Modernization of the CHAIN code for fission gas release calculation

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Abstract. The issue of a quantitative assessment of the fission products release from the fuel is one of the important part of the analysis of the radiation consequences of accidents. In the reactor fission products that accumulate as a result of the fission reaction are contained within the fuel elements. The article discusses the modification of the CHAIN code for the calculation of the isotopic composition of the fuel, which was added a subroutine calculating the fission gas release from the fuel. The subroutine is based on the diffusion equation, the modification of which makes it possible to take into account the release of radionuclides from uranium dioxide of fuel elements. The coefficients in the diffusion equation were selected experimentally. The results of the calculations for two verification tests of the modified code are also presented. Comparisons of the calculation results of the code were carried out for emergency fuel heating and during normal operation of the reactor.

1. Introduction and CHAIN code background
The development of the CHAIN code was started in 1995. The program is used to calculate the radiation characteristics of neutron-irradiated materials and to form a source for further use in dose rate calculations. The code also has the ability to simulate the burnup of fissile materials, to calculate the residual energy release and the intensity of the spontaneous fission neutron source. It has been modernized to increase the capabilities of the code. The modernization consisted in the inclusion of a special unit for calculating the release of radionuclides from the fuel.

The CHAIN code is designed to simulate changes in the nuclide composition of materials under irradiation with neutrons and in the process of radioactive decay of unstable nuclei included in their composition. The program uses a point model with the conditions of uniformity of the properties of the medium and the neutron flux density in the considered volume.

In combination with the attached library of neutron-physical cross-sections, the CHAIN code is intended for calculations of thermal neutron reactors.

As the initial data, in addition to the multi-group neutron flux density, the initial nuclide composition and irradiation time, as well as the list of exposure times after the end of irradiation, are specified. The code uses both for modeling the fuel fission process and for irradiation of non-fissile materials.

The radiation characteristics of irradiated materials are uniquely determined by their nuclide composition. The CHAIN code uses an analytical solution to the nuclear transition equation to calculate changes in nuclear concentrations over time.

At the moment of reactor stop and for each of the specified exposure times, the following characteristics are calculated:
• nuclide composition of the material;
• the depth of fuel burnup;
• $\alpha$, $\beta$, and $\gamma$-activity;
• full activity;
• residual energy release;
• line spectrum of $\gamma$-radiation;
• spontaneous fission neutrons source.

The program was modernized in terms of assessing the release of fission radionuclides from the fuel matrix. The results of calculating the nuclear concentrations of the main dose-forming nuclides in the irradiated fuel, if necessary, are used in a separate subroutine of the program for these calculations.

This article describes the CHAIN code subroutine that calculates the fission gas and products release from uranium dioxide.

2. Method

The problem of the nuclides release from dioxide has been intensively studied in the past decades in parallel with the development of nuclear power. Despite the complexity of the release process it has been proposed for a long time to use the equation of nuclides diffusion inside the fuel pellet to describe the release. In this case, the computational model should take into account that, in addition to the nuclides release through the geometric surfaces of the pellet, there is also release through the porosity, and this release can exceed the geometric one.

To calculate the fission products release from the fuel a cylindrical uranium dioxide fragment is considered (one fuel pellet is simulated). Nuclides are generated in the volume of the fragment and diffuse to the surface. Diffusion is described by the equation:

$$\frac{\partial N}{\partial t} = \nabla D \nabla N - \lambda N - \Sigma N + q$$

(1)

where:

$N$ : the concentration of the radionuclide of interest;
$D$ : the diffusion coefficient;
$\lambda$ : decay constant;
$\Sigma$ : takes into account the leakage of the radionuclide out of the grains of uranium dioxide into pores and then outside the dioxide boundaries;
$q$ : the accumulation rate of the radionuclide on on precursor fission and decay.

In Eq. (1), $\Sigma N$ describes internal absorption, for which the radionuclide egresses from the fuel and beneath the cladding.

To obtain the numerical value of $\Sigma$, the following expressions are used.

The flux of radionuclide $f_{in}$ from grain to pores (per unit volume) are given by the equation 2:

$$f_{in} = \frac{S_1}{R_z} \cdot D \cdot \xi (N - N_0) = \frac{S_1}{R_z} \cdot D \cdot \xi (1 \cdot \frac{N_0}{N}) \cdot N = \frac{S_1}{R_z} \cdot D_{eff} \cdot N$$

(2)

where

$S_1$ : square of grains for unit volume;
$R_z$ : the size of a dioxide grain;
$\xi$ : the relative fraction of the pores with open egress;
$N_0$ : the concentration of the radionuclide in the open space;
$D_{eff}$ : the effective diffusion coefficient.

