ROMANIAN JOURNAL OF INFORMATION SCIENCE AND TECHNOLOGY
Volume 27, Number 3–4, 2024, 310–322

Statistical Computational Model of Fission Products Composition of Irradiated Nuclear Fuel and Their Contribution to Gas-aerosol Emissions of Nuclear Power Plants

Sergey TRAVIN[1], Oleg B. GROMOV[2], Gheorghe DUCA[3], and Radu-Emil PRECUP[4, 5, *]

[1]Russian Academy of Sciences, Semenov Federal Research Center for Chemical Physics, Kosygina Street 4, Building 1, 119991 Moscow, Russian Federation

[2]A. A. Bochvar High-Technology Research Institute of Inorganic Materials, Rogova Street 5a, 123098 Moscow, Russian Federation

[3]Institute of Chemistry, Research Center of Physical and Inorganic Chemistry, Str. Academiei 3, 2028 Chisinau, Republic of Moldova

[4]Politehnica University of Timisoara, Department of Automation and Applied Informatics, Bd. V. Parvan 2, 300223 Timisoara, Romania

[5]Romanian Academy – Timisoara Branch, Center for Fundamental and Advanced Technical Research, Bd. Mihai Viteazu 24, 300223 Timisoara, Romania

Email: travinso@yandex.ru, ollgromov@mail.ru, ggduca@gmail.com, radu.precup@aut.upt.ro[*]

[*] Corresponding author

Abstract. Based on the assumption that the radionuclide composition of gas-aerosol emissions from nuclear power plants (NPPs) into the environment is determined by the composition of the fuel in the reactor (but not identical to it), an analysis of the probability of the occurrence of various isotopes in possible emissions is conducted in this paper. It is confirmed that the activity of nuclear fuel, and consequently the emissions, are determined by uranium fission fragments, proving that the decay rate of radionuclides is proportional to the probability of the appearance of radionuclides with certain mass numbers. It is also shown that there is no functional relationship between the mass yield of radionuclides and their atomic mass, as the variability (fluctuations) in the content of radionuclides in VVER-440 reactor fuel and consequently in NPP emissions reaches 20 orders of magnitude, which prevents the application of computational methods for processing the results. This paper proves that, in order to determine the yield of radionuclides from their observable activity, applying a Zipf-Mandelbrot type distribution in "rank - size" coordinates, it is possible to carry out the mathematical modeling based on the appropriate processing of the obtained results.

Key-words: Dose load; isotope; nuclear fuel; nuclear power plant gas-aerosol emission; radionuclide; uranium fission fragments.

1. Introduction

The interest in the composition of radioactive emissions from nuclear power plants is associated not only with public attention to ecology and environmental protection from pollution but also with the need for state regulation of their permissible levels. According to Article 23 of the Federal Law of the Russian Federation on Environmental Protection, emissions of radioactive substances into the environment are permitted within established norms based on permits issued by executive authorities that exercise state management in the field of environmental protection [1], [2]. Traditionally, for most nuclear power plants (NPPs), emissions are regulated by inert radioactive gases (IRG, including gaseous isotopes of argon, krypton, and xenon); ^{131}I (gaseous and aerosol compounds); ^{60}Co (aerosol); ^{134}Cs (aerosol); ^{137}Cs (aerosol). Some concern is also caused by the emissions of radiocarbon and tritium. According to the results of radiation-technical surveys conducted at Russian NPPs, ^{14}C is among the fifteen radionuclides that account for at least 99% of the radiation dose to the critical group of the population from NPP emission sources [3]. However, it is justified in [4] and [5] to consider the non-criticality of radiocarbon in NPP emissions.

In recent years, the number of publications on the study of the composition of aerosol emissions from NPPs has decreased somewhat. The main reason for this seems to be that within the established paradigm, the problem is considered sufficiently studied, while new ideas for the theoretical understanding of isotopic composition arise relatively rarely. Even in the recent papers including [6], tables are provided that contain only twenty-two radionuclides and empirical coefficients for converting their radiological equivalence to ^{131}I.

