ESTIMATION OF RADIONUCLIDE COMPOSITIONS OF ACCUMULATED HIGH LEVEL RADIOACTIVE WASTE: FEATURES AND SOLUTION METHOD

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1. INTRODUCTION

The problem of determining the radionuclide compositions of spent nuclear fuel (SNF) and RW emerges in all the stages of their treatment lifecycle. However, knowledge of different radionuclide groups is vital in different stages. For example, at the treatment stage, when the personnel has to be immediately close to the sources of ionizing radiation, especially significant become the radionuclides, which produce high-energy gamma-quants (Cs-137, Co-60, Nb-94 et al.) and neutrons (trans uranium elements) during the decay. Knowledge of the content of the latter is also important for estimation of criticality and maintaining the nuclear security during storage and disposal. Determining the content of such radionuclides for most types of RW is not as complex and is performed by methods of nondestructive testing. However, with long-term safety estimation (LTSE) of RW or SNF disposal, quickly migrating radionuclides (C-14, Cl-36, Se-79, Tc-99, I-129 et al.) are significant, content measuring of which can be performed using the destructive testing.

When analysing the radionuclide composition of RW produced during the SNF processing, objective difficulties in determining the radionuclides significant for LTSE. The nomenclature of such waste is determined by ratio of different types of RW processed (power (VVER-440, BN-600), research et al.), technological processing peculiarities and exposition time [1]. Considering these factors with the accuracy level required for the tasks of disposal safety estimation both during exploitation period and after the closure of deep geological waste repository (GWR), requires developing the special attitudes, methods et al., including the performing the interconsistent measures of wide spectrum first of all radiation characteristics and calculation research in order to precise defining and predicting the RW activity on the whole and uncertainty decrease for specific RW containers, physical and chemical characteristics of glass, planning the transport and logistical operations with HLRW containers.

There were earlier trials of radionuclide composition of RW in similar context. Thus, within the scope for the assessment work, initial radionuclide composition of vitrified HLRW of FGUE Mayak was formed basing on the expert analysis of available measure result and data on glass composition of analogous foreign manufactures.

To confirm the modelling results, there emerges the necessity for verification and validation of SNF and HRW nuclide composition [2] based on the comparison of calculation results benchmark experiments. Groups of international experts are working on the issues of selection and compilation of integral experiments on determining the nuclear physical RW characteristics and a number of results of their activity are presented in papers [3-5]. These publications describe experiments on determining the RW radiation characteristics and accumulation of decay products and actinides, including SNF for Russian VVER-440 reactors [2].

This report pays special attention to the next two aspects: correctness of RW composition computational research, being a base for the further analysis of composition and characteristics of vitrified HLRW (VHLWR) and prioritization by significance for LTSE radionuclides probably composing the VHLRW.

2. PROGRAM VERIFICATION FOR CALCULATION OF VHRW COMPOSITIONS

Main amount of the SNF processed in the RT-1 factory is VVER-440 fuel, so the VHRW radionuclide composition will be mostly determined by its characteristics. To test the correctness of the RW composition computational research, it is necessary to perform the series of verification research. One of this is the “МНТЦ-2670р” project [6, 7].

Within the project scope the research of isotope composition of a typical working assembly TVS135 from the 4th block of Novovoronezh NPP having worked for four fuel campaigns were performed. The fuel assembly researched was exploited in the emergency protection of the VVER-440 reactor for 1109 effective days; cooling time (from the start of irradiation to the radiochemical analysis) was 12.34 years (4518 days).

For each considered specimen, neutron flux and burn-up fraction were determined in two ways:

— By measured fuel isotope composition before and after the irradiation using computational and analytical correlations (heavy atoms method - HAM);
— By measured ratio of burn-up monitor fission product atoms to the number of heavy atoms in the dissolved fuel specimen (method of fission products accumulation - FPA).

