Physical processes in the fuel containing masses interacting with aqueous solutions in the "Shelter" object. Inhomogeneous diffusion of ions UO$_2^{2+}$, Cs$^+$ in the system "glassy nuclear magma – water"

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The influence of water on the glassy nuclear magma of the Chornobyl reactor is discussed. To describe it the simplest model for the "glassy nuclear magma–water" system is formulated. Inhomogeneous position- dependent diffusion coefficients are calculated near the interface of the system

1. Introduction

The nuclear and ecological safety of the "Shelter" object at the Chornobyl Nuclear Power Plant is determined by the state of nuclear fuel and water [1-5]. During the last 6 years, a large number of investigations was made on the Chornobyl's problem, some of the results were published [6-14]. All of them point to cardinal changes of the melted fuel masses, i.e. the nuclear magma. The nuclear magma (or fuel containing masses) is a glassy composite, which is formed from the melt of CaO, MgO, Al$_2$O$_3$, Fe$_2$O$_3$, ZrO$_2$, SiO$_2$, H$_2$O and up to 18% of UO$_2$. Now it is being destroyed and transforms into a nuclear dust. One of the important factors of the destruction is the decrease of fuel containing masses temperature, that enabled water to penetrate magma body and actively interact with it [15].

The processes of glassy nuclear magma destruction are very complicated. First, it is necessary to say that the magma, which consists of SiO$_2$ to the extent of 30-60%,

is non-equilibrium amorphous medium with long relaxation time, and thus, external factors, in particularly water, can rapidly alter the state of the medium. Even in 1986, when the nuclear magma was formed, one could state it was steadily destroyed versus time. This was due to active processes of induced radioactive decay of ^{235}U, ^{239}Pu, ^{241}Pu, products of which are hydrogen, tritium, inert gases, such as neon, crypton, xenon and the whole range of unstable elements. The latter transform to stable isotopes and radioactive elements with large half-lives (more than 10^5 years) in response to neutron irradiation. The radioactive decay of nuclei of ^{235}U, ^{239}Pu, ^{241}Pu or α-, β- transformations is accompanied by considerable yields of energy. This causes intensive energy transfer in the medium, and thus an inhomogeneous temperature distribution. Besides, the products of radioactive decays are in fact nonequilibrium impurities in the glassy nuclear magma structure and according to the theory of dislocations in solids, it can bring about substantial structural transformations, recrystallization. Much more complicated processes are connected with the gaseous products of decay, namely, hydrogen, tritium, helium, neon, crypton, xenon. Due to the small solvability of inert gases they gather inside the material of fuel elements, for example UO_2, and produce gaseous cavities. The formation and growth of these cavities lead to swelling of the material. The amount of gas, accumulated in irradiated magma can be large enough to result in its deformation and destruction. The swelling depends on different factors such as temperature change, conditions and intensity of irradiation, properties of a material. The growth of the gaseous pores is a very important part of the swelling processes. Experimental investigations [9] point to the significant porosity of the glassy nuclear magma. The process of pore formation is promoted by the diffusion of gas atoms in various defects, cracks, dislocations, diffusion of hydrogen to metals, etc. As a result of the macroscopic diffusion because of temperature fluctuations gaseous pores may combine leading to a large-scaled deformation.

The investigations made in 1989–1991 [9, 12, 13, 15] showed that glassy masses of nuclear magma in the object were loosing their firmness. Obviously, this is a result of irreversible physico-chemical processes in magma body.

After the decrease of surface temperature of glassy masses to 40–60^{0}C in the average, pores and cracks in the nuclear magma were filled with water, which find its way within through numerous holes in the roof and in the walls of the object, due to the condensation from the interior air and sprinkling of nuclear dust. We can recognize the following four major types of water interaction with the nuclear fuel. First, the interaction with active core fragments. Second, the interaction with persistently flooded nuclear magma. In view of high magma porosity the area and intensity of the interaction are very large. Third, parts of nuclear magma, covered by water seasonally. Fourth, the interaction of water with the nuclear dust.

Water in nuclear magma pores undergoes radiolysis because of α-, β-decays and γ-radiation. Radiolysis products and water molecules at the interface "nuclear magma – water" participate in complicated hydration [16, 17] of uranium UO_2, plutonium PuO_2 and other oxides of radioactive elements [4, 18, 19]. An intensive leaching of uranium from nuclear magma and egress into indoor water basins at the

"Shelter" object occur. Experimental studies [3, 5, 15] indicate a significant variation of uranium concentration in "Shelter" basins in time.

