Use of Information Theory Concepts for Developing Contaminated Site Detection Method: Case for Fission Product and Actinides Accumulation Modeling

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ABSTRACT

Information theory concepts and their fundamental importance for environmental pollution analysis in light of experience of Chernobyl accident in Belarus are discussed. An information and dynamic models of the radionuclide composition formation in the fuel of the Nuclear Power Plant are developed. With the use of code DECA numerical calculation of actinides (58 isotopes are included) and fission products (650 isotopes are included) activities has been carried out and their dependence with the fuel burn-up of the RBMK-type reactor have been investigated.

INTRODUCTION

In the event of nuclear or radiological emergency an objective estimation of the site contamination characteristic is a principal factor for correct protection decision. Belarus has not its own nuclear facilities but the danger of radioactive contamination exists from Nuclear Power Plants (NPP) which are situated in Lithuania, Ukraine and Russia immediately close to its border. It is quite possible that online estimation of radiological situation on the base of native data acquisition and by means of national monitoring system will be necessary.

However in a fresh accidental release, direct knowledge of magnitude and radionuclide composition is not available with high degree of certainty. There is good reason to think that neither the means of on-site NPP Safety nor the off-site environment monitoring are not all sufficient to assess radioactive situation after the accident correctly. So default data of site contamination must be identified by indirect estimations.

Republic of Belarus just faced the above challenge after the Chernobyl accident. We consider this practical experience of the radionuclide contamination analysis to be very useful for environmental monitoring strategy enhancing. After Chernobyl accident a large-scale γ-spectrometry measurement of early site contamination of the area close to Chernobyl Nuclear Power Plant (ChNPP) was carried out by specialists of the Institute of Power and Nuclear Research Belarusian Academy of Sciences (JIPNR BAS) in 1986. Basic radiation-dangerous long-lived fission products (the isotopes Cs-134, Cs-137, Ru-106, Ce-144) and the short-lived as well (the isotopes Zr-95, Nb-95, I-131, Ce-141, Ru-103, Cs-136) were identified in the samples and the data bank of fission products of early data acquisitions was created at the institute [1]. Over the next period of years number of studies in the fields of radiation protection, radioecology and radiobiology were undertaken to reconstruct reliable radiological situation and population doses in the early stage of the accident in Belarus [2,3]. As opposite to research made in Ukraine [4] that was aimed to establish relationship between physical or chemical properties of the dispersed fuel particles (nonvolatile hot particles) and conditions of particle formation during the accident, we would like to refer to results obtained in Belarus and presented in paper [2]. These investigations were concerned with the main regularities of early site contamination of Belarus as the result of fallout of condensate forms of radionuclides. Volatile isotopes of Cs, Te, I, and Ru, Ce (in a lesser degree) as well, were released during high-temperature nuclear fuel annealing, then they were caused by atmospheric scattering and deposition. To reconstruct real radioactive situation and assess retrospectively the total short-lived isotope contribution to population dose an integrative technique known as isotope correlation methods [5] were used by the authors. By means of mathematical statistics applied to collected data, the authors proved the presence of an essential indication in a form of activity relationships between the volatile radionuclides and refractory one (for example relationship Cs-137 – Zr-95), stated quantitative correlation measures and established functional regression relations between relative activities of radionuclides that differed in their volatile properties (Ru-106, Ce-144, I-131) and Cs-137 contamination density.

Turning back to the problem of radiation contamination after nuclear accident we suppose that the most reasonable approach for radionuclide composition formation modeling may be obtained in terms of information theory. To develop a proper model for this complex process we need a general measure. It is attractive to use concepts of information theory and to represent the process of radionuclide composition formation in terms of coding, transfer and decoding of information. The activity ratios of different pairs of radionuclides that were identified in the samples with respect to corresponding activity ratios in nuclear facility are the basic informative indications for this purpose.

Information theory was developed by C. Shannon for technical systems of communication [6]. At present there is a tendency to enhance information theory to environment investigation [7]. As to radiation protection problems just valuable information characterization is distinctive feature of radionuclide composition for successful applying of information approach.

While solving radioecology problems after Chernobyl accident a demand arose for determination of complete composition and radionuclide activities of the facility (Unit 4 of ChNPP) at the moment of accident [2,3]. In fact radionuclide composition not only presents initial data. It provides the base of informative indications to study the process. As the result of nuclear fission an accumulation of fission products and actinides in nuclear fuel may be considered as a well-ordering process. Exactly the unique structure of fission product decay chains and actinide transmutation network enable natural information coding of radionuclide composition data. We suppose that the first step to realize an information approach is a proper model for fission products and actinide accumulation in accidental facility [8]. Taking in account the above mention we applied an information theory concepts for developing a model of radionuclide composition formation in nuclear fuel.