The value of $N_0$ takes into account the possibility of nonzero boundary conditions at the outer grain boundary and has significant values for long-lived radionuclides in a sealed fuel element, since for short-
lived radionuclides (due to decay) or for all nuclides in a leaky fuel element (due to a large open space) it was taken equal to zero. In the case of long-lived radionuclides, the value of $N_0$, increasing with time, will slow down further release. The value of $\Sigma$ are calculated using the equation 3:

$$\Sigma = \frac{S_1 \cdot D_{\text{eff}}}{R^2_{\text{eff}}} \ , \quad D_{\text{eff}} = D \cdot \sqrt{n} \left( 1 - \frac{N_0}{N} \right)$$ (3)

The key parameter is the diffusion coefficient, which is described by the equation 4 [1,2]:

$$D = A \exp\left(-\frac{B}{T}\right) + 1.41 \cdot 10^{25} \sqrt{n} \exp\left(-13800/T \right) + 2 \cdot 10^{40} n$$ (4)

where

$T$ : the temperature of the uranium dioxide;

$n$ : the fission density.

The initial values of the constants $A$ and $B$, taken from [1], were adjusted due to the wide spread of the values of these coefficients in the literature.

The diffusion coefficient it is proposed to use in the form of a sum of three components, the equation 5 [3].

$$D = D_1(T) + D_2(T, n) + D_3(n)$$ (5)

It is assumed that $D_1$ prevails at temperatures above 1200°C, which corresponds to classical diffusion in a solid and is described by the Arrhenius law, where $D_1 = A \cdot e^{-B/\tau}$ [1,4].

$D_2 (T, n)$ - makes the main contribution at temperatures of 800-1200°C. $D_2 = C \cdot \sqrt{n} \cdot e^{-13800/T}$. This component corresponds to radiation-stimulated diffusion and is associated with the formation of vacancies for diffusing atoms under irradiation.

$D_3 (n)$ - makes the main contribution at low temperatures $T<800^\circ$C [5] and is represented as $D_3 = D_0 \cdot n$. This component is responsible for the release processes of nuclides due to recoil and knock-out during the fission.

It should also be noted that in concerning temperature ranges where this or that diffusion mechanism dominates the clause "roughly" is given. So it can be assumed that the indicated temperature limits are not generally accepted constants.

The equation (1) is solved in two-dimensional $r-z$ geometry by the finite-difference method with boundary conditions $N = 0$ at the ends and lateral surface of the cylinder (including the central hole in the tablet, if any) and the initial condition $N(r, 0) = 0$.

At each time moment after obtaining the distribution $N(r, t)$ he leakage is calculated at the cylinder boundaries, and the number of radionuclide nuclei escaping from the uranium dioxide through geometric surfaces is determined. To this is added internal leakage, defined as the volume integral of $\Sigma N$. The value of $N_0$ is determined in the course of the calculation, taking into account the given fraction of the open space in the fuel element, which can change over time (for example, due to the reduction of the fuel-cladding gap). The accumulated radionuclides in uranium dioxide and outside it are determined by summation over the entire exposure time, taking into account radioactive decay.

The input data for the code are the radionuclide number, half-life (hours), fission rate, dioxide grain size, calculation geometry, radius and height of a pellet, number of axial zones in a fuel element, energy release distribution, time step, number of time steps, dependences power of a fuel element versus time, radial and axial partitioning of a dioxide tablet into nodes, and a number of other parameters. The fuel

1 In ANS5.4 [6] it is proposed to solve the problem of radionuclides release from a certain sphere (Booth's model) in each computational node of the fuel pellet using the temperature corresponding to this node. The size of the sphere should be chosen according to the results of experiments. Then the output is summarized across all nodes. In our model, a similar approach is used, but instead of considering the Booth sphere, we consider one grain, the leakage from which under the clad is calculated using equations 2, 3.
element is subdivided in the axial direction into a number of zones, for each of which the relative energy release is specified. Also, the dependence of the fuel assembly power on burnup is set, which makes it possible to take into account the change in fuel temperature along the fuel rod campaign. At each point in time along the campaign, at each axial section of the fuel element, using a special program, the temperature distribution along the radius of the fuel pellet is calculated. It also takes into account the change in thermal conductivity and the size of the fuel-cladding gap with fuel burnup. Instead of using a thermophysical unit, fuel temperatures can be programmed manually.

Taking into account the above, the diffusion equation is taken as the basis of our model, the diffusion coefficients in the form of the sum of 3 components were selected according to experimental data in order to ensure the solution of 2 problems:

1. Prediction of release from the uranium dioxide in sealed fuel elements during normal operation;
2. Prediction of the radionuclides release from the dioxide during emergency fuel heating.

The model also empirically describes an increase in the yield of fission products at high burnup. A possible reason for the increased yield at high burn-ups is some process leading to a significant refinement of uranium dioxide grains. The grain size decreases by about an order of magnitude. This effect is taken into account in the model by reducing \( R_s \) in the ratio of equation 3 when the burnup exceeds the level of 50 MW·day / kg.

