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STUDY OF THE RADIONUCLIDE COMPOSITION OF RADIOACTIVE WASTE STREAMS IN THE NUCLEAR POWER PLANT

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RADIONUKLIDINĖS SUDĖTIES TYRIMAS ATOMINĖS ELEKTRINĖS RADIOAKTYVIŲJŲ ATLIEKŲ SRAUTUOSE

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INTRODUCTION

Relevance of the work. Continually increasing consumption of energy, expansion of new industrial branches and technology unavoidably cause problems related to the harmful impact of industrial waste on the environment and people. Due to the increasing technogenic load and declining self-regulation capabilities of the environment, the society is obliged to invest in the environment preserving development ways. Nuclear energy has no alternatives from this point of view. However, it is essential to ensure high nuclear safety and radiation protection level, resolve technological tasks of radioactive waste management, understand mechanisms of radionuclide migration in the environment and better conceive aspects of ionizing radiation impact on the environment and people. It is relevant to optimisation of occupational exposure and radiation protection of public during operation of nuclear facilities as well as processing, storing and disposal of radioactive waste. These issues are particularly relevant to decommissioning of nuclear facilities, because many new technological and radiation safety aspects concerning large radioactive waste streams are not fully clear.

The work is closely related to the main task of energy in Lithuania during this decade – safe decommissioning of the Ignalina NPP. Modern radioactive waste management, utilization and disposal technologies shall be used, ensuring long-term safety and minimum impact of ionizing radiation on the environment and people.

During operation of nuclear power plants and their decommissioning, dismantling of installations and buildings, tens or more thousands cubic meters of radioactive wastes are generated. Those wastes are not equally hazardous from radiation safety and nuclear safety viewpoint due to different specific activities of radionuclide and other physicochemical characteristics. Hence, the first step in the assessment of potential radiological impact of radioactive waste is the estimation of the nuclide composition. It is an essential characteristic of operational radioactive waste and contamination of buildings and equipment for dividing radioactive waste to the streams for subsequent processing and disposal. Lists of radionuclides, indicating significant radionuclides in assessment of ionizing radiation impact on the environment and people, are compiled in various countries. The united list of significant radionuclides suitable for the whole variety of nuclear facilities does not exist. Therefore, it is relevant to assess the nuclide composition of nuclear fuel and activated materials, considering materials of nuclear fuel and reactor structures and characteristics of neutron flux, and to define safety-relevant radionuclides from the radiation safety viewpoint. It is particularly relevant to nuclear power plants with RBMK-1500 reactors, as the spent nuclear fuel operational and other radioactive waste treatment, storage and disposal technologies during decommissioning of the plant are implemented for the first time. Accumulated scientific knowledge on the theoretical and experimental evaluation of the nuclide composition of operational and decommissioning radioactive waste would be useful when selecting optimal technologies, assessing possible scenarios of radionuclide migration from repositories, predictions and outcomes of unwanted radioactive pollution. The increasing demand for nuclear energy worldwide stimulates the relevance of these problems and solution of arisen tasks requires new scientific knowledge and its creative practical implementation.

The aim of the work was to develop a model of formation of radioactive waste streams in the Ignalina NPP technological media and methodology for assessment of the radionuclide composition. To achieve this aim the following **objectives** were set up:

1. to develop methodology of analysis for compilation of the list of radiation protection safety-relevant radionuclides, suitable for all Ignalina NPP radioactive waste streams and radioactive waste disposal options.
2. to investigate the sequence of radionuclide composition variance, encompassing technological processes from radioactive waste generation to disposal, and to create the radioactive waste generation scheme, explaining the nature of formation of radioactive waste streams with a different radionuclide composition.
3. to assess possibilities of direct and indirect radioactive waste characterization methods and develop a nuclide vector application scheme for assessment of the nuclide composition of radioactive waste streams at nuclear power plants with RBMK type reactors.
4. to establish nuclide vectors of the main Ignalina NPP radioactive waste streams and evaluate the accuracy of the method, optimal application conditions and limits.

Novelty of the work. The novelty of this work is governed by the particularity of the research object – Ignalina NPP – and lack of comprehensive scientific knowledge of processes ongoing in the RBMK reactor and formation of radioactive waste streams. In the work, a complex of computer modeling, radiochemical analysis and nuclear spectroscopy methods was applied. It is a new indirect methodology of estimation of difficult-to-measure nuclides for the nuclear power plant with the RBMK type reactor proposed and implemented for the first time.

In this work scaling factors for the RBMK reactor operational radioactive waste (conditioned liquid radioactive waste and filters used for water purification, and solid radioactive waste) were established for the first time. Moreover, the nuclide composition of contamination of nuclear power plant equipment was estimated. By analyzing experimental data, intermediate key radionuclides for estimation of actinides and fission products were proposed. In the work recommendations on application of scaling factors for characterization of different radioactive waste streams are given.

Statements presented for defence

1. The scaling factor method is suitable for characterization of RBMK-1500 radioactive waste. As a key nuclide gamma emitter (^{60}Co or ^{137}Cs) is selected, in evaluating its correlation with the difficult-to-measure radionuclide.
2. Scaling factors of fission products to ^{60}Co depend on the radioactive waste stream, scaling factors of actinides depend slightly and dependence of scaling factors of corrosion products is not observed at the existing experimental accuracy.
3. One nuclide vector for characterization of RBMK radioactive waste is not sufficient – establishment of separate nuclide vectors is necessary for different waste streams, which are governed by NPP technological peculiarities.

4. Solid radioactive waste stream is not homogeneous. Due to different ways of equipment contamination and ongoing technological processes solid radioactive waste stream splits into several streams with a different radionuclide composition.

Structure of the dissertation. The dissertation consists of introduction, list of original scientific publications, three chapters, conclusions and the list of references (125 entries). Materials of the dissertation are presented in 127 pages, 28 figures and 16 tables.

METHODOLOGY

Experimental determination of scaling factors

The most obvious method for the radionuclide activity determination is the direct measurement. However, not all safety-relevant radionuclides can be measured directly. This is due to the fact that activity concentrations of some long-lived radionuclides are low, there are no medium or high energy gamma lines in their decay schemes as well as due to the absorption of ionizing radiation (first of all of alpha and beta particles) in the waste materials and packages. Therefore, for the determination of the radionuclide concentration in waste, indirect methods, both semi-empirical and analytical, are used. In this work the methodology for the radionuclide composition determination is based on the experimental measurements of specific activities of various radionuclides and on the computer modeling results of nuclear fuel composition and the reactor construction material activation in the reactor neutron flux.

The scaling factor method is widely used worldwide for characterizing the radioactive waste nuclide composition. This method allows characterizing the waste radiologically by using nondestructive methods. Some of radionuclides are easy-to-measure due to emission of high energy gamma rays (^{60}Co and ^{137}Cs). Having measured activity of easy-to-measure (key radionuclides) and difficult-to-measure radionuclides by direct destructive laboratory measurements one can easily calculate the activity ratios, called scaling factors. Later on the determined scaling factors are used to determine activities of difficult-to-measure radionuclides from measured activities of key radionuclides. The most frequently used linear dependence between specific activities of nuclides in the investigated sample is:

$$A_i = k_i A_{key}, \tag{1}$$

where A_i is the specific activity of the difficult-to-measure radionuclide, A_{key} is the specific activity of the easy-to-measure key radionuclide, k_i is a constant, called the scaling factor.

The scaling factors of radionuclides, the specific activity of which can be measured by α, β, and γ spectrometric methods, are determined by the measurement runs, statistically processing the results according to the correlation of the investigated radionuclide with the key nuclides. To ensure that results are reliable, the sample should meet the Grubb's test and the measured specific activities should cover the whole possible interval. In this case, the upper and lower boundaries of the scaling factor are directly obtained from the correlation function confidence interval.

Gamma spectrometry was applied using gamma-ray spectrometers, stationary and portable, with high purity germanium (HPGe) semiconductor detectors. The stationary spectrometer comprised three Ge detectors, with relative efficiencies of 38%, 30% and 15% and the respective energy resolution of 2.05 keV, 1.80 keV and 1.80 keV at 1333 keV. Detection limit of ^{137}Cs and ^{60}Co is 0.012 Bq and 0.020 Bq, respectively, for the detector with the relative efficiency of 38%, 0.13 Bq and 0.15 Bq for the detector with the relative efficiency of 30% and 0.15 Bq and 0.18 Bq for the detector with the relative efficiency of 15%. This spectrometer ensures the measurement of absolute activity of all radionuclides in the 122-1461 keV energy range with the uncertainty not exceeding 6%. The coincidence-summing corrections were applied when measuring activities of radionuclides whose decay schemes incorporated the cascade transitions (^{60}Co, ^{94}Nb and ^{134}Cs). The counting efficiency was also corrected for the sample aliquot density; the final result was decay-corrected to the sampling date. The activity of short-lived radionuclides originated in water of the main circulation circuit was measured with the portable spectrometer immediately after the coolant sampling. The activity of these radionuclides was determined with the uncertainty not larger than 30%. The relative efficiency of the portable detector was 20%, while the energy resolution was 1.80 keV at 1333 keV.

A radioanalytical method for the determination of ^{63}Ni, ^{55}Fe, ^{90}Sr, ^{241}Am, $^{242,243,244}Cm$, $^{238,239,240}Pu$ and ^{99}Tc in nuclear waste based on decomposition of samples, precipitation and liquid–liquid extraction followed by extraction and anion exchange chromatography was applied. Iron hydroxides, calcium oxalate, calcium phosphate and nickel dimethylglyoxime were used to precipitate and separate radionuclides from the matrix elements after their leaching from samples. The separated radionuclides were further purified to remove interfering radionuclides using the Bio-Rad anion exchange resin from the Bio-Rad Laboratory (USA) and/or commercially available Eichrom Ni, Sr and TRU, UTEVA and TEVA resins (Eichrom Technologies, Inc., IL 60561, USA) (Horwitz et al., 1993, 2005; Maxwell, 2006; Hou et al., 2005; Horwitz et al., 1995).

β spectrometry was applied using the liquid scintillation beta spectrometer Quantulus-1220. The measurement uncertainty was not higher than 10%. The radionuclide detection limits at the measurement duration of 1 hour for ^{55}Fe were 0.036 Bq, for ^{63}Ni – 0.022 Bq, for ^{90}Sr – 0.027 Bq, for ^{241}Pu – 0.032 Bq. The element radiochemical extraction yield is 30% for ^{55}Fe, 70% for ^{63}Ni, 85% for ^{90}Sr and 35% for ^{241}Pu.

