THE IMPACT OF NEUTRON FLUENCE ON THE NUCLIDE VECTOR OF SPENT NUCLEAR FUEL FOR THE GEOLOGICAL DISPOSAL PURPOSES

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Abstract

The analysis of impact of different neutron fluence on the isotopic composition of spent nuclear fuel was performed. In this paper, two burnup levels of VVER-440 fuel assembly with an average enrichment 4.87 wt % of ^{235}U are chosen to examine the changes in the isotopic composition of spent nuclear fuel. Subsequently, the impact of individual concentration differences on release rates from the engineered barrier system (EBS) to the surrounding host rock is evaluated. The results on release rates were obtained from the radionuclide migration simulations which were performed for hypothetical geological disposal system. The calculation of release rates of individual radionuclides was carried out for one disposal container using the simulation software GoldSim. Based on the results, changes in concentration affect mainly release rates of radionuclides which do not exceed their solubility limits.

1. Introduction

Spent fuel assemblies discharged from the VVER nuclear reactors in Slovakia are cooled several years (3-5 years at the Bohunice site, 6-7 years at the Mochovce site) in storage basins in the vicinity of the reactor [1]. After this period, the spent fuel is transported from the nuclear power plant to the interim spent nuclear fuel storage facility in Jaslovské Bohunice [2]. The spent fuel assemblies are in this facility stored in the pools filled with water (so called wet type of storage) [1].

Nowadays, as for the final stage of the spent fuel management, a direct disposal of spent nuclear fuel in a deep geological repository within the country after a certain period of interim storage is the preferred option.

A deep geological repository as an alternative of spent fuel management is expected to provide sufficient protection of human health and the environment for at least several thousand years. To ensure protection of human health and the environment, it is necessary to design a complex system of protective measures as well as to perform a comprehensive safety analysis in every period of the repository lifetime.

The spent fuel characteristics are a necessary initial condition for the waste disposal repository design. It is considered that in the geological repository there will be disposed various spent fuel assemblies from the operation of Slovak nuclear power plants and also radioactive wastes which are not suitable for a near-surface type of repository.

2. Isotopic composition of VVER-440 spent fuel assembly

Changes in the isotopic composition of spent nuclear fuel were calculated using the computational code MCNP5. Calculations were performed for the radial profiled VVER-440 fuel assembly with the initial average enrichment 4.87 wt % of ^{235}U. In the 2nd generation fuel

assemblies, there are used fuel pins containing gadolinium oxide (3.35 wt % of the fuel pin) as a burnable absorber integrated into the fuel pellets [3]. Fuel assemblies with gadolinium support the safe handling of the fresh fuel with higher initial enrichment. The use of gadolinium pins also allows reducing the concentration of H_3BO_3 at a campaign with a higher enrichment fuel loading. In comparison with the 1st generation fuel, fuel content in the 2nd generation fuel assemblies was increased and water/fuel ratio was optimized to improve the fuel efficiency [3]. Hafnium content in the cladding material was also lowered (less than 0.01%) in order to reduce parasitic neutron absorption [3].

Within this analysis, two different nuclide vectors of spent nuclear fuel corresponding to two burnup levels were investigated. Calculations were performed for the fuel assembly with the average enrichment of 4.87 wt % of ^{235}U whereby the differences in the isotopic composition were evaluated for two burnup cases:

- 43 MWd/kgU,
- 60 MWd/kgU.

. In general, the time-dependent change of nuclide concentration during depletion calculations is influenced by fission, neutron transmutation and radioactive decay. As the fuel assembly is a heterogeneous system, concentrations were calculated for the spent fuel which consists of fuel pins with different enrichments (4.95 wt % ^{235}U, 4.6 wt % ^{235}U and 4.4 wt % ^{235}U + 3.35 wt % Gd_2O_3) and the structural material (claddings and spacing grids made from the zirconium alloys E-110 and E-125). Chemical composition of the fuel materials and zirconium alloy was defined including the impurities to obtain the results on the concentration of possible activation products.

