beta - \frac{\pi}{2}\frac{\sigma_\beta^2}{1-\eta}P_m}{1+\Gamma\sigma_\beta},$$

$$P_m = \frac{\sum_\beta \frac{\rho_\beta \sigma_\beta Z_\beta}{1+\Gamma\sigma_\beta}}{1+\frac{\pi}{2(1-\eta)}\sum_\beta \frac{\rho_\beta\sigma_\beta^3}{1+\Gamma\sigma_\beta}}, \quad \eta = \frac{\pi}{6}\sum_\beta \rho_\beta\sigma_\beta^3, \quad \rho_\beta = N_\beta/V,$$

wherein $\beta$ stands for active ions including negative $OH^-$-groups with the density to satisfy the general electroneutrality condition for the system

$$\sum_\beta \rho_\beta Z_\beta^2 = 0.$$

Then the solution of (3.6) under the condition $f_\alpha(\infty)=1$ is the following

$$f_\alpha(z) = \exp(-U_1^\alpha(z)). \tag{3.9}$$

For pair distribution function the superposition approximation yields:

$$F_2^{\alpha\beta}(z_1,z_2,r) = f_\alpha(z_1)f_\beta(z_2)F_2^{\alpha\beta}(r) = \\ = \exp(-U_1^\alpha(z_1) - U_1^\beta(z_2) + g(z_1,z_2,r)) \tag{3.10}$$

Figures 1, 2 show qualitative profiles for ions $UO_2^{2+}$, $Cs^+$, $Sr^{2+}$ to be estimated at various concentrations with the aid of (3.9). It can be seen that small concentrations of active elements result in essential wall effect even at long distance while large ones bring about screening due to which the system quickly attains bulk properties. Moreover in the case of small concentrations, the ionic behavior is determined by their valences only– curves for bivalent uranilum and strontium coincide but differ from that for univalent cesium. At high concentrations ionic sizes reveal themselves –uranilum and strontium assume individual distribution features at the interface "aqueous radioactive solution–clayey wall".

## 4. Calculation of inhomogeneous diffusion coefficient for ions $UO_2^{2+}$, $Cs^+$, $Sr^{2+}$ in the system "water radioactive solution – clayey ground"

The obtained distribution functions (33), (34) for ions $UO_2^{2+}$, $Cs^+$are qualitatively correct as to a structural distribution near the surface "water radioactive solution – clayey ground". Using them we are going to



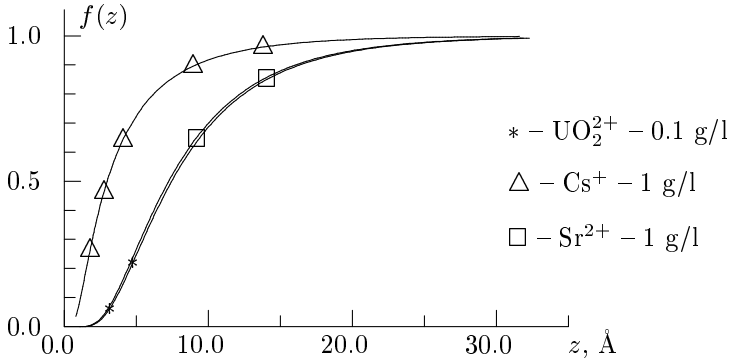


Figure 2. The same as in Fig.1 but for distinct concentrations.

perform the calculation of inhomogeneous diffusion coefficients for ions $UO_2^{2+}$, $Cs^+$, $Sr^{2+}$ in the aqueous solution interacting with glassy nuclear magma. For this purpose only Markovian transport processes of ions in solutions will be considered. Then from equations (11)-(12) the mean flux of $k$-species particles in the phase $l$ is represented by the following expression:

$$\boldsymbol{j}^\alpha(\boldsymbol{r}_l;t) = -\sum_\beta \sum_{l'} \int_{V_{l'}} d\boldsymbol{r}_l' \, D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_l') \frac{\partial}{\partial \boldsymbol{r}_l'}\delta n^\beta(\boldsymbol{r}_l';t), \tag{4.1}$$

where $D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_l')$ are normalized inhomogeneous diffusion coefficients to be related to generalized coefficients $D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_l';t,t')$ (18) in the way:

$$D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_l') = \int_0^\infty d\tau D^{\alpha\beta}(\boldsymbol{r}_l,\boldsymbol{r}_l';\tau). \tag{4.2}$$