GENERAL PRINCIPLES OF INFORMATION MODEL AS APPLIED TO RADIONUCLIDE COMPOSITION FORMATION PROBLEM

The process of quantitative and qualitative changes of the radionuclide composition formation in accidental nuclear reactor starts up with a neutron irradiation of uranium nuclei resulting in their fission and radioactive fragment yields. This process is accompanied by subsequent radioactive transformation of fission products (known as decay chains) and actinides transmutation. During the radiation accident at NPP different fractions of fission products escape into environment terminating radionuclide composition formation process. So the process of the radionuclide composition that forms at the accidental conditions can be regarded as a complex technical-environmental system. On account of insufficient information governing the severe accident an attempt to construct mathematical model of the system by traditional methods in terms of matter and energy transformation faces difficulties and compels to look for an alternative approach to the problem. This conceptual framework is a system analysis as a more common methodology of the construction and application of information models of the complex systems with the different physical nature [9]. Its general position is that the system integrity and ordering attains owing to dynamic interaction of the parts of the system.

So the problem of a system complexity includes structural and functional factors [10]. A set of system elements connecting by a set of relations means structural complexity. Structural complexity enlarges as the rate of the interrelated elements or their intensity increase. Quantitative measure of this type of complexity as a degree of the system ordering is information. Communicative connection governing the behaviour of the system (or of its parts) has more common nature. Being accompanied every physical interaction of the systems and controlling their common functioning, this characteristic of the communicative connection is comprehended as information (following C. Shannon). This type of the complexity can frequently be related to the lack of knowledge about the investigated phenomena. So models imitating transformation of matter, energy and information supplement each other. This means that studying of the information features of the system by indirect analysis one may attempt to recognise causal dependence and principles of its functioning. We may also expect that the changes of radionuclide composition resulting from the accidental processes are to be correlated to the structural changes of the system.

Let us consider a process of a thermal neutron irradiation of uranium fuel primary consisting of U-235 and U-238 and accumulation of fission-products and actinides in a power reactor facility. To receive more information about radionuclide composition at the facility, a well-ordering structural-information model of the radionuclide composition formation is developed by the author. As a base of the dynamic model simulating the radionuclide composition formation in a nuclear facility first of all a conceptual model of a system has been constructed. Conceptual model defines required information about the process reflecting internal structural interaction between subsystems and external relationships of the system. Considering a neutron flux as an external system we assume following subsystems and hierarchical construction of the structure as a whole:

- the first-level subsystem is a fissile nuclei of U-235;
- the second-level subsystem is a set of actinides, which itself is complex and consists of
- the transuranium isotope network including a new fissile nuclei accumulation (we consider here more significant Pu-239 and Pu-241 nuclei) ;
- the second-level subsystem of fission products that consists of a set of fission product decay chains; they present the third-level subsystems.

Principal processes caused the change of the radionuclide composition during burn up are the following:

- fission of primary nuclear fuel - U-235 by a neutrons results in fission product yields;
- neutron capture by U-238 initiating the actinide formation by a subsequent neutron captures competitive with the radioactive α- or β-decays and possible fission as well;
- secondary nuclear fuel (Pu-239 and Pu-241) accumulation and fission as well;
- β-decay transformation of fission products in 94 decay chains with the mass numbers from 72 to 166;
- possible neutron capture by some fission products;
- delay-neutron emission by a delay-neutron precursors.

Thus having established the conceptual model then the information-structural model of the radionuclide composition formation is to be realised. A detailed network of all possible actinide and fission-product transmutations has complex structure that included branching, feedback and cycle loops [11]. As applied for the systems with a complex structure the graph theory presents a powerful method of an object ordering [12]. To describe a correct structure of the radionuclide formation network we introduced into consideration bond graph $G(V,E)$ consisting of a set of a vertexes V and a set of an edges E. Every of v_1, v_2, $\ldots v_n$ vertexes is associated with the transmuting nuclide (fission product or actinide) and pair of the vertexes (v_i,v_j) is the edge indicating radioactive transformation from v_i to v_j. Let $|V|$ is a number of vertexes and $|E|$ is a number of an edges, then $|V|$ is equal to $r = 708$ as the sum of $p = 650$ and $q = 58$, where p,q is quantity of fission products and actinides correspondingly. The bond structure of the oriented graph $G(V,E)$ is described by adjacency matrix M in such a way that matrix coefficient $m_{ij}=1$ only if i,j is the edge of the graph, otherwise $m_{ij}=0$.