To determine the burn-up using the FPA method, products of heavy nuclei fission 137Cs, 148Nd, and summary accumulation of 145Nd and 146Nd were used as monitors. In addition, there were obtained more accurate data on the initial isotope 235U enrichment for all the specimens. Estimated neutron fluxes and energy spectra for emergency protection of VVER-440 were obtained using the code using the Monte Carlo method using the ENDF/B-VII.0 neutron data library. The calculations were performed in the 2-D geometry with geometry parameters according to the VVER-440 project. As an example, Fig. 1 shows calculated neutron flux distribution for the fuel element compared to the experimental data. One can see that the biggest result deviation of the calculation from the experiment achieves 18% in the lower part of the fuel element. At the same time, the error of neutron flux distribution by height reaches the value of about 24%.

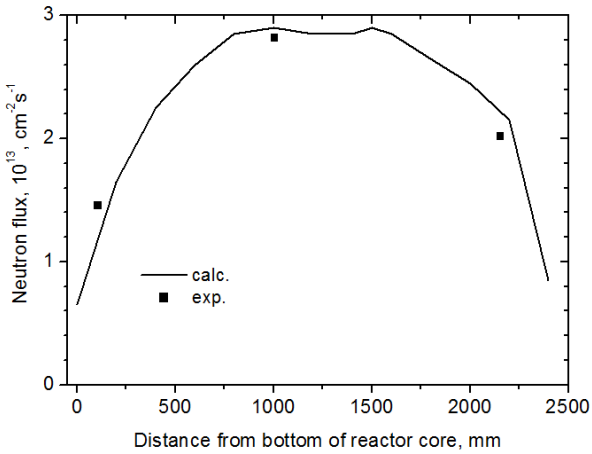

FIG. 1. Neutron flux destiny distribution by the height of fuel element 65.

For the abovementioned SNF specimens, actinide and fission products mass outputs were measured with different fuel burn-up ratios from 22.6 to 47.5 GW*day/tUO2 depending on the considered specimen. Analogous calculated data were obtained using TRACT computation complex in the burn-up areas 20 to 48 GW*day/tUO2. The calculation took into consideration the neuron fluxes levels for the specimen positions in

the FE taken from abovementioned emergency protection neutron characteristics calculations. Mass outputs for 16 actinides and 32 fission products were defined, such as:

— For actinides: U-234, 235, 236, 238, Np-237, Pu-238, 239, 240, 241, 242, Am-241, 242m, 243, Cm-244, 245, 244;
— For fission products: Mo-95, Tc-99, Ru-101, Pd-105, 108, Ag-109, Cs-133, 134, 135, 137, Ce-140, 142, 144, Nd-142, 143, 144, 145, 146, 148, 150, Sm-147, 148, 149, 150, 151, 152, 154, Eu-151, 153, 154, 155, Gd-155.

Figures 2–5 show experimental and calculation data on the actinide content: isotopes U-235, -236, and -238, isotopes Pu-239, -241, -240, and -242, isotopes Am-241 and -243 in the selected SNF specimens. For the U-235 and -236 isotopes, measured and calculation results agree up to 10%. For U-238, practically on the whole considered area one can observe exceeding of the calculation data up to 10% which may be caused by understatement of U-238 neutron radiation capture cross section.

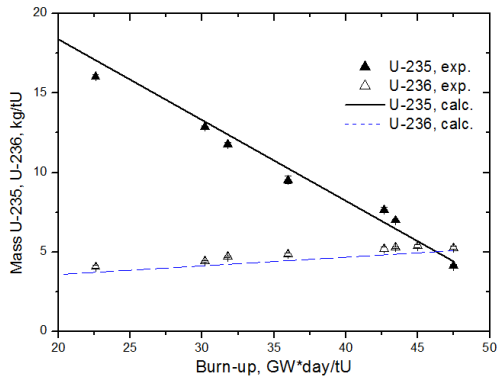

FIG. 2. Dependency of ^{235}U and ^{236}U content on fuel burn-up.

