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ANALYSIS OF NUCLIDE CONTENT IN IGNALINA NPP RADIOACTIVE WASTE STREAMS

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Statistical analysis of radionuclide specific activities in Ignalina NPP technological media and radioactive waste was performed, and the corresponding scaling factors were calculated. The results indicate that radionuclide scaling factors are directly influenced by the radionuclide generation and migration processes in the reactor as well as by chemical properties of radionuclides. Correlations of difficult-to-measure radionuclides to the key radionuclide ^{60}Co show that radioactive waste streams of Ignalina NPP can be distinguished according to the calculated scaling factor values. Results indicate that the scaling factor method can be applied to the RBMK–1500 reactor waste characterization.

Keywords: RBMK radioactive waste, scaling factor, specific activities, modelling

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1. Introduction

One of the essential activities of the decommissioning of Ignalina Nuclear Power Plant (Ignalina NPP) is the radioactive waste management. Huge amounts of radioactive waste were generated at Ignalina NPP during the whole operation period since the start of the first Unit in 1983. Requirements for radioactive waste management changed in 2001, when new radioactive waste management norms were set up. Besides other technological issues of the radioactive waste management, it is required to measure and declare the nuclide content in radioactive waste which must be disposed of in disposal facilities. Several theoretical nuclide determination techniques such as modelling of activation in structural components of the reactor core and prediction of the fuel nuclide composition in nuclear fuel using codes such as MCNP or SCALE can be applied [1]. However, it is difficult to predict the nuclide content in very low level, and low and intermediate level radioactive waste by purely theoretical methods due to the physical and chemical processes involved in the radioactive waste generation. Therefore, experimental measurement techniques have to be implemented. Moreover, experimental measurements of the nuclide content are a basic tool for validation of theoretical predictions [2–5]. However, the practical problem of experimental determination of the nuclide content in radioactive waste is due to a huge amount of radioactive wastes to be characterized. Therefore, some optimization principles have to be implemented in order to make the whole characterization process feasible and practicable [6].

The aim of this work is to show how the application of experimental techniques and implementation of analysis of waste generation at Ignalina NPP make it possible to group the radioactive waste into several streams which can be characterized by characteristic scaling factors. In particular, experimental results of the radioactive waste activity measurement demonstrate the possibility of the scaling factor application to characterization of the RBMK type reactor radioactive waste streams.

2. Radioactive waste generation at Ignalina NPP

A simplified scheme of main streams of the Ignalina NPP radioactive waste generation is presented in Fig. 1. The reactor core is the main polluting source due to generation of radionuclides during neutron capture or nuclear fission and activation of reactor core components. These radionuclides can be released to the technological media of NPP due to fuel cladding defects and corrosion of metal structures of reactor core components and contamination of the main circulation circuit (MCC) coolant due to direct contact. The contaminated coolant

