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DEVELOPMENT OF AN ALGORITHM FOR ESTIMATING THE RADIONUCLIDE COMPOSITION OF VITRIFIED HLW OF FSUE "PA "MAYAK" FOR THE PURPOSE OF THEIR SAFE DISPOSAL

Remizov M. B.[1], Kozlov P. V.1,[2], Borisenko V. P.[1], Dementeva I. I.[1], Blokhin P. A.[3], Samoylov A. A.[3]

[1]FSUE "PO Mayak", Ozyorsk, Chelyabinsk region, Russia

[2]OTI National Research Nuclear University MEPhI, Ozyorsk, Chelyabinsk region, Russia

[3]Nuclear Safety Institute (IBRAE RAN), Moscow, Russia

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The paper focuses on the development of a method enabling to calculated the activity of relevant radionuclides in vitrified RW Class 1 from FSUE "PA Mayak". This method is supposed to be used in the safety case for radioactive waste disposal in geological formations. The algorithm is based on the calculated isotopic composition of spent nuclear fuel derived from its characteristics (initial enrichment of ^{235}U, burnup and cooling time after irradiation) and process parameters responsible for radionuclide redistribution among material streams at various stages of SNF reprocessing and during RW vitrification.

Key words: *spent nuclear fuel, radioactive waste, aluminophosphate glass, vitrification, relevant radionuclides.*

Introduction

Since 1987, RT-1 plant at FSUE PA "Mayak" has been operated to vitrify radioactive waste (RW) resulting from spent nuclear fuel (SNF) reprocessing by its immobilization into aluminophosphate glass matrix. A total of over 6,200 tons of vitrified high-level waste (HLW) with a total activity of some 640,000,000 Ci (at the time of waste vitrification) have been accumulated during the entire operational life of RT-1 four electric furnaces (EP-500/2, EP/500/1-r, EP-500/3 and EP-500/4) [1]. In late 2017, EP-500/5 was commissioned providing for some 3,800 tons of vitrified RW with a total activity of over 570,000,000 Ci to be generated during its operation.

According to the classification system presented in [2], vitrified HLW are categorized as RW Class 1 and should be disposed of in deep disposal facilities (DDF RW). Relevant safety case development process should involve simulation of repository evolution considering relevant characteristics of the vitrified RW and the safety barrier system, both engineered and natural. This requires certain data on the evolution of glass material properties during their long-term storage under particular conditions (thermal, radiation and other impacting factors being taken into account). As far as RW disposal strategy was proposed rather recently [3] no measures had been taken in the past to identify RW characteristics considering this very context. Relevant characterization process was narrowed down to identifying a set of parameters required to ensure the operational safety of their storage.

The translation is done by N. S. Cebakovskaya

A qualitatively different approach to the initial data contents evaluation is required for the long-term safety assessment.

Radionuclide composition is viewed as an essential RW characteristics and relevant requirements for its identification are set forth in relevant provisions of general waste acceptance criteria [4]. However, these provisions suggest no specific requirements concerning the list of radionuclides to be identified. This should be changed, apparently, via the development of specific acceptance criteria for a particular RW disposal facility. Moreover, international practice shows that data on the majority of radionuclides contained in RW are not required ipso facto [5].

Thus, integrated task of characterizing very high-level waste (VHLW) physical and chemical properties and its radionuclide composition should be viewed as quiet a relevant one under the design development of the first Russian DDF RW. Taking into account large RW inventory accumulated to date and the projected future RW arisings, a data base summuarizing information on RW package characteristics should be viewed as a tool enabling to address this task. Due to intrinsic problems associated with direct measurements of VHLW and inconsistency of available records with current requirements, an algorithm enabling to determine VHLW composition based on PA Mayak data should be developed at the first stage of this project. The PA Mayak data should firstly involve process monitoring data from SNF reprocessing and LRW vitrification, as well as relevant data from modern computational and analytical systems.

