Computational study of high nuclear fuel burnup radiotoxicity for WWER-1000 reactor

J A Korchova, N V Harbachova and N D Kuzmina

Joint Institute for Power and Nuclear Research-Sosny, National Academy of Sciences of Belarus, Minsk, Belarus

Abstract. Radiotoxicity behavior of WWER-1000 reactor high burnup spent nuclear fuel is discussed by the authors. Radionuclides concentrations as function of nuclear fuel burn up to 68 MWt day/kg for 4.81 % initial enrichment have been calculated on the base of approximation relations of Regulation RB-093-14 (Moskow, 2014). Time dependences of fission products and actinides activities and radiotoxicity behavior were derived at different stages: in case of spent nuclear fuel is disposed in intermediate storage facility and when it will transmit in geological repository.

1. Introduction
At present Belarusian Nuclear Power Plant (NPP) is constructed in Grodno region. It consists of two Units of the WWER-1200 reactors by the AES-2006 Project.

Strategy of Spent Nuclear Fuel (SNF) Management of Belarussian NPP is developed by National Government. The objective is to provide safe, economical and ecology acceptable SNF management option.

Fission products activities as well as actinides radiotoxicity indices represent basic characteristics that determine spent nuclear fuel hazard to the environment during its storage in the intermediate SNF storage facility and in a geological repository for many millennia. In the papers [1-3], a radiation migration equivalence principle for spent nuclear fuel hazard assessment in relation with natural uranium one was introduced into consideration.

The objective of this paper is computational study of SNF radiotoxicity in relation with its burnup up to 68 MWt day/kg for the WWER-1000 reactor.

Radionuclides concentrations as function of nuclear fuel burnup for different kinds of initial fuel enrichments have been calculated on the base of approximation relations of Regulation RB-093-14 [4].

Fission products and actinides activities as well as radiotoxicity indices have been calculated. The results of radiotoxicity behaviors for the SNF at 68 MWt day/kg burnup (4.81 % of the $^{235}$U – initial enrichment) are represent in this paper.

Computational tools in the MS Excels environment for and the DB “The WWER1000 Radiotoxicity Indices” were developed by the authors.

2. Radiotoxicity indices computation and analyses
Radiotoxicity indices of SNF describe an environmental hazard measure of the SNF in relation to water and air contaminations ways at different stages of SNF management.
Definitions
Denote a DOA, (Bq · m⁻³) is average annual volumetric activity of the RNᵢ – radionuclide in the air as a permissible value for human intake of contaminated air inhalation in accordance with [5]. The radiotoxicity index of the RNᵢ – radionuclide, RTᵢ.air, is an air volume, which is necessary to reduce its specific activity to a safe level by dilution (in case if it’s determined by an air contamination way) in accordance with the formula:

\[
RTᵢ.air = \frac{Aᵢ}{DOAᵢ}
\]

where \(Aᵢ\), Bq · m⁻³, is specific activity of RNᵢ-radionuclide rating at 1 t of SNF.

Denote an intervention level ILᵢ (Bq · kg⁻¹) is average annual activity of the RNᵢ – radionuclide in a ground water as permissible value for human intake of contaminated water in accordance with [5]. The radiotoxicity index RTᵢ.water of the RNᵢ – radionuclide is a water mass needed to reduce its specific activity dilution to a safe level (in case if it’s determined by a water contamination way) in accordance with the formula:

\[
RTᵢ.water = \frac{Aᵢ}{ILᵢ}
\]

where \(Aᵢ\) ( Bq · t⁻¹) is specific activity of RNᵢ-radionuclide rating at 1 t of SNF.

Total air radiotoxicity index \(RTₐᵢr\) of 1 t of SNF is a sum of each of RNᵢ-air – radiotoxicity indices in according determined by the formula:

\[
RTₐᵢr = \sum RTᵢ.air
\]

Total water radiotoxicity index \(RTₜᵢw\) of 1 t of SNF is a sum of each of RNᵢ-water – radiotoxicity indices in according determined by the formula:

\[
RTₜᵢw = \sum RTᵢ.water
\]

3. The results of the SNF radiotoxicity calculations for the WWER-1000 reactor
Radionuclides concentrations as function of nuclear fuel burnup for different kinds of WWER-1000 initial fuel enrichments reactor have been calculated on the base of approximation relations, which were derived in Regulation RB-093-14 [4].