The activity of actinides was determined with the alpha spectrometer, the detection limit at the measurement duration of 100000 s being 0.001 Bq. This corresponds to the specific activity of 0.4 Bq/kg at the 22% measurement uncertainty. The chemical yield of the analytical procedure changes from 70 to 90% for Pu, from 60 to 80% for Am, from 70% to 90% for U. The minimal detection limit for Pu is 0.001 q/sample, for Am it is 0.0015 Bq/sample, for U – 0.001 Bq/sample.

Computer modeling

Modeling of the radionuclide generation in nuclear fuel can also be used in the cases when it is complicated and expensive to make measurements. As the main source of the nuclide origin in waste is the nuclear reactor, so the nuclide formation in the nuclear fuel and construction materials can be simulated using computer codes such as

SCALE, MCNP coupled with ORIGEN. Applicability of the codes to the RBMK reactor is demonstrated (Ancius et al., 2005; Remeikis and Jurkevicius, 2004; Remeikis et al., 2007). The nuclear fuel composition and activation of the fuel cladding, the fuel assembly construction materials and fuel channel are evaluated using the Origen-ARP program, which solves differential equations of the nuclide amount variation in the neutron flux and due to natural decay. The neutron interaction with the material is characterized by capture and fission cross-sections of one group, which are taken from the libraries compiled specially for the RBMK-1500 reactor. The libraries were compiled using the SAS2 program of the SCALE 4.4a package for the 2.0÷3.0% enrichment ^{235}U fuel without or with the appropriate amount of burnable Er admixture. For characterization of some radioactive waste streams modeling was performed by applying 2D depletion sequence TRITON from the program package SCALE 5 (DeHart et al., 2005). The fuel burnup can vary from 0 to 45 MWd/kgU, and the coolant density – in the range of (0.1÷1.0) g/cm^3. The initial 2.4% enrichment ^{235}U fuel with 0.41% Er admixture, whose amount at present is the largest, was chosen for modeling. The fuel composition at the 12 MW d/kgU burnup (for the average fuel burnup in the Ignalina NPP reactors) has been calculated. The results of comparative calculations for 2.0% and 2.6% fuel enrichment differ insignificantly from the calculations for 2.4% fuel enrichment. Comparative calculations where performed for 5, 16 and 21 MWd/kgU fuel burnup. Decay of radionuclides while fuel channel and assembly are in the reactor was taken into account.

Radionuclide generation and migration pathway from fuel to transfer medium was taken into account for calculation of the correlation dependence. In case the pathway of analyzed radionuclides was significantly different due to different origin of radionuclides (fission or activation product), different chemical properties (e.g., ^{134}Cs, ^{90}Sr, actinides, and key nuclide ^{60}Co), the additional key nuclides, whose calculated activities in nuclear fuel and the MCC coolant or in radioactive waste correlated better, were selected, e.g., $^{239+240}Pu$ in case of actinides and ^{137}Cs in case of fission products. Then the scaling factor was calculated by an equation:

$$k_i = k_{ij} k_j, \qquad (2)$$

where k_i is the scaling factor of the investigated radionuclide, k_{ij} is the scaling factor of the investigated radionuclide in respect of the intermediate key radionuclide, k_j is the scaling factor of the intermediate radionuclide.

The data of measured radionuclide activity were approximated by means of linear fitting procedure. The data clearly show the linear dependence function between specific activities when data are presented on a logarithmic scale. A linear function can be used for fitting of data:

$$\lg(A_i) = \lg(k_i) + b \lg(A_{key}), \qquad (3)$$

where b is the line slope and other terms are described in Eqs. (1) and (2). When a fit is good enough as presented in Fig. 3 (a and b), b is close to unity and fit function is identical to Eq. (1). From confidence bands it is possible to obtain uncertainty of the scaling factor and from upper prediction band one can find the upper limit of the nuclide activity in the radioactive waste when the key nuclide activity is known.

Fit function confidence bands are evaluated by equation:

$$\overline{Y}_{x_0} \pm t(1-\alpha/2, n-2)\cdot s\{\overline{Y}_{x_0}\}, \tag{4}$$

where $\overline{Y}_{x_0}$ is the most probable specific activity value of the difficult-to-measure radionuclide at value x_0 (specific activity of key radionuclide, t is the Student coefficient, n is the number of data, *(1-(1-α)/2)* significance level, where α is the confidence level (applied value was 0.95),

$$s^2\{\overline{Y}_{x_0}\} = S_{\overline{Y}_{x_0}} \cdot \left[1/n + (x_0 - \overline{x}_i)^2 / \sum (x_i - \overline{x})^2\right], \tag{5}$$

where $S_{\overline{Y}_{x_0}}$ is the mean square error of $\overline{Y}_{x_0}$.

The fit function confidence interval indicates how correctly values of the fitting function are estimated for independent variables x_i. With 100·α% confidence it can be assured that the correct fitting function value is within the confidence interval.

The prediction interval is also evaluated for confidence level α. 100·α% values of the independent repeated measurement would be within the prediction interval. Prediction bands are evaluated by equation:

$$\overline{Y}_{x_0} \pm t(1-\alpha/2, n-2)\cdot s\{pred\}, \tag{6}$$

where
$$s^2\{pred\} = S_{Y_{X_0}} + s^2\{\overline{Y}_{x_0}\}. \tag{7}$$

For application of scaling factors a correlation coefficient between difficult-to-measure and key nuclides is evaluated by equation:

$$R = \frac{n\sum xy - \sum x \sum y}{\sqrt{[n\sum x^2 - (\sum x)^2][n\sum y^2 - (\sum y)^2]}}, \tag{8}$$

where n is the number of samples, x and y are measured values.

RESULTS AND DISCUSSION

Compilation of a list of safety-relevant radionuclides

The methodology for compiling the list of critical radionuclides, present in the Ignalina NPP radioactive waste, relevant from the point of view of the radioactive waste disposal safety has been developed. At first a long general list of radionuclides is obtained by calculations of the nuclear fuel evolution in the reactor and the activation of the fuel channel performed using the Origen-ARP computer code. Short-lived radionuclides, the half-life of which is shorter than a few days, make up the largest part of the total activity. All these radionuclides decay in a quite short period and are not important to the long-term safety.

The suggested screening criteria to leave only safety-relevant radionuclides in the list are: half-life (half-life longer than 0.5) and the ratio of the relative activity concentration to Co^{60} R_i / R_{Co} larger than 10^{-5}. Here $R_i = C_i^* / C_i$ is the relative activity concentration of the i radionuclide, i.e. the ratio of the activity concentration of i radionuclide to its unconditional clearance level (uncontrollable activity), $R_{Co} = C_{60\text{-}Co}^*$/

$C_{60\text{-}Co}$ is the relative activity concentration of Co^{60}, i.e. the ratio of activity concentration of Co^{60} to its unconditional clearance level.

Hence, the general list is shortened by eliminating the radionuclides, the half-life of which is too short and which decay fast. Thus, after a rather short time they do not impose hazards to people and the environment because they do not reach the biosphere. The chosen half-life criterion is short enough and would suit even the disposal type when waste is not isolated from the environment by engineering barriers and can migrate over some tens of years.

Waste containing radionuclide concentrations below unconditional clearance levels are considered as not radioactive and can be reused without any restrictions. Unconditional clearance levels for particular radionuclides can vary by up to several orders of magnitude (e.g. ^{3}H and ^{63}Ni relative activity concentration ratios are 10^4 times larger than those of ^{60}Co, ^{134}Cs, ^{137}Cs, ^{94}Nb. The applied clearance levels are presented in Table 1. Radionuclides are divided into nine groups. It reflects relation of unconditional clearance levels to the radionuclide radiotoxicity. Unconditional clearance levels (activity concentration [Bq/g]) of radionuclides which were not included in the list (LAND 34-2000, 2000) were equated to the one tenth of the exempt level expressed in Bq/g (HN 73:2001). Coefficients for other radionuclides were evaluated according to (Radiation Protection Guidance No. 113, 1999; Radiation Protection Guidance No. 114, 2000).

Table 1. Unconditional clearance levels

Radionuclide	Clearance level, Bq/g
^{241}Am, ^{242m}Am**, ^{243}Am**, ^{243}Cm**, ^{244}Cm, ^{237}Np, ^{238}Pu**, ^{239}Pu, ^{240}Pu, ^{242}Pu**, ^{232}Th ^{234}U, ^{235}U	0.1
^{110m}Ag, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{54}Mn, ^{94}Nb, ^{226}Ra, ^{65}Zn	0.4
^{236}U*, ^{238}U*	1
^{90}Sr, ^{126}Sn** ^{133}Ba**, ^{166m}Ho**	4
^{40}K*, ^{241}Pu*	10
^{129}I, ^{182}Hf**	40
^{135}Cs*, ^{93m}Nb*, ^{59}Ni*, ^{93}Zr*	100
^{14}C, ^{36}Cl, ^{41}Ca*, ^{55}Fe, ^{79}Se*, ^{99}Tc	400
^{45}Ca, ^{3}H, ^{63}Ni	4000

* - radionuclides included in the table on the basis of (HN 73:2001).

** - radionuclides included in the table on the basis of (Radiation Protection Guidance No. 113, 1999; Radiation Protection Guidance No. 114, 2000).

The activity concentration measurements of natural radionuclides in soil at the boundary of the Ignalina NPP sanitary zone show that ^{40}K activity concentration is higher than 0.2 Bq/g, while that of ^{226}Ra and ^{232}Th is higher than 0.01 Bq/g. Taking into account clearance levels of these natural radionuclides, the lowest ratio of their relative activity concentration to Co^{60} (0.02) was determined in case of potassium. Therefore, even with the conservative assumptions to possible modeling uncertainties, it is not rational to include into the list of safety-relevant radionuclides those whose relative activity concentration in waste is 10^5 times lower than that of ^{60}Co.

Radionuclides remaining in the list after screening against the proposed criteria are presented in Fig. 1. The figure shows time dependence of the radionuclide relative activity concentration ratio to the relative activity concentration of ^{60}Co at the fuel assembly unloading from the reactor core. ^{93}Zr does not meet the second criterion. However, its relative activity concentration was close to the criterion and it was included into the list in order to better consider RBMK-1500 peculiarity. Natural potassium level is shown in the figure for illustration.