However, after the publication of the Ministry of Natural Resources Order No. 579 [7], which prescribed the regulation of emissions for each isotope of 92 chemical elements, the interest in the topic of regulating radionuclide emissions and answering the question about the composition of radioactive emissions became more in demand than ever before since some elements, such as iodine, have over forty radioactive isotopes. Accordingly, the total list of proposed regulated radionuclides exceeded 3000 names, including isotopes of the rarest element astatine, the total amount of which in the atmosphere of the planet Earth does not exceed one gram. This order was canceled shortly after its issuance, other standards were released (mainly for calculating the contribution of each radionuclide to the dose load for the population as, for example, in [8] and [9]), but the question of the necessary and sufficient regulation of radionuclides released into the air remains open.

The aim of this paper is to systematize the probability of gas-aerosol emissions from NPPs and provide a theoretical description of the radionuclide composition of spent fuel, which is undoubtedly the most important determining factor of radioactive environmental pollution. In the course of numerous discussions of the material of this paper, the authors were repeatedly pointed out that the total radioactive emissions from NPPs consist not only of the fission products of the nuclear fuel itself but also of radionuclides formed from the structural materials of the NPP reactor under irradiation (so-called induced radioactivity). This is undoubtedly a contributing

factor, but even in the old paper of Ponomarev-Stepnoi and co-authors [10], it was shown that during reactor operation, along with the formation of new radioactive elements, part of the initial radioactive nuclei of the fuel load burns out. This paper also provides a quantitative comparison of the balance of destroyed and newly appearing radioactive decays and their behavior over time. The fuel cycle includes various stages, during which the fuel is concentrated with the formation of corresponding tailings, so it is important to evaluate the balance of radioactivity in relation not only to the initial fuel in the reactor but also to the production tailings. As criteria for assessing radioecological safety, data on their radioactive decays (total number of decays, decay rate), energy release, radiation, and chemical toxicity should be used.

The ratio of the number of emerging decays to the number destroyed in the reactor $P_{\text{new}}/P_{\text{destroyed}}$ is less than unity, and after the decay of the main mass of fission products, it is 0.2 for VVER-thorium, 0.3 for VVER, 0.4 for BN. Thus, even for the most "unfavorable" reactors, the proportion of induced radiation is many times less than its level determined by the fission products of nuclear fuel. For this reason, the main attention was paid to the statistical analysis of the composition of fission products, while the issues of induced radiation were left without attention in this paper.

This paper is structured as follows: source data and research methodology are treated in the next section. Section 3 is focused on results and discussion. The conclusions are drawn in Section 4.

2. Source Data and Research Methodology

As a source of initial data for subsequent analysis and systematization of emitted radionuclides, this paper starts with the handbook [11], where the radionuclide composition of spent nuclear fuel and the patterns of ongoing campaigns are presented quite fully. The main hypothesis underlying this paper is based on the law of conservation of matter, which can be formulated as follows: the radionuclide composition of gas-aerosol emissions from NPPs into the atmosphere is determined by the composition of the working fuel in the reactor mixture, and the variability of measurable radionuclide compositions in the emission is determined by the efficiency of the filters used at NPPs and the time delay of their release into the atmosphere [12], [13]. For example, for short-lived isotopes, including inert gases, significant activity reduction is achieved precisely by the time delay in the filters.