Fuel containing masses suffer considerable changes because of nuclear, physical and chemical processes. A result of their interaction with water is the emergence of yellow spots on the black ceramics [7, 8]. For the first time such yellow spots were observed in 1991 [12]. They were investigated by means of electronic spectroscopy, X-ray phase and X-ray spectral analysis. As a result, it was cleared up that yellow spots are the needle-like crystals of the uranium minerals with the main phases such as: epiyantynite $UO_3 2H_2O$, studtite $UO_3 16CO_3 1.91H_2O$, two modifications of $UO_4 4H_2O$, retzerfordine $UO_2 CO_3$, and unnamed phases $Na_4 UO_2(CO_3)_3$ and $Na_2 UO_2(CO_3)_2 2H_2O$ [7, 8]. All of these minerals have one very significant feature – they are well soluble in water. These mineral combinations along with UO_2, $UO_2 + UO_3$, PuO_2, $(ZrU)O_2$, $(UZr)SiO_4$ are nuclear hazardous materials with a large amount of neutron moderators SiO_2, $Al_2(SiO_3)(OH)_4$, $Na_3 PO_4$, $C_x H_y O_z$, $CaMg(CO_3)_2$, $CaCO_3$, K_2O, CaO,MgO, Fe_2O_3, ZnO_2, H_2O, C.

Summarizing the results of previous investigations, we can say that irreversible processes are running inside the object "Shelter" increasing tendency of nuclear fuel egress to the water basins of the object. This is one of nuclear hazard factors inside the object premises. The problem of water interaction with fragments of the destroyed active core of reactor, nuclear magma and nuclear dust is extremely important because:

- Water interaction with nuclear magma causes a substantial increase of effective neutron multiplication factor K_{eff}, and as a result, local self-sustaining nuclear chain reaction is possible [1-5].

- Due to the complicated processes of radiolysis, the eventual egress of the radioactive materials is observed, that in its turn causes uncontrollable transfer of radioactive materials indoors and outdoors.

- When the nuclear magma is destroyed radioactive dust is produced (containing uranium, plutonium, americium). It is another factor of ecological hazard to the object.

Aqueous solutions of the salts in radioactive elements inside the object are interacting with various construction materials, among them are concrete and glassy masses of nuclear fuel. It is clear, that the character of such an interaction depends very much on the state of these solutions. The presence of uranium, plutonium, americium, curium in aqueous solutions and also their interaction with the fuel-containing masses lead to intensive processes of water radiolysis as a result of α- and β-decays and γ-radiation [3-5, 18, 19]. In the process of radiolysis chemically active radicals are formed: hydrated electrons and ions H^+, groups OH^-, molecular compounds H_2, HO_2, H_2O_2, that largely determines the character of interaction of aqueous solutions with the different materials inside the object.

A particular attention should be paid to the interaction of aqueous solutions with the glassy masses of nuclear fuel. And here we have to give the answer what

is the role of the products of radiolysis of water in the process of destroying the masses, what is the character of their interaction as well as to investigate their influence on the processes of diffusion of uranium, plutonium from the nuclear magma into solutions [4, 18]. Another important question is how the products of radiolysis influence the chemical reactions in which possible reactants are the ions of uranyl UO_2^{2+}, plutonyl PuO_2^{2+}, and other complexes containing radioactive elements [19]. To utterly comprehend nuclear magma destruction and the egress of uranium into water as well as to predict these processes, it is important to research radionuclide diffusion in the "nuclear magma–water" system. Inhomogeneous equations for such a system have been studied in [20, 23]. In so doing an important problem consists in calculating the inhomogeneous diffusion coefficients for ions UO_2^{2+}, PuO_2^{2+}, Cs^+, Sr^{2+} at the interface "nuclear magma – water" as far as temperature, concentration and pH vary. To do this qualitatively we now consider a statistical model taking into account interactions between particles and their structural distribution, and apply it to the phase "aqueous solution".

