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Evaluation of radioactive material leakage through the fuel cladding as result of diffusion processes during the long-term storage of spent nuclear fuel

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A B S T R A C T

Diffusion is regarded as one of mechanisms, which leads to leakages through the fuel cladding during the long-term storage of spent nuclear fuel. The schematization of the diffusion process through a circular cylindrical cladding of fuel elements, where axial and circumferential diffusion flows are neglected, is considered. Fundamental properties of leakages through a cylindrical cladding of fuel elements associated with the effect of sorption, desorption, and diffusion are investigated; it is shown that the diffusion through the cladding material is the main limiting factor for leakages, whereas sorption and the effect of desorption are limited due to the saturation property. A mathematical model how to assess the maximum possible leakage of radioactive materials through the cladding of fuel elements due to diffusion processes during the long-term storage of spent nuclear fuel is proposed.

This model is presented in the form of a system of differential equations with initial and boundary conditions that make it possible to determine the concentration of radioactive materials inside the fuel cladding taking into account radioactive decay, the concentration of radioactive material in the fuel cladding material during diffusion, and the concentration of radioactive material in the volume outside the fuel cladding. The solution of differential equations, which represent a mathematical model of the leakage of radioactive materials through the cladding of a fuel element during the long-term dry storage of spent nuclear fuel, was performed using the straight line method. A qualitative analysis of the patterns of leakage of radioactive materials was carried out and the influence of the initial concentration and half-life of radioactive materials was established. A quantitative analysis of possible leakages of radioactive material through the cladding of a fuel element during the long-term storage of spent nuclear fuel has been carried out on the basis of computer modeling using specially developed original software.

1. Introduction

The increase of electricity generation production by nuclear power plants leads to the accumulation of spent nuclear fuel, which, in accordance with the IAEA requirements, should either be reprocessed or stored for a long time in compliance with all safety requirements (e.g. described by Sorenson (Sorenson, 2015), Mahfuth et al. (Mahfuth et al., 2020), Swift (Swift, 2017) and in other works). At present, the dry method of storage is the most widespread one for the long-term storage of spent nuclear fuel before its disposal or reprocessing (Ojovan and Lee, 2013). Unfortunately, along with the obvious advantages of this method of the storage (such as economy and simplicity), there is a number of disadvantages. Indeed, the processes that occur in fuel and storage facilities during a long period of time (100 years or more) have not been sufficiently studied yet. The proven mathematical models of the storage process, taking into account the necessary set of factors (for example, described by Alyokhina et al. (Alyokhina et al.,

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2018; Alyokhina et al., 2009), Sheng et al. (Sheng et al., 2019), Gutiérrez et al. (Gutiérrez et al., 2018) and others), are currently not sufficiently developed, which does not allow fully predicting the state of the fuel at the time of completion of intermediate storage, which, in its turn, is necessary in order to ensure its preparation for disposal or reprocessing.

It is known that dry storage systems are based on the usage of multilevel protection (Lee et al., 2013) of spent nuclear fuel from the spread of radioactive materials contained in it. The fuel cladding is the one of protective barrier separating radioactive materials from the environment during the long-term storage of spent nuclear fuel. Taking this into account, the possibility of the long-term storage of spent nuclear fuel and its subsequent disposal are also depend on fuel cladding failure detection. Thus, the study of physical processes associated specifically with the fuel cladding is a priority task when justifying and assessing the safety criteria for the long-term dry storage and subsequent management of spent nuclear fuel.

2. Analyses of problem state

The problem of the integrity of the fuel cladding is considered in many scientific works (Standring, 2015). In particular, physical factors, which affect the fuel cladding and lead to its destruction are considered and analyzed by Khattak et al. (Khattak et al., 2019). The strength characteristics of fuel rods and the models that allow them to be identified are discussed in detail by Yefimov et al. (Yefimov et al., 2015) and Pelykh et al. (Pelykh et al., 2009).

A detailed analysis of the thermal state of spent nuclear fuel during the long-term dry storage for the final stages of the nuclear fuel cycle was carried out by Alyokhina (Alyokhina, 2019) and (Alyokhina and Kostikov, 2017). It should be noted that considerable attention is paid to the problem of the diffusion of radioactive substances through the fuel cladding. For example, the results of experimental studies of the penetration of uranium and cesium into a single-crystal cladding of fuel elements with carbonitride fuel are presented by Vasil'ev et al. (Vasil'ev et al., 2014). Mentioned experimental studies have been performed at a temperature of about 1500 °C, however during the storage the temperatures are much smaller. The study of Keiser (Keiser, 2012) analyzed the diffusion between the metallic fuel of a sodium reactor, its cladding and fission products from the point of view of the possibility of the formation on the inner surface of the cladding zones, which may become brittle or contain relatively low-melting phases and subsequently result in cracking and failure of the fuel cladding.

It should be noted that the research methods from the above and many other works (for example, Jiang et al. (Jiang et al., 1997)), which are devoted to the study of the physics of the diffusion process in the reactor core for a relatively short time, cannot be directly applied to the study of the problem of the long-term dry storage of spent nuclear fuel. Considerable attention is also paid to the experimental definition of the diffusion properties of fuel (Park et al., 2021), various materials of the fuel cladding (Degueldre et al., 2001; Khatamian, 1997), as well as the coatings of the cladding (Khatkhatay et al., 2013; Firouzdor et al., 2012). The above mentioned and many other studies consider either "fresh" fuel or fuel operated in the reactor, and therefore, such studies cannot solve the problem of the long-term storage of spent nuclear fuel; they can be useful only partially and only at the initial stages of spent nuclear fuel management.