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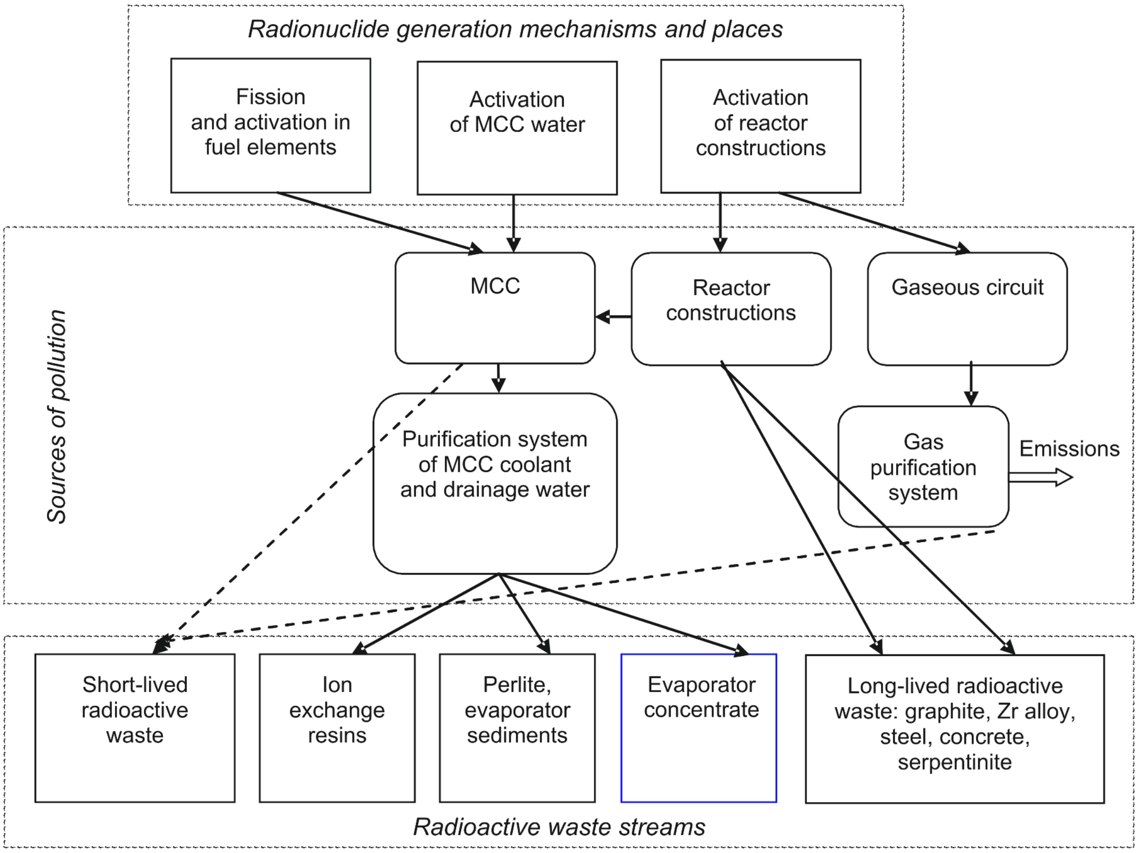

Fig. 1. Simplified scheme of Ignalina NPP radioactive waste streams. MCC is main circulation circuit.

from MCC can be further transferred to the final waste by three main routes: (i) direct contamination due to direct contact with structural materials of NPP, (ii) the loss of a coolant through leakages to the drainage system, and (iii) the chemical purification of a coolant by ion exchange resins and perlite. Each step of radionuclide transfer from one media 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 a nuclear fuel, reactor core components, and final radioactive waste. This implies that isotopes of elements with some common chemical properties (solubility in water, chemical activity) can be expected to behave similarly during transport through technological media of NPP.

3. Experimental methods and modelling

Ignalina NPP technological media – the main circulation circuit and radioactive waste – were investigated by theoretical and experimental techniques. A list of radiologically relevant radionuclides was compiled taking into account their unconditional release limits [7], half-life (>0.5 years) and relative radiotoxicity: ^{54}Mn, ^{55}Fe, ^{60}Co, ^{65}Zn, ^{90}Sr, ^{93}Zr, ^{93m}Nb, ^{94}Nb, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{244}Cm [8].

Measurements of the radioactive waste specific activity were performed in a broad specific activity range. Samples for measurements were taken for characterization of waste streams (Fig. 1): cemented waste, bituminized waste, solid waste as well as the main circulation circuit, turbine hall equipment, emergency core cooling system. Measurement techniques were described in detail in [8].

In order to extend the range of activity correlation, theoretical modelling of the spent nuclear fuel nuclide composition was carried out using T_DEPL sequence from SCALE 5 code package [9]. One quarter of fuel assembly with the fuel channel and the surrounding graphite moderator was modelled using 2D NEWT geometry for the neutron cross-section calculation. Mirror boundary conditions were used for 4 sides. In order to account for the axial neutron leakage, the stack height was set to 6.84 m. The 2.6% ^{235}U initial en-