2. Calculation of inhomogeneous diffusion coefficient for ions UO_2^{2+}, Cs^+ in the "nuclear magma – water" system

We have investigated distribution functions for ions UO_2^{2+}, Cs^+ at the interface "nuclear magma – water" in [20, 21]. They appear to describe this structural distribution in a qualitatively correct manner. Using them we are going to perform the calculation of inhomogeneous diffusion coefficients for ions UO_2^{2+}, Cs^+ in the aqueous solution interacting with glassy nuclear magma. For this purpose only Markovian transport processes of ions in solutions will be considered. The mean flux of k-species particles in the phase l is represented by the following expression:

$$\boldsymbol{j}^k(\boldsymbol{r}_l; t) = -\sum_{k'} \sum_{l'} \int_{V_{l'}} \mathrm{d}\boldsymbol{r}_l' \, D^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) \frac{\partial}{\partial \boldsymbol{r}_{l'}} \delta n^{k'}(\boldsymbol{r}_{l'}; t), \tag{1}$$

where $D^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'})$ are normalized inhomogeneous diffusion coefficients to be related to generalized coefficients $D^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}; \tau)$ in the way:

$$D^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) = \int_0^\infty \mathrm{d}\tau D^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}; \tau), \tag{2}$$

$$D^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}; \tau) = \tag{3}$$

$$= \sum_{l''} \sum_{k''} \int_{V_{l''}} \mathrm{d}\boldsymbol{r}_{l''} \langle (1 - \mathcal{P}_0) \hat{\boldsymbol{j}}^k(\boldsymbol{r}_l) T_0(\tau)(1 - \mathcal{P}_0) \hat{\boldsymbol{j}}^{k''}(\boldsymbol{r}_{l''}) \rangle_0 \left[\tilde{F}^{-1}(\boldsymbol{r}_{l''}, \boldsymbol{r}_{l'}) \right]_{k''k'}.$$

are generalized diffusion coefficients of ions. Here

$$\hat{\boldsymbol{j}}^k(\boldsymbol{r}_l) = \sum_{j=1}^{N_k} \boldsymbol{p}_j \delta(\boldsymbol{r}_j - \boldsymbol{r}_l) \tag{4}$$

is the momentum for ions in the respective phase l,

$$\mathcal{P}_0 A = \langle A \rangle_0 + \sum_{\substack{l,l' \\ k,k'}} \int\limits_{V_l} \mathrm{d}\boldsymbol{r}_l \int\limits_{V_{l'}} \mathrm{d}\boldsymbol{r}_{l'} \, \langle A(\boldsymbol{r}) \delta \hat{n}^k(\boldsymbol{r}_l) \rangle_0 \left[\tilde{F}^{-1}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) \right]_{kk'} \delta \hat{n}^k(\boldsymbol{r}_{l'}) . \quad (5)$$

is the Mori projection operator, $T_0(\tau) = \exp\{(1 - \mathcal{P}_0) \cdot \tau \, \mathrm{i} L_N\}$ is the time evolution operator, $\mathrm{i} L_N$ is the Liouville operator appropriate to the system's Hamiltonian to have been given in [20,21]. The functions $\left[\tilde{F}^{-1}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) \right]_{kk'}$ make up the matrix $\tilde{F}^{-1}(\boldsymbol{r}, \boldsymbol{r}')$ which is inverse to $\tilde{F}(\boldsymbol{r}, \boldsymbol{r}')$. This latter consists of the pair equilibrium distribution functions:

$$F_{nn}^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) = \langle \delta \hat{n}^k(\boldsymbol{r}_l) \cdot \delta \hat{n}^{k'}(\boldsymbol{r}_{l'}) \rangle_0 . \quad (6)$$

We omit phase indices, because only the phase of aqueous solution will be treated. At last, the time correlation function [22, 23] is assumed to be a Gaussian one, therefore we arrive at

$$D^{kk'}(\boldsymbol{r}, \boldsymbol{r}') = \int\limits_0^\infty \mathrm{d}\tau \lambda_0^{kk'}(\boldsymbol{r}, \boldsymbol{r}') \exp \left\{ -\frac{\overline{\lambda}_2^{kk'}(\boldsymbol{r}, \boldsymbol{r}')}{2!} \tau^2 \right\} \quad (7)$$

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

wherein $\lambda_0^{kk'}(\boldsymbol{r}, \boldsymbol{r}')$ is the zeroth moment, $\overline{\lambda}_2^{kk'}(\boldsymbol{r}, \boldsymbol{r}')$ is normalized second one for the time correlation function

$$\overline{\lambda}_2^{kk'}(\boldsymbol{r}, \boldsymbol{r}') = \frac{\lambda_2^{kk'}(\boldsymbol{r}, \boldsymbol{r}')}{\lambda_0^{kk'}(\boldsymbol{r}, \boldsymbol{r}')}. \quad (8)$$