The problem of evaluating the long-term diffusion of fission products through the fuel cladding has been little studied as a whole, however some of its aspects have been considered. Thus, work (Sureda et al., 2010) analyses the sorption of strontium in case of the disposal of high-level hazardous radioactive waste. In study (Majorana and Salomoni, 2004), the diffusion of radioactive materials contained in disposed low-level radioactive waste into the surrounding underground environment was studied. However, the above and many other studies do not take into account the influence of radioactive decay on the processes of diffusion, which is incorrect to neglect when simulating long-term dry storage of spent nuclear fuel containing fission products with relatively long half-lives.

3. Aim and objectives of research

The aim of this work is to develop a simulation model of the leakage of fission products as a result of the diffusion through the fuel cladding during the long-term storage of spent nuclear fuel under normal conditions for further study of their environmental effect. In order to achieve this goal, the following tasks were solved.

1. On the basis of the analysis of typical designs of fuel cladding, hypotheses about the mechanism of possible leakage of fission products through the fuel-element cladding are formulated with the account of the results of existing studies.
2. The effect of sorption, diffusion, and desorption processes on the leakage of radioactive materials through the fuel cladding is investigated by passage to the limit of solution of a specially formulated problem of stationary diffusion.
3. A mathematical model for the conservative evaluation of the leakage of radioactive material through the fuel cladding due to diffusion has been developed, taking into account radioactive decay during the long-term storage of spent nuclear fuel.
4. By means of specially developed original software, computer simulations were performed and some qualitative and quantitative regularities of the leakage of radioactive material through the fuel cladding during the long-term dry storage of spent nuclear fuel were obtained.

4. Methodology

4.1. Hypotheses on the mechanism of leakage of radioactive materials through the fuel cladding

The fuel cladding of a currently widespread design can be schematically represented in the form of a long circular cylinder with the inner radius a, outer radius b and length L (Fig. 1-a). Taking this circumstance into account, in order to determine the position of the points of the fuel cladding, let us further use the cylindrical coordinates r, θ, z with the basis unit vectors $\vec{e}_r$, $\vec{e}_\theta$ and $\vec{e}_z$, which determine, respectively, the radial, circumferential

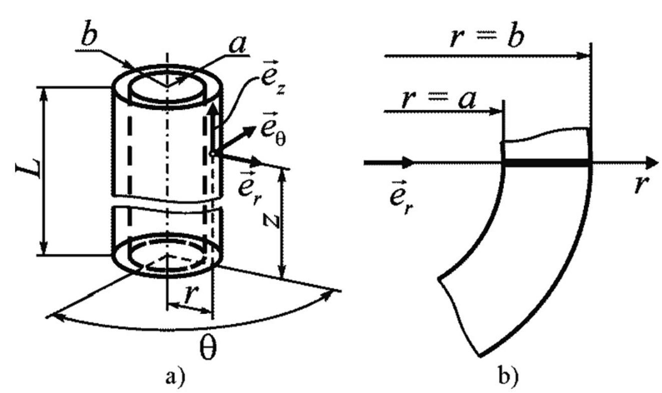

Fig. 1. Assembly representation (a) and schematization (b) of the fuel cladding.

and axial directions in the fuel cladding (Fig. 1-a). The coordinates of the points of the fuel cladding are limited by the following inequalities:

$$a \leqslant r \leqslant b, \quad 0 \leqslant \theta \leqslant 2\pi, \quad 0 \leqslant z \leqslant L \tag{1.1}$$

For typical nuclear power reactors the outer radius of the fuel cladding is $b \sim 10\text{mm}$ and the length of the cladding is $L \sim 4500\text{mm}$, i.e it satisfies the inequality

$$b \ll L \tag{1.2}$$

Radioactive materials are accumulated in the area surrounded by the fuel cladding $r < a$ and in the area $a \leqslant r \leqslant b$ of the fuel cladding material during the operation in the reactor core. During the long-term storage of spent nuclear fuel, radioactive materials accumulated in fuel elements can penetrate outside the cladding into the area $r > b$. In order to assess the leakage of radioactive material, a mechanism for the formation of such leakage should be proposed. Further, let us accept the hypothesis that the leakage of radioactive materials through the fuel cladding during the long-term storage of spent nuclear fuel occurs due to sorption through the boundary surface $r = a$ of the fuel cladding, diffusion in the area $a \leqslant r \leqslant b$ of the material of the fuel cladding and the desorption at the outer surface $r = b$ of the fuel cladding. Such a hypothesis coincides with available researches, including (Sureda et al., 2010; Majorana and Salomoni, 2004).

The accepted hypothesis on the mechanism of leakage of radioactive materials through the fuel cladding during the long-term storage of spent nuclear fuel requires the development of mathematical models of the sorption processes on the inner surface of the cladding, the diffusion process in the cladding material, and the desorption process on the outer surface of the cladding. The development of such mathematical models and their application for solving problems will be greatly simplified if we take into account the relation (1.2) of the characteristic dimensions of the cladding. Taking into account the relation (1.2), let us assume the hypothesis that the circumferential and axial diffusion flows are negligible in comparison with the radial diffusion flow. The assumed hypothesis makes it possible to reduce the assessment of the leakage of radioactive materials through the fuel cladding to the study of a one-dimensional diffusion flow along the radial direction of the fuel cladding, and represent this cladding itself as a segment limited by the thickness of the cladding wall (see Fig. 1-b)

$$a \leqslant r \leqslant b \tag{1.3}$$

The accepted hypothesis introduces some errors, but at the same time it greatly simplifies the study of the processes of sorption, diffusion, and desorption for the assessment of the leakage of radioactive materials through the fuel cladding during the long-term storage of spent nuclear fuel. This hypothesis seems to be suitable for the initial study of leakages due to sorption, diffusion and desorption in order to establish the most general qualitative regularities and to obtain preliminary quantitative estimates. Thus, based on the analysis of typical designs of fuel-element cladding and the results of existing studies, hypotheses on the mechanism of possible leakage of fission products through the fuel-element cladding are formulated.