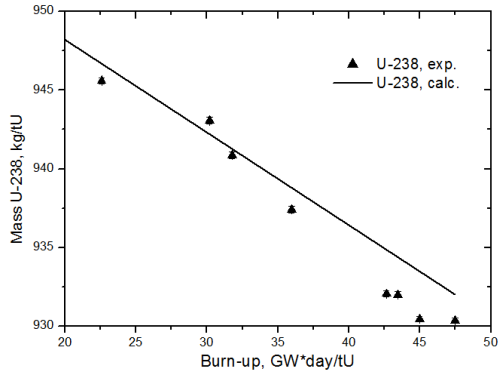

FIG. 3. Dependency of ^{238}U content on fuel burn-up.

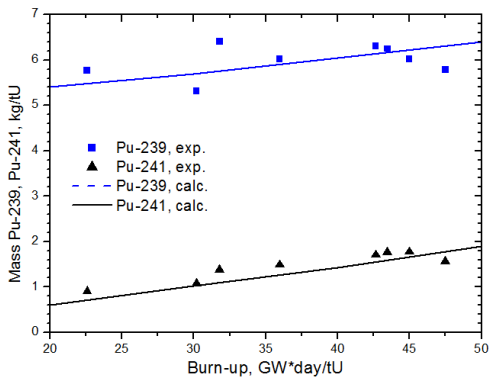

FIG. 4. Dependency of ^{239}Pu and ^{241}Pu accumulation on fuel burn-up.

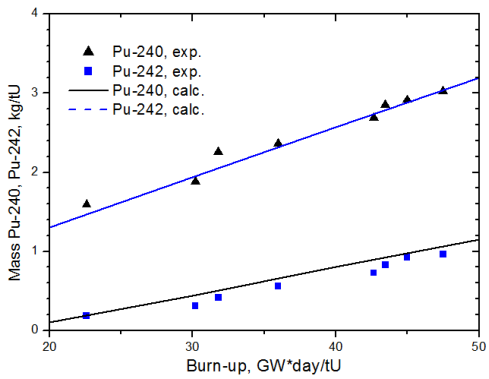

FIG. 5. Dependency of ^{240}Pu and ^{242}Pu accumulation on fuel burn-up.

For most actinides, deviation of calculated and measured data lies between 10%. For U-234, accumulation of which is ~100 g/tU, the deviation reaches about +24%. For Np-237 and Cm-244, there exist significant deviations up to -60%. It should be noted that a cause of such deviations might be also errors in experimental results. As notes, for the specimen 57, content of Np-237 is determined with error of 39%, and of Cm-244 – with error 33%. Let us note overall, that the output of most products is described with an accuracy of ~20%.

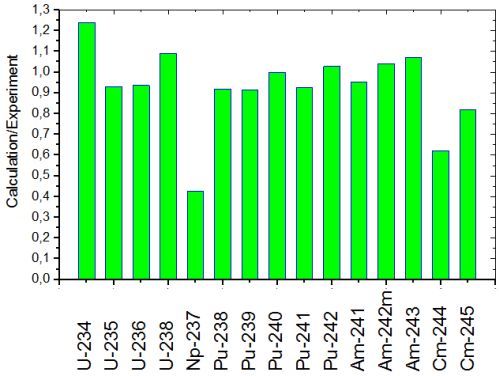

FIG. 6. Calculation/experiment ratio for accumulation of actinides in the specimen 57 with fuel burn-up level 36 GW day/tU.

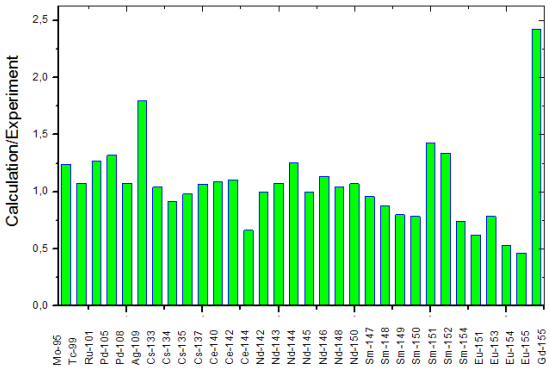

FIG. 7. Calculation/experiment ratio for accumulation of fission products in the specimen 57 with fuel burn-up level 36 GW day/tU.