These moments have the following structure

$$\lambda_0^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) = \frac{3}{m_k} kT \, f_1^k(\boldsymbol{r}_l) \left[\tilde{F}^{-1}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) \right]_{kk'} , \quad (9)$$

$$\lambda_2^{kk'}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) = \sum_{l''} \sum_{k''} \int\limits_{V_{l''}} \mathrm{d}\boldsymbol{r}_{l''} \left(\langle \hat{\vec{j}}^k(\boldsymbol{r}_l) \hat{\vec{j}}^{k''}(\boldsymbol{r}_{l''}) \rangle_0 + \frac{N_k}{V_l} \frac{N_{k''}}{V_{l''}} \frac{(kT)^2}{m_k m_{k''}} \right) \quad (10)$$

$$\times \frac{\partial}{\partial \boldsymbol{r}_l} \frac{\partial}{\partial \boldsymbol{r}_{l''}} \left\{ f_1^k(\boldsymbol{r}_l) f_1^{k''}(\boldsymbol{r}_{l''}) \left[\tilde{F}^{-1}(\boldsymbol{r}_l, \boldsymbol{r}_{l''}) \right]_{kk''} \right\} \left[\tilde{F}^{-1}(\boldsymbol{r}_{l''}, \boldsymbol{r}_{l'}) \right]_{k''k'} .$$

in which m_k is the mass of k-species particle, $f_1^k(\boldsymbol{r}_l) = \langle \hat{n}^k(\boldsymbol{r}_l) \rangle_0$ is the profile of k-species particle in a phase l, $\langle \ldots \rangle_0$ means the averaging with the total equilibrium function ρ_0.

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

$$\left[\tilde{F}^{-1}(\boldsymbol{r}_l, \boldsymbol{r}_{l'}) \right]_{kk'} = \frac{\delta_{kk'} \delta(\boldsymbol{r} - \boldsymbol{r}')}{\langle \hat{n}^{k'}(\boldsymbol{r}') \rangle_0} - c_2^{kk'}(\boldsymbol{r}, \boldsymbol{r}'). \quad (11)$$

Along with (11) the expression for fluxes (1) takes the following form:

$$\hat{\boldsymbol{j}}^k(\boldsymbol{r};t) = D^{kk}(\boldsymbol{r})\frac{\partial}{\partial \boldsymbol{r}}\delta n^k(\boldsymbol{r};t) + \sum_{k'}\int \mathrm{d}\boldsymbol{r}'\,\overline{D}^{kk'}(\boldsymbol{r},\boldsymbol{r}')\frac{\partial}{\partial \boldsymbol{r}'}\delta n^{k'}(\boldsymbol{r}';t), \qquad (12)$$

where

$$D^{kk}(\boldsymbol{r}) = D^k(\boldsymbol{r}) = \frac{kT}{m_k}\sqrt{\frac{\pi}{2\overline{\lambda}_2^{kk}(\boldsymbol{r})}} \qquad (13)$$

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

$$\overline{D}^{kk'}(\boldsymbol{r},\boldsymbol{r}') = \frac{kT}{m_k}f_1^k(\boldsymbol{r})c_2^{kk'}(\boldsymbol{r},\boldsymbol{r}')\sqrt{\frac{\pi}{2\lambda_2^{kk'}(\boldsymbol{r})}} \qquad (14)$$

is the inhomogeneous interdiffusion coefficient for k- and k'-species particles with respect to the interface.
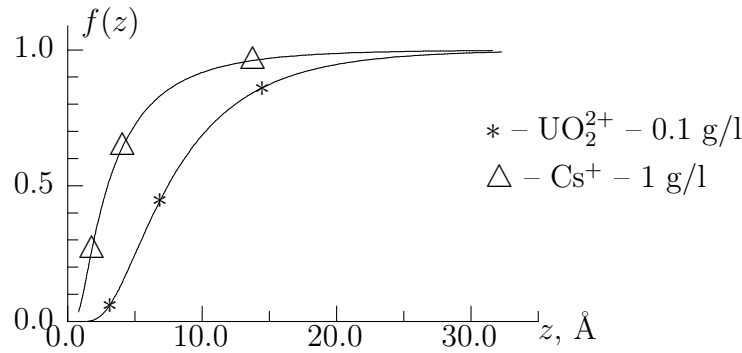

Figure 1. Qualitative density profiles of radioactive particles near the interface "glassy nuclear magma – water".