DYNAMIC MODEL OF RADIONUCLIDE COMPOSITION FORMATION IN A NUCLEAR FACILITY

Dynamic model described fission product and actinide accumulation in nuclear fuel irradiated by a neutrons has been constructed as a set of flows of matter on the graph $G(V,E)$.

Let us denote x_i as concentration of i-radionuclide and a set of actinide and fission product concentrations by vector $\vec{x}(t) = [x_1, ..., x_p, x_{p+1}, ... x_{p+q}]$. Then we associate with the edge (i,j) of graph $G(V,E)$ the flow of i-radionuclide matter and denote it as $s_{ji}(t) \cdot x_i(t)$, where s_{ij} is a constant of formation j-nuclide from i- one. Then we can write the set of the ordinary differential equations described radionuclide transmutation in a form of

$$\frac{dx_i(t)}{dt} = -\sum_{j=1}^{m} s_{ij}(t)x_i(t) + \sum_{k=1}^{n} s_{ki}(t)x_k(t) \quad , \qquad \text{(Eq. 1)}$$

were i=1,708; m,n denote, correspondingly, number of all possible receivers and sources of i – nuclide that belong to graph $G(V,E)$.

We write Eq (1) in vector form as

$$\frac{d\vec{x}(t)}{dt} = \hat{S}(t) \cdot \vec{x}(t) \qquad \text{(Eq. 2)}$$

with the time-dependent transmutation matrix $\boldsymbol{S(t)}$ and initial concentration at a moment t_o $\boldsymbol{x_o}$. The dimension of matrix $\boldsymbol{S}$ is equal to $r = p + q = 708$.

To complete the dynamic model the nuclear data were introduced. Nuclear database of 650 fission products and of 58 actinide nuclei has been constructed. It consists of: λ_i,- decay constants; y_i - fission product yields of ^{235}U, ^{239}Pu, ^{241}Pu; $\sigma(E)$ - neutron capture cross sections [13].

We consider here a reactor of RBMK-type. This type of reactor acts on thermal neutrons. The neutron-induced transmutation rates have been calculated by the use of code TRIFON [14] and inserted here in a model in accordance to the formulas:

$$s_{ij}(t) = \int dV \int_0^\infty \sigma_{ij}(E)\Phi(\vec{R}, E, t)dE \qquad \text{(Eq. 3)}$$

$$s_{ij} = \sigma_{ij}^{\ av}\Phi_T \quad , \qquad \text{(Eq. 4)}$$

and

$$\Phi_T = \frac{w}{E^f{}_k \sum_k (\sigma^f{}_k x^f_k)} \quad , \qquad \text{(Eq. 5)}$$

where $\Phi(\mathbf{R},E,t)$ is energy and spatial-distribution of the neutron density, w is specific power of the facility, $\sigma_{ij}{}^{av}$ *is* the energy-averaged cross section, Φ_T is the thermal-neutron flux, E^f_k is fission energy and x^f_k is concentration of fission nuclei of k-type (where k =1,2,3 and denotes U-235, Pu-239, Pu-241, correspondingly).

If for a time interval $\tau_j = t_j - t_{j-1}$ matrix $\boldsymbol{S}^j$ can be regarded as invariable we can write formal solution of the Eq.(1) as the exponential function of matrix $\boldsymbol{S}^j$:

$$\vec{x}(t_j) = \exp[\hat{S}^j \tau_j]\vec{x}(t_0) = \hat{B}^j \cdot \vec{x}(t_0) \qquad \text{(Eq. 6)}$$