4.2. *Influence of the processes of sorption, diffusion and desorption on the leakage of radioactive materials through the fuel cladding*

In order to study the effect of sorption, diffusion, and desorption on the leakage of radioactive materials through the fuel cladding during the long-term storage of spent nuclear fuel, let us consider the mathematical models of these processes taking into account the accepted simplifying hypotheses. All the listed processes are considered here as stationary.

Let us represent the mathematical model of the sorption of radioactive materials through the inner surface $r = a$ of the fuel cladding in the following form:

$$-D\frac{dc}{dr}\bigg|_{r=a} = -\alpha_a\left(c|_{r=a} - c_a\right) \tag{2.1}$$

where $D = const$ is the diffusion coefficient of the material of the fuel cladding; $c = c(r)$ is the concentration of radioactive material in a point of the fuel cladding; $\alpha_a = const$ is the sorption coefficient of radioactive material through the inner boundary of the fuel cladding; $c_a = const$ is the concentration of radioactive material in the area $r < a$ which is restricted be the fuel cladding.

The relation (2.1) gives the proportional link between the flow of radioactive materials through the surface $r = a$ of the fuel cladding and the difference between the concentrations of radioactive materials on the boundary surface $r = a$ of the fuel cladding and in the area $r < a$.

Obviously, in general, the mathematical model taken in the form (2.1) reflects the main properties of the sorption process, in particular, the saturation property, which corresponds to the equality $c|_{r=a} = c_a$ between the corresponding concentrations. It is assumed that the selection of the coefficient value α_a will make it possible to obtain sufficiently accurate quantitative estimations of the sorption process of various materials.

The mathematical model of stationary diffusion through the material of the fuel-element cladding is represented by the well-known diffusion equation, which with account of the accepted hypotheses takes the following form in cylindrical coordinates:

$$\frac{d^2c}{dr^2} + \frac{1}{r}\frac{dc}{dr} = 0, \quad a < r < b. \tag{2.2}$$

The equation (2.2) is well-known and studied in mathematical physics.

Let us represent the mathematical model of the desorption of radioactive materials on the outer surface $r = b$ of the fuel cladding as:

$$-D\frac{dc}{dr}\bigg|_{r=b} = -\alpha_b\left(c_b - c|_{r=b}\right), \tag{2.3}$$

where $\alpha_b = const$ is the desorption coefficient of the radioactive material on the outer surface $r = b$ of the fuel cladding; $c_b = const$ is the concentration of the radioactive material on the outside of the fuel cladding in the area $r > b$.

The mathematical model taken in the form (2.3) in general reflects main properties of the desorption process, in particular the property of saturation, which complies the equation $c|_{r=b} = c_b$ between corresponding concentrations. It is assumed that the choice of the value of the coefficient α_b will allow obtaining quite precise quantitative evaluations of the desorption process of different materials.

A set of mathematical models (2.1)–(2.3) of the processes of sorption, diffusion and desorption can be considered as a boundary-value problem for the diffusion equation (2.2), boundary conditions (2.1) and (2.3) of which correspond to the accepted mathematical models of sorption and desorption. The boundary conditions of the type (2.1) and (2.3) are known in mathematical physics. The solution of the diffusion equation (2.2), taking into account the boundary conditions (2.1) and (2.3) will allow detecting the leakage of the radioactive material through the fuel cladding within a given period of time:

$$Q = -D\frac{dc}{dr}\bigg|_{r=b} \cdot 2\pi bLt \tag{2.4}$$

where Q is the leakage of the radioactive material through the fuel cladding during the time t.

The solution of the equation (2.2), as it is known, has the following form:

$$c(r) = A_1 \ln r + A_2 \tag{2.5}$$

where A_1 and A_2 are the integration constants, that must be defined by means of boundary conditions of the task.

In order to define the integration constants A_1 and A_2 in the solution (2.5) let us use the mathematical models (2.1) and (2.3) of sorption and desorption, which are considered as the boundary conditions for the diffusion equation (2.2). Taking into account the solution (2.5), the relations (2.1) and (2.3) will lead to the system of two linear algebraic equations in order to define A_1 and A_2:

$$\begin{cases} \left(\frac{D}{a} - \alpha_a \ln a\right)A_1 - \alpha_a A_2 = -\alpha_a c_a, \\ \left(\frac{D}{b} - \alpha_b \ln b\right)A_1 - \alpha_b A_2 = \alpha_b c_b. \end{cases} \tag{2.6}$$

The solution of the equation (2.6) has the following form:

$$A_1 = \frac{c_b - c_a}{\ln\frac{b}{a} + D\left(\frac{1}{a\alpha_a} + \frac{1}{b\alpha_b}\right)}, \quad A_2 = \frac{c_a \ln b - c_b \ln a + D\left(\frac{c_b}{a\alpha_a} + \frac{c_a}{b\alpha_b}\right)}{\ln\frac{b}{a} + D\left(\frac{1}{a\alpha_a} + \frac{1}{b\alpha_b}\right)}. \tag{2.7}$$