It can be noted that at the beginning ^{54}Mn, ^{60}Co, and ^{137}Cs have the largest relative activity ratios and slightly lower values are for ^{134}Co, ^{94}Nb and ^{90}Sr. Ratios of other radionuclides are more than 100 times lower. However, in a couple of decades ^{54}Mn and ^{134}Cs decay, and ^{60}Co decays in 100 years. Then ^{94}Nb and ^{137}Cs are dominating radionuclides. At that time the ^{241}Am relative activity ratio increases due to production of ^{241}Am by decay of ^{241}Pu and becomes equal to that of ^{90}Sr. Relative activity ratios of ^{63}Ni, ^{93}Zr, ^{94}Nb, ^{239}Pu and ^{240}Pu do not change and the ratio of ^{238}Pu decreases slowly. It shows the importance of these radionuclides to the long-term safety.

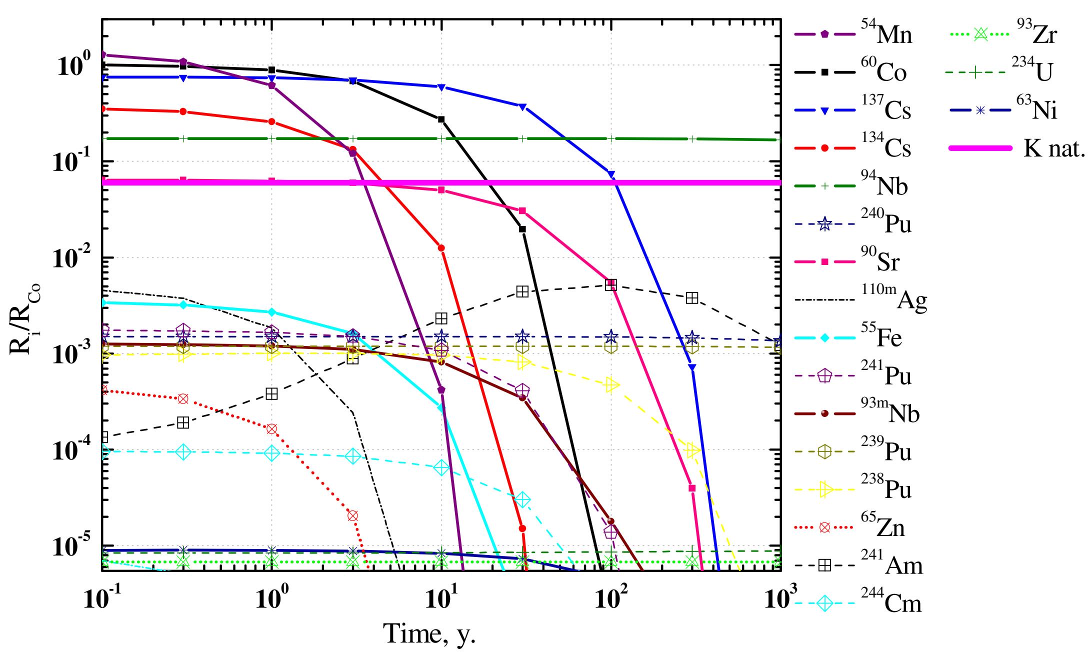


Fig.1. Ratios of radionuclide relative activity concentration to initial Co^{60} relative activity concentration R_i/R_{60Co}.

The obtained list of radionuclides cannot be regarded as fully representing all radioactive waste streams of RBMK reactors because the activation of the reactor active zone materials (e.g. of graphite) in the neutron flux was not modeled. Therefore, the derived list of radionuclides is more applicable to operational waste polluted due to contact with the coolant and containing very low activity concentration of radionuclides.

During 300 years the relative activity ratio to ^{60}Co for part of radionuclides, eliminated from the list, remains almost the same. Hence, some eliminated radionuclides can be important for to long-term safety of repositories depending on the radionuclide mobility in engineered and natural barriers and a shortened list of safety-relevant radionuclides can be supplemented with additional radionuclides obtained from the safety assessment of repository and regarded as safety-relevant ones. Based on analysis of safety-relevant radionuclides according to the international practice, preliminary waste acceptance criteria for near-surface disposal in Lithuania and judgments on relevance of some radionuclides for investigated Ignalina NPP radioactive waste streams, a shortened list of safety-relevant radionuclides was supplemented with ^{3}H, ^{14}C, ^{59}Ni, ^{63}Ni, ^{99}Tc, ^{129}I, ^{237}Np, ^{234}U, ^{235}U, and ^{238}U.

Analysis of methods for determination of radionuclide content of the Ignalina NPP radioactive waste

A preferred option for determination of scaling factors is use of experimental methods. In case the precision of methods and equipment is unsatisfactory, analytical methods shall be applied. Activity concentrations of α emitters ^{238}Pu, $^{239+240}Pu$, ^{241}Am, $^{243+244}Cm$, β emitters ^{3}H, ^{14}C, ^{55}Fe, ^{63}Ni, ^{90}Sr, ^{129}I, ^{241}Pu and γ emitters ^{54}Mn, ^{60}Co, ^{65}Zn, ^{94}Nb, ^{110m}Ag, ^{134}Cs, ^{137}Cs were measured by the α, β and γ spectrometry methods. The scaling factors with key nuclides were determined by performing a series of measurements and statistically processing the results. In cases when the method of scaling factors cannot be applied or it is not expedient because of weak correlation of the radionuclide activity with the activities of key radionuclides, the average concentration of radionuclides in the waste stream is estimated. Scaling factors for not measured radionuclides were estimated using intermediate radionuclides and applying modeling methods and computer codes. The methods of the radionuclide activity determination were chosen after evaluation of the possibilities to determine activity concentrations of all radionuclides from the list and the rationality of these evaluations.

Table 2. Methods of determination of radionuclide activity concentrations in waste of the Ignalina NPP

Method	Radionuclides
Direct measurement	^{60}Co, ^{137}Cs (key nuclides)
Scaling factor	^{14}C, ^{54}Mn, ^{55}Fe, ^{63}Ni, ^{65}Zn, ^{90}Sr, ^{94}Nb, ^{95}Zr, ^{110m}Ag, ^{129}I, ^{134}Cs, ^{238}Pu, $^{239+240}Pu$, ^{241}Pu, ^{241}Am, $^{243+244}Cm$
Mean concentration in the waste stream	^{3}H, ^{14}C (in case of weak correlation with the key nuclide)
Computer modeling	^{59}Ni (according to ^{63}Ni), ^{99}Tc (according to ^{90}Sr or ^{60}Co), ^{93}Zr (according to ^{95}Zr or ^{94}Nb), ^{93m}Nb (according to ^{94}Nb), ^{129}I (according to ^{131}I or ^{137}Cs), ^{235}U, ^{238}U and ^{237}Np (according to ^{239}Pu) $^{239}Pu/^{240}Pu$, $^{243}Cm/^{244}Cm$

Analysis of the Ignalina NPP radioactive waste streams

A general scheme of the Ignalina NPP radioactive waste generation is presented in Fig. 2. The scheme shows radionuclide routes from the radionuclide generation to the disposal and explains the formation of radioactive waste streams. The crucial point is that each step of radionuclide transfer from one medium to another (fuel matrix – fuel to clad gap, MCC coolant–surface of structural materials) imposes some change of activity concentrations of isotopes of different chemical elements due to different physical and chemical properties. Therefore, the ratios of isotope activities in general will be different in nuclear fuel and the final waste product. One can conclude that depending on the complexity of radionuclide transfer several nuclide vectors are needed for characterization of all radioactive waste.

The radionuclides are generated during nuclear fission and due to activation of the reactor core components as well as the Main Circulation Circuit (MCC) water and gas circuit gases. These radionuclides can be released from nuclear fuel and reactor components to the technological media of NPP due to present fuel cladding defects and corrosion of metal structures of the reactor core components and contamination of the MCC coolant due to direct contact. Contaminated coolant from the MCC can be further transferred to the final waste by three main routes: direct contamination due to direct contact with structural materials (thermal insulation, metal components such as pipes, etc.); loss of coolant through leakages to the drainage system; and cleaning of coolant by ion-exchange resins and perlite. Activated reactor components are the source of long-lived radioactive waste. MCC coolant and NPP drainage water purification system and gas purification system are also sources of pollution because of contamination of filters used for purification of water and gases. A dotted line from MCC to solid short-lived waste stream in Fig. 2 shows that solid waste is formed not directly from MCC water but also during repair works contaminating tools, materials, etc. Another dotted line indicates connection of MCC and gas circuit in case of rupture of the fuel channel.

In the scheme five radioactive waste streams are identified: short-lived solid radioactive waste, evaporator concentrate (bituminized waste), ion-exchange resins, perlite and sediments of evaporator concentrate (cemented waste), long-lived solid radioactive waste and spent nuclear fuel. These radioactive waste streams should have a different radionuclide content depending on the radioactive waste generation way - direct generation (e. g. in case of long-lived waste) or pollution due to contact with technological media (water and gas). Short-lived solid radioactive waste stream is not homogeneous because of generation of radioactive waste from different pollution sources and ongoing technological processes in NPP equipment (e.g. evaporation of MCC water and separation of vapour). Several nuclide vectors are necessary for characterization of long-lived solid radioactive waste stream due to different composition of activated material: graphite, steel, Zr-Nb alloy, serpentine. In the purification system of coolant and contaminated water two radioactive waste streams are formed: evaporator concentrates and spent filtre aid materials.

Finally all materials, contaminated by radionuclides, are treated as radioactive waste, solidified if needed (cemented or bituminized) and directed to the disposal facility. Waste meeting clearance levels is released from control. Other wastes are temporarily stored at storage facilities. Industrial, very low-activity (ionizing radiation dose rate at the 10 cm distance from the surface does not exceed 0.6 μSv/h) Ignalina NPP waste is accumulated at a special dumping site on the territory of the Ignalina NPP. Transformation of bituminized waste storage facility to repository is foreseen, if the long-term safety of this facility is justified. Planned repositories for disposal of very low-level waste and low- and intermediate-level waste as well as a geological repository suitable for disposal of long lived waste are shown in the scheme.