The quantity of each type of radionuclide entering the atmosphere as aerosols is obviously proportional to the radionuclide content in the working fuel, its penetration capability through the filter system, and the time of radionuclide accumulation, *i.e.*, the interval between purges. Such dependence can be represented by the formula

$$N_{aerosol} = N_{fuel} \cdot D(radionucleide) \cdot \Delta t(reactor), \tag{1}$$

where: $N_{aerosol}(pcs)$ – the number of radionuclide atoms entering the atmosphere, $N_{aerosol}(pcs/t)$ – the radionuclide content of this type in the fuel, $D(radionucleide)(t/d)$ – the radionuclide penetration coefficient (depends not only on the type of radionuclide itself but also on the type of reactor and, most importantly, the filtration system), and $\Delta t(reactor)(d)$ – the filter delay time (for IRG) or the time between purges.

In turn, data on the radionuclide composition of nuclear reactor fuel at any given moment of their operation would allow creating a predictive model for dozens and even hundreds of potentially dangerous isotopes based on measurements of only a few most characteristic components.

Currently, the primary control is conducted for the sum of IRG and isotopes ^{131}I, ^{60}Co, ^{134}Cs, ^{137}Cs. Although the emission of IRG accounts for over 99% of the activity, these gases are easily mixed in the atmosphere, after which they cease to pose a significant danger, whereas aerosol-type radionuclides settle and tend to accumulate in plants, soils, and bottom sediments of water bodies.

Given the above, the quantitative ratio of aerosol element isotopes is of independent interest. It should be noted that in various European countries, NPPs with PWR reactor installations control from 1 to 53 radionuclides (without considering total indicators), and in total up to 83 radionuclides [10], while at Russian NPPs, usually no more than seven radionuclides are controlled in emissions. Obviously, if a "spectrum of presence" of radionuclides can be obtained, *i.e.*, a functional dependence of their quantity on some objective indicator, it would be possible to extend the control data of the selected number of isotopes to a much broader set. One of the problems that is tried to solve in this paper is precisely the search for such an objective indicator. As will be seen later, neither the atomic number nor the atomic mass of the radionuclide can serve as such an indicator.

The diagram given in Fig. 1 shows the distortions that the permeability coefficient that can be derived using (1) can introduce into the original distribution of isotopes in the fuel, changing it beyond recognition in the aerosol composition. As can be seen in Fig. 1, the mass yield of isotopes in aerosols is determined not only by their quantity in the fuel but also by their penetrating capability. It is emphasized that this diagram is for demonstration purposes and does not refer to any specific set of elements.

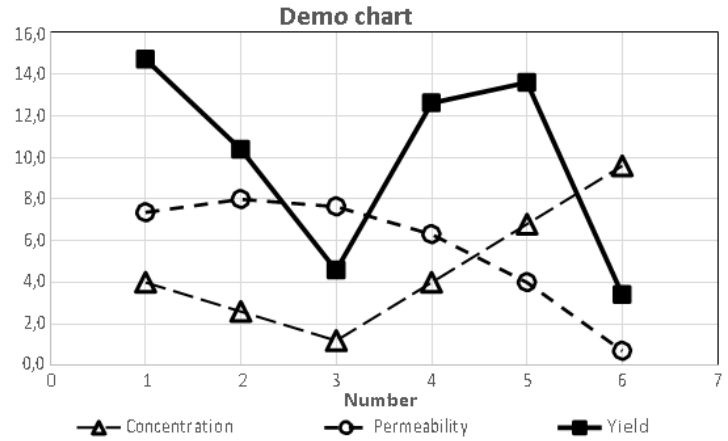

Fig. 1. Demonstration model of radionuclide yield calculated using (1).

The final composition of emissions (controlled after passing through filters) can and should differ significantly from the radionuclide composition in the reactor core. However, the efficiency of the filters only makes adjustments to the primary composition of the products striving to escape from the fuel. Simply put, only those radionuclides that showed a noticeable number of decays in the active zone can exhibit noticeable radioactivity after passing through the filters.