The paper discusses the development of an algorithm enabling to evaluate VHLW radionuclide composition to ensure their safe disposal in the DDF.

Challenges associated with radionuclide composition of PA "Mayak" VHLW

SNF reprocessing (considering the part responsible for VHLW characteristics) and HLW vitrification involves a number of stages (Figure 1).

Two key challenges associated with the identification of RW radionuclide composition should be noted: compiling the list of monitored radionuclides and development of an algorithm to determine their contents in RW.

A quite obvious statement suggesting that the data on the radionuclide composition included into RW passport should cover all the needs arising during the development of a safety case, including the long-term safety assessment, and should comply with waste acceptance criteria, was considered as the basis for the list of monitored radionuclides. Consequently, the lists of radionuclides important for the following assessments should be previously compiled:

- Long-term safety assessment;
- Nuclear safety assessment;
- Assessment of thermal impact on DDF engineered safety barriers and geological formations.

Initial steps addressing this task considering DDF reference evolution scenario already performed in a previous study [6] resulted in a list of radionuclides relevant in terms of the long-term safety. The following parameters have been taken into account:

- Radionuclide activity in SNF initial composition;
- Radionuclide half-life;
- Water solubility limit for chemical species with investigated radionuclides contained in RW;
- Radionuclide migration velocity (in case of continuous porous medium, the phase distribution coefficient during physical and chemical interactions occurring within RW — ground water — rocks

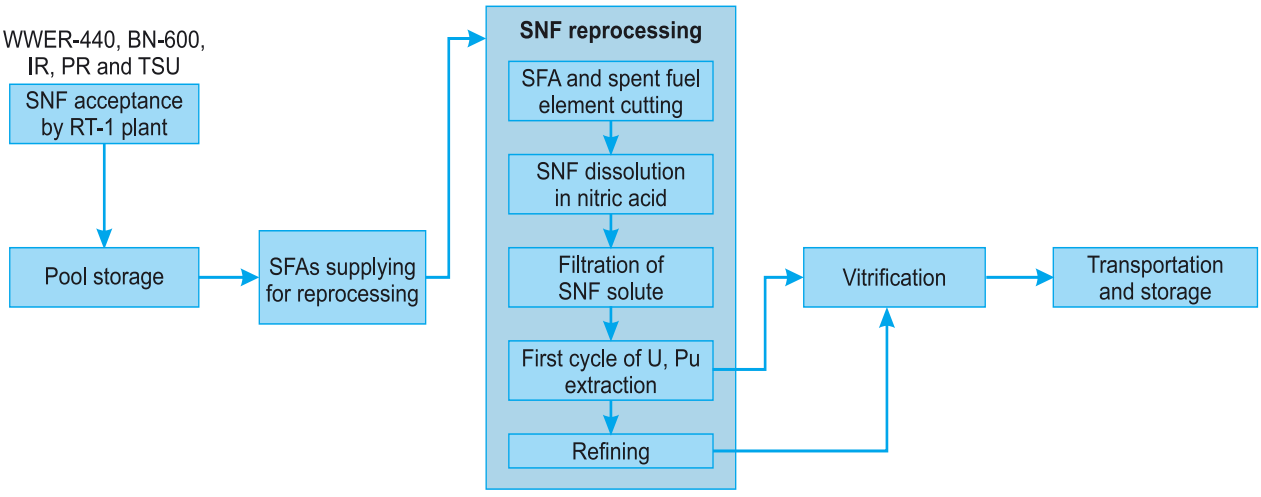

Figure 1. SNF management stages at RT-1

system and characteristics of the geological environment);
- Dose factors for drinking water intake (the reference scenario discussing potential impact on the population suggests drinking water supply with contaminated groundwater).

Considering the fact that no common agreement exists regarding the issues associated with the mechanism of radionuclide migration inside DDF and their release into the environment, list of radionuclides shown in Table 1 [6] was proposed as a reference one corresponding to the iteration step with no migration characteristics being accounted for.

