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ANALYSIS OF RW RADIONUCLIDE COMPOSITION RELEVANT FOR THE LONG-TERM SAFETY OF ITS DISPOSAL

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At present time, active development of the Unified State System for Radioactive Waste Management is underway: existing RW management practice is being revised in keeping with newly approved requirements that had been set forth in the Federal Law on Radioactive Waste Management and Amendments to Some Regulations of the Russian Federation. A fundamental change introduced therein suggests that disposal of generated RW should be now considered as a must. Thus, it imposes certain requirements on all RW management processes including those associated with their certification. In Russia, the practice of radionuclide control mostly addresses personnel radiation safety, nuclear safety and the losses of products at operational stages. Bearing in mind that the list of radionuclides determining waste hazard level at different time periods and waste management stages may differ, existing waste management practice should be evaluated in terms of its compliance with the new RW management setting.
This paper studies the list of radionuclides contained in RW that should be measured at NPPs based on international experience of applying radionuclide vector method and safety case development for RW disposal facilities.

Key words: *radioactive waste (RW), disposal of radioactive waste, legal framework, long-term safety of radioactive waste disposal facilities (RW DF), unified state system for radioactive waste management (USS RW), radionuclide composition.*

Introduction

Radionuclide composition and activity are seen as most important parameters describing RW. Moreover, at different RW management stages (storage, transportation, disposal and etc.) the level of RW potential hazard is associated with different radionuclides. Historically, radionuclide composition control practice has been guided by operational safety concerns [1]. Due to a number of reasons, for a long time, issues associated with RW disposal, including thorough characterization of RW radionuclide content, haven't been properly accounted for.

However, in 2011, a regulation [2] introducing the concept of compulsory disposal for all RW types was approved providing for essentially new requirements on radionuclide content control process. This issue is also relevant in terms of economic feasibility as the classification of retrievable RW [3] based on which RW disposal tariffs are set [4] considers two criteria, namely, radionuclide specific activity and half-life. In general requirements to the list of radionuclides subject to control and certification should be identified based on waste acceptance criteria set on individual basis for a particular facility.

Legal changes in RW management concept produced no drastic alterations in terms of the certification process which is still aimed at addressing three key objectives:

- Radiation safety of personnel during RW management;
- Control over product losses;
- Nuclear safety.

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The above-mentioned concerns have resulted in a situation when no measurements are performed for some long-lived and particularly hardly detectable radionuclides. Even though such data is required for the long-term safety assessment of RW disposal facilities.

The first stage in the development of the Unified State System for RW Management (USS RW) involved the inventory taking campaign covering the whole amount of already accumulated waste. Data obtained including those on RW radionuclide composition and content has been put into the State System for Radioactive Substance and Radioactive Waste Accounting and Control (SGUK RV RAO). This paper evaluates the adequacy of existing data in terms of addressing the identified tasks in the field of RW management under new legal and regulatory setting.

It should be noted that literature sources have already mentioned some possible negative effects of data gaps associated with radionuclide composition that can be produced on RW processing activities [5] and classification of retrievable RW for disposal [6]. For this reason, these issues are not being discussed in detail here with particular focus placed on the sufficiency of data for adequate safety assessment.

Analysis of international experience in RW characterization and disposal safety assessment

The task of determining radiologically relevant radionuclides requiring some state regulatory actions is viewed as an important one for all nuclear power countries. To date a big amount of knowledge addressing this issue has been gained worldwide. For decades, RW disposal concepts have been implemented in a number of European countries and extensive knowledge has been gained in the field of long-term safety assessment. Thus, this knowledge can provide a basis for further investigations aiming at the development of requirements on RW radionuclide composition in Russia.

It's worth mentioning that there are some significant differences in terms of the existing requirements discussing the way in which the list of radionuclides contained in RW should be developed.

In some cases, these differences are due to different RW characteristics resulting from different nuclear technologies applied in these countries. As RW DF safety assessment determines the requirements for measured radionuclides, different requirements may be set forth for different RW. This is the case of radionuclide inventories that had been proposed in RW DF safety assessment developed in Sweden for low- and intermediate waste [10] and spent nuclear fuel [11].