By applying the formula (2.4) and solutions (2.5) and (2.7), let us define the leakage of radioactive material through the fuel cladding:

$$Q = 2\pi LD \frac{c_b - c_a}{\ln\frac{b}{a} + D\left(\frac{1}{a\alpha_a} + \frac{1}{b\alpha_b}\right)} t. \tag{2.8}$$

It is possible that the formula (2.8) can be applied for the quantitative evaluation of the leakage of radioactive materials through the fuel cladding during the long-term storage of spent nuclear fuel; however, its main value is that the qualitative evaluation can be done. According to the formula (2.8) in case when the coefficients are $\alpha_a = 0$ and $\alpha_b = 0$, we receive that the leakage is $Q = 0$; if the coefficients α_a and α_b increase, the leakage will increase as well. It is interesting that the maximum leakage will correspond to the limit value (2.8) when $\alpha_a \to \infty$ and $\alpha_b \to \infty$:

$$Q_{\max} = 2\pi LD \frac{c_b - c_a}{\ln b - \ln a} t \tag{2.9}$$

It is evident that final leakage in the case when $\alpha_a \to \infty$ and $\alpha_b \to \infty$ correspond to the saturation properties of sorption and desorption. The formula (2.9) is the conservative evaluation of the leakage of radioactive materials through the fuel cladding during the long term storage of spent nuclear fuel. The mathematical models (2.1) and (2.3) of sorption and desorption in the case when $\alpha_a \to \infty$ and $\alpha_b \to \infty$ will obviously converged to simple relations, which correlate with the saturation conditions of the corresponding processes:

$$c|_{r=a} = c_a \tag{2.10}$$

$$c|_{r=b} = c_b \tag{2.11}$$

The relations (2.10) and (2.11) represent the class of boundary conditions of the diffusion equation known in mathematical physics, provided that the use of these boundary conditions will give conservative estimates of the leakage of radioactive materials through the fuel cladding. Thus, the influence of sorption, diffusion, and desorption processes on the leakage of radioactive materials through the fuel cladding is investigated by passing to the limit of solving a specially formulated problem of stationary diffusion.

4.3. Mathematical model for the conservative evaluation of radioactive material leakage through the fuel cladding as a result of diffusion taking into account radioactive decay

For the conservative evaluation of possible leakage of radioactive materials during the long-term dry storage of spent nuclear fuel, let us use a mathematical model in the form of differential equations, initial and boundary conditions of related processes of changes in the concentration of fission products inside the volume $0 \leqslant r < a$ confined by the fuel cladding, in the volume $a \leqslant r \leqslant b$ of the fuel cladding material, and also in the volume $r > b$ outside the fuel cladding (see Fig. 1).

Let us give the mathematical model of the concentration of fission products in the internal volume $0 \leqslant r < a$ confined by the fuel cladding in the form of an ordinary differential equation with the initial condition:

$$\frac{dc_a}{dt} = \frac{2D}{a}\frac{\partial c}{\partial r}\bigg|_{r=a} - \frac{\ln 2}{T}c_a + \frac{p_a \mu_f}{\pi a^2 L}\frac{\ln 2}{T_f}e^{-\frac{\ln 2}{T_f}t}, \quad c_a(0) = c_{a(0)}, \tag{3.1}$$

where $c_a = c_a(t)$ is the averaged concentration of the fission product in the area limited by the fuel cladding which depends on the time $t \geqslant 0$; $c = c(r,t)$ is the concentration of the fission product in the fuel cladding material, $a \leqslant r \leqslant b$; T is a half-life of fission product; μ_f and T_f are the molar mass and half-life of fissile materials inherent in spent nuclear fuel; p_a is the probability of the formation of the fission product in the course of radioactive decay which is capable of penetrating the fuel cladding; $c_{a(0)}$ is the averaged concentration of the fission product in the area $0 \leqslant r < a$ confined by the fuel cladding at the beginning of the storage at time $t = 0$.

The differential equation (3.1) represents the expression of the mass balance of the fission product material in the volume $a \leqslant r \leqslant b$ restricted by the fuel cladding, taking into account its penetration (sorption) into the fuel cladding with respect to its radioactive decay and its formation during the radioactive decay of other fissile materials which are originally present in spent nuclear fuel. This differential equation (3.1) should be considered taking into account the initial concentration of the fission product initially present in spent nuclear fuel as well as with respect to the mathematical model of the concentration change $c = c(r,t)$.

Let us represent the mathematical model of the concentration change $c = c(r,t)$ in the volume $a \leqslant r \leqslant b$ of the material of the fuel cladding in the form of a partial differential equation with initial and boundary conditions:

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial r^2} + \frac{1}{r}\frac{\partial c}{\partial r}\right) - \frac{\ln 2}{T}c, \quad c(r,0) = c_{(0)}, \quad a < r < b, \tag{3.2}$$

$$c(a,t) = c_a, \quad c(b,t) = c_b, \tag{3.3}$$

where $c_{(0)}$ is the concentration of the fission product initially present at the time of the beginning of the storage in the volume of fuel cladding material; $c_b = c_b(t)$ is the concentration of the fission product material in the volume $r > b$ outside the fuel cladding.

It is well-known, the oxide layers are actually presented on the inner as well as on the outer surface of the cladding and to take into account the effect of these oxide layers it is necessary to consider the diffusion of the radioactive materials through these layers by using the partial differential equations similar to (3.2) with the required initial and boundary conditions as well as the conjugation conditions on the contacts between these oxide layers and the cladding. These complications are not suitable in this particular research, because it is necessary to estimate firstly principal possibility of leakages due to the diffusion mechanisms, but the future researches must have considering the presence of the oxide layers on the cladding.