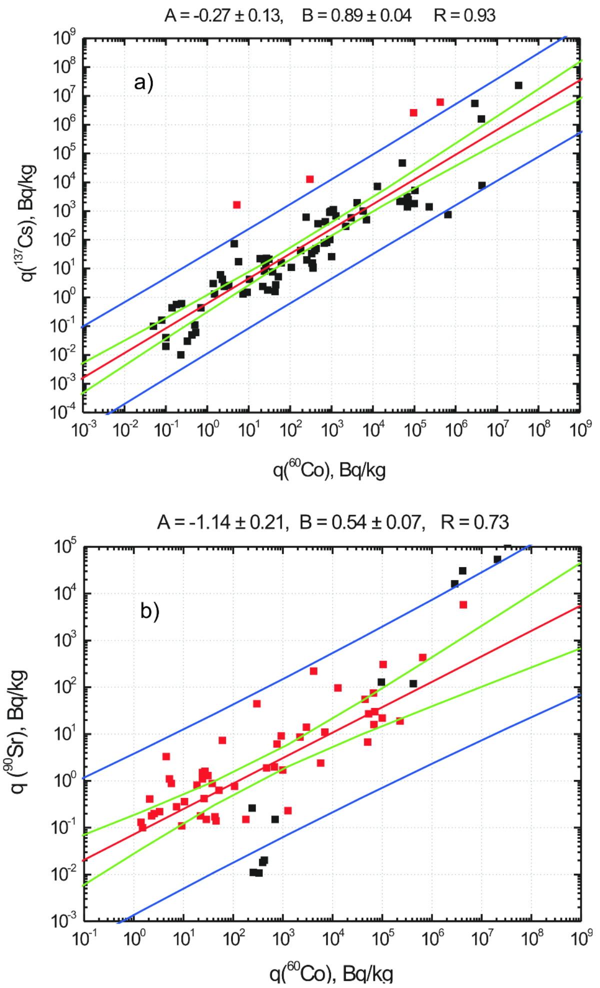

Fig. 2. Correlation of ^{137}Cs and ^{90}Sr specific activities with the key radionuclide ^{60}Co specific activity in Ignalina NPP radioactive waste.

richment of fuel was used as most common in the Ignalina NPP reactor core. The average burnup was taken 12 MW d/kg U. During refuelling of the core only fuel assemblies are changed, the fuel channel and graphite remain in the core. Thus, the fuel channel activation was modelled for 20 years of continuous operation. Coolant density was 0.5 g/cm^3 and temperature 550 K.

4. Results and discussion

The correlation analysis of the measured specific activity of above indicated radionuclides was performed. As the measured activities cover several orders of magnitude, the logarithmic scale has been used. Analysis results provide information on the coefficient of proportionality (scaling factor) between the key nuclide ^{60}Co and the difficult-to-measure radionuclide. To evaluate uncertainty of scaling factors the confidence bands were calculated for each correlation line. The upper limit of nuclide specific activity in the sample was determined from the upper prediction band (Fig. 2). The correlation coefficient of radionuclides with the key one in most cases was better than 0.9.

Results presented in Fig. 2 indicate the applicability range of determined correlations of specific activities between the difficult-to-measure and key radionuclides. For some radionuclides, e. g., ^{137}Cs, activation products ^{54}Mn, ^{55}Fe, ^{65}Zn, ^{93}Zr, ^{93m}Nb, ^{94}Nb specific activities correlate well with ^{60}Co specific activity due to the same generation process in the reactor core (Fig. 2(a)). However, as one can see in Fig. 2(b) the linear correlation of ^{90}Sr and ^{60}Co is not very good in the lower range of activities. When specific activities are low, the influence of systematic measurement errors (e. g., lower detection limit) probably becomes more relevant, and thereby affects the final results. This can be taken into account when fitting experimental data by applying some weighting of data points which are suspected to have higher experimental uncertainties.

Analysis of radionuclide activity correlations shows how radionuclides are transported in the technological media of Ignalina NPP. The scaling factor value of a particular radionuclide reflects its mobility in comparison to the selected key radionuclide ^{60}Co. One can see in Fig. 2 that the scaling factor of ^{137}Cs is higher than that of ^{90}Sr, therefore ^{137}Cs is more mobile than ^{90}Sr, which corresponds to the chemical properties of these elements. Moreover, Fig. 3 demonstrates how migration pathway from the generation source to the waste influences the resulting scaling factor of radionuclides. One can clearly see in Fig. 3 that the activity ratio of ^{137}Cs to ^{60}Co is by about two orders of magnitude higher in the liquid waste compared to the one in the solid waste. This can be understood as the influence of radionuclide transport processes on the final waste inventory. The liquid waste is directly contaminated by fission products due to the presence of fuel cladding defects, whereas the path of radionuclides to the solid radioactive waste is longer, therefore the concentration of fission products like ^{137}Cs is relatively lower (Fig. 3).