Table 1. List of radionuclides relevant for the long-term safety assessment

Radionuclide	$T_{1/2}$, years	Radionuclide	$T_{1/2}$, years
^{93}Zr	$1.53 \cdot 10^6$	^{234}U	$2.46 \cdot 10^5$
^{93m}Nb	16.1	^{235}U	$7.04 \cdot 10^8$
^{126}Sn	$2.11 \cdot 10^5$	^{236}U	$2.34 \cdot 10^7$
^{129}I	$1.57 \cdot 10^7$	^{238}U	$4.47 \cdot 10^9$
^{226}Ra	$1.60 \cdot 10^3$	^{238}Pu	87.7
^{228}Ra	5.75	^{239}Pu	$2.41 \cdot 10^4$
^{227}Ac	21.8	^{240}Pu	$6.57 \cdot 10^3$
^{228}Th	1.91	^{242}Pu	$3.74 \cdot 10^5$
^{229}Th	$7.34 \cdot 10^3$	^{241}Am	433
^{230}Th	$7.54 \cdot 10^4$	^{243}Am	$7.37 \cdot 10^3$
^{231}Pa	$3.28 \cdot 10^4$	^{245}Cm	$8.50 \cdot 10^3$
^{233}U	$1.59 \cdot 10^5$	^{246}Cm	$4.73 \cdot 10^3$

As most part of SNF reprocessed at RT-1 plant (by volume) had resulted from VVER-440 reactor operations, the study [6] was based on the data discussing the radionuclide composition of VHLW from the spent fuel reprocessing. To demonstrate the capabilities of the proposed algorithm, this paper was based on a similar assumption. However, the opportunities for incorporating other SNF types to the batch (BN-350, BN-600, production, research reactors and transport ship units) have been also accounted for in the basic algorithm for radionuclide composition identification.

Moreover, the resulted list (Table 1) accounts only for the need of long-term safety assessments and should be extended based on the needs for assessing the power output, contents of fissile material and, possibly, the application of the radionuclide vector method.

Based on the evaluation of VVER-440 SNF radiation characteristics performed earlier it was concluded [7] that ^{137}Cs and ^{90}Sr with their daughter products should be considered as the radionuclides contributing to the largest extent to the energy output during the first 300 years. Among long-lived radionuclides, the biggest contribution to the energy output is associated with ^{241}Am. To assess nuclear safety in terms of radionuclide content, information on the contents of fissile isotopes is required. Such information is to the fullest extent possible presented in Table 1. For this reason, the list was not extended. As the result, the final list updated to ensure nuclear safety and the required thermal output includes 26 radionuclides (^{90}Sr, ^{93}Zr, ^{93m}Nb, ^{126}Sn, ^{129}I, ^{137}Cs, $^{226, 228}$Ra, ^{227}Ac, $^{228, 229, 230}$Th, ^{231}Pa, $^{233, 234, 235, 236, 238}$U, $^{238, 239, 240, 242}$Pu, $^{241, 243}$Am, $^{245, 246}$Cm).

As for the development of an algorithm enabling to determine VHLW radionuclide content, it should be noted that it will directly depend on a number of parameters:
- characteristics of initial SFAs batch transferred for reprocessing;
- degree of U and Pu extraction (was altered during technology development);
- degree of additional isotope extraction (extraction of Np and other isotopes was carried out periodically);
- redistribution of radionuclides along process flows during SNF reprocessing and RW vitrification.

Most relevant in this case is the impact of the latter factor being the least studied and potentially having a significant impact on the composition of relevant radionuclides in VHLW. In the first approximation, this process can be accounted for by introducing distribution coefficients. These coefficients indicate the relative proportion of the chemical element that remained in the material and is taken to the next stage of SNF reprocessing or RW vitrification process. Such coefficients should be determined individually for each relevant radionuclide with due account of process parameters ant its specific characteristics.