Calculated isotopic compositions of spent nuclear fuel (two burnup cases) were used as input parameters for modelling the radionuclide migration through the near field of geological disposal system (two radionuclide migration cases).

3. Near field modelling

Spent fuel is a complex and heterogeneous system and therefore was within the model conceptually divided into the structural material, UO_2 matrix and instant release fraction (IRF). IRF is a fraction of inventory which is after water contact released rapidly, in the term of long-term safety instantaneously [4]. According the international safety assessments and certain assumptions as high solubility limits, relatively long half-lives, poor sorption on bentonite buffer, significant IRF values, 36 radionuclides were identified as relevant for the safety analysis. Inventory of ^{14}C was divided between the structural material and UO_2 matrix. It is considered that ^{14}C originated from the structural material is presented in organic compounds and ^{14}C originated from UO_2 matrix is a part of inorganic compounds [5].

The geological repository design is based on the principle of multi-barrier concept - combination of engineering barriers and a suitable geological formation. Engineering barriers are designed to provide reasonable protection of environment against the radiation for a long period of time (thousands to millions of years). One of the barriers is the very form of spent fuel which dissolution rate in the contact with the groundwater is very low. Another barrier is represented by disposal container which parameters and physical properties are based on [6]. The disposal capacity of one container is 7 fuel assemblies [6].

Disposal container is surrounded by a bentonite buffer with the wall thickness of 300 mm. It provides a self-sealing, low permeable barrier because of its swelling potential.

Conceptual model of radionuclide migration through the engineered barrier system takes into account the following main assumptions:

- Disposal container fails after 1000 years,
- Spent nuclear fuel is divided into the UO_2 matrix and structural materials,

- Certain inventory fraction is after water-contact released instantaneously, long-term release occurs congruently with the dissolution of individual materials,
- Concentration of radionuclides is in the void volume of disposal container and in the bentonite buffer limited by elemental solubility,
- It is considered that the bentonite buffer is fully water-saturated that is why nuclides migrate through the buffer by radial diffusion,
- Contaminants are linearly sorbed on the material of bentonite buffer.

Based on these assumptions, release rates (activity rates) from the EBS to the surrounding host rock were calculated using the simulation tool GoldSim for the two isotopic compositions of spent nuclear fuel. For better understanding, Fig. 1 illustrates the time-dependent release rates of activation and fission products which represent the largest contribution to the total release rate in the early years of analysis. The calculated release rates are related to higher burnup case (60 MWd/kgU).