It is necessary to note, the different kinds of cracks are naturally existed in the cladding due to the operational damaging for example. Influencing the micro cracks on the diffusional processes leading to leakages of fission products can be imagined in changing the diffusion coefficient of the cladding which must be greater comparing with the diffusion coefficients of the cladding with more little cracks. So, considering the micro cracks can be reduced to defining the effective diffusion coefficient. At the same time, using the concept about the effective diffusion coefficient is restricted for considering the only some dimensions micro cracks. Really, existing of the micro cracks of some dimension can make the cladding as the porous and the leakages will be defined by the motion of the fission products thru the porous medium, but not by the diffusion. It is obviously, that diffusional mechanism is not main in the case of existing the macro cracks, because the leakages due to the motion of the fission products thru these macro cracks will be significantly greater all others possible leakages. However, the long term storage is firstly for the spent fuel with the claddings without the significant damages so the diffusional mechanism seems as the most possible for such claddings.

Let us represent the mathematical model of the concentration $c_b = c_b(t)$ of the fission product in the volume $r > b$ outside the fuel cladding in the form of an ordinary differential equation with the initial condition:

$$\frac{dc_b}{dt} = -\frac{2D}{k_b b}\frac{\partial c}{\partial r}\bigg|_{r=b} - \frac{\ln 2}{T}c_b, \quad c_b(0) = 0 \tag{3.4}$$

where k_b is the ratio of the volume of the storage cask for the spent fuel assemblies to the volume taken by all the fuel elements situated in the cask. Thus, (3.1)–(3.4) present the mathematical model for the conservative evaluation of radioactive material leakage through the fuel cladding in the consequence of the diffusion, taking into account radioactive decay during the long-term storage of spent nuclear fuel.

4.4. Computer simulation of the radioactive material leakage through the fuel cladding during the long storage of spent nuclear fuel

Despite the fact that the differential equations of the mathematical model (3.1) - (3.4) of radioactive material leakage during the long-term storage of spent nuclear fuel are linear, their analytical solution for obtaining quantitative evaluations of possible leakage is a rather time-consuming task. Hereafter, let us consider a computer simulation of the leakage of fission products based on the model (3.1) - (3.4) using numerical methods.

Let us use the idea (Hoffman and Frankel, 2001) about possibilities of semi-discrete finite differences approximations, so to reduce the partial differential equation (3.2), (3.2) with the initial and the boundary conditions to the ordinary differential equations with the initial conditions. Such reducing will allow considering the system (3.1)–(3.4) of the initial-value and initial-boundary-value problems as the approximate initial-value problem, which can be solved by the typical numerical methods, like the Runge-Kutta methods for example. It is really, the finite element method seems as the more perfected comparing with the finite differences, but we need having also the finite differences for future benchmarking the finite element method; besides, the finite differences is more simple for programming so that they are useful for the initial estimating the proposed approaches.. Following this method, let us introduce a uniform grid (Fig. 2) with the coordinates of the nodes in the investigated area $a \leqslant r \leqslant b$ of the fuel cladding:

$$r_k = a + k\Delta r, \quad \Delta r = \frac{b-a}{n+1}, \quad k = 0,1,2,\ldots,n,n+1 \tag{4.1}$$

where $n \geqslant 1$ is the given number of the grid nods in the inner area $a < r < b$ of the fuel cladding; Δr is a grid step.

Instead of the concentration $c = c(r,t)$, the grid (4.1) allows considering its nodal values:

$$c_k(t) = c(r_k, t), \quad k = 0,1,2,\ldots,n,n+1 \tag{4.2}$$

Further, in order to define nodal values (4.2) let us obtain ordinary differential equations with initial conditions and solve this problem by means of a numerical method – the Runge-Kutta method (Butcher, 2016; Baag et al., 2017).

The first boundary condition (3.3) and the grid (4.1), (4.2) allow writing:

$$c_a(t) = c_0(t) \tag{4.3}$$

Let us use approximate relationship $\frac{\partial c}{\partial r}\big|_{r=a} \approx \frac{c_1 - c_0}{\Delta r}$ and with respect to the relationship (4.3) let us represent the mathematical model (3.1) in the form:

$$\frac{dc_0}{dt} = -\left(\frac{2D}{a\Delta r} + \frac{\ln 2}{T}\right)c_0 + \frac{2D}{a\Delta r}c_1 + \frac{p_a\mu_f}{\pi a^2 L}\frac{\ln 2}{T_f}e^{-\frac{\ln 2}{T_f}t}, \quad c_0(0) = c_{a(0)} \tag{4.4}$$

In order to transform the differential equation (3.2) at interior points of the grid, let us use the well-known approximate relationship:

$$\frac{\partial^2 c}{\partial r^2}\bigg|_{r=r_k} \approx \frac{c_{k-1} - 2c_k + c_{k+1}}{\Delta r^2}, \quad \frac{\partial c}{\partial r}\bigg|_{r=r_k} \approx \frac{c_{k+1} - c_{k-1}}{2\Delta r} \tag{4.5}$$

As a consequence, we shall obtain differential equations and boundary conditions:

$$\frac{dc_k}{dt} = \frac{D}{\Delta r}\left(\frac{1}{\Delta r} - \frac{1}{2r_k}\right)c_{k-1} - \left(\frac{2D}{\Delta r^2} + \frac{\ln 2}{T}\right)c_k + \frac{D}{\Delta r}\left(\frac{1}{\Delta r} + \frac{1}{2r_k}\right)c_{k+1}, \quad c_k(0) = c_{(0)} \tag{4.6}$$

where $k = 1,2,\ldots,n$.