Most of these radionuclides are present in both lists, however some differences can be noted. For example, the long-term safety assessment for a near-surface disposal facility [10] involves data on a big number of radionuclides. The reason behind is that it also involves short-lived radionuclides and radionuclide release from a near-surface repository will occur faster as compared to a deep SNF repository. Moreover, ^{41}Ca not accounted for in deep RW DF safety assessments should be considered in case of a near-surface repository as its significant amounts can be present only in irradiated concrete structures subject to disposal in near-surface repositories.

Considering this, international best practices can be evaluated using two approaches: the first one suggesting RW certification practices in terms of measured (controlled) radionuclides, the second one — considering safety case development practices for RW disposal facilities. Both options may provide a list of radionuclides that should be measured in RW.

One type of RW was provided as an example in order to derive a correct list of radionuclides. RW generated from NPP operation seems to be the most appropriate one: their management practice is currently considered as a most elaborated one. On the other hand, as any country with well-developed nuclear power has such waste, it enabled to obtain a most representative selection for our analysis.

The first approach suggests the use of radionuclide vector method [12] — practice widely used abroad as RW generated by NPPs contain a significant amount of hardly detectable radionuclides. This method is based on the identified rate between specific activities of reference radionuclides that can be detected using relatively simple gamma-spectrometry methods and hardly detectable radionuclides that can be identified only via destructive analysis. 9 countries were selected to perform this evaluation (US, Belgium, Slovakia, Spain, Grate Britain, France, Lithuania, Germany and Canada) [13]. Also data on Novovoronezh NPP was used in this assessment [14].

Evaluation was performed via iterative comparison of radionuclide lists aiming to identify and to exclude from further evaluations those of rare occurrence. Iterations continued till the list of radionuclides measured by the majority of the countries mentioned above has emerged. Python-library was used as an evaluation tool for the data analysis (Pandas) [15].

High rates for correlation matrixes in the lists of different countries were used as criteria for radionuclide selection. Furthermore, during the final iteration multiple correlation factor for a given list of radionuclides was calculated for each country as a final check of compliance. It should be noted that when relatively small selections are used for evaluations, multiple correlation factors are generally overestimated. In such cases the results obtained can hardly reflect the actual state of affairs. For this reason, standard criteria defining the high correlation rate has been increased from $R_{y|x_1,x_2\dots} = 0.7$ to $R_{y|x_1,x_2\dots} = 0.8$ [16].