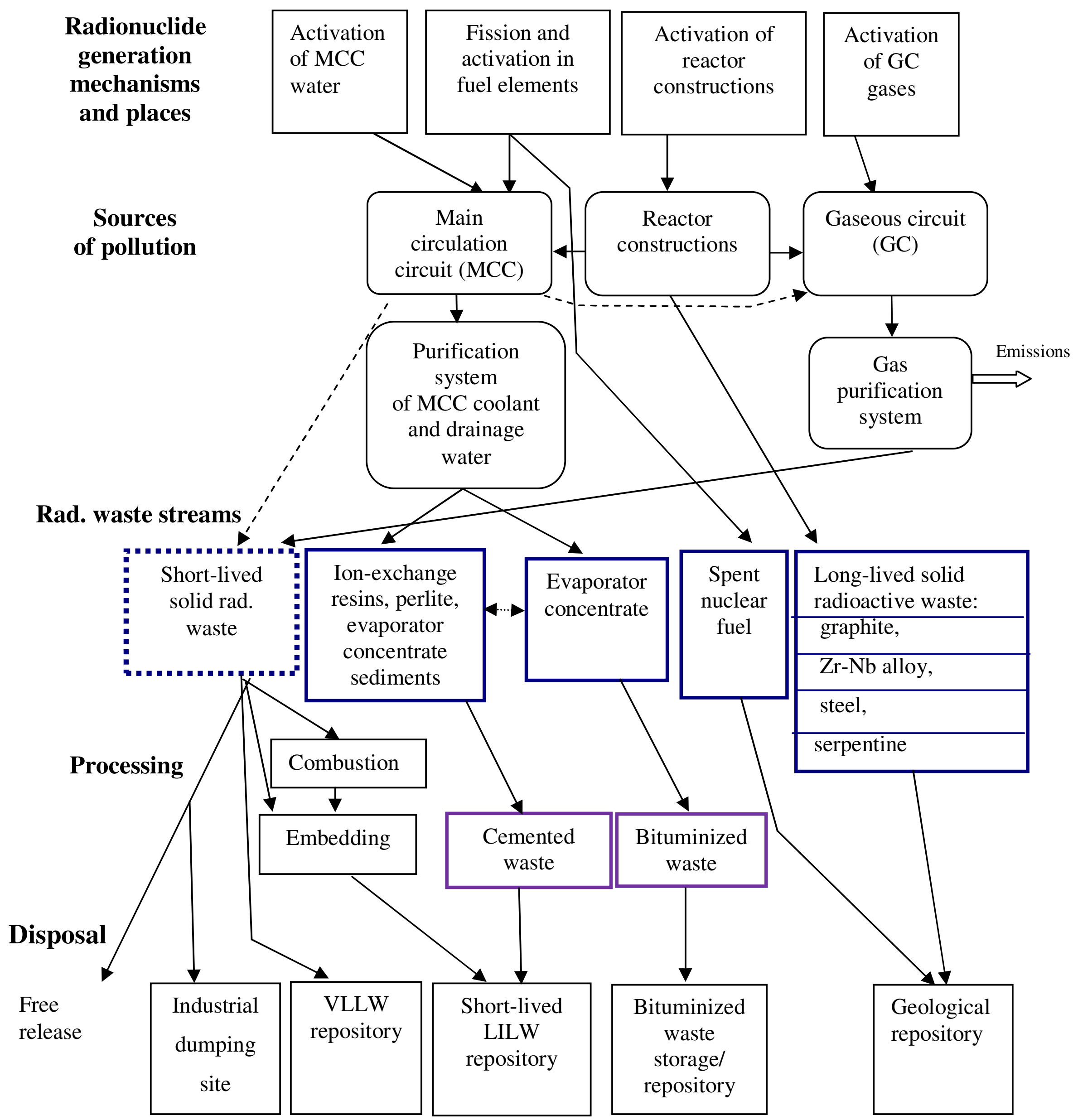


Fig.2. Scheme of radioactive waste generation at the Ignalina NPP and formation of waste streams.

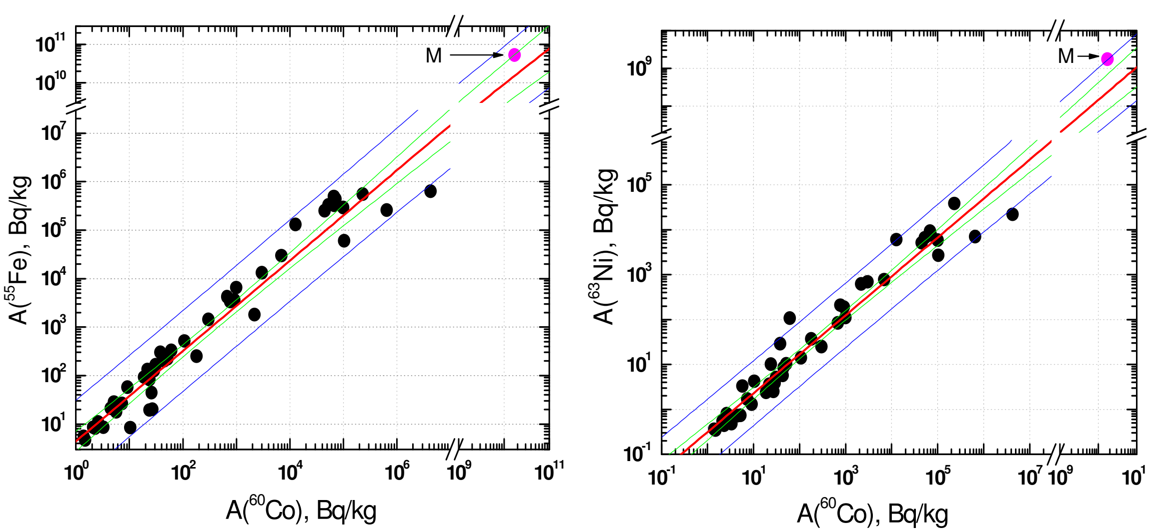
In this work three radioactive waste streams were investigated: short-lived solid radioactive waste stream cemented and bituminized waste streams. For characterization of solid radioactive waste stream several nuclide vectors are necessary. In case of decontamination of NPP equipment during the decommissioning nuclide vectors of waste streams can change. Nuclide vectors shall be corrected during decommissioning of NPP because half-lifes of key nuclides ^{60}Co and ^{137}Cs are quite short in comparison to those of other safety-relevant radionuclides.

Analysis of nuclide vectors in Ignalina NPP radioactive waste streams

In order to apply and show feasibility of the scaling factor method for waste characterization of the RBMK-1500 reactor of the Ignalina NPP, correlations of radionuclide activities with key radionuclides in various technological media were investigated. Scaling factors for MCC, turbine hall equipment, emergency core cooling system, gas circuit equipment, ventilation system equipment, gas circuit filters and three waste streams – cemented waste, bituminized waste, solid very-low-level-activity (industrial) waste – are presented in Table 3.

The correlation coefficients between specific activities of radionuclides of the same origin (corrosion) and ^{60}Co were large. In the industrial waste, emergency core cooling system equipment and gas circuit correlation coefficients of ^{54}Mn with ^{60}Co were 0.95 – 0.96 and in the turbine hall equipment – 0.84. The correlation coefficient between activities of ^{55}Fe and ^{60}Co in the same waste was 0.93 – 0.98 and 0.83, accordingly. Correlation coefficients of ^{63}Ni activity with ^{60}Co were: 0.7 – in the turbine hall equipment, 0.94 – in gas circuit equipment, 0.96 – in gas filters, 0.97 – in emergency core cooling system, 0.99 – in bituminized waste and 1 – in cemented waste. Correlation coefficients of ^{94}Nb activity with ^{60}Co in industrial waste were 0.92, in gas circuit equipment – 0.96. Fig. 3 demonstrates correlation of ^{55}Fe and ^{63}Ni with ^{60}Co.

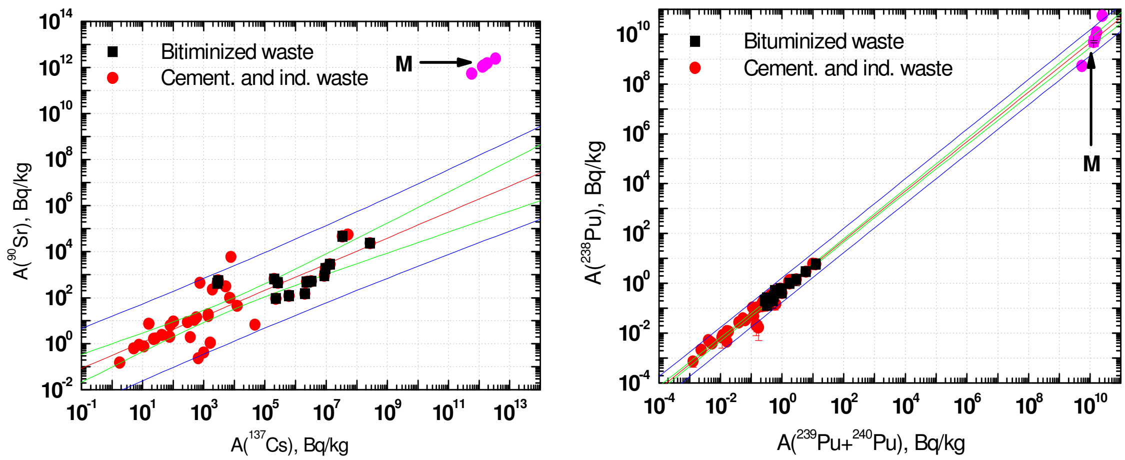
The correlation between specific activities of different origin radionuclides depends on the waste stream. The correlation between fission product ^{137}Cs and corrosion product ^{60}Co in the industrial waste, cemented waste and in the emergency core cooling system equipment was good, with the correlation coefficients R=0.93, R=0.97 and R=0.84, respectively. However, there is weak correlation in turbine hall equipment (R=0.23). It shows different migration of corrosion and fission products within the technological media.



(a) a=0.64±0.10, b=0.93±0.03, R=0.97 (b) a=-0.5±0.09, b=0.87±0.03, R=0.97

Fig.3. Correlations between measured (a) ^{55}Fe versus ^{60}Co and (b) ^{63}Ni versus ^{60}Co specific activities in industrial radioactive waste. Dots, experimental points, M, modeled SNF composition.