3. FORMING THE LIST OF RADIONUCLIDES SIGNIFICANT FOR LONG-TERM SAFETY

In order to plan the measures and experiments with VHLRW specimens, the research of radionuclide significance for RW disposal safety (long-term, in particular) estimation is performed.

At the stage of preliminary estimation of the list of significant radionuclides, one can neglect the redistribution of radionuclides in the technological production flows and RW. With such approximation, the composition of RW will correspond the initial composition of the RW processed (with a correction to the ratio between the quantities of the SNF processed and obtained RW). Radionuclide behaviour in the near zone of GWR is rather difficult to predict. I such a situation, let us use a conservative assumption that after the expiration of defensive properties of engineered safety barriers, the RW matrix and other barriers lose their defensive properties completely. In that case, their dissolution process will determine the content of radionuclides in underground waters, and maximal possible concentration will be conservatively correspondent to the solubility limit of the radionuclide compounds. The ratio of radionuclide transition through geological environment is determined by combination of migration parameters of the corresponding chemical element, characteristics of the geological environment and underground waters. Let us also act on premise that the main parameter determining the radiation effect on a human will be determined by dose coefficients of the corresponding radionuclide.

Thus, to determine the radiologically significant nuclides we are going to use the following parameters:

- Radionuclide activity in the initial RW composition;
- Half-life;
- Water solubility limit of chemical compounds containing considered radionuclides in the RW composition;
- Parameters determining the radionuclide migration ratio (in case of a solid porous environment – a coefficient f phase distribution in the system “RW – underground waters – geological materials” and the characteristics of a geological environment);
- Dose coefficients for drinking water consumption (as a main scenario of potential exposure of population, we imply using the contaminated underground waters for drinking).

Determining the radiologically significant nuclides, we shall perform stage wise exclusion of radionuclides from the list basing on the above-listed parameters.

At the preparation stage, one should obtain a full list and activity of radionuclides containing in RW (list «0»). In the first stage of radionuclide selection, change of radionuclide composition due to radioactive fission and the characteristics of the object as well. For this aim, a recalculation of RW composition at the time moment T0 is performed, which is determined as a time of performing the insulation from the environment by engineering barriers. If the specific activity of some radionuclide at the specified time moment would be less than the RW criterion, we can obviously eliminate it from further consideration. Thus, we get list «1». At that,

one should take in account the accumulation of daughter radionuclides, activity of which by time may grow, so a calculation of radionuclide composition dynamics for selected radionuclides for a significant time (more than 1 million years). In case, if the content of some daughter radionuclide exceeds the RW criteria, parent and daughter radionuclides are included to the further analysis i.e. added to the list «1».

Second stage of nuclide elimination – taking into account the solubility of chemical compounds, containing radionuclides, in water considering physical and chemical properties of RW and underground waters in the supposed GWR place. After descending arraigning of radionuclides from list «1» by product of activity, solubility limit and dose coefficient with water consumption by organism, let us discard the last N nuclides, satisfying the next ratio:

$$V_{H_2O} \sum_i \varepsilon_i \cdot C_i \prec D_{\lim} \; mSv / year \qquad (1)$$

where,

V_{H2O} – average annual drinking water volume, consumed by human liter/year

ε_i – dose coefficient with consumption of its radionuclide into the organism of adult people with water;

C_i – specific activity of its radionuclide in underground waters, corresponding to the solubility limit

D_{im} – boundary value of dose rate for population.

In the other words, the radionuclides eliminated from further consideration have a dose rate specified by their presence in drinking water, which even with the maximal solubility will not exceed Dim. This way, list «2» is formed.

At the third stage, time of radionuclides to reach the human i.e. migration properties of radionuclides and characteristics of geological environment and underground waters. This stage is most complex considering the present uncertainties and depends on the scenario of radioactive contamination spreading through the geological environment. In case, if the considered GWR is in the solid porous environment, consideration of radionuclide migration parameters may be performed this way:

- Determining the biosphere reaching time by the front of neutral tracer agent;
- Determining for each radionuclide a delay of the radionuclide migration front compared to the neutral tracer agent;
- Performing the radionuclide activity calculation in the underground waters, corresponding to the solubility limit per time of potential arrival of the radionuclide to the human.