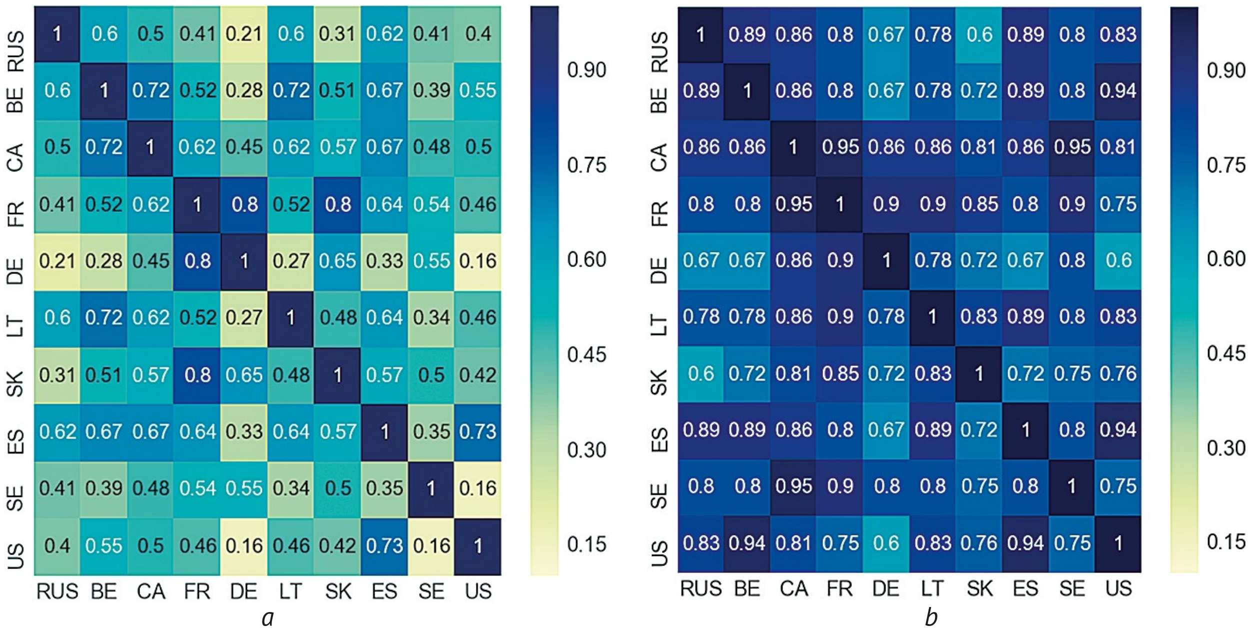

Figure 1. Comparison of final RW lists

Equivalency of initial radionuclide list contents was evaluated at the preliminary stage based on Pearson correlation coefficient [17]. Correlation matrix presented at the thermal map (figure 1a) shows that most part of considered radionuclide lists have little common in between. In particular, this could be explained by differences in reactor technologies applied and RW streams, as well as RW disposal conditions. As data on measurements of particular actinides was not available in some countries, these were excluded from further analysis at the preliminary stage already. Basically, this enabled to improve the general picture, however, the final goal of developing a complete list was yet not achieved. A number of iterations aiming to exclude rarely occurring radionuclides was carried out.

The final iteration enabled to exclude the radionuclides appearing in less than 50 % of country lists evaluated. Results of multiple correlation estimations are presented in figure 1b. Correlation factor values have grown considerably and multiple correlations for each country have reached the values derived from criteria accounting for 0.85—0.99. This enables to conclude that the radionuclide list derived may be viewed as a basis enabling to assess the adequacy (sufficiency) of data in SGUK RV and RAO system for the long-term safety assessment of RW disposal facilities.

As the result, 14 radionuclides were present in the baseline list derived: ^{3}H, ^{14}C, ^{36}Cl, ^{55}Fe, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{93}Zr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{135}Cs and ^{137}Cs.

Two groups of radionuclides can be singled out from this list:

1. Markers (^{137}Cs, ^{60}Co and ^{94}Nb);

2. Hardly detectable (^{3}H, ^{14}C, ^{36}Cl, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{93}Zr, ^{99}Tc, ^{129}I, ^{135}Cs).

Radionuclides assigned to group 1 can be measured using quite simple instrumental monitoring methods (gamma-spectrometry). Content of group two radionuclides is defined based on radionuclide vector method.

Yet another interesting example to note and associated with the second approach to the development of measured radionuclide lists is the long-term safety assessment for a near-surface disposal facility in Sweden. In [18] and [19] it was stated that radiological significance of radionuclides depends on RW management stage. A similar conclusion can be drawn about considered repository evolution scenarios. The table below presents the evaluations for relative contribution of different radionuclides into annual effective doses for different SFR (storage facility for low- and intermediate-level waste) evolution scenarios. Radionuclides contributing to less than 5 % of annual effective dose for all listed evolution scenarios have been excluded from these evaluations [10].

Evolution scenarios for the repository itself and site vicinities covering a period of 1,000,000 years serves a basis for repository long-term safety assessment in Sweden. Evolution scenarios are based on the following points:

- Baseline conditions at repository site;
- External impacts affecting the repository at the post-closure stage. External impacts involve both climate changes and climate related processes: permafrost, global warming, shoreline displacement. Human activities can also impact the repository.
- Internal processes occurring inside the repository. Internal processes involve thermal, hydraulic, mechanical and chemical processes occurring inside the repository. These processes include ground water flow and chemical degradation affecting engineered safety barriers.