Computational tools in the MS Excels environment have been developed by the authors. The results of the SNF radiotoxicity indices calculations are represented at the DB “The WWER1000 Radiotoxicity indices”.

Time dependence of the total SNF activities for the SNF at 68 MWt day/kg burnup at different periods of SNF storage are represented in the table 1. The results indicates that total SNF activity after first several hundreds years is decreased smoothly and reaches about 5 TBq · t⁻¹ after 10 000 years exposure at the intermediate storage facility.
Table 1. Specific activity of the SNF for 68 MWt day/kg burnup (4.81 % of initial enrichment).

| Exposure time, years | Specific SNF activities, Bq/t |
|----------------------|-------------------------------|
| 1.99·10^3           | 2.12·10^{17}                 |
| 5.20·10^4           | 7.78·10^{12}                 |
| 1.02·10^5           | 2.52·10^{12}                 |
| 1.52·10^5           | 1.36·10^{12}                 |
| 2.02·10^5           | 1.01·10^{12}                 |
| 2.52·10^5           | 8.71·10^{11}                 |
| 2.92·10^5           | 8.02·10^{11}                 |

The SNF fission products and actinides activities for the 4.81 % of initial enrichment fuel at the 68 MWt·day/kg burnup have been calculated as function of SNF storage duration. Then the radiotoxicity indices were calculated for this kind of SNF with the use of 1–4 formulas.

Similar calculations of radiotoxicity indices were carried out for SNF of 4.4% of initial enrichment for 40 MWt·day/kg of fuel burnup by the authors in the paper [5-9].

Let’s consider the radiotoxicity behavior at intermediate SNF storage stage. The results of time dependences of the SNF radiotoxicity indices for long-lived fission products and actinides, which are accumulated in the SNF of the 68 MWt·day/kg burnup (4.81 % of initial enrichment), are represent in figure 1–4.

At the initial storage stage (after exposure in the fuel storage pool) lasting till 100 years the SNF radiotoxicity is determined by main fission products (FPs) such as ^{106}Ru, ^{144}Ce, ^{90}Sr, ^{137}Cs, ^{134}Cs, ^{147}Pm, ^{154}Eu (figure 1–2). Total FPs air radiotoxicity index $RT_{FP\;air}$ is equals to $3.4\cdot10^{15}$ m$^3$·t$^{-1}$ and then it is reduced to $3.0\cdot10^{14}$ m$^3$·t$^{-1}$. The FPs water radiotoxicity index $RT_{FP\;w}$ is equals to $5.4\cdot10^{15}$ kg·t$^{-1}$ and then decreased to $6.7\cdot10^{14}$ kg·t$^{-1}$.

Figure 1. Spent nuclear fuel storage stage: radiotoxicity of fission products by air and by water.
Main radiotoxic actinides are: $^{238}\text{Pu}$, $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$, $^{242}\text{Cm}$, $^{244}\text{Cm}$. Total air radiotoxicity index $RT_{\text{act\_air}}$ of actinides is decreased from $4,9\cdot10^{18}$ to $2,35\cdot10^{17} \text{ m}^3\cdot\text{t}^{-1}$. Total water radiotoxicity index $RT_{\text{act\_w}}$ of actinides is decreased from $2,4\cdot10^{17}$ to $1,4\cdot10^{16} \text{ kg}\cdot\text{t}^{-1}$. It should be mentioned that for $^{241}\text{Am}$ there are exception from common behavior: its activity and toxicity indices are increased on the period under consideration (figure 2).