Arrival time of contaminated underground water of the control point is determined basing on the equation describing the delay factor:

$$T_i = T_0 \cdot (1 + \frac{\rho}{\varphi} \cdot K_d^i) \qquad (2)$$

where:

ρ – density of the most penetrable ground in the radionuclide migration way;

φ – porousness of the most penetrable grounds in the radionuclide migration way;

K_d^i – distribution coefficient of the its radionuclide for the most penetrable grounds in the radionuclide migration way. T_0 – time of arrival for neutral tracer to the potential water intake point.

In case of placing the object on the fractured geological environment, it is necessary to perform conservative migration calculations for list “2” radionuclides, and then perform recalculation of radionuclide activity.

To consider the migration mechanism for radionuclide being in the same radioactive decay chain, a number of simplifications is used. In case, if a half-life of the daughter radionuclide is relatively small, one can neglect its independent migration and, therefore, migration parameters of parent radionuclide are crucial. In opposite case, when the parent radionuclide is relatively small migration parameters of the daughter nuclide are crucial. For comparable radionuclide half-lives, solution may be simplified by using the next conservative approximation: the radionuclide moves by single front (without regard of different ratios of forming sources), at that, as a parameter determining the migration ratio (distribution coefficient for a geological material), one chooses the one which corresponds the fastest migration.

After decreasing arrangement of list «2» radionuclides by product of activity (moment TI), solubility limit, and dose coefficient of human consumption of the radionuclide with water, let us disregard the last N nuclides, for which equation (1) is true. This way, the list «3» is formed.

At the next stage, one can consider the RW processing technologies, considering redistribution of nuclides during their conditioning. The final radionuclide list may be used hereinafter to form the list of checked radionuclides and development of admissibility criteria. Table 1, as an example of approbation of the suggested method, shows an example of stage wise evolution of the list of significant radionuclides. The initial RW composition is obtained basing on the radionuclide composition calculation of RW of VVER-440.

TABLE 1. FORMING OF THE LIST OF RADIOLOGICALLY SIGNIFICANT RADIONUCLIDES

Stage	Stage 1 «Decay»	Stage 2 «Solubility + dose»	Stage 3 «Migration +dose» *(example)*	Final list, considering parent nuclei *(example)*
Quantity of radionuclides «Before»/«After»	<100/24+2	26/23	*23/10*	*10/14*
Radionuclide list	Zr-93, Nb-93m, Tc-99, Pd-107, Sn-126, I-129, Ra-226, **228** Ac-227, Th-**228**, 229,230, Pa-231, U-233, 234, 235, 236, 238, Np-237, Pu-239, 240, 242, Am-241, 243, Cm-245, 246	Zr-93, Nb-93m, Sn-126, I-129, Ra-226, 228, Ac-227 Th-228, 229, 230, Pa-231, U-233, 234, 236, 236, 238, Pu-239, 240, 242 Am-241, 243, Cm-245, 246	*I-129,* *Ra-226, 228* *Ac-227,* *Th-228, 230,* *Pa-231,* *U-234, 235, 238*	*I-129,* *Ra-226, 228,* *Ac-227* *Th-228, 230,* *Pa-231,* *U-234, 235,* ***236,*** *238* *Pu-**238, 239, 240**.*

4. CONCLUSION

To estimate correctness of estimation results of VVER-440 reactor SNF, we performed the verification of a specialized code based on the international project experimental data. The comparison of the obtained calculated data demonstrated overall adequacy of the considered physical processes description. Similar comparison with calculation results, obtained by other programs, demonstrated stable correlation with them and experimental data.

In order to plan the measurements of long-term radionuclides content in the vitrified RW, we suggest an approach allowing to reduce the common radionuclide list (more than 100) down to 14, most significant. The approach offered allows considering their main characteristics influencing the population and environment: half-life, solubility, migration parameters, and dose coefficients.

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