Based on such evolution scenarios future radiological impact of the repository on public and the

Table 1. Relative contribution of different radionuclides most significantly impacting public exposure rates under various repository evolution scenarios

Radionuclides	Scenarios						
	Basic		Low-probability		Very low-probability		
	Global warming	Early periglacial climate	Waste disposal in case of permissible limit exceedance	High concentration of complexation agents	Altered Eh conditions	Open storage, uncontrolled	
						SFR	SFR+SFL*
	Contribution to the annual dose, %						
^{129}I	5.8	71.1	7.7	1.4	1.8		
^{99}Tc				1.7	11		
^{59}Ni			1.5	75.7	18.8		
^{63}Ni						28.1	93.3
^{93}Mo	57.7	7.8	47.3				1.1
^{14}C			8.4				5.1
^{79}Se			15.1				
^{239}Pu				8.5	53.8		
^{137}Cs						63.3	1.6

*this scenario considers an option when waste subject to disposal in HLW repository (SFL) are temporally stored in SFR.

environment can be evaluated. More detailed discussion of modelling issues associated with each evolution scenario presented in the table can be found in [10].

Table 1 clearly shows that no more than 5 radionuclides provide an important contribution to the public exposure rate for each scenario evaluated. At the same time the list of radionuclides may change depending on the scenario in question.

It should be noted that in case of long-term safety assessment considering a normal evolution scenario most decisive contribution into the public exposure is made by long-lived radionuclides having high migration rate. ^{129}I, ^{93}Mo and ^{14}C may be considered as such. There are much more strict requirements on their content than those associated with ^{137}Cs and ^{60}Co. For example, as specified in [10], ^{129}I activity that can be disposed of in the repository is 6 orders of magnitude smaller than the one of ^{137}Cs even though specific activity limits differ only by an order of magnitude (10^2 and 10 respectively). Safety assessment performed for the near-surface repository showed that depending on scenario, ^{129}I may contribute to up to 71% of public exposure value [10]. Thus, these radionuclides should be definitely accounted for.

^{93}Mo and ^{79}Se were added to the list of measured radionuclides based on the evaluation of safety assessment case studies for near-surface repositories.

Evaluation of RW inventory taking campaign results carried out in Russia

To evaluate the sufficiency of existing data on the radionuclide composition of Russian RW, resulting list should be compared with RW inventory taking results. Their evaluation has shown that only 15 radionuclides had been indicated for RW generated by NPPs. However, this number should not give a wrong idea as the level of detail for data on radionuclide composition seams to be quite heterogeneous. Figure 2 shows the "occurrence" rate for radionuclides accumulated in RW from NPPs (amount of RW indicating if the given radionuclide is present).

Data presented indicates that for the base amount of RW only the content of four radionuclides was specified (^{54}Mn, ^{60}Co, ^{134}Cs, ^{137}Cs). This results from the common RW characterization practice at NPPs implemented using gamma-spectrometry. This method, on the one hand, is viewed as a quite simple one, and on the other, enables to obtain the data concerning the radionuclides most significantly contributing to personnel exposure.

Comparison of data presented in the table and the list of radionuclides derived from the inventory taking campaign demonstrates that for RW from Russian NPPs, ^{137}Cs is the only radionuclide singled out of the total number of radionuclides most significantly contributing to repository evolution scenarios. Moreover, in the long-term safety assessment ^{137}Cs contribution is considered to be significant only in case of very low-probability scenario. Namely, in case of open uncontrolled storage. This clearly demonstrates that currently available data on the radionuclide content of waste generated by NPP is not sufficient enough for a comprehensive safety assessment of its disposal.

As for radionuclide characterization of RW disposed of in currently operated repositories for RW class 3 and 4 [20], Russian waste acceptance criteria provide no requirement to the contents of the abovementioned radionuclides. This may be due to the absence of accurate data on the contents of relevant radionuclides in RW which is not being considered as an unsolvable problem at the initial stages of RW management. In principle, for RW disposed of in the same repository different waste acceptance criteria accounting for their specific features can be set. As the knowledge on actual RW characteristics grows, conservative interval estimates on radionuclide contents in RW already disposed of in the repository may be accounted for