Stages of SNF reprocessing and RW vitrification process

Let's take a closer look at SNF reprocessing (the part impacting VHLW characteristics) and RW vitrification process to single out particular stages that may result in radionuclide redistribution. This process involves a number of main stages presented below:

1. Supplying a complete set of spent fuel assemblies (SFAs);
2. SNF dissolution and filtering;
3. Extraction of target components — U, Pu and Np at the first extraction cycle;
4. Supplying a batch of HLW and its evaporation;
5. HLW vitrification.

1 Supplying a complete set of spent fuel assemblies (SFAs)

This stage involves formation of SFA set transferred for reprocessing with due account of characteristics impacting the nuclear safety. Compliance with relevant guidelines on the contents of impurities in the dissolved SNF is considered as an important factor enabling to ensure routine reprocessing and accounting the isotopic composition of fuel target components. One batch is composed of a number of SFA types. Most part of already reprocessed SNF accounted for VVER-440 SNF. However, in separate batches the ratio between different types of SFAs may differ significantly. On average, over the year, the range of changes in the quantity of various types SFAs sent for processing is as follows: VVER-440 — from 55.4 to 98.0 %; BN-350, 600 — from 0.0 to 34.5 %; IR — from 0.0 to up to 2.6 %; TSU — from 0.8 to 12.9 %.

Radionuclide content in VHLW may be determined based on the radionuclide composition of reprocessed SNF, which can in turn be evaluated based on the research data from [8, 9]. This paper is based on the results obtained from TRACT nuclide kinetics code calculations [7].

Further on, under certain approximations (neglecting unrecoverable residues of the process chain and the continuity of extraction/re-extraction cycle, etc.) we will assume that the radionuclide composition of the initial HLW is determined specifically by the radionuclide composition and the number of different SFA types, as well as the subsequent process redistribution of radionuclides among various material streams (including the product of SNF radiochemical processing).

Types and the amount of SFAs being reprocessed as one batch, the time of their cooling can be determined based on archive records stored at RT-1 plant. Relationship between the specific activity of the *i* radionuclide in VHLW with no account of radionuclide redistribution among different streams can be described as follows:

$$A_i(t) = \frac{\sum_k m_k \sum_n A_i^n(t)}{M_{\text{OBAO}}}, \qquad (1)$$

where $A_i^n(t)$ is the specific activity of i radionuclide in the n SFA;
m_k is the mass of *k*-type SFA;
M_{OBAO} — resulting VHLW mass.

Calculating the activity of different radionuclides in each particular SFA is believed to be a quite complex task resulting from existing uncertainties associated with SFA operational conditions and neutron physical characteristics of the reactor facility during its operating time. For this reason, it's seems more practical to shift the focus from considering functional dependence for a specific SFA to a single dependence for SFA type between the exposure time after the irradiation was completed and the burnout depth. In this case, activity of VHLW at the time of its generation can be presented as follows:

$$A_i = \frac{\sum_k m_k \cdot \sum_j A_i^k(t^j_{\text{выд}}, \text{B}^j)}{M_{\text{OBAO}}}, \qquad (2)$$

$A_i^k(t^j_{\text{выд}}, B^j)$ — i-radionuclide specific activity in *k*-type SFA with burn-up and cooling time *j*;
m_k is the mass of *k*-type SFA;
N_k — number of *k*-type SFAs;
M_{OBAO} — resulting VHLW mass.

Further redistribution of radionuclides during reprocessing and HLW vitrification can be accounted for by introducing radionuclide redistribution factors. In this case the equation will be as follows:

$$A_i = \frac{\sum_k m_k \cdot \sum_j A_i^k(t^j_{\text{выд}}, \text{B}^j)}{M_{\text{OBAO}}} \cdot K_i, \qquad (3)$$

where K_i is redistribution/extraction factor for *i*-radionuclide.