It is specified in [6] that the multistep mechanism of the leakage of radioactive components from the reactor into the environment. When defects of the type of gas-tightness occur in fuel rods, the mechanism of radionuclide transfer from irradiated fuel into the coolant is determined (simplified) by the following processes: the diffusion of gaseous, volatile, and low-melting *fission*

products (FP) from the fuel matrix (mainly from narrow micro-areas in the matrix, representing the boundaries between grains; from the grains themselves - orders of magnitude less) into the space under the fuel rod cladding, the diffusion of these nuclides from under the cladding through gaps into the coolant. Through this mechanism, mainly FP can enter the coolant. Alpha-active radionuclides, *i.e.*, exclusively various isotopes of actinides, which are not FP, are immobilized in the fuel matrix and cannot enter the coolant in significant quantities through diffusion in the fuel and through the fuel rod cladding.

Although this paper is focusing on the permeability of barriers between the reactor and the outside world (which should be the subject of a separate study) and the composition of FP, the authors express their disagreement with some of the conclusions of [6]. In that paper, it is stated that the coefficients characterizing the share of radionuclide activity "penetrating" through physical barriers are determined only by permissible coolant leaks from the primary to the secondary circuit and emissions from turbine ejectors, *i.e.*, it is assumed (without any justification) that the "leakage" of any radionuclide does not depend on its nature or on the type and chemical compound it is part of.

Such an assumption has no chemical basis and contradicts well-known experimental facts. Thus, the release of IRG into the atmosphere is at least two orders of magnitude higher than the total emission of all other elements, and it is IRG that contribute 99% to the overall radioactivity of aerosols. In our profound conviction, this is precisely because IRGs do not form any condensed products, exist exclusively in a gaseous state, and practically do not bind (neither adsorptively nor chemically) with filter materials. For this reason, cleaning emissions from IRG is particularly difficult and relatively ineffective. Fortunately, most IRG isotopes are short-lived, so even the simplest delay in flow retention chambers (a sealed volume inside which a labyrinth is organized for the gas flow) provides a noticeable reduction in the radioactivity of emissions into the environment.

Therefore, this paper emphasizes once again that fission products of nuclear fuel formed during NPP operation are of primary interest in terms of radiation safety. In turn, data on measurements of only a few most characteristic components of nuclear reactor fuel at any given moment of their operation would allow creating a predictive model of the radionuclide composition for dozens and even hundreds of potentially dangerous isotopes. Currently, primary control is conducted for the sum of IRG and isotopes ^{131}I, ^{60}Co, ^{134}Cs, ^{137}Cs. Although IRG emissions account for over 99% of the activity, these gases are easily mixed in the atmosphere, after which they cease to pose a significant danger [13], [14], whereas aerosol-type radionuclides settle and tend to accumulate in plants, soils, and bottom sediments of water bodies [15].

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In the initial stage of the research conducted in this paper, the characteristics of a spent fuel assembly of the VVER-440 reactor with loaded fuel enriched to 3.6% (by ^{235}U) were selected.

The main conclusions of the paper will also be valid for VVER-1000 and RBMK-1000 reactors.

The handbook [11] provides data on the activity (Bq/t) of irradiated fuel. Therefore, to obtain the mass of each isotope and the number of its atoms, the well-known formulae are used for recalculation:

$$\begin{aligned} N(t) &= N_0 \cdot e^{-kt}, \\ A(t) &= -\frac{dN(t)}{dt} = k \cdot N_0 \cdot e^{-kt}, \\ k &= \frac{\ln 2}{T_{1/2}}, \end{aligned} \tag{2}$$

where: t – the time variable, $N(t)$ – the calculated number of radionuclide atoms per ton of fuel, N_0 – the initial number of radionuclide atoms per ton of fuel, – the radionuclide decay rate constant, $A(t)$ – the radionuclide activity, and $T_{1/2}$ – the half-life period. Relation (2) results in

$$N(t) = A(t) \cdot \frac{T_{1/2}}{\ln 2}. \tag{3}$$

3. Results and Discussion

As assumed previously, there was no clear pattern or correlation between the mass (actually quantitative) yield of uranium fission products and their atomic mass as illustrated in Fig. 2, where $1, E + X$ generally indicates 10^X.