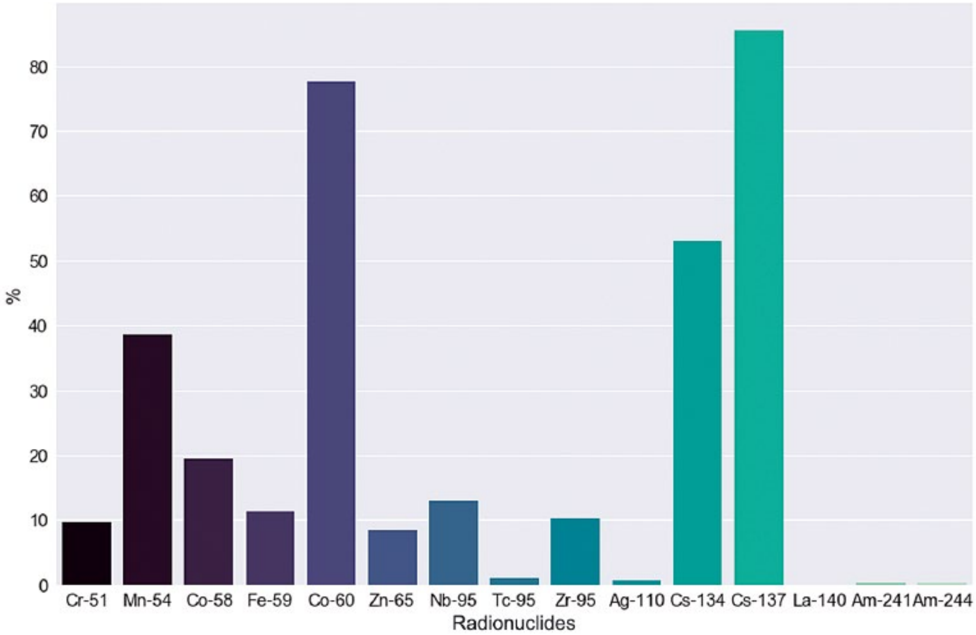

Figure 2. Distribution of radionuclides depending on RW amount

under the review of waste acceptance criteria. At the same time, stricter requirements may be imposed on newly disposed waste. In fact, this approach is similar to the one used in France — near-surface disposal facility (Aube) accepted short-lived LLW along with long-lived LLW in form of irradiated graphite. This resulted in extremely strict requirements on the contents of ^{36}Cl considered as a fast migrating radionuclide. In keeping with waste acceptance criteria specified for this disposal facility, the maximum content set for this radionuclide accounts for less than 5 Bq/g (maximum specific activity — less than 10^4 Bq/g) which is explained by its relatively high contents in graphite RW.

To avoid imposing excessively harsh requirements, data on the extended as compared to the existing RW radionuclide composition should be as soon as possible included into repository safety assessment and accounted for during the elaboration of waste acceptance criteria. The sooner this is implemented the less probable is the ineffective use of repository capacity.

It should be noted that the abovementioned challenge is considered to be topical not only for near-surface repositories, but also for deep geological disposal facility planned to be built in the Nizhnekansk rock mass. However, considering the plans on RW disposal in near-surface repositories (over 450,000 m^3 until 2035), extended evaluation of radionuclide composition for RW to be disposed of in near-surface repositories is considered as a more urgent task.

Conclusions

Statistical analysis of data on RW (generated at NPPs) certification in a number of countries has enabled to develop a basic list of most frequently occurring radionuclides which has been compared to similar data present in SGUK RV and RAO. Sufficiency of data available in SGUK RV and RAO on radionuclide composition of accumulated and generated RW was analyzed in the context of repository long-term safety assessment. It was demonstrated that the majority of radiologically relevant radionuclides is not measured. Data on radionuclide composition of RW available to date can not be considered sufficient for a comprehensive safety assessment of RW disposal facility and setting optimized waste acceptance criteria.

This circumstance should not be considered critical for the current stage of USS RW development as the use of interval assessments indicating the contents of radiologically relevant radionuclides enables to comply with safety requirements at the initial stage. However, international experience shows that later this approach can result in significantly stricter waste acceptance criteria for newly disposed waste and/or ineffective use of disposal capacity.

More detailed information on RW radionuclide composition is required for the long-term safety assessment. An in-depth evaluation of available knowledge should be performed at an early stage. It should be also indicated which method may be used for a certain type of waste.

Radionuclide vector method applied in a number of countries can be considered as a feasible one to provide the identification of such radionuclides in RW. It should be noted that investigations addressing the implementation of this method at NPPs have been started in Russia. This practice shall be continued and expanded. RW certification practice "to the extent possible" should be replaced by the one "as required" which is considered as a global challenge in this field.

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Bibliographic description

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