Then nuclide concentration $\boldsymbol{x}\ (t_n)$ and activity $A_i(t_n)$ at the moment t_n can be calculated as

$$\vec{x}(t_n) = \hat{B}^n \cdot \ldots \cdot \hat{B}^j \cdot \ldots \cdot \hat{B}^1 \cdot \vec{x}(t_0) \qquad \text{(Eq. 7)}$$

$$A(t) = \lambda_i \vec{x}_i(t) \qquad \text{(Eq. 8)}$$

In view of common problems of the system complexity, namely, large number of system dimension and the presence of highly dynamic processes flowing simultaneously with slowly variable ones, we develop an improved numerical algorithm. To calculate numerically Eq. (6-7) we expand exponential function of matrix ***S*** into series with a small number K and short interval *h* by a formula

$$R(\hat{S}(h)) = \sum_{k=1}^{K} \frac{(h\hat{S})^k}{k!} \qquad \text{(Eq. 9)}$$

and then we obtain the solution at a moment $t=t_n$, as consistent with the following formula:

$$x_n = \left(\sum_{k=1}^{K} \frac{\left(h\,\hat{S}\right)^k}{k\,!} \right)^n x_0 \qquad \text{(Eq. 10)}$$

The calculation procedure expressed by eq.(6-10), nuclear database and the bond graph information as well has been realised by authors in code DECA. A sparse matrix technology that raises a processing speed and economises computational resources has been employed [15].

RESULTS AND DISCUSSION

With the use of code DECA simulation of the nuclear fuel irradiation and accumulation of 58 actinide isotopes and of 650- fission product isotopes during the lifetime at RBMK-1000 were carried out. This type of a reactor is the same as at Chernobyl NPP. There are two NPPs with RBMK-type reactor beyond the bound of Belarus that are situated in about of 10 km neighbourhood to the territory of our Republic.

They are Ignalina NPP in Lithuania, where one of a two facilities is shutting off and the other is functioning, and Chernobyl NPP in Ukraine, where all of a three nuclear facilities at present are shut off. Keeping in mind considerable radioactive capacity presenting at Sarchofagus (destructed 4th reactor of ChNPP) it is clear that these nuclear objects present a high radiation emergency to the territory of Belarus in a case of possible accidental releases.

One of the important characteristics of RBMK-type reactors is their online refuelling capability. Under normal operation and nominal reactor power up two fuel assembly changes per day are carried out. As a result there are a set of fuel channels in a core with a fuel burn-up differed from 0 to 20 MWt·day/kgU and various radionuclide activities. To obtain definitely a total core activity it is necessary to sum up the specific activities stored up at each assembly. A specific power of a fuel assembly decreases during the burn-up lifetime, while the neutron flux on average in a core is constant.

The basic core parameter of RBMK-1000 are the following: the power is 3200 MWt, the load of uranium fuel with 2% of ^{235}U enrichment, fuel mass is equals to 190 tons, fuel life-time is 3 years.

In a case of radiation accident the greatest danger present radionuclides with the volatile properties such as isotopes of rare gas, isotopes of iodine, caesium, ruthenium and their compounds as well. The high specific activity of short-lived isotopes defines their radiological danger at the early stage of the accident. In fig.1,2 the numerical calculation of the radionuclide specific activities depending on the fuel burn-up are shown. There are the short-lived fission products I-131 ($T_{1/2}$=8,04 d), Sr- 89 ($T_{1/2}$=50,5d), Ru- 103 ($T_{1/2}$=39,4d) and Zr-95 ($T_{1/2}$=64 d) with the daughter Nb-95 ($T_{1/2}$=35,2d) and Nb-95m ($T_{1/2}$=3,6days), Sr-91 ($T_{1/2}$=9,5h), Y-91 ($T_{1/2}$=58d). The short-lived isotopes have typically bell-shaped characteristic with a maximum at approximately of 10 MWt·day/kg U. This fact indicates that when the equilibrium between the radionuclide yield and decay is being reached then the activities decrease as far as the specific power in the assembly is reduced. As a whole the activities of the short-lived fission products strongly depend on the reactor power and their behavior may indicate transient and accidental processes of the NPP.