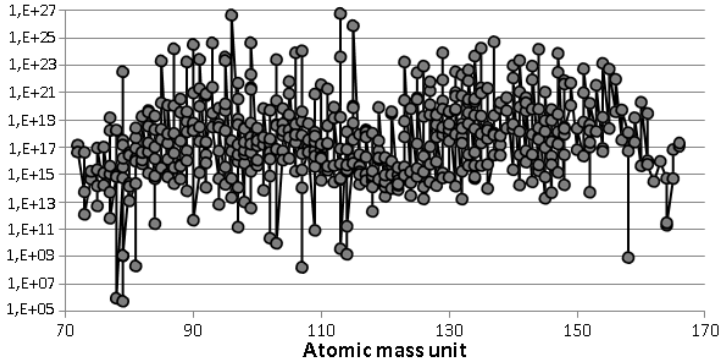

Fig. 2. Mass yield of element isotopes according to the data given in [9].

The scatter in radionuclide content is about 20 orders of magnitude, and the graph resembles a highly noisy signal: the noise level significantly exceeds possible information-containing trends highlighted in Fig. 2 and Fig. 3.

It is noted that uranium fission fragments with a mass less than 70 amu are practically absent. For the same reason, the second uranium fission fragment with a mass greater than 170 amu is also absent. A logarithmic scale was chosen for the ordinate axis since the variation in the content of different isotopes exceeds 20 orders of magnitude.

Fig. 3 shows a fragment of Fig. 2 with the indication of all isotopes and their atomic masses recalculated according to (2). It is outlined that in the atomic mass range from 94 to 99, there are over 40 isotopes.

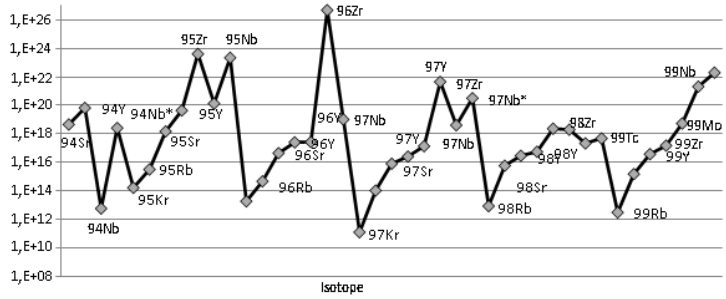

Fig. 3. Quantitative yield (in pieces of atoms per ton of fuel) of radioactive isotopes from strontium ^{94}Sr to niobium ^{99}Nb in irradiated nuclear fuel VVER-440.

It is also well known that under the influence of neutron irradiation of the fuel material (mainly a mixture of ^{238}U and ^{235}U), the fission of ^{235}U atoms occurs into two fragments of different masses [17]. Since these fragments, in turn, may be unstable, *i.e.*, subjected to radioactive decay, it can be assumed that unlike concentration, their decay rate will follow known physical laws. Consequently, in steady-state conditions, uranium atoms in the fuel assembly are "fragmented" at a constant rate. The rate of fragment formation with different atomic masses follows the known "double-hump" distribution law [18] and equals the rate of their disappearance, *i.e.*, the activity corresponding to the isotope with the relevant mass number. Therefore, the decay rate (not the steady-state concentration) is proportional to the primary probabilities of the appearance of fragments with certain mass numbers.

Based on the above, it is tried in this paper to compare not the mass yield but the experimentally observed activity, *i.e.*, the decay rate of radionuclides with their atomic weight (mass).