The second boundary condition (3.3) and the grid (4.1), (4.2) allow writing:

$$c_b(t) = c_{n+1}(t) \tag{4.7}$$

Let us use the evident approximate relationship $\frac{\partial c}{\partial r}\big|_{r=b} \approx \frac{c_{n+1} - c_n}{\Delta r}$ and with respect to the relationship (4.7) let us represent the mathematical model (3.4) in the form:

$$\frac{dc_{n+1}}{dt} = \frac{2D}{k_b b\Delta r}c_n - \left(\frac{2D}{k_b b\Delta r} + \frac{\ln 2}{T}\right)c_{n+1}, \quad c_{n+1}(0) = 0. \tag{4.8}$$

Thus, in (4.4), (4.6), and (4.8), we have a system $n+2$ of ordinary differential equations with initial conditions, which we will further solve approximately using the Runge-Kutta method.

In order to develop programs that implement computer simulation of radioactive material leakages during the long-term storage of spent nuclear fuel, differential equations and initial conditions (4.4), (4.6), and (4.8) are conveniently represented in a matrix and vector form:

$$\frac{d\boldsymbol{c}}{dt} = \boldsymbol{A}\cdot\boldsymbol{c} + \boldsymbol{f}(t), \quad \boldsymbol{c}(0) = \boldsymbol{c}_0, \tag{4.9}$$

where $\boldsymbol{c}$ is the vector of the nodal values of the concentration of the fission product in the material of the fuel cladding; $\boldsymbol{A}$ and $\boldsymbol{f}(t)$, $\boldsymbol{c}_0$ are the matrix and vectors, given with respect to the differential equations and initial conditions (4.4), (4.6) and (4.8).

The vector $\boldsymbol{c}$, matrix $\boldsymbol{A}$ and vectors $\boldsymbol{f}(t)$, $\boldsymbol{c}_0$ take the following form:

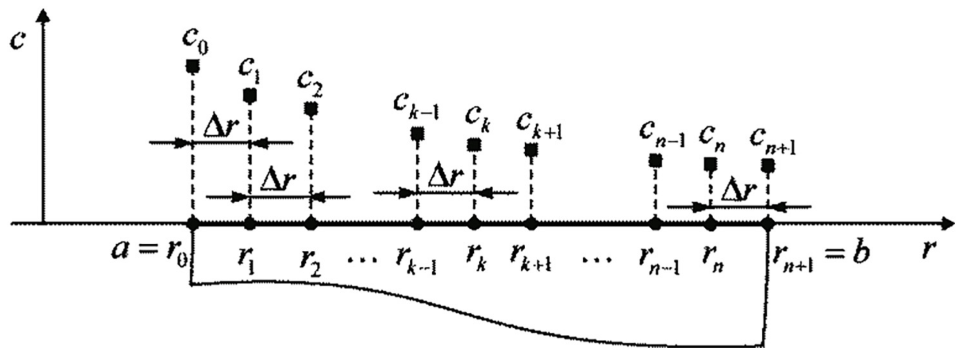

Fig. 2. Grid over the thickness of the fuel cladding and nodal values.

$$\boldsymbol{c} = \begin{pmatrix} c_0 \\ c_0 \\ c_0 \\ \vdots \\ c_{n-1} \\ c_n \\ c_{n+1} \end{pmatrix}, \quad \boldsymbol{c}_0 = \begin{pmatrix} c_{a(0)} \\ c_{(0)} \\ c_{(0)} \\ \vdots \\ c_{(0)} \\ c_{(0)} \\ 0 \end{pmatrix},$$

$$\boldsymbol{A} = \begin{pmatrix} Y_0 & Z_0 & 0 & 0 & \dots & 0 & 0 & 0 & 0 \\ X_1 & Y_1 & Z_1 & 0 & \dots & 0 & 0 & 0 & 0 \\ 0 & X_1 & Y_1 & Z_1 & \dots & 0 & 0 & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \dots & X_{n-1} & Y_{n-1} & Z_{n-1} & 0 \\ 0 & 0 & 0 & 0 & \dots & 0 & X_n & Y_n & Z_n \\ 0 & 0 & 0 & 0 & \dots & 0 & 0 & X_{n+1} & Y_{n+1} \end{pmatrix},$$

$$\boldsymbol{f}(t) = \left(\tfrac{p_a \mu_f}{\pi a^2 L} \tfrac{\ln 2}{T_f} e^{-\frac{\ln 2}{T_f} t} \quad 0 \quad 0 \quad \dots \quad 0 \quad 0 \quad 0 \right)^T. \tag{4.10}$$

Let us solve the differential equations (4.9) and (4.10) using the Runge-Kutta method of the fourth order, choosing the integration step so that the results, corresponding to the step divided by two do not greatly differ from the results corresponding to this step. The convergence of approximate solutions with an increase in the number of nodes n will be established by comparing the solutions corresponding to different n.

Differential equations arising in physics including diffusion analysis usually solve by usage of standard programs such as Mathematica, Maple, MathLab etc. (Al-Jawary et al., 2020; Aliev et al., 2016). However, these programs are for commercial usage and require special skills to work with them. Special original software for computer modeling of the leakage of fission products through the fuel cladding was developed in the FORTRAN-90 programming language; it provides the input of the initial calculation data, construction of matrices (4.10) and the numerical solution of the Cauchy problem (4.9) and (4.10) using the Runge-Kutta method of the fourth order.