Results of correlation analysis provided in Fig. 3 indicate that waste streams generated by the RBMK–1500 reactor can be distinguished by the scaling factor (e. g., for ^{137}Cs in Fig. 3). This is the answer to the question whether the scaling factor method can be applied to the RBMK–1500 type reactor that is currently operating in NPP. The results clearly show that the waste generated in different waste streams (liquid and solid waste in the case presented in Fig. 3) can be separated due to different correlation of the radionuclide of interest with the

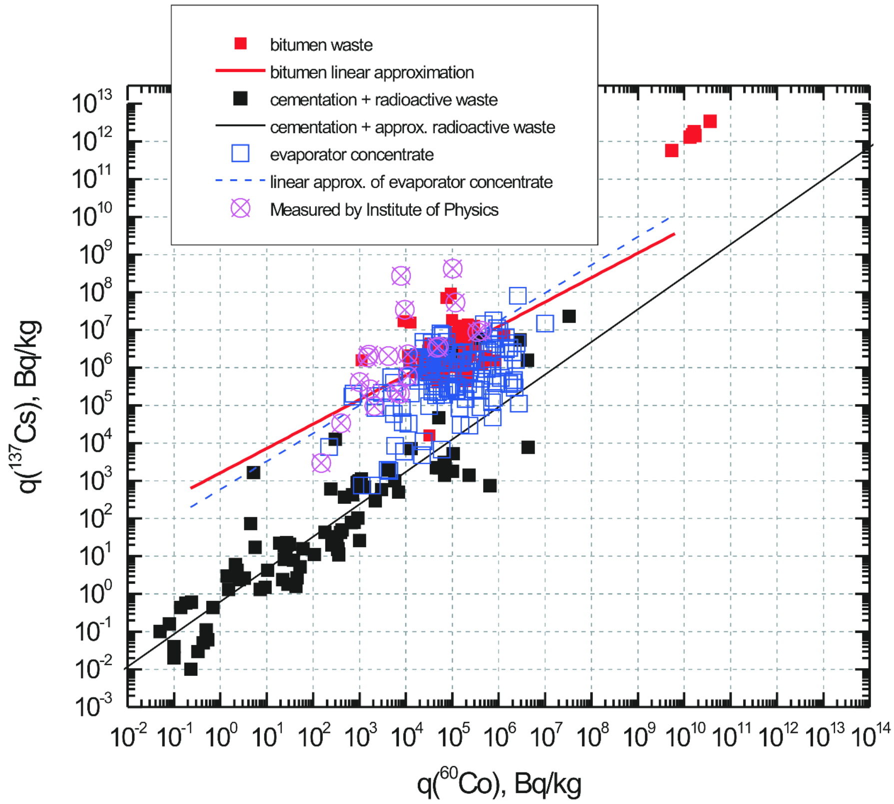

Fig. 3. Correlation of ^{137}Cs specific activity with the key radionuclide ^{60}Co specific activity in Ignalina NPP solid and liquid radioactive waste.

key radionuclide. Moreover, measured specific activities of ^{137}Cs and ^{60}Co in waste samples in both cases correlate rather well (the correlation coefficient in most cases is more than 0.9). This means that activity values are close to the correlation line in a broad activity range and uncertainties of activity predictions are kept within an acceptable range, known from the analysis (Fig. 2).

5. Conclusions

Correlation analysis of experimental data of radiologically relevant radionuclides in various technological media and radioactive waste of the RBMK–1500 reactor of Ignalina NPP was performed. The analysis results show that data on logarithmic scales can be approximated by a linear dependence of the activity of difficult-to-measure radionuclides to the key radionuclide with the correlation coefficient in most cases better than 0.9. The calculated correlation is sufficient to predict the activity of difficult-to-measure radionuclides using the key radionuclides. Experimental data of the radionuclide specific activity in Ignalina NPP technological media and radioactive waste show that the radionuclide inventory and resulting scaling factors of waste streams are influenced by the radionuclide transport processes governing the waste generation and chemical properties of radionuclides.