Fig. 4 shows the dependence of the observed activity of spent nuclear fuel (*i.e.*, the decay rate) on the atomic weight of the radionuclide. The envelope line of observed activity peaks was obtained by numerically substituting two Gaussian distributions of the form

$$f_i(x) = \frac{1}{\sigma_i\sqrt{2\pi}} e^{-0.5\frac{(x-\mu_i)^2}{\sigma_i^2}}, \quad i \in \{1,2\}, \tag{4}$$

with the parameters: $\mu_1 = 97.5$, $\sigma_1 = 6$, $\mu_2 = 138.5$, $\sigma_2 = 6$, $\mu_2/\mu_1 = 1.42$, and $\mu_1+\mu_2 = 236$, where: x (amu) – the actual mass of one of the fragments (larger or smaller, does not matter), μ_1 (amu) – the average mass of fragment 1 (smaller), μ_2 (amu) – the average mass of fragment 2 (larger), σ_1 and σ_2 – the standard deviations for two peaks of the distribution. Obviously, $\sigma_1 = \sigma_2$ because the masses of the fragments are not independent, but complement each other to the mass of the fissioning nucleus.

The-least squares approach was applied to derive the values of the parameters in (4) accounting for the above constraint. However, metaheuristic algorithms are also popular recently as, for example, quantum annealers [21], ant colony optimization [22], slime mould algorithms [23], particle swarm optimization algorithms [24], and string theory algorithms [25]. These algorithms are efficient, however the mathematical guarantee of their convergence is generally not solved.

The sum of the abscissas of the maxima practically coincides with the mass of the fissioning element (uranium), and the widths of the distributions are the same. Thus, this paper deals not with two but essentially with one Gaussian distribution, with the second one being the com-

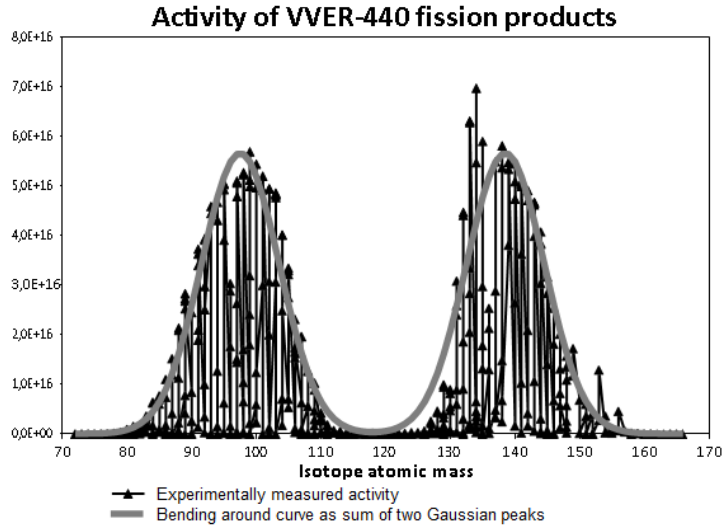

Fig. 4. Distribution of VVER-440 fission products' activity by isotope mass numbers and envelope as the sum of two Gaussian peaks.

plement of the mass of the other fragment to the whole atom. The amplitudes of the Gaussian distributions also coincide. The slight deviation of the sum of the peak mass centers from 235, *i.e.*, from the mass of the fissioning uranium atom, is likely caused by the fact that the heavier isotope ^{238}U also participates in the fission. In Fig. 4, a regular linear scale was used for the ordinate axis. Therefore, less than 10% of the points (about 50 out of 513 presented in the handbook [11]) are distinguishable on the graph. The rest of the points are indistinguishable due to their graphical coincidence with the abscissa axis.

The same data is presented more clearly in a logarithmic ordinate scale in Fig. 5, although the Gaussians look unusual. Nevertheless, the experimental points themselves are clearly distinguishable. This graph once again confirms the impossibility of functionally-analytically correlating the radioactivity (decay rate) of isotopes with their atomic mass.