2 Dissolution and SNF filtration

This reprocessing stage involves SNF cutting operations, its dissolution and segregation of non-soluble residues. This process involves redistribution of both volatile (^{3}H, ^{14}C, ^{129}I, etc.) and slightly soluble radionuclides (or of those absorbed on the solid phase) under conditions typical for SNF dissolution process (uranium, plutonium, cesium isotopes) occurs. The content of Pu and Cs isotopes in undissolved shells and sediments is extremely low [10]. This accounts for less than 0.5 % from those present in the SNF, therefore, these losses can be neglected in relevant calculations.

At the dissolution stage, almost all the amount of ^{14}C evolves into gaseous form as dioxide. Some part of it present in the form of elemental carbon stays at the stage of dissolved SNF filtration and remains in the residues. Thus, ^{14}C is not available at the next stage of extraction.

Moreover, 99.5 % of ^{129}I evolves into gaseous phase in the form of molecular iodine or organic iodine compounds.

Tritium redistribution is also worth noting. Although, tritium is not considered as radiologically relevant radionuclide for HLW (due its relatively

short half-life), its redistribution should be of interest in terms of near-surface disposal safety. Thus this radionuclide is considered relevant for other RW resulting from SNF reprocessing (owning to its high migration capacity).

A significant amount of ^{3}H (about 40 % from the amount generated during the fission process) remains within the zirconium cladding structure [11]. The remaining ^{3}H is excreted in the form of tritiated water (HTO) during the dissolution process. Thus, at this stage, about 60 % of the generated tritium remains in the SNF solution.

As for the other radionuclides contained in the spent fuel, almost all their amount is passed to the extraction stage.

3 Extraction of SNF target components

As a result of this redistribution high-level raffinates of the first extraction cycle (liquid HLW) are formed including nitrate solutions containing fission products, corrosion products and transplutonium elements, as well as trace amounts of under-extracted uranium, plutonium and neptunium [11—13].

At the first extraction cycle, the following amounts of isotopes are transferred into organics:

- 99.9 % of U isotopes;
- 99.5 % of Pu isotopes;
- 98 % of Np isotopes;
- 90 % of Th isotopes.

Other radionuclides remain in the raffinate almost entirely.

4 Supplying a batch of HLW and its evaporation

At this stage raffinates resulting from the first cycle of highly-enriched and low-enriched SNF are transferred to RW processing unit. There they are mixed with other liquid radioactive waste (LRW), mostly intermediate-level waste (ILW) (for example, raffinates resulting from the second cycle). This waste contains significant amount of stable macro components (ions of sodium, iron, sulfate, etc.) and insignificant amount (as compared to those contained in HLW) of radioactive ones. LRW batch is supplied within large tanks — up to 1,500 m^3. Thus, the initial LRW mixture is formed. This mixture is being sent to the evaporation unit located in the same shop of the plant. After being evaporated by factor 20, these evaporated liquid HLW are collected into storage reservoirs with a capacity of 285 m^3. The contents of one reservoir after relevant preparations is transferred for vitrification. This activity is conventionally called operation. Each operation is given its ID number. After being completed, the data on its main characteristics, chemical and radiochemical analysis are being registered in a particular act. This act provides the calculations of chemical and radiochemical compositions of vitrified HLW resulting from this portion.

^{3}H carry over is observed at the evaporation stage. In the first approximation, this process can be quantitively evaluated based the proportion of water distilled from the vapor phase, omitting that water molecules with different hydrogen isotopes have different evaporation capacities. As the initial LRW amount subject to evaporation typically amounts to some 6,400 m^3 resulting in some 275 m^3 of concentrate, the coefficient of tritium transition into solution at this stage is estimated to be equal to some 0.05. This evaporation stage also involves some additional iodine distillation.