We omit phase indices, because only the phase of aqueous solution will be treated. In Gaussian approximation (2.20) for $D^{\alpha\beta}(\boldsymbol{r},\boldsymbol{r}')$ we have

$$D^{\alpha\beta}(\boldsymbol{r},\boldsymbol{r}') = \int_0^\infty d\tau \lambda_0^{\alpha\beta}(\boldsymbol{r},\boldsymbol{r}') \exp\left\{-\frac{\overline{\lambda}_2^{\alpha\beta}(\boldsymbol{r},\boldsymbol{r}')}{2!}\tau^2\right\} = \tag{4.3}$$

$$= \lambda_0^{\alpha\beta}(\boldsymbol{r},\boldsymbol{r}')\sqrt{\frac{\pi}{2\overline{\lambda}_2^{\alpha\beta}(\boldsymbol{r},\boldsymbol{r}')}}$$

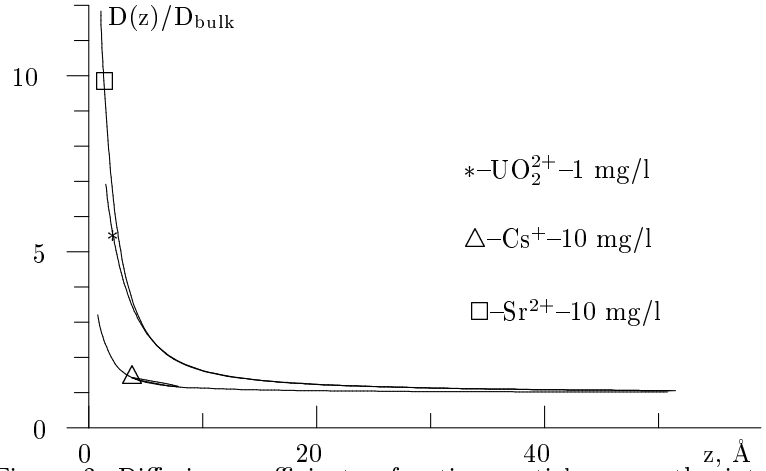


Figure 3. Diffusion coefficients of active particles near the interface "aqueous solution–clayey wall".

The function $\left[\tilde{F}^{-1}(\boldsymbol{r}_l, \boldsymbol{r}_{l'})\right]_{kk'}$ in (2.18) is defined in terms of direct correlation function $c_2^{\alpha\beta}(\boldsymbol{r}, \boldsymbol{r}')$:

$$\left[\tilde{F}^{-1}(\boldsymbol{r}, \boldsymbol{r}'_l)\right]_{\alpha\beta} = \frac{\delta_{\alpha\beta}\delta(\boldsymbol{r} - \boldsymbol{r}')}{f_1^{\beta}(\boldsymbol{r}')} - c_2^{\alpha\beta}(\boldsymbol{r}, \boldsymbol{r}'). \tag{4.4}$$

Along with (38) the expression for fluxes (35) takes the following form

$$\hat{\boldsymbol{j}}^{\alpha}(\boldsymbol{r}; t) = D^{\alpha\alpha}(\boldsymbol{r})\frac{\partial}{\partial \boldsymbol{r}}\delta n^{\alpha}(\boldsymbol{r}; t) + \tag{4.5}$$
$$+ \sum_{\beta}\int_V d\boldsymbol{r}'\, \overline{D}^{\alpha\beta}(\boldsymbol{r}, \boldsymbol{r}')\frac{\partial}{\partial \boldsymbol{r}'}\delta n^{\beta}(\boldsymbol{r}'; t),$$

where

$$D^{\alpha\alpha}(\boldsymbol{r}) = D^{\alpha}(\boldsymbol{r}) = \frac{k_{\mathrm{B}}T}{m_{\alpha}}\sqrt{\frac{\pi}{2\overline{\lambda}_2^{\alpha\alpha}(\boldsymbol{r})}} \tag{4.6}$$

is inhomogeneous selfdiffusion coefficient for $\alpha$-species particle with respect to nuclear magma–aqueous solution interface, whereas

$$\overline{D}^{\alpha\beta}(\boldsymbol{r}, \boldsymbol{r}') = \frac{k_{\mathrm{B}}T}{m_{\alpha}}f_1^{\alpha}(\boldsymbol{r})c_2^{\alpha\beta}(\boldsymbol{r}, \boldsymbol{r}')\sqrt{\frac{\pi}{2\lambda_2^{\alpha\beta}(\boldsymbol{r}, \boldsymbol{r}')}} \tag{4.7}$$