Although in [18] this type of distribution is called "by masses", it is obvious that the author meant "by mass numbers". Its characteristic feature is outlined, namely the asymmetry of the fragment mass distribution. For the fission of ^{235}U, the ratio of the average masses of the heavy and light fragments is approximately 1.46, which practically coincides with the calculated curve in Fig. 3. With an increase in excitation energy, the probability of symmetrical fission increases, while the probability of asymmetrical fission changes very little. For this reason, the depth of the "trough" is maximal for low-energy, *i.e.*, thermal neutrons (Fig. 4). This case is realized in VVER and RBMK reactors.

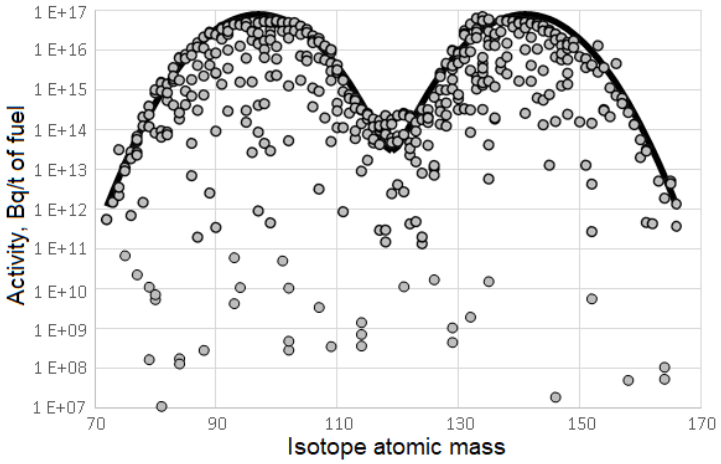

Fig. 5. Distribution of fission fragments by isotope mass numbers during the fission of ^{235}U under the action of thermal neutrons and the envelope of two Gaussians along with experimental points in [11] (values are the same as in Fig. 4, but the ordinate scale is logarithmic).

The ratio of probabilities in the minimum and maximum for ^{235}U fission products is equal to 600, and similarly, the ratio of activities in Fig. 5 also reaches approximately three orders of magnitude. At the same time, individual downward outliers related to more stable isotopes can reach 13-14 orders of magnitude. It is clear that from the point of view of analyzing radioactive pollution and aerosol emissions, they neither represent a danger nor interest.

Finally, another way of presenting the dependence of radioactivity on the atomic mass of the fission product is shown in Fig. 6. The data used are similar to those in Fig. 4 and Fig. 5, but normalized to the total radioactivity and expressed as a fraction of the overall radioactive background.

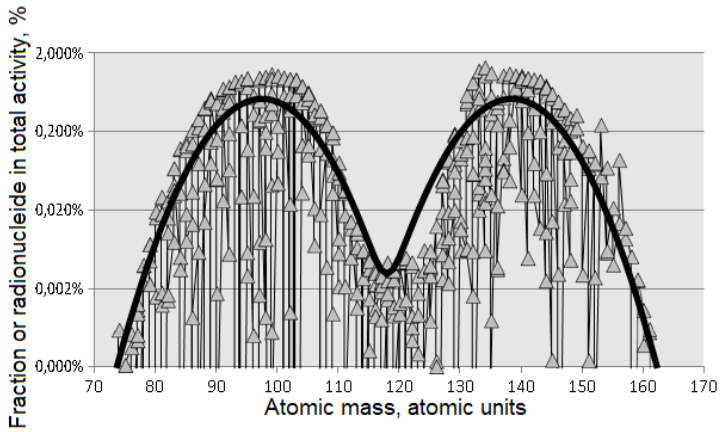

Fig. 6. The share of radioactivity for VVER-440 fuel fission products depending on atomic mass.

Analyzing Fig. 6, it is evident that even for the first two or three dozen most radioactive fission fragments, none of their contributions exceed 2%. Therefore, for a representative analysis, it is necessary to take into account significantly more radionuclides. This fact suggests that all fission fragments should be ranked by activity and that a functional relationship should be sought not with the physical parameters of the fragment but with its position in the rank distribution according to the quantitative sample from the total emission in "rank-size" coordinates.