5 HLW vitrification

At this stage, the supplied and evaporated HLW batch is fluxed with liquid additives (phosphoric acid, sodium nitrate solution) and processed in a direct electric heating furnace. Phosphate glass melt is poured into steel 200 l cans packaged into steel canisters. The canisters are hermetically sealed by remote argon welding and placed into a storage facility for vitrified waste [14]. This stage involves process redistribution of tritium, as well as iodine, cesium and technetium isotopes.

Dehydration and calcination of vitrified HLW suggests that complete distillation of the residual amount of water (and therefore ^{3}H) and I occurs in the surface layer of the electric furnace's operating zone. Thus, these radionuclides are not introduced into the vitrified waste being transferred to intermediate-level waste streams.

Calcination and glass melting also involves the carry-over of relatively volatile Cs (about 1.5 %) and solid phase (about 0.4 %). The solid phase carries along the equal amount of all radionuclides contained in VHLW. These indicated values are confirmed by systematic chemical and radiochemical analysis of the condensate collected from bubbler cooler installed at the outlet of electric furnace's gas duct.

Tc carry-over in the form of Tc_2O_7 at the stage of glass melting heavily depends on relevant process conditions (degree of element's oxidation, temperature, presence of a relatively cold calcinate crust at the surface of the melt and etc.) and may range from 45 % to 95 % [15]. Under conservative assumptions, the estimated amount of Tc transferred to the vitrified RW at this stage is equal to 50 % (as compared to the initial one).

Owning to limited solubility in glass, radioactive ^{107}Pd along with stable $^{106,108}Pd$ isotopes (as well as

other platinum group metals — Ru and Rh) are segregated into metal-like phase. This phase, owning to design feature of EP-500 furnace, are collected in its bottom part in purposely arranged pits. At this stage of research, it seems to be impossible to get a quantitative estimation of these element redistribution during furnace operation. In the first approximation, a value equal to 50% of ^{107}Pd amount discharged into VHLW cans can be set in calculations.

Based on the abovementioned assumptions relevant matrix of radionuclide redistribution factors for each reprocessing stage (Table 2) was compiled.

Table 2. Matrix of radionuclide redistribution factors depending on VVER-440 SNF reprocessing and HLW vitrification stage

Nuclide	Redistribution factor for a particular stage, %				Resulting factor, %
	2	3	4	5	
^{90}Sr	100	100	100	99.6	99.6
^{93}Zr	100	100	100	99.6	99.6
^{93m}Nb	100	100	100	99.6	99.6
^{126}Sn	100	100	100	99.6	99.6
^{129}I	0.5	100	0	0	0
^{137}Cs	100	100	100	98.5	98.5
^{226}Ra	100	100	100	99.6	99.6
^{228}Ra	100	100	100	99.6	99.6
^{227}Ac	100	100	100	99.6	99.6
^{228}Th	100	10	100	99.6	9.96
^{229}Th	100	10	100	99.6	9.96
^{230}Th	100	10	100	99.6	9.96
^{231}Pa	100	100	100	99.6	99.6
^{233}U	100	0.1	100	99.6	0.1
^{234}U	100	0.1	100	99.6	0.1
^{235}U	100	0.1	100	99.6	0.1
^{236}U	100	0.1	100	99.6	0.1
^{238}U	100	0.1	100	99.6	0.1
^{238}Pu	100	0.5	100	99.6	0.5
^{239}Pu	100	0.5	100	99.6	0.5
^{240}Pu	100	0.5	100	99.6	0.5
^{242}Pu	100	0.5	100	99.6	0.5
^{241}Am	100	100	100	99.6	99.6
^{243}Am	100	100	100	99.6	99.6
^{245}Cm	100	100	100	99.6	99.6
^{246}Cm	100	100	100	99.6	99.6

It should be noted that this problem has never been investigated before. For this reason, all the abovementioned distribution factor values should be considered as preliminary requiring further elaboration.