Let’s conceder the radiotoxicity behavior at repository stage of the SNF management. At the long-lived of the SNF management stage lasting from 1000 years till 300 thousands years there are following main toxic radionuclides: $^{99}\text{Tc}$, $^{93}\text{Zr}$, $^{129}\text{I}$, $^{135}\text{Cs}$, $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{242}\text{Pu}$ и $^{243}\text{Am}$ [10]. Total FPs air radiotoxicity index $RT_{\text{FPs\_air}}$ is decreased to $1,7\cdot10^{11} \text{ m}^3\cdot\text{t}^{-1}$ and their water radiotoxicity index is decreased to $2,5\cdot10^{10} \text{ kg}\cdot\text{t}^{-1}$. Total air radiotoxicity index $RT_{\text{act\_air}}$ of actinides is decreased to $7,8\cdot10^{14} \text{ (m}^3\text{t}^{-1})$ and their water radiotoxicity index is decreased to $3,6\cdot10^{12} \text{ kg}\cdot\text{t}^{-1}$. The results are presented on figure 3–4.

![Figure 2](image-url)  
**Figure 2.** Spent nuclear fuel storage stage: radiotoxicity of actinides by air and by water.

![Figure 3](image-url)  
**Figure 3.** Spent nuclear fuel repository stage: radiotoxicity of fission products by air and by water.
4. Conclusion
Computational tool in the MS Excels environment was developed by the authors. The results of calculations of the SNF radiotoxicity indices for high burnup SNF (68 MWt·day/kg; 4.81% of $^{235}\text{U}$ – initial enrichment) of the WWER-1000 reactor are presented at the DB "The WWER-1000 Radiotoxicity indices".

The main radiotoxic FPs and actinides at initial stage of the SNF management are: $^{90}\text{Sr}$, $^{106}\text{Ru}$, $^{137}\text{Cs}$, $^{134}\text{Cs}$, $^{144}\text{Ce}$, $^{147}\text{Pm}$, $^{154}\text{Eu}$, $^{238}\text{Pu}$, $^{240}\text{Pu}$, $^{244}\text{Cm}$, $^{241}\text{Am}$, $^{99}\text{Tc}$, $^{129}\text{I}$, $^{135}\text{Cs}$, $^{239}\text{Pu}$, $^{242}\text{Pu}$, $^{243}\text{Am}$. Main contribution in the air and water radiotoxicity indices are the actinides toxicity and equal to $2.35 \times 10^{17} \text{m}^3\text{t}^{-1}$ and to $1.4 \times 10^{16} \text{kg}\text{t}^{-1}$, correspondingly.

The results are of value for decision making on ecology acceptable SNF management option for the Belarusian NPP.

References
[1] Lopatkin A, Velichkin V, Nakipelov B and Poluektov P 2002 Nuclear energy 92(4) 308 -317
[2] Alexakhin R, Spirin E, Solomatin V and Spiridonov S 2016 Nuclear energy 120(6) 312-318
[3] Bergelson B, Gerassimov A, Zaritskaya T, Kiselev G and Myrtsymova L 2000 Nuclear energy 89(3) 215-220
[4] Nuclear and Radiation safety 2014 RB-093-14 119 p 51
[5] Radiation impact assessment criterias 2012 The Hygienic standard of the Health Ministry of Belarus 213 p 230
[6] Harbachova N, Kulich N, Korchova J 2016 Calculated studies of radiotoxicity and specific activity of spent nuclear VI Conferences Nuclear technologies of the XXI centures (Belarus: Minsk) p 79
[7] Beresneva N, Gorbachev N, Kulich N and Skurat V 2008 Radiotoxicity of spent nuclear fuel from a VVER and RBMK reactor during long-term storage Preprint gr-qc/34
[8] Korbut T, Rudak E and Petrovskii A 2018 Statistical description of the emitters-nuclei ensemble decay within the framework of the sub-Poisson distribution Bulletin of the Russian Academy of Sciences: Physics 10 (82) 80–86
[9] Petrovskii A, Korbut T and Rudak E 2018 Analytical ways of determining the activity of fission products in the core of a VVER-1200 reactor and their applications Bulletin of the Russian Academy of Sciences: Physics 10 (82) 1335-1341
[10] A basic toxicity classification of radionuclides 1963 Technical reports series 15 (Vienna:IAEA) p 39