It is convenient to select ^{60}Co as a key nuclide because according to the measurements its contribution to the total gamma radiation intensity is the largest due to its high concentration in radioactive waste and high energy of emitted gamma rays. An exception is bituminized waste, where the concentration of ^{60}Co is only 2.5% of ^{137}Cs specific activity. Hence, ^{137}Cs should be taken as a key nuclide for characterization of bituminized waste.



(a) a=-1.94±0.34, b=1.01±0.06, R=0.93 (b) a =-0.27±0.03, b=0.99±0.01, R=0.998

Fig.4. Correlations between measured (a) ^{90}Sr versus ^{137}Cs and (b) ^{238}Pu versus ^{239}Pu + ^{240}Pu specific activities in radioactive waste. Markings as in Fig.3.

^{137}Cs was selected as a suitable intermediate key nuclide for the determination of scaling factors of ^{90}Sr and ^{134}Cs nuclide activities, and afterwards the corresponding scaling factor for these nuclides and ^{60}Co was recalculated. Usually, the correlation of ^{60}Co and ^{137}Cs is much better due to the relative simplicity of measurements (activities of both ^{60}Co and ^{137}Cs of one sample are evaluated in one gamma activity measurement), therefore considerably more samples can be measured, which reduces uncertainty of the scaling factor between ^{60}Co and ^{137}Cs due to significantly better statistics of a data set. Moreover, correlation of ^{137}Cs with fission products is also better due to the same physical process of generation in a reactor. Then, the uncertainty and correlation of the recalculated scaling factor of the nuclide activity of interest, e.g., ^{90}Sr, are better than if we tried to calculate it directly with ^{60}Co. This is the practical basis for using intermediate key nuclides in scaling factor analysis. The scaling factors of actinide activities were estimated by applying correlation to sum activity of ^{239}Pu + ^{240}Pu measured by alpha spectrometry as intermediate key nuclides. Examples of using ^{137}Cs and sum of ^{239}Pu and ^{240}Pu activity as intermediate key nuclide are presented in Fig. 4.

Scaling factors for different waste streams and technological media are presented in Table 3. In order to compare scaling factors for NPP equipment and operational waste, the factors were corrected taking into account decay of radionuclides after shutdown of unit 1 and established for the unit shutdown date 31 December, 2004.

The list of radionuclides was adjusted to the radioactive waste disposal option. For the waste to be disposed in the near-surface disposal facilities, having a long maintenance period, short-lived radionuclides such as ^{54}Mn, ^{55}Fe and ^{65}Zn, the half-live of which is a few years, are not important. Solid radioactive waste arising from dismantling of the equipment of the turbine hall and emergency core cooling system, ventilation system and gas circuit belong to both types of waste which can be released from control and to the waste to be disposed off in the near-surface repository. Hence, a full list of safety-relevant radionuclides is applicable. ^{3}H is not a safety-relevant nuclide for solid and bituminized radioactive wastes due to low quantity of moisture. The average concentration of ^{3}H in the evaporator concentrate and the amount of water in the cement matrix during a cementation process are used for the estimation of ^{3}H in the cemented waste.

Table 3. Scaling factors k_{60Co}

Nuclide	Nuclear fuel*	MCC coolant	Ce-men-ted waste	Bitu-mini-zed waste	*Indus-trial waste*	*Turbine hall equip.*	*ECC equip.*	*Venti-lation system equip.*	*Gas circuit equip.*	*Gas filters*
^{14}C	$6{,}9\cdot10^{-4}$		$7\cdot10^{-2}$	$2\cdot10^{-2}$		$7\cdot10^{-4}$	$2\cdot10^{-3}$	$8\cdot10^{-2}$	17	$8\cdot10^{-2}$
^{54}Mn	0,16	2			0,3	0,7	0,7	0,6	0,6	0,7
^{55}Fe	4,12	0.4			3	5	13	3	3	1
^{59}Ni	$5{,}7\cdot10^{-4}$		$1\cdot10^{-3}$	$4\cdot10^{-4}$		$2\cdot10^{-3}$	$1\cdot10^{-4}$	$5\cdot10^{-4}$	$5\cdot10^{-4}$	$5\cdot10^{-4}$
^{63}Ni	$7{,}2\cdot10^{-2}$	$1\cdot10^{-2}$	0,2	$4\cdot10^{-2}$		0,2	0.1	$7\cdot10^{-2}$	$7\cdot10^{-2}$	$7\cdot10^{-2}$
^{65}Zn	$5{,}2\cdot10^{-4}$	$1\cdot10^{-1}$			$1\cdot10^{-2}$	$1\cdot10^{-3}$	$2\cdot10^{-3}$	$3\cdot10^{-3}$	$3\cdot10^{-3}$	$3\cdot10^{-3}$
^{90}Sr	77	$1\cdot10^{-4}$	$4\cdot10^{-3}$	$2\cdot10^{-2}$	$8\cdot10^{-3}$	$1\cdot10^{-3}$	$1\cdot10^{-3}$	$1\cdot10^{-4}$	0,2	0,4
^{93}Zr	$2{,}6\cdot10^{-4}$				$1\cdot10^{-4}$	$3\cdot10^{-5}$	$1\cdot10^{-5}$	$1\cdot10^{-4}$	$1\cdot10^{-4}$	$3\cdot10^{-6}$
^{93m}Nb	$4{,}9\cdot10^{-1}$				0.1	0.3	0.1	0,2	0,2	$8\cdot10^{-3}$
^{94}Nb	$6{,}2\cdot10^{-2}$		$7\cdot10^{-4}$	$3\cdot10^{-3}$	0.01	$2\cdot10^{-2}$	$1\cdot10^{-3}$	$1\cdot10^{-2}$	$1\cdot10^{-2}$	$3\cdot10^{-4}$
^{99}Tc	$1{,}2\cdot10^{-2}$		$1\cdot10^{-5}$	$1\cdot10^{-2}$		$2\cdot10^{-5}$	$1\cdot10^{-5}$	$1\cdot10^{-5}$	$1\cdot10^{-5}$	$1\cdot10^{-5}$
^{110m}Ag	$6{,}8\cdot10^{-1}$				$3\cdot10^{-2}$	$7\cdot10^{-3}$	$1\cdot10^{-2}$	$2\cdot10^{-2}$	$2\cdot10^{-2}$	$2\cdot10^{-2}$
^{129}I	$2{,}4\cdot10^{-5}$		$2\cdot10^{-7}$	$2\cdot10^{-5}$			$2\cdot10^{-7}$	$5\cdot10^{-8}$		
^{134}Cs	44				0.1		$5\cdot10^{-2}$	$7\cdot10^{-3}$		
^{137}Cs	91	$6\cdot10^{-1}$	1	40	0.2		0,5	$9\cdot10^{-2}$		
^{234}U	$2{,}5\cdot10^{-3}$		$3\cdot10^{-8}$	$1\cdot10^{-7}$		$3\cdot10^{-7}$	$1\cdot10^{-7}$	$9\cdot10^{-7}$	$9\cdot10^{-7}$	$2\cdot10^{-8}$
^{235}U	$6{,}2\cdot10^{-5}$		$7\cdot10^{-10}$	$3\cdot10^{-9}$		$7\cdot10^{-9}$	$3\cdot10^{-9}$	$2\cdot10^{-8}$	$2\cdot10^{-8}$	$4\cdot10^{-10}$
^{238}U	$7{,}5\cdot10^{-4}$		$9\cdot10^{-9}$	$3\cdot10^{-8}$		$1\cdot10^{-7}$	$4\cdot10^{-8}$	$3\cdot10^{-7}$	$3\cdot10^{-7}$	$7\cdot10^{-9}$
^{237}Np	$9{,}4\cdot10^{-5}$		$1\cdot10^{-9}$	$4\cdot10^{-9}$		$2\cdot10^{-8}$	$8\cdot10^{-9}$	$6\cdot10^{-8}$	$6\cdot10^{-8}$	$1\cdot10^{-9}$
^{238}Pu	$2{,}8\cdot10^{-1}$	$2{,}6\cdot10^{-5}$	$5\cdot10^{-6}$	$2\cdot10^{-5}$	$7\cdot10^{-5}$	$1\cdot10^{-4}$	$5\cdot10^{-5}$	$2\cdot10^{-4}$	$2\cdot10^{-4}$	$4\cdot10^{-6}$
^{239}Pu	$3{,}6\cdot10^{-1}$	$2\cdot10^{-5}$	$4\cdot10^{-6}$	$2\cdot10^{-5}$	$5\cdot10^{-5}$	$5\cdot10^{-5}$	$2\cdot10^{-5}$	$1\cdot10^{-4}$	$1\cdot10^{-4}$	$3\cdot10^{-6}$
^{240}Pu	$4{,}6\cdot10^{-1}$	$2\cdot10^{-5}$	$6\cdot10^{-6}$	$2\cdot10^{-5}$	$6\cdot10^{-5}$	$9\cdot10^{-5}$	$4\cdot10^{-5}$	$2\cdot10^{-4}$	$2\cdot10^{-4}$	$6\cdot10^{-6}$
^{241}Pu	66	$7\cdot10^{-3}$	$2\cdot10^{-4}$	$2\cdot10^{-3}$	$1\cdot10^{-2}$	$7\cdot10^{-3}$	$2\cdot10^{-3}$	0,1	0,1	$8\cdot10^{-4}$
^{241}Am	$4{,}0\cdot10^{-2}$	$2\cdot10^{-5}$	$1\cdot10^{-5}$	$4\cdot10^{-5}$	$1\cdot10^{-4}$	$1\cdot10^{-4}$	$1\cdot10^{-4}$	$3\cdot10^{-5}$	$3\cdot10^{-5}$	$3\cdot10^{-6}$
^{244}Cm	$4{,}6\cdot10^{-2}$	$3\cdot10^{-5}$			$2\cdot10^{-4}$	$8\cdot10^{-5}$	$1\cdot10^{-4}$	$1\cdot10^{-4}$	$1\cdot10^{-4}$	$2\cdot10^{-6}$

* - ratio to ^{60}Co calculated at fuel discharge moment.