As the object of study, we shall consider the fuel cladding of the WWER-1000 nuclear reactor in a cask used for the long-term dry storage of spent nuclear fuel assemblies, which are characterized by the following dimensions (Alyokhina, 2019):

$$a = 3.9 \text{ mm}, \quad b = 4.55 \text{ mm}, \quad L = 4500 \text{ mm}, \quad k_b = 1.2. \tag{4.11}$$

Due to the variety of possible compositions of fission products in spent nuclear fuel and their diffusion properties, let us take the diffusion coefficient of the fission product in fractions of the hydrogen diffusion coefficient D_H in zirconium as:

$$D = D_H, \quad D_H/100, \quad D_H/1000. \tag{4.12}$$

The value (4.12) corresponds to a conservative estimate of the leakage of fission products through the fuel cladding, since hydrogen has very high diffusion capacity.

Initial concentrations $c_{a(0)}$ and $c_{(0)}$ of the fission product in the inner cavity of the cladding and in the fuel cladding itself is determined by their total molar masses μ_a and μ_0 of the fission product in the corresponding volumes:

$$c_{a(0)} = \frac{\mu_a}{\pi a^2 L}, \quad c_{(0)} = \frac{\mu_0}{\pi \left(b^2 - a^2 \right) L}. \tag{4.13}$$

Taking into account the variety of possible data on the composition of fission products in spent nuclear fuel let us have

$$\mu_a = 5 \text{ mole}, \quad \mu_0 = 2 \text{ mole}, \quad T = 25 \text{ years}. \tag{4.14}$$

Due to the variety of possible data on the composition of fissile materials in spent nuclear fuel, let us assume the following characteristics of fissile materials:

$$\mu_f = 7 \text{ mole}, \quad T_f = 10 \text{ years}, \quad p_a = 0.75. \tag{4.15}$$

The results obtained in the computer modeling are approximate and their error associated with the applied solution method is determined by the size of the grid step Δr, i.e. the number of grid nodes n (see Fig. 2). The selection rationale of a sufficient number of grid nodes n is carried out by comparing the solutions obtained for different n numbers. The results of such a comparison (Fig. 3-a)

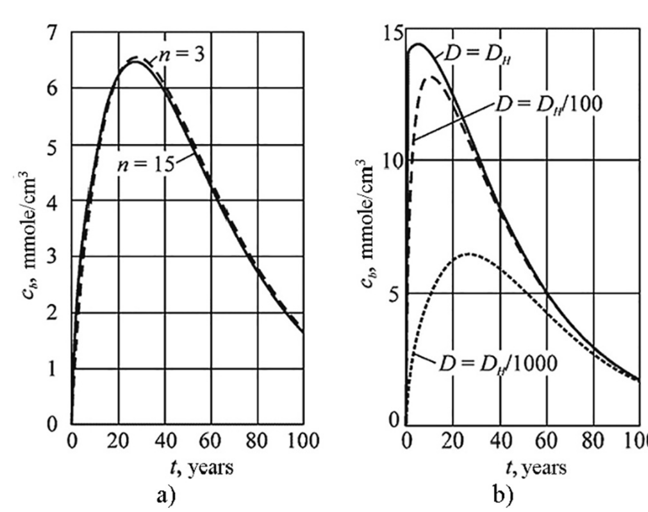

Fig. 3. Research results of the convergences of (a) approximate solutions and influence of diffusion coefficient of the fuel cladding; b) leakage of the fission product through the fuel cladding.

show that the solutions corresponding to the numbers $n = 3$ and $n = 15$ are somewhat different, but this difference cannot be considered significant; the calculations show that the solutions corresponding to the numbers $n = 15$ and $n = 33$ are visually indistinguishable on the scale of Fig. 3-a). Taking into account the circumstances noted above, let us further use the results corresponding to the number $n = 15$, since the computer computation time is approximately 10 times less than for the number $n = 33$. Besides the numerical convergence, it is necessary using the benchmark data for validating the results, but the problem about storage the waste nuclear fuel is not fully researched now and we had no find the similar results of other authors to use for validating ours results. So, it is permissible to assume the results are valid due to the established numerical convergence which is the serious independent argument too in this case.

The calculations show (Fig. 3-b) that during the storage the value of the diffusion coefficient significantly influences the maximum concentration of the fission product outside the fuel cladding in a long-term storage cask of fuel assemblies. It is the maximum value of the concentration of the fission product that creates the danger of radioactive contamination of the environment around the storage facility of spent nuclear fuel.

Thus, computer simulations have been performed by means of specially developed original software and some qualitative and quantitative regularities of radioactive material leakage through the fuel cladding during the long-term dry storage of spent nuclear fuel have been obtained.

5. Discussion of the research results

The storage of spent nuclear fuel must exclude the release of fission products accumulated in this fuel during their radioactive decay into the environment. It is assumed that at the end of the established period of the long-term storage, the content of fission products accumulated in spent nuclear fuel due to radioactive decay will be small enough for relatively safe handling of such fuel in the process of its subsequent processing.

It is evident that the process of reducing the mass of fission products materials in nuclear fuel due to their radioactive decay should be taken into account when considering the diffusion processes of such fission products in order to assess their leakage during the long-term storage of spent nuclear fuel. It should be noted that a special group of radioactive materials could also be present in spent nuclear fuel; such materials do not have a noticeable diffusion property, but can form other fission products during radioactive decay, capable of penetrating through the walls of fuel cladding due to diffusion.