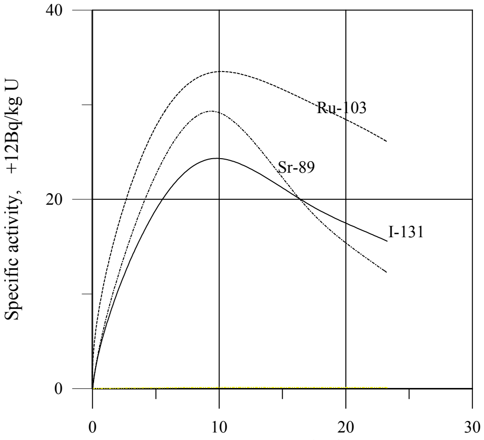

Fig. 1. Sr-89, Ru-103, I-131 specific activity burn-up dependence

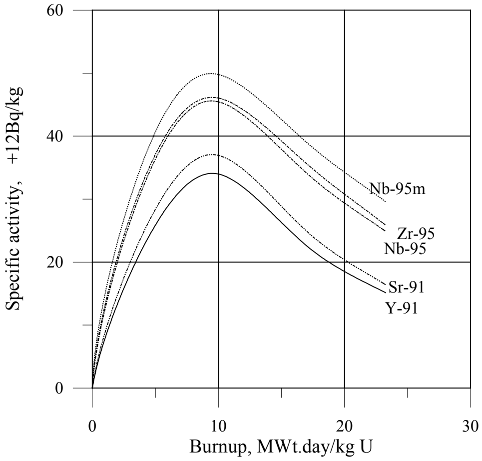

Fig. 2. Zr-95+Nb-95, Nb-95m, Sr-91+Y-91specific activity burn-up dependence

As to the long-lived isotopes, their activities constantly increase as a fuel burn-up is enlarged. In fig.3 the numerical calculation of the radionuclide specific activities of Cs-134 ($T_{1/2}$=2,06 y), Cs-137 ($T_{1/2}$=30,17 y) and Sr-90 ($T_{1/2}$=28,5y) with the daughter Y-90 ($T_{1/2}$=32,5 d), depending on the fuel burn-up are shown.

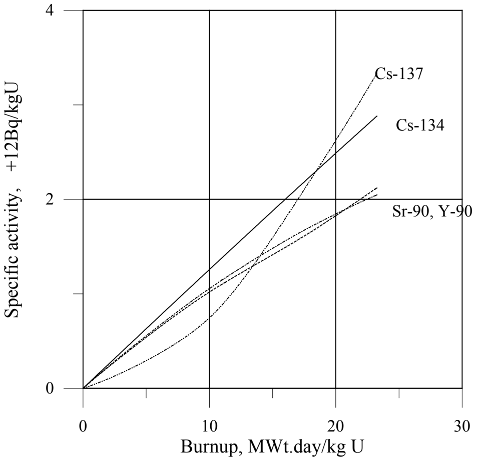

Fig. 3. Sr-90+Y-90, Cs-134, Cs-137 specific activity burn-up dependence

The burn-up dependence of plutonium, curium, americium isotopes reveals truly complex nature caused by the features of their transmutation chains. The burn-up characteristics of the isotopes Pu-239, Pu-240, Pu-241, Pu-242 are represented in fig. 4.

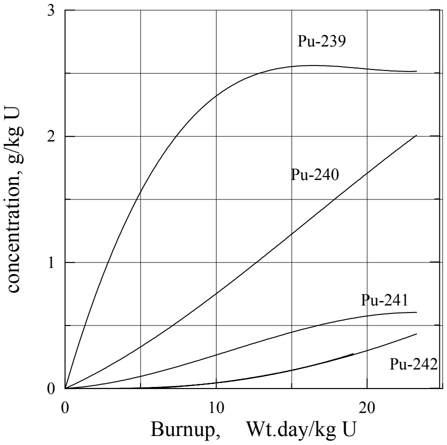

Fig. 4. Pu-239, Pu-240, Pu-241, Pu-242 concentration burn-up dependence

CONCLUSIONS

Lessons learnt after Chernobyl accident in Belarus and summarizing of the experience obtained in neighbor regions as well enable us to offer an information theory approach to radionuclide composition formation of contamination sites. We considered the radionuclide composition formation as well-ordering process. We developed its information model and investigated in details the most essential factors that caused the regularity of its formation. In case of neutron-irradiated nuclear fuel they are power of the NPP and fuel burn-up. Since a correct structure model of a system was reproduced by the bond graph one may define rigorously the data set of the radionuclide activities and to calculate their correlations. With the use of code DECA we calculated radionuclide activity and investigated their dependences of fuel burn up for of RBMK-type reactor. We consider just the set of radionuclide correlation data contains valuable diagnostic information about the acting processes in nuclear fuel.

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NOMENCLATURE

x_i	concentration of i-radionuclide, kg^{-1} U
s_i	constant rate, c^{-1}
y_i,	fission product yield
E^f	fission energy, Mev
w	specific power, Vt/kgU

Greek Letters

Φ	neutron flux density, $cm^{-2} \cdot c^{-1}$
σ_i	neutron capture cross section, barn
λ_i,	decay constant, c^{-1}

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