There were not enough experimental data for determination of good correlation between ^{14}C and a key nuclide. So, the arithmetical mean value is used to estimate ^{14}C concentration in the waste. Activity concentrations of ^{65}Zn and ^{110m}Ag in the turbine hall and emergency core cooling installations were below the detection limits. Hence, scaling factors were derived by the computer modeling. Concentration of ^{94}Nb in bituminized waste was also below the detection limit and the scaling factor has been overestimated assuming that the concentration of ^{94}Nb is equal to the detection limit. The same approach has been applied to the estimation of the scaling factor for ^{90}Sr in the emergency core cooling system equipment. Experimental data were insufficient for determination of correlation between ^{94}Nb and ^{60}Co in the turbine hall equipment, so the arithmetical mean value was used.

Results of correlation analysis provided in Fig. 5 indicate that waste streams generated by the RBMK-1500 reactor can be distinguished by the scaling factor for ^{137}Cs. One can clearly see in Fig. 5 that the activity ratio of ^{137}Cs to ^{60}Co is by about two orders of magnitude higher in liquid waste compared to that in solid waste. This can be understood as the influence of radionuclide transport processes on the final waste inventory and solubility of Cs chemical formations.

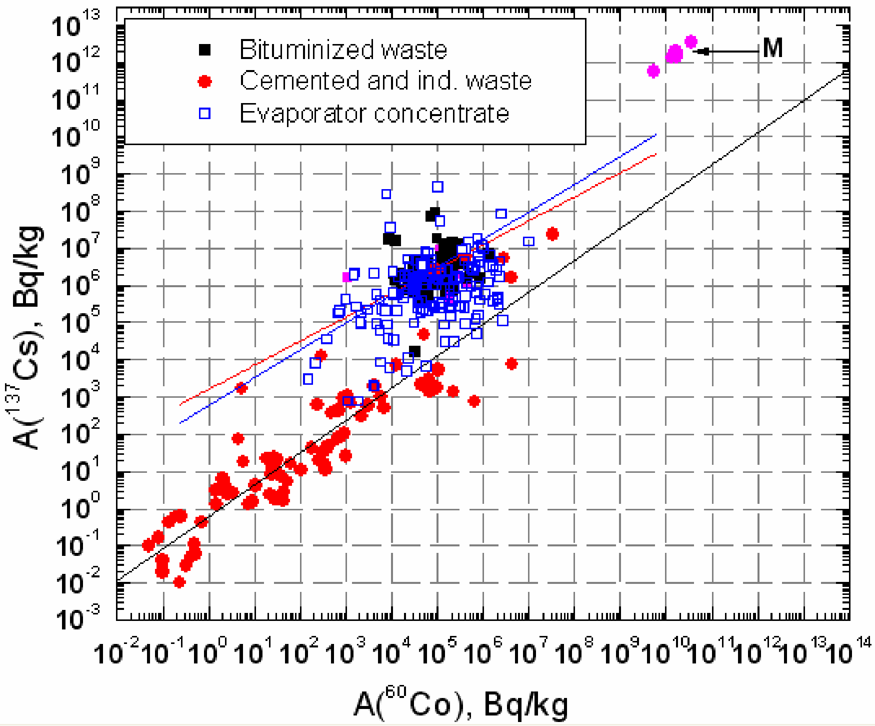


Fig.5. Correlation of ^{137}Cs with key radionuclide ^{60}Co in INPP solid and liquid radioactive waste.

Scaling factors of ^{14}C vary up to 10^4 in different nuclide vectors. Scaling factors of fission products and actinides depend on the radioactive waste pollution source (MCC coolant or gas circuit gases). Scaling factors of ^{14}C and ^{90}Sr are hundreds and thousands times larger than in other waste streams and factors of actinides are up to hundreds times smaller. Dependence of scaling factors of these radionuclides on the radioactive waste stream is presented in Fig. 6.

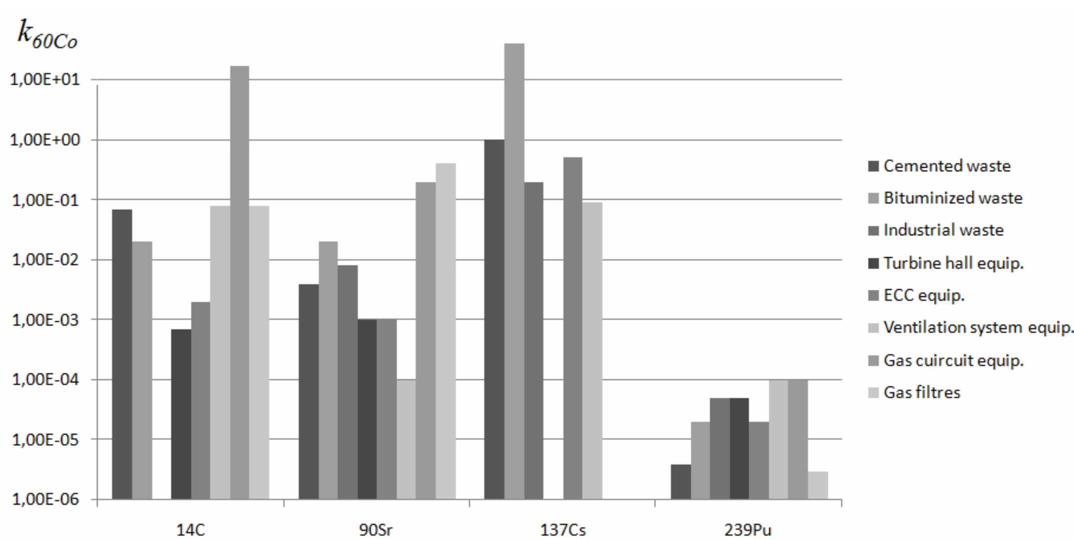
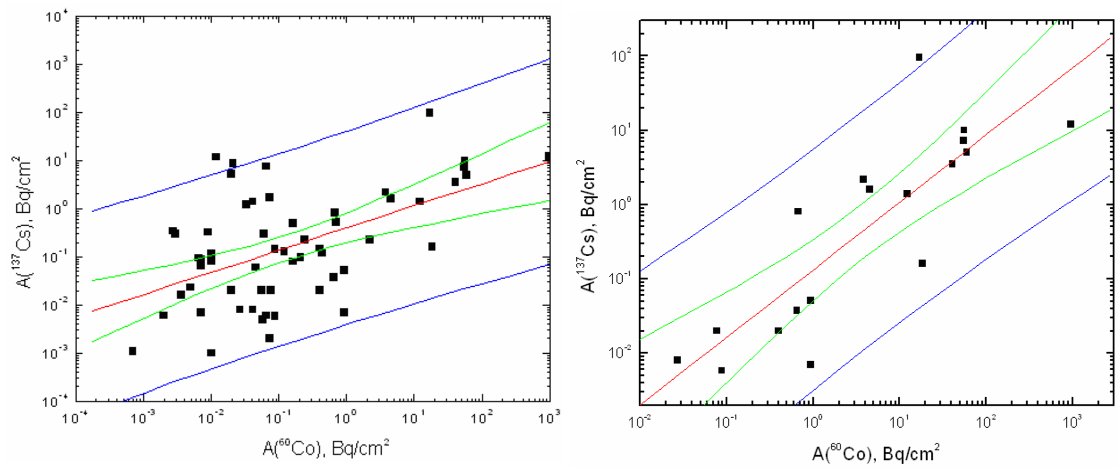


Fig.6. Dependence of ^{14}C, ^{90}Sr, ^{137}Cs and ^{239}Pu scaling factors on the waste stream.

Low correlation coefficients by analyzing the equipment of the whole turbine hall (R=0.23) and building V1 (R=0.53), where gas circuit equipment and ventilation system equipment are located, indicate possibility of mixture of different radioactive waste streams. Experimental results of derivation of the nuclide vectors for equipment of turbine hall and building V1 confirmed that solid radioactive waste stream is not homogeneous and several nuclide vectors are needed for characterization of the solid radioactive waste stream.

The correlation between ^{137}Cs and ^{60}Co is weak and the coefficient b is far from 1 for the whole equipment in building V1 (Fig. 7 (a)). Under such conditions the scaling factor method is not applicable and ^{129}I and ^{134}Cs scaling factors are determined to the key nuclide ^{137}Cs. However, the correlation of activation products ^{54}Mn, ^{55}Fe, ^{63}Ni, ^{94}Nb and ^{60}Co is good =0.95, R=0.93, R=0.94 and R=0.96. Besides, the correlation is good between $^{239+240}$Pu and ^{60}Co R=0.93 and between actinides ^{238}Pu, ^{238}Pu, ^{241}Am, $^{243+244}$Cm and $^{239+240}$Pu – correlation coefficients are R=0.97, R=0.8, R=0.9 and R=0.89, accordingly.



(a) a=-0.4, b=0.46, R=0.53 (b) a=-0.88, b=0.91, R=0.84

Fig.7. Correlation of ^{137}Cs with key radionuclide ^{60}Co (a) for building V1 equipment and (b) for ventilation system equipment.

Even a worse correlation of ^{137}Cs with ^{60}Co is for turbine hall equipment (1.14±0.36, b = 0.18±0.13, R = 0.23). However, correlation coefficients of ^{54}Mn, ^{55}Fe, ^{63}Ni with ^{60}Co are quite good R=0.84, R=0.83 and quite good R is about 0.7. Correlation between transuranic radionuclides is also good R=0.75-0.98. However, the correlation coefficient of $^{239+240}$Pu and ^{60}Co is low R=0.51.

Building V1 equipment was divided into two groups: ventilation system and maintenance cooling system assigned to the first group and the remaining equipment - to the second group, except gas purification systems. Due to such grouping a better correlation between ^{60}Co and ^{137}Cs (Fig. 7 (b)) is observed for the first group of equipment, which allows application of the scaling factor method. The correlation between ^{60}Co and ^{137}Cs is weak (R=0.25) in the second group of V1 equipment. Hence, ^{137}Cs shall be chosen as a key radionuclide for the determination of ^{134}Cs and ^{129}I. k_{137Cs} for ^{129}I is $4.7 \cdot 10^{-7}$ and for ^{134}Cs – 0.03. An individual nuclide vector shall be established for gas purification filters. Turbine hall equipment can be divided into three groups to obtain a better correlation of ^{60}Co with ^{137}Cs. In the first group (equipment of main steam, steam pipeline drainage, main condensate and feed water systems) ^{137}Cs was not measured. In the second group (detonating gas combustion facility) ^{60}Co was absent. In the third group (the remaining equipment) both ^{137}Cs and ^{60}Co were measured. Hence, both ^{60}Co and ^{137}Cs shall be used as key radionuclides. ^{137}Cs is a key nuclide for ^{134}Cs and ^{129}I, ^{60}Co – for others. The scaling factor k_{137Cs} for ^{129}I is $4.7 \cdot 10^{-7}$, for ^{134}Cs – 0.09. Specific activities of ^{134}Cs, ^{137}Cs and ^{129}I in the first group equipment and specific activities of all radionuclides except, ^{134}Cs, ^{137}Cs and ^{129}I in the second group equipment are below the clearance levels.