As we can see from the results (2.8) and (2.9), the leakage of radioactive materials is proportional to the diffusion coefficient of the cladding material, so that infinite increase in the coefficient leads to infinite increase in leakage. Thus, the appropriate value of the diffusion coefficient of the material ensures the leakage tightness of the fuel cladding. This result is consistent with the results of published scientific papers, for example (Keiser, 2012); (Jiang et al., 1997); (Degueldre et al., 2001); (Khatamian, 1997); (Khatkhatay et al., 2013); (Firouzdor et al., 2012), which study the process of fuel diffusion through fuel claddings. However, these and most of other existing studies are of little use for the solution of the problem of the long-term storage of spent nuclear fuel, as they consider the diffusion of fresh fuel during a short period of time through undamaged fuel claddings in the reactor core, although such results are of interest to solve spent nuclear fuel management problems at the initial stages of the long-term storage. The results obtained in this work (see Fig. 3) correspond to the diffusion process over a long period of time and can be used to assess the possible damage to the environment during the long-term storage of spent nuclear fuel as well.

The result (2.9), substantiating conservative estimates of diffusion leakage during the long-term storage of spent nuclear fuel is very important, because conservative estimate of leakage of radioactive materials are of interest for predicting the possible impact of storage facilities for spent nuclear fuel on the environment. In addition, the definition of the sorption and desorption coefficients α_a and α_b is a difficult task, as demonstrated by Sureda et al. (Sureda et al., 2010). In this case, the use of boundary conditions of the form (2.10) and (2.11), corresponding to the saturation of sorption and desorption processes, will allow obtaining reliable conservative estimates of possible leakage of fission product through the fuel cladding during the long-term storage of spent nuclear fuel; as it was shown before, the actual leakages will not exactly exceed the estimates obtained and are likely to be much smaller. At the same time, the results of studies (Sureda et al., 2010) of sorption and desorption processes are of great importance for a more accurate determination of possible leakages, and the results of such studies can be taken into account due to the refined formulation of boundary conditions (3.3).

The authors (Majorana and Salomoni, 2004) and many other studies do not take into account the mutual influence of diffusion and radioactive decay processes, which is quite reasonable for studying the long-term storage of low-level radioactive waste, as well as for solving problems of high-level radioactive waste and spent nuclear fuel management at the initial stages of its long-term storage. The mathematical model proposed in this paper allows considering diffusion processes during the long-term storage of highly radioactive spent nuclear fuel, taking into account the processes of radioactive decay.

In fact, the differential equation (3.2) is a known diffusion equation, supplemented by the corresponding terms in order to take into account the decrease in the concentration in the elementary volume due to the radioactive decay the fission product. The initial condition (3.2) allows taking into account the concentration of the fission product accumulated in the fuel cladding material. The boundary conditions (3.3) correspond to the limiting cases (2.10) and (2.11) of sorption and desorption of the fission product through the inner and outer boundary surfaces of the fuel cladding. The use of boundary conditions (3.3) will make it possible to obtain conservative estimates of the possible leakage of the fission product through the fuel cladding during the long-term storage of spent nuclear fuel; as it was shown before, actual leakages will not exactly exceed the estimates obtained and are likely to be much smaller.

6. Conclusions

Based on the results of this study, the following conclusions can be drawn.

1. It is accepted that the processes of sorption, diffusion and desorption can be considered as possible mechanisms for the formation of leakages of fission products through the fuel cladding during the long-term storage of spent nuclear fuel. It is recommended to continue studies of possible leakages of fission products through fuel cladding due to sorption, diffusion and desorption during the long-term storage of spent nuclear fuel.
2. It is shown that due to their inherent saturation properties the sorption and desorption processes limit the maximum possible leakage of fission products through the fuel claddings during the long-term storage of spent nuclear fuel. Given this circumstance, it is recommended to use conservative estimates of the

leakage of fission products through fuel claddings during the long-term storage of spent nuclear fuel, which meet the limit case of infinite sorption and desorption coefficients.

3. A mathematical model for a conservative assessment of the leakage of radioactive material through the fuel cladding due to diffusion has been developed. This model is based on differential equations for the balance of the concentration of radioactive material, taking into account sorption, diffusion and desorption flows, as well as decrease in the concentration of fission products due to its radioactive decay. It should be emphasized that the differential diffusion equation used in the proposed mathematical model contains an additional term that allows taking into account decrease in the concentration of fission products due to its radioactive decay.
4. It is proposed to use the straight line method (half-sampling) for computer simulation of the leakage of radioactive materials through the cladding of fuel elements during the long-term storage of spent nuclear fuel. This method reduces the considered problem to the integration of ordinary differential equations with initial conditions, which can be carried out using well-developed numerical methods. The experience of the application of the Runge-Kutta method has shown that this method allows obtaining the required solutions if the time step is correctly chosen considering with the step of the spatial grid. The choice of the time integration step for the given grid was carried out manually by comparing the obtained results with different time steps. The application of integration methods with automatic step selection, which will be done in further research, seems to be more promising.
5. The results of computer simulating the leakages of the fission product during the long-term storage of spent nuclear fuel from WWER-1000 reactors show that the diffusion coefficient of the fuel cladding significantly affects the maximum concentration of the fission product outside the fuel cladding. This maximum is observed in first third of the spent nuclear fuel storage period and exactly this maximum creates the danger of fission products entry into the cask cavity. In the considered problem, the concentration of the fission product behind the fuel cladding after 100 years of storage depends little on the diffusion coefficient of the cladding and is determined by the radioactive decay of the fission product.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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