The problem of fission products release from the dioxide of leaking fuel elements and their movement inside the fuel element, as well as the problem of additional release with power fluctuations, is not considered at this stage.

3. Some results and discussion

The model was verified using experimental data and similar (certified) programs. Of the entire set of radionuclides, only twelve are considered in the model, namely: \(^{85}\)Kr, \(^{87}\)Kr, \(^{88}\)Kr, \(^{131}\)Xe, \(^{133}\)Xe, \(^{131}\)I, \(^{132}\)I, \(^{133}\)I, \(^{134}\)I, \(^{135}\)Cs, \(^{137}\)Cs. Exactly these radionuclides that are of primary interest when analyzing the radiation consequences of accidents at a reactor. Below, the results of calculations and verification are briefly presented for the case of emergency fuel heating and during normal operation of the reactor.

3.1 Emergency fuel heating

In case of emergency fuel heating, a sharp temperature jump is observed. A sharp increase in fuel temperature will lead to an abrupt increase in the yield of fission products. At such temperatures, in addition to the release of gaseous and volatile fission products, a noticeable release of medium and weakly volatile radionuclides will also begin (especially at high burnup).

It should be noted that verification of such codes is always difficult. Most experiments are not carried out under ideal conditions and have significant errors. Some of the most successful, according to the authors, experiments and comparison with them are given below.

Table 1 gives a brief description of experiments VI and VR [7], on the basis of which one of the verification tests of the code was carried out.

Table 2 shows the experimentally obtained release for iodine and cesium and the corresponding calculated values.

**Table 1.** Brief description of experiments on measuring the radionuclides release for heating fuel fragments.

| No | Experiment designation | Experiment content |
|----|------------------------|-------------------|
| 1  | VI – 2                 | Heating up to \( T_{\text{max}} = 2300K \) and exposure at \( T_{\text{max}} \) during 60 min |
| 2  | VI – 3                 | Heating up to \( T_{\text{max}} = 2700K \) and exposure at \( T_{\text{max}} \) during 20 min |
| 3  | VR – 3                 | Heating up to \( T_{\text{max}} = 2570K \) and exposure at \( T_{\text{max}} \) during 15 min |
| 4  | VR – 4                 | Heating up to \( T_{\text{max}} = 2570K \) and exposure at \( T_{\text{max}} \) during 30 min |
Table 2. Experimental and calculated values of radionuclide release, %.

| No | Nuclide | Experiment | Calculated values | Deviation from experiment, % |
|----|---------|------------|-------------------|-----------------------------|
| 1  | I       | 40.0       | 35.9              | -11.4                       |
|    | Cs      | 67.0       | 36.0              | -86.1                       |
| 2  | I       | 79.0       | 72.4              | -9.1                        |
|    | Cs      | 99.9       | 72.6              | -37.6                       |
| 3  | I       | 70.0       | 78.7              | 11.1                        |
|    | Cs      | 70.0       | 78.9              | 11.3                        |
| 4  | I       | 87.0       | 90.1              | 3.4                         |
|    | Cs      | 93.0       | 90.4              | -2.9                        |

From the results shown in Table 2, it can be seen that the maximum deviation of our calculation from the experiment is 86.1%, which is an acceptable error value for such calculations.

It should be noted that it is proposed to use the diffusion coefficient (and so it is used in the model) for all isotopes of iodine or cesium, as well as for all inert radioactive gases.

3.2 Reactor operation under normal conditions

When the reactor is operating under normal conditions, the fuel is at significantly lower (operating) temperatures. By the end of the fuel campaign, the yield can reach only 1% for individual isotopes.

Table 3 shows the calculated data for some radionuclides in terms of the rate of escape under the cladding of a hermetically sealed medium-stress fuel element of a VVER reactor with a burnup of 43 MW day / kg.

The calculation results are in good agreement with the conclusions, including those from article [1], where the results of measurements of the fission products release from PWR fuel elements during their normal operation are discussed. On French PWRs, the fission products release is at a level of less than 1% with burnup 10-30 MW day / kg, increases to 1-2% to burnup 40 MW day / kg and up to 5% to burnup 60-70 MW day / kg. It is noted that the linear load of fuel elements on French PWRs is usually at the level of 150-200 W / cm.

Table 3. Release rate (R/B) calculated using the CHAIN program for a VVER fuel element.

| Nuclide | Release rate R/B |
|---------|------------------|
| Xe-133  | 0.395            |
| Cs-134  | 0.97             |
| Cs-137  | 1.057            |
| I-131   | 0.49             |
| I-133   | 0.1              |
4. Conclusion
A description is given of a calculation model designed to calculate the release of radionuclides from uranium dioxide during normal operation or during an accident. The semi-empirical model is based on the diffusion equation. The diffusion coefficient, including its temperature dependence, was selected according to the experimental data. The resulting model was then verified according to the data of other experiments on the release of radionuclides with fuel heating and according to the data of experiments during normal operation.

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