Fig. 8 presents dependence of scaling factors on technological media analyzed for one of fission products, ^{137}Cs, one of corrosion nuclides, ^{63}Ni and one of actinides, ^{239}Pu.

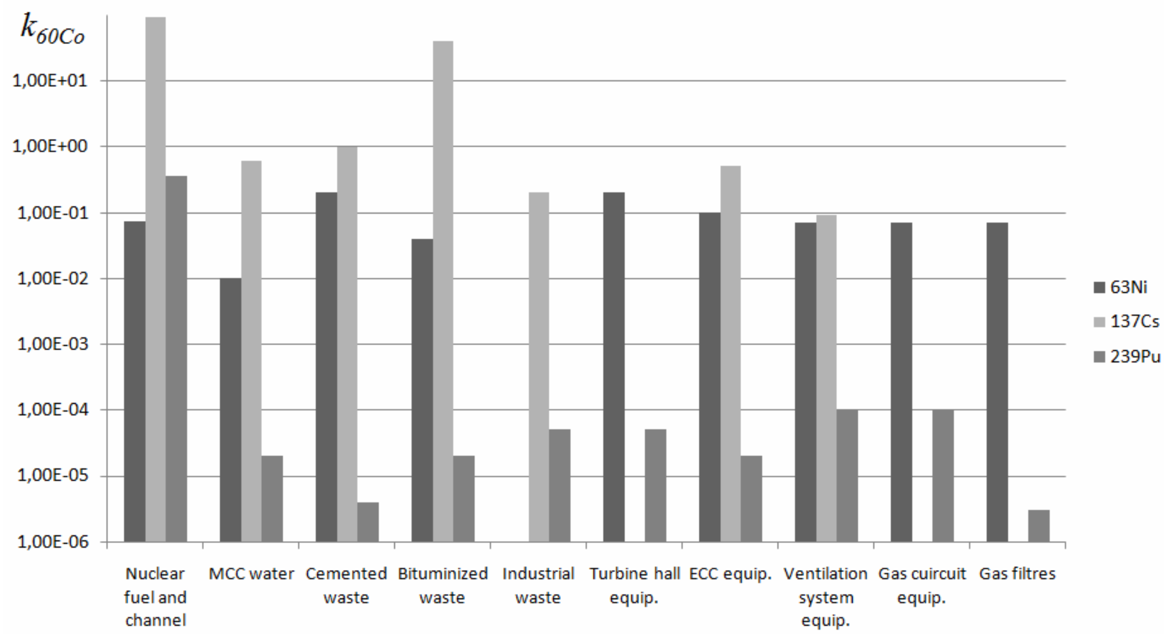


Fig.8. Scaling factors of ^{63}Ni, ^{137}Cs and ^{239}Pu in the Ignalina NPP technological media.

By comparing scaling factors in MCC coolant to calculated ratios one can conclude that the ^{63}Ni release rate to coolant is similar to that of ^{60}Co (difference is less than 10 times), the release rate of ^{137}Cs is by two orders of magnitude lower than that of ^{60}Co and the release rate of ^{239}Pu is by four orders of magnitude lower. The scaling factor of ^{63}Ni in the cemented and bituminized waste changes insignificantly (increases less than 10 times) compared to MCC coolant. The scaling factor of ^{137}Cs almost does not change (increases almost twice) in bituminized waste and increases about 100 times in the cemented waste. It can be explained that very soluble ^{137}Cs chemical forms remain in evaporator concentrate. The scaling factor of ^{239}Pu does not change significantly in cemented (decreases 5 times) and bituminized waste. In the solid radioactive waste stream – investigated NPP systems equipment – the scaling factor of ^{63}Ni changes insignificantly (less than 10 times). The scaling factor of ^{239}Pu changes insignificantly (less than 5 times) in the industrial waste and investigated NPP systems. Only in gas filters the scaling factor of ^{239}Pu decreases more than 10 times. The scaling factor of ^{137}Cs for solid radioactive waste stream depends on technological NPP processes. As mentioned above, there is weak correlation between specific activities of ^{137}Cs and ^{60}Co in the turbine hall equipment, where three waste streams can be distinguished and two key nuclides ^{60}Co and ^{137}Cs shall be used. The correlation between specific activities of ^{137}Cs and ^{60}Co in the gas circuit equipment and the gas filter is weak as well. Hence, ^{137}Cs is also a key nuclide for estimation of ^{134}Cs and ^{129}I activities. The scaling factor of ^{137}Cs for ventilation system is similar (less than 10 times) to that of other solid radioactive waste (industrial and emergency core cooling system) and is about 10 times less than in cemented waste.

Determined scaling factors of radioactive waste generated by the RBMK-1500 reactor could be compared with those of other reactors involving some technological similarities, as BWR. RBMK and BWR are both boiling light water reactors, however RBMK is a channel-type reactor, while BWR is not. Therefore, inventory of the reactor core of these reactors is different: RBMK contains Zr–Nb alloy fuel channel tubes, while in BWR a relative amount of this material is considerably less. It is confirmed by the data provided in (Müller, 2001; Masui et al., 2003) for the scaling factor of ^{94}Nb to ^{60}Co, which is from one to two orders of magnitude higher in RBMK case depending on the origin of waste (Table 4).

Table 4. Scaling factors k_{60Co}

Nuclide	RBMK-1500	BWR
^{14}C	$10^{-3}\div10^{-1}$	$\sim6\cdot10^{-4}$
^{63}Ni	$10^{-2}\div10^{-1}$	10^{-1}
^{94}Nb	$10^{-3}\div10^{-2}$	$10^{-5}\div10^{-4}$
Total α (k_{137Cs})	10^{-4}	$\sim3\cdot10^{-2}$

Analogous situation is for the scaling factor of ^{14}C to ^{60}Co. This can also be explained by the presence of a large amount of graphite in RBMK-1500 used as moderator as well as due to the gas circuit of RBMK-1500 where nitrogen activation yields ^{14}C. The scaling factor of all alpha emitting radionuclides (total alpha) for the RBMK-1500 reactor is by about two orders of magnitude lower than that for BWR due to the fact that defective fuel assemblies of RBMK-1500 are exchanged by refueling machine just after the defect occurrence, while in BWR case fuel reload campaigns are relatively rare and long-lived alpha emitters accumulate in technological media thus increasing the ratio to the key nuclide. However, for ^{63}Ni no significant difference could be identified between RBMK and BWR as results are comparable for both reactor types. Therefore, it can be concluded that radionuclide behavior in technological media of NPP and the resulting nuclide composition of radioactive waste depend on many factors and each case deserves individual analysis.

CONCLUSIONS

1. The methodology of compiling a list of safety-relevant radionuclides is based on two screening criteria – radionuclide half-life criterion (larger than 0.5 y.) and radionuclide specific activity and clearance level ratio – R_i comparison to Co^{60} specific activity and clearance level ratio R_{Co} (R_i/R_{Co} larger than 10^{-5}). Afterwards, the list is supplemented with safety-relevant radionuclides with reference to preliminary radioactive waste acceptance criteria for its disposal. Based on this methodology a general list of safety-relevant radionuclides has been compiled and adjusted considering radioactive waste stream characteristics and disposal option.
2. Correlation coefficients derived from experimental data between the corrosion product nuclides (^{54}Mn, ^{55}Fe, ^{60}Co, ^{63}Ni, ^{65}Zn), correlation coefficients between fission products (^{90}Sr, ^{134}Cs, ^{137}Cs), between actinides (^{241}Am, $^{239+240}$Pu, ^{241}Pu, $^{243+244}$Cm) and correlation coefficients of individual nuclides with key nuclides in the Ignalina NPP radioactive streams are mostly between 0.8 and 0.98 (regression coefficient b is close to 1) and rarely between 0.7 and 0.8. This substantiates the nuclide vector method suitability for characterization of the RBMK radioactive waste.
3. Good correlation (correlation coefficient is between 0.88-0.98, except one case, when it is 0.8) of different Pu isotopes measured activities (alpha emitters $^{239+240}$Pu with beta emitter ^{241}Pu) demonstrates a reliability of the applied method to derive nuclide vectors.
4. It is reasonable to select ^{60}Co and/or ^{137}Cs as key nuclides to characterize radioactive waste of the RBMK type reactor. When the specific activity of ^{60}Co is approximately equal or larger than the specific activity of ^{137}Cs and good correlation between activities of these radionuclides is observed, it is reasonable to select ^{60}Co as a key nuclide because of its lower detection limit in radioactive waste packages that of ^{137}Cs and good correlation of ^{60}Co activities with activities of actinides is observed (correlation coefficient is between 0.8-0.93). If the specific activity of ^{60}Co is much lower than the specific activity of ^{137}Cs, it is reasonable to select ^{137}Cs as a key nuclide. In case the correlation of these radionuclides is weak (e. g., radioactive waste stream encompassing a filter system), two key nuclides shall be selected: ^{60}Co and ^{137}Cs.
5. Ratios of corrosion products and ^{60}Co specific activities (scaling factors k_{60Co}) are almost equal (within one order of magnitude) in all radioactive waste of the RBMK type reactor. Variations of activity ratios of actinides and ^{60}Co in different RBMK radioactive waste streams are slight (e. g., difference between the lowest scaling factors k_{60Co} of ^{239}Pu in gas filters and cemented waste and the largest in the gas circuit equipment and ventilation system is around 30 times). However, activity ratios of fission products and ^{60}Co in different RBMK radioactive waste streams vary significantly (e. g., scaling factors of ^{137}Cs k_{60Co} vary from $9{\cdot}10^{-2}$ in ventilation system to 40 in bituminized waste and those of ^{90}Sr – from $1{\cdot}10^{-4}$ to $2{\cdot}10^{-2}$). Activity ratio between ^{14}C and ^{60}Co varies up to tens of thousands times (from $7{\cdot}10^{-2}$ in turbine hall equipment to 17 in gas circuit equipment). Hence, it is reasonable to distinguish separate

RBMK radioactive waste streams where scaling factors vary insignificantly, and characterize these streams by different nuclide vectors.