References

[1] W. Müller, Activity determination and declaration – An overview, in: *Determination and Declaration of Nuclide Specific Activity Inventories in Radioactive Wastes* (International Workshop, Cologne, Germany, 2001) pp. 1–10.

[2] V. Remeikis and A. Jurkevičius, Evolution of the neutron sensor characteristics in the RBMK–1500 reactor neutron flux, Nucl. Eng. Des. **231**(3), 271–282 (2004).

[3] V. Remeikis, G. Klevinskas, L. Juodis, A. Plukis, and R. Plukienė, Analysis of the relative release rate of radionuclides from the RBMK–1500 reactor fuel elements, Nucl. Eng. Des. **237**(8), 840–847 (2007).

[4] D. Ancius, D. Ridikas, V. Remeikis, A. Plukis, R. Plukienė, and M. Cometto, Evaluation of the activity of irradiated graphite in the Ignalina Nuclear Power Plant RBMK–1500 reactor, Nukleonika **50**(3), 113–120 (2005).

[5] V. Remeikis, A. Plukis, L. Juodis, A. Gudelis, D. Lukauskas, R. Druteikienė, G. Lujanienė, B. Lukšienė, R. Plukienė, and G. Duškesas, Study of the nuclide inventory of operational radioactive waste

for the RBMK–1500 reactor, Nucl. Eng. Des. 2008 (submitted).

[6] H. Masui, M. Kashiwagi, W. Müller, and B. Lantés, Suggestion to waste classification for scaling factor method, in: *ICEM'03, Proc. of the 9th International Conference on Environmental Remediation and Radioactive Waste Management*, eds. G. Benda and F. Sheil (American Society of Mechanical Engineers, New York, 2003) pp. 1–6.

[7] *Clearance Levels of Radionuclides, Conditions of Reuse of Materials and Disposal of Waste*, Normative document of the environmental protection of the Republic of Lithuania LAND 34-2000 (Ministry of Environment of the Republic of Lithuania, 2000).

[8] D. Lukauskas, R. Plukienė, A. Plukis, A. Gudelis, G. Duškesas, L. Juodis, R. Druteikienė, G. Lujanienė, B. Lukšienė, and V. Remeikis, Method to determine the nuclide inventory of low-activity waste of the RBMK–1500 reactor, Lithuanian J. Phys. **46**(4), 497–503 (2006).

[9] M.D. DeHart, *TRITON: A Two-Dimensional Depletion Sequence for Characterization of Spent Nuclear Fuel*, Vol. I, Book 3, Sect. T1 (ORNL/TM-2005/39, USA, 2005).

NUKLIDINĖS SUDĖTIES ANALIZĖ IGNALINOS AE RADIOAKTYVIŲJŲ ATLIEKŲ SRAUTUOSE

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Santrauka

Eksperimentiškai įvertinta Ignalinos AE technologinių terpių ir radioaktyviųjų atliekų nuklidinė sudėtis bei SCALE 5 programiniu paketu teoriškai sumodeliuota panaudoto branduolinio kuro nuklidinė sudėtis. Išanalizuotos sunkiai matuojamų ir atraminių radionuklidų savitųjų aktyvumų, nustatytų eksperimentiškai ir teorinio modeliavimo būdu, koreliacijos, apskaičiuoti proporcingumo daugikliai įvairiems Ignalinos AE atliekų srautams. Analizės rezultatai rodo, kad proporcingumo daugiklių vertės tiesiogiai atspindi radionuklidų susidarymo ir pernašos procesus reaktoriuje bei radionuklidų chemines savybes. Sunkiai matuojamų radionuklidų savitųjų aktyvumų koreliacija su atraminio ^{60}Co radionuklido savituoju aktyvumu rodo, kad Ignalinos AE radioaktyviųjų atliekų srautai gali būti atskiriami pagal išmatuotas ir / ar apskaičiuotas proporcingumo daugiklių vertes. Gauti rezultatai rodo, kad proporcingumo daugiklių metodas gali būti naudojamas Ignalinos AE atliekoms charakterizuoti.