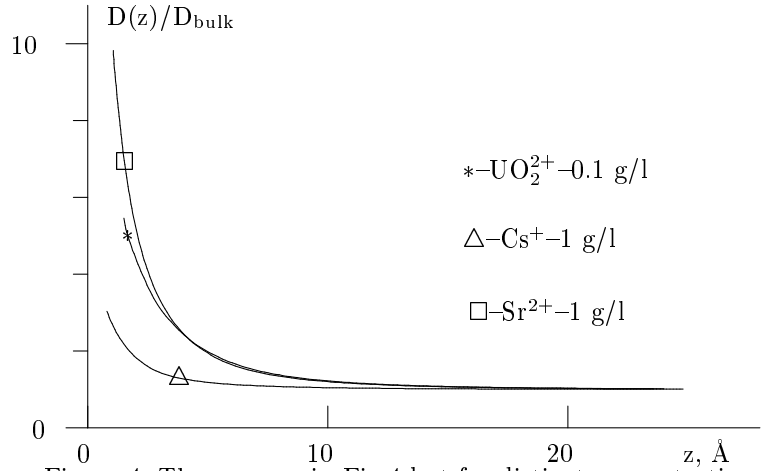


Figure 4. The same as in Fig.4 but for distinct concentrations.

is the inhomogeneous interdiffusion coefficient for $\alpha$- and $\beta$-species particles with respect to the interface.

The interdiffusion coefficient is determined by direct correlation function $c_2^{\alpha\beta}(\boldsymbol{r}, \boldsymbol{r}')$ which is an auxiliary one in the theory of integral equations. It is usually accepted in some form. To avoid supplement approximations we have calculated only selfdiffusion coefficients for the same concentrations at which distribution functions were evaluated. The ratio of selfdiffusion coefficient to bulk value is plotted in figures 3, 4 as a function of the distance to ground wall. The general behaviour of the coefficient is determined by the valency like it was for density profiles (Figures 1 and 2). Far from the interface the coefficient tends unity, namely, the system attains bulk properties. For ions of cesium this occurs faster. Near the surface the coefficients increase, ionic mobility is greater. At the same time, the electrostatic repulsion prevents the approach of radioactive ions to the interface even if water can penetrate through clayey wall. Due to large diffusion coefficient in this region, the mobility of ions is high, hence, they easily return to water solution.

## 5. Conclusion

We have considered the modern picture of radionuclide migration from the "Shelter" object and temporary burials of radwastes into soil and ground water of Chernobyl zone. This problem was very actively dis-

cussed in recent papers [Kivva S.L. (1997), Zheleznjak M.I. (1997)], where mathematical models proposed to predict the radionuclide migration, depend on parameters, namely, coefficients of diffusion, adsorption, desorbtion to be accepted as constants for individual homogeneous layers composing all the system. As a rule, these parameters are determined experimentally at fixed temperature, concentrations and pH of medium. This restricts the mathematical modeling of migration in wide region of parameters' values to a great extent.

We have proposed a statistical diffusion model to calculate structural distribution and inhomogeneous diffusion coefficients for ions $UO_2^{2+}$, $Cs^+$, $Sr^{2+}$ in the system "clayey ground–water solution", taking into account a spatial inhomogeneity, electrostatic effects, in particular ionic images, and dimension effects related to particle sizes. The model can be said to be still a qualitative one as it does not treat explicitly the interaction of solution ions and molecules with ground particles that would correspond the consideration of its absorption and desorption properties. In our approach, the problem can be solved by the introduction of appropriate potentials for ground-solution interaction in the Hamiltonian (2.1). In so doing, it is clear that the behavior of inhomogeneous diffusion coefficients $D_\alpha(z)$ near the very surface would differ from that plotted in figures 4 and 5. We are going to perform such calculations in future papers that is important in view of selection of a clayey screen to terminate the radionuclide migration. The formulas obtained for inhomogeneous diffusion coefficients may be computed versus temperature, concentration and provide the consideration of details of particle interaction and structural distribution in the system. The analytical expressions for inhomogeneous diffusion coefficients might be used as input parameters in equations of diffusion and hydrodynamics for inhomogeneous mediums, in particular those described in [Kivva S.L. (1997), Zheleznjak M.I. (1997)]. At the same time, both inhomogeneity and particle interaction will be considered.

In future papers we hope to complicate the diffusion including the solvent, i.e. rotational and translational motions of water molecules as well as the interaction of solution molecules with ground surface, in particular bentonite clay.

## Acknowledgements

This work has been supported by International Association for Promotion of Cooperation between NIS countries and European Community (the grant INTAS - Ukraine-95-0133).



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Статистична теорія процесів дифузії радіонуклідів у грунтових та підземних водах

Роботу отримано 7 грудня 1998 р.

Затверджено до друку Вченою радою ІФКС НАН України

Рекомендовано до друку семінаром відділу теорії нерівноважних процесів

Виготовлено при ІФКС НАН України