6. Total radionuclide activity values in radioactive waste govern its storage and disposal options, and these govern, which radionuclides in the particular waste shall be declared. Hence, when distinguishing radioactive waste streams characterized by different nuclide vectors it shall be considered which radionuclides in these streams shall be declared. Three radioactive waste streams of the Ignalina NPP were investigated: cemented waste, bituminized waste, short-lived solid waste. 8 streams were distinguished in the short-lived solid radioactive waste stream characterized by individual nuclide vectors: industrial waste stream; equipment of the reactor emergency core cooling system; equipment of turbine hall, excluding equipment of the main vapour system and detonating gas combustion installation; equipment of the main vapour system (where ^{137}Cs is absent); detonating gas combustion installation (where corrosion products and actinides are absent); equipment of the gas circuit, excluding ventilation system equipment and gas filters; equipment of the gas circuit ventilation system; gas circuit filters.

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RADIONUKLIDINĖS SUDĖTIES TYRIMAS ATOMINĖS ELEKTRINĖS RADIOAKTYVIŲJŲ ATLIEKŲ SRAUTUOSE

Reziumė

Nuolat augančios energijos sąnaudos bei naujų pramonės šakų ir technologijų plėtra neišvengiamai kuria problemas, susijusias su žalingu gamybos atliekų poveikiu supančiai aplinkai ir žmogui. Augant technogeniniam krūviui ir senkant aplinkos savireguliacijos galimybėms, visuomenė priversta investuoti į aplinką tausojančių plėtros kelių paieškas. Šiuo požiūriu alternatyvų neturi branduolinė energetika, tačiau būtina užtikrinti aukštą branduolinės saugos bei radiacinės apsaugos lygį, išspręsti radioaktyviųjų atliekų tvarkymo technologinius uždavinius, suprasti radioaktyviųjų izotopų sklaidos gamtinėje aplinkoje dėsningumus bei geriau suvokti jonizuojančiosios spinduliuotės poveikio aplinkai ir žmogui aspektus. Tai aktualu optimizuojant profesinę ir gyventojų radiacinę saugą eksploatuojant branduolinius įrenginius, taip pat perdirbant, saugant ir laidojant radioaktyviąsias atliekas. Šie klausimai ypač aktualūs nutraukiant branduolinių objektų eksploataciją, nes daugybė naujų technologinių bei radiacinės saugos aspektų, susijusių su dideliais radioaktyviųjų atliekų srautais, iki šiol galutinai nėra aiškūs. Todėl darbo tema glaudžiai siejasi su svarbiausiu šio dešimtmečio Lietuvos energetikos pramonės uždaviniu – saugiai nutraukti Ignalinos atominės elektrinės eksploataciją. Tam turi būti naudojamos šiuolaikinės radioaktyviųjų atliekų tvarkymo, utilizacijos ir laidojimo technologijos, kurios garantuoja ilgalaikę saugą ir minimalų jonizuojančiosios spinduliuotės poveikį aplinkai ir žmogui.

Pirmasis žingsnis, siekiant įvertinti galimą radioaktyvių atliekų radiologinį poveikį, - nustatyti jų radionuklidinę sudėtį. Vieningo reikšmingų radionuklidų, vertinant jonizuojančiosios spinduliuotės poveikį aplinkai ir žmogui, sąrašo, tenkinančio visą branduolinių įrenginių gausą, nėra. Todėl aktualu, atsižvelgiant į branduolinio kuro ir reaktoriaus struktūrinių medžiagų sudėtį bei neutronų srauto charakteristikas, remiantis teoriniais ir eksperimentiniais vertinimais, nustatyti radionuklidinę kuro ir aktyvuotų medžiagų sudėtį bei apibrėžti radiacinės saugos požiūriu reikšmingų radionuklidų sąrašą. Sukauptos mokslinės žinios apie eksploatacinių ir eksploatacijos nutraukimo metu susidarančių radioaktyviųjų atliekų nuklidinės sudėties teorinius ir eksperimentinius vertinimus, būtų naudingos pasirenkant optimaliausias technologijas, įvertinant galimus radionuklidų sklaidos iš kapinynų scenarijus, prognozes ir nepageidautinos radioaktyviosios taršos pasekmes.

Pagrindinis šio darbo tikslas - sukurti radioaktyviųjų atliekų srautų susidarymo Ignalinos AE technologinėse grandyse modelį ir srautų radionuklidinės sudėties vertinimo metodiką. Svarbiausi šio darbo rezultatai, atspindintys jo naujumą ir svarbą:

Parengtas reikšmingų radionuklidų sąrašo sudarymo metodas, kuris remiasi dvejais atrankos kriterijais - radionuklidų pusėjimo trukmės (didesnė nei 0,5 m.) ir radionuklidų savitųjų aktyvumų ir jų nebekontroliuojamųjų lygių santykių - R_i lyginimo su ^{60}Co savitojo aktyvumo ir jo nebekontroliuojamojo lygio santykiu - R_{Co} (R_i/R_{Co} didesnis nei 10^{-5}). Po to sąrašas papildomas kapinyno saugos vertinimui svarbiais radionuklidais, remiantis preliminariais priimtinumo laidoti atliekas kriterijais. Remiantis šia metodika sudarytas reikšmingų radionuklidų sąrašas, tikslintas kiekvienam srautui atsižvelgiant į radioaktyviųjų atliekų srauto savybes ir atliekų šalinimo būdą.

Ištirta radioaktyviųjų atliekų sudėties kitimo seka, aprėpianti technologinius procesus nuo radioaktyviųjų atliekų susidarymo iki jų laidojimo, ir sudaryta radioaktyviųjų atliekų susidarymo schema, paaiškinanti skirtingos radionuklidinės sudėties radioaktyviųjų atliekų srautų susidarymo esmę.

Pirmą kartą pasiūlyta ir įdiegta nauja sunkiai matuojamų nuklidų netiesioginio vertinimo metodika atominei elektrinei su RBMK reaktoriumi. Darbe kompleksiškai taikyti kompiuterinio modeliavimo, radiocheminės analizės ir branduolinės spektroskopijos eksperimentiniai metodai.

Eksperimentiniais rezultatais pagrįstas nuklidinio vektoriaus metodo tinkamumas charakterizuoti RBMK radioaktyviąsias atliekas. Pateikiamos rekomendacijos dėl proporcingumo daugiklių taikymo charakterizuojant skirtingus radioaktyviųjų atliekų srautus. Atraminiais nuklidais charakterizuoti RBMK reaktoriaus radioaktyviąsias atliekas tikslinga pasirinkti ^{60}Co ir/arba ^{137}Cs. Kai radionuklido ^{60}Co savitasis aktyvumas yra apytikriai lygus arba didesnis negu radionuklido ^{137}Cs savitasis aktyvumas ir stebima gera šių radionuklidų aktyvumų koreliacija, atraminiu nuklidu tikslinga pasirinkti ^{60}Co, nes šio radionuklido aptikimo slenkstis radioaktyviųjų atliekų pakuotėse yra mažesnis negu ^{137}Cs ir stebima gera ^{60}Co ir aktinoidų aktyvumų koreliacija (koreliacijos koeficientas yra tarp 0,8-0,93). Jei radionuklido ^{60}Co savitasis aktyvumas yra daug mažesnis negu radionuklido ^{137}Cs savitasis aktyvumas, atraminiu nuklidu tikslinga pasirinkti ^{137}Cs. Tais atvejais, kai šių radionuklidų aktyvumų koreliacija silpna (pvz., kai nagrinėjamas apimantis filtrų sistemą radioaktyviųjų atliekų srautas), reikia pasirinkti du atraminius nuklidus: ^{60}Co ir ^{137}Cs. Analizuojant eksperimentinius rezultatus, pasiūlyti pagalbiniai atraminiai radionuklidai aktinoidų ir dalijimosi produktų proporcingumo daugikliams nustatyti.

Darbe pirmą kartą sudaryti RBMK reaktoriaus eksploatacinių radioaktyviųjų atliekų srautų - apdorotų skystųjų radioaktyviųjų atliekų bei vandens valymui naudojamų filtrų bei kietųjų radioaktyviųjų atliekų - proporcingumo daugiklių rinkiniai (nuklidiniai vektoriai). Taip pat įvertinta atominės elektrinės įrangos radioaktyviosios taršos nuklidinė sudėtis. Kietųjų atliekų sraute išskirti 8 srautai, apibūdinti atskirais nuklidiniais vektoriais.

Nustatyta, kad korozinių radionuklidų ir ^{60}Co aktyvumų santykiai (proporcingumo daugikliai $k_{60Co)}$ praktiškai (dydžio eilės tikslumu) yra vienodi visose RBMK reaktoriaus radioaktyviosiose atliekose. Aktinoidų ir ^{60}Co aktyvumų santykiai įvairiuose RBMK reaktoriaus radioaktyviosiose atliekose kinta nedaug (pvz., mažiausi ^{239}Pu proporcingumo daugikliai k_{60Co} dujų filtruose ir cementuotose atliekose skiriasi nuo didžiausiųjų dujų kontūro įrangoje ir ventiliacijos sistemoje apie 30 kartų). Tačiau dalijimosi produktų ir ^{60}Co aktyvumų santykiai įvairiuose RBMK reaktoriaus radioaktyviosiose atliekose gali žymiai skirtis (pvz., ^{137}Cs proporcingumo daugikliai k_{60Co} kinta nuo $9{\cdot}10^{-2}$ ventiliacijos sistemoje iki 40 bitumuotose atliekose, ^{90}Sr - nuo $1{\cdot}10^{-4}$ iki $2{\cdot}10^{-2}$). ^{14}C ir ^{60}Co aktyvumų santykis skirtinguose atliekų srautuose kinta nuo $7{\cdot}10^{-2}$ turbinų salės įrangoje iki 17 dujų kontūro įrangoje. Todėl tikslinga RBMK reaktoriaus radioaktyviosiose atliekose išskirti atskirus atliekų srautus, kuriuose proporcingumo daugikliai kinta nedaug, ir šiuos radioaktyviųjų atliekų srautus charakterizuoti skirtingais nuklidiniais vektoriais.