The obtained dependence, essentially analogous to the graphs of fission product yields depending on their activity according to Fig. 2 to Fig. 5, is presented in Fig. 7. Analyzing Fig. 7, it is clear that, unlike the unpredictable "jumps" in the case of choosing atomic mass or atomic number of the radionuclide as the independent variable (abscissa), the rank distribution shows a smooth functional dependence. Unlike the mass range with a width of no more than 100 amu, the rank distribution covers all positions given in [10].

It is claimed in this paper that in this case, it is dealt with one of the varieties of the Zipf-Mandelbrot distribution. A more detailed consideration of this distribution concerning the activity distribution of VVER-440 reactor fuel fission products will be presented as part of the future research in the framework of another paper.

Concluding, the sum of the mass numbers of the "light" and "heavy" peaks accurately falls within the range of 235-238 amu, indicating that the accumulation of deeper products is insignificant, and all activity of spent fuel is gained by uranium fission fragments.

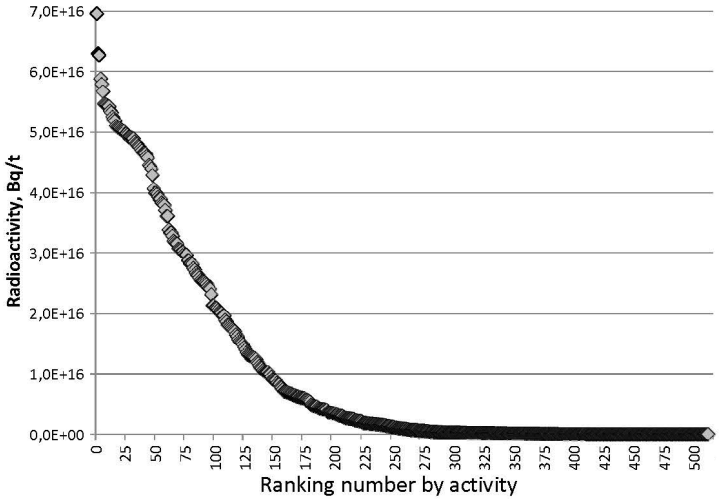

Fig. 7. Rank distribution by activity of fission products of reactor VVER-440.

4. Conclusions

This paper developed a statistical computational model of fission products composition of irradiated nuclear fuel and their contribution to gas-aerosol emissions of nuclear power plants (NPPs). The following hypothesis is proposed: the primary (before the application of filters and gas purification systems) radionuclide composition of gas-aerosol emissions from NPPs with

VVER-440 reactors into the environment corresponds to the composition of the fuel in the reactor, with the activity of spent fuel being determined by uranium fission fragments.

The fluctuations in the content of radionuclides in VVER-440 reactor fuel, and consequently in NPP emissions, are up to 20 orders of magnitude. The graphical results show that the noise level significantly exceeds the information-content trends, which prevents strict mathematical analysis of the radionuclide mass yield into the environment. Furthermore, it is proven that the decay rate of radionuclides is proportional to the probability of the appearance of fragments with certain mass numbers, and the ratio of activities of emitted radionuclides representing real danger reaches three orders of magnitude in absolute terms.

Future research will be focused on the continuation of the description of this modeling approach. Being relatively easily understandable, the approach will be applied to data measured in other representative applications including well-being [26], evolving systems [27] and telesurgical applications [28] involving several models as, for example, fuzzy cognitive maps in path planning for multi-agent systems [29], finite element formulation [30], resource provisioning [31], cognition process modeling [32], and data streams [33].

Acknowledgement. This work was supported by a grant of the Ministry of Research, Innovation and Digitization, CNCS/CCCDI - UEFISCDI, project number ERANET-ENUAC-e-MATS, within PNCDI IV, and by the national project of Republic of Moldova number 20.80009.5007.27.

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