The use of relevant factors enables to get a more precise VHLW radionuclide composition with due account of the process factors involved. It's worth mentioning that vitrification results in a weight increase ratio ranging from 1.5 to 1.8.

Results and conclusions

The algorithm developed to calculated the radionuclide composition of VHLW should provide a basis for the data base. This data base compiling achieved records on SNF reprocessing at RT-1 plant, performed measurement and other available information should enable to calculate RW compositions. Further on, the results obtained should be verified at least for the following experimental data:

- Measurements of package temperature;
- Radiochemical analysis of radionuclide composition for a given sample of know origin;
- Spectrometric investigations of RW packages.

At the present stage of research, due to a big number of assumptions and simplifications applied, we were able to obtain only some approximate data on vitrified HLW characteristics. At least 3 ways enabling to decrease the resulting errors can be considered:

- Stepwise refinement of parameters at all calculation stages (taking into account the ratio between different SNF types and their compositions; taking into account other types of waste subject to vitrification; refinement of radionuclide redistribution factors by stages, etc.);
- Verification of calculated results based on the available analytical control data on HLW subjected to vitrification (the list of radionuclides is short and is mostly composed of readily detectable gamma-emitting radionuclides);
- Verification of calculation results based on new VHLW analysis data using modern analytical methods and software.

The abovementioned efforts suggest that certain labor and financial resources are to be applied. Moreover, the latter one requires maximum efforts and resources (a full-fledged research laboratory equipped with hot cells and costly equipment is required, issues associated with real VHLW canister measurements should be addressed, sampling of glass from the cans is required and etc.). However, after a considerable time (not less than 5 years) this approach will enable to obtain a comprehensive set of information on the investigated matter. It should be noted that radionuclide vector method is considered to be quite promising one in

dealing with the analysis of batch VHLW composition [16].

At the same time, in the nearest future the proposed calculational method after relevant refinements are made will enable to obtain quite reliable information on a large massive of VHLW packages that can be used in the development DDF safety case, as well as to amend some design solutions (if deemed necessary).

It should be noted that the list of relevant radionuclides has been modified based on the algorithm developed. The evaluation of SNF reprocessing flow chart has shown that ^{129}I being considered relevant for long-term safety perspective practically does not get into VHLW composition. However, on the other hand, the list has been extended by introducing ^{137}Cs and ^{90}Sr — these are necessary to be accounted for to determine the heat output of RW packages. The impact of different SNF types on the final VHLW radionuclide content should be evaluated at the next stage.

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Information about the authors

Remizov Mikhail Borisovich, Ph.D in Science, Head of Radioactive Waste Management Laboratory, Central Plant Laboratory, FSUE "PA "Mayak", (18, Ermolaeva st., Ozyorsk, 456780, Russia), e-mail: mirem@yandex.ru.

Kozlov Pavel Vasilevich, head of group Central Plant Laboratory, FSUE "PA "Mayak" (18, Ermolaeva st., Ozyorsk, 456780, Russia), e-mail kozlov_pavel@inbox.ru.

Dementyeva Irina Ivanovna, engineer-technologist-chemist-reseacher of Central Plant Laboratory, FSUE "PA "Mayak", (18, Ermolaeva st., Ozyorsk, 456780, Russia), e-mail: iidementeva@yandex.ru.

Borisenko Vladimir Pavlovich, head of group Central Plant Laboratory, FSUE "PA Mayak", (18, Ermolaeva st., Ozyorsk, 456780, Russia).

Blokhin Pavel Anatol'evich, junior researcher. Nuclear Safety Institute of Russian Academy of Sciences (52, Bolshaya Tul`skaya st., Moscow, 115191, Russia), e-mail: blokhin@ibrae.ac.ru.

Samoylov Andrey Anatol'evich, senior researcher, Nuclear Safety Institute of Russian Academy of Sciences (52, Bolshaya Tul`skaya st., Moscow, 115191, Russia), e-mail: samoylov@ibrae.ac.ru.

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