Here we have restricted our computations to the only selfdiffusion coefficient (13) on the basis of suitable moments (9), (10) for the ions UO_2^{2+}, Cs^+. For this purpose, density profiles from figure 1 were used. The ratio of selfdiffusion coefficient to bulk value is plotted on figure 2 as a function of distance to nuclear magma surface. Concentrations are accepted to be average for the "Shelter".The calculations are qualitative as far as a simple ionic model of solution is considered. As is expected the diffusion coefficients for ions reach their bulk values far from the surface. For ions of cesium this occurs faster. Near the surface,the coefficients increase, ionic mobility is greater. It means that after UO_2^{2+}, Cs^+ egress from nuclear magma as a result of complicated leaching (combination of ion exchange and silicon-oxygen network depolymerization [4]), they are intensively repulsed away from the surface by complex electrostatic forces. The larger distance from the surface, the less influence of electrostatic inhomogeneity is observed. The leaching of ions UO_2^{2+}, Cs^+ results
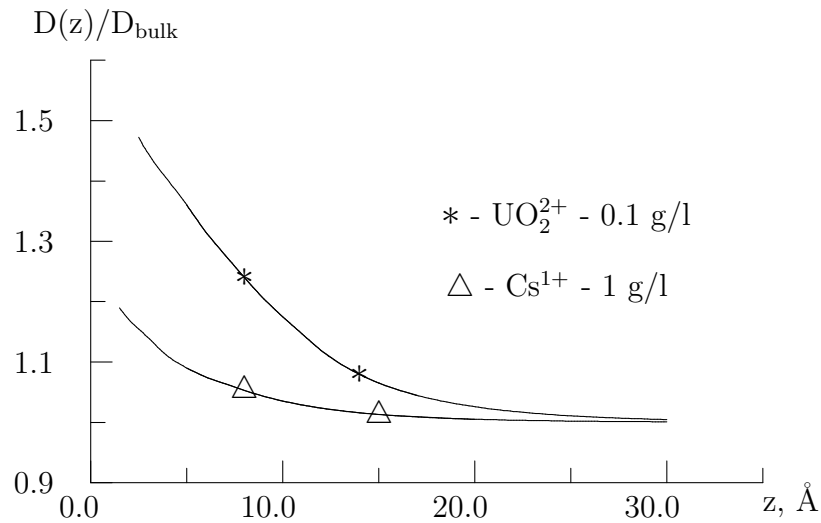
Figure 2. Selfdiffusion coefficients for radioactive ions near the interface "glassy nuclear magma – water".

in a pulverization of nuclear magma surface and in a loss of firmness. In its turn, it enlarges contact with aqueous solutions and promotes a more intensive egress of radioactive elements into water basins of "Shelter". The phenomenon is observed experimentally [5]. Thus, there is a problem of radionuclide migration in these basins and penetration into the outdoor ground water.

To calculate inhomogeneous interdiffusion coefficient we need to know direct correlation functions $c_2^{kk'}(\boldsymbol{r}, \boldsymbol{r}')$. This is a separate problem for a nonequilibrium system. We are going to perform such investigations in future. They are important in view of describing the correlation effects at the interface "nuclear magma – water". It is the region where active magma surface destruction occurs. We'll also investigate the behaviour of $D^{kk}(\boldsymbol{r})$, $\overline{D}^{kk'}(\boldsymbol{r}, \boldsymbol{r}')$ as temperature and concentration vary.

Acknowledgements

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Фізичні процеси взаємодії паливомісних мас з водними розчинами в об'єкті "Укриття". Неоднорідна дифузія іонів UO_2^{2+} та Cs^+ у системі "склоподібна ядерна магма – вода"

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Обговорюється вплив води на склоподібну ядерну магму чорнобильського реактора. З метою теоретичного опису розвинуто найпростішу модель для системи "склоподібна ядерна магма – вода". Обчислено неоднорідні позиційно-залежні коефіцієнти дифузії біля межі поділу системи.

Ключові слова: *Чорнобиль, склоподібні відходи, корозія, вилужування, радіоліз, радіоактивні елементи, функція розподілу, коефіцієнт дифузії*

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