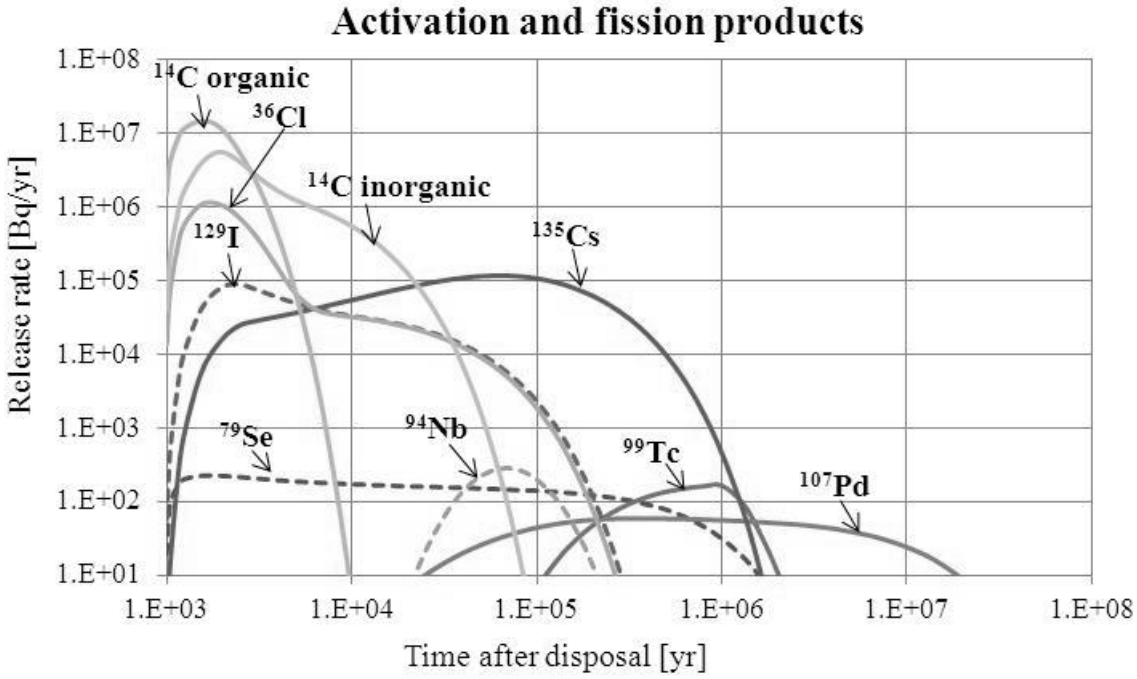

Fig.1: *Release rates of activation and fission products from the EBS per one disposal container.*

4. Results analysis

Differences in the nuclide concentrations and differences in maximum release rates of relevant radionuclides are given in Tab. 1 and Tab. 2 in percentage terms. A positive percentage value means that a concentration of an individual radionuclide has increased in respect of the lower burnup case.

According to concentration results (concentrations of isotopes right after the discharge from reactor) it can be observed that the concentrations of ^{94}Nb, ^{126}Sn, ^{129}I, ^{36}Cl originated from fuel matrix and concentrations of activation products originated from structural material are roughly proportional to the burnup.

The fuel burnup has a significant impact on actinides. The depletion process results in burning of fissile material and accumulation of transuranic elements by neutron

transmutation. It can be seen that ^{239}Pu saturates at the first burnup level and its change in concentration is very small in comparison with the higher burnup case. The change in the concentration of ^{238}U is characterised by a slow descent with increasing burnup. The decrease in concentration of ^{235}U is exponential.

The percentage differences in concentrations of relevant radionuclides (Tab. 1) were compared with the percentage differences resulted from the comparison of maximum release rates between two migration calculation cases (Tab.2). Based on this comparison, the following conclusions can be done:

- ^{99}Tc, ^{107}Pd, ^{237}Np, ^{79}Se – the percentage difference in release rates for the two migration cases is much smaller than the difference in concentrations after the discharge from the nuclear reactor. This can be related to their significant inventories and low solubility limits,
- ^{226}Ra and ^{210}Pb occur in 4n+2 decay chain (^{242}Pu → ^{238}U →^{234}U → ^{230}Th → ^{226}Ra → ^{210}Pb). The concentration of ^{226}Ra is highly limited by its elemental solubility and therefore the difference in release rates is negligible. On the other hand, the solubility limit of ^{210}Pb is much higher,
- Due to the fact that radionuclides like ^{126}Sn, ^{135}Cs, ^{129}I, ^{36}Cl, ^{14}C originated from the fuel matrix and ^{14}C originated from the structural material do not reach their solubility limits, the percentage difference in concentrations is relatively the same as the difference in release rates which correspond to the individual burnup cases,
- Decrease in the release rates of ^{231}Pa and ^{227}Ac for higher burnup level is connected with the decrease in the concentration of ^{235}U.
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Tab. 1. *Percentage differences in concentrations between two burnup cases.*

		δ [%]		δ [%]		δ [%]
Fuel matrix	^{240}Pu	28	^{239}Pu	1	^{107}Pd	61
	^{236}U	11	^{235}U	-48	^{126}Sn	49
	^{237}Np	42	^{94}Nb	52	^{135}Cs	30
	^{242}Pu	100	^{93}Zr	27	^{129}I	42
	^{238}U	-1	^{79}Se	24	^{36}Cl	48
	^{234}U	64	^{99}Tc	24	$^{14}C_{inorg}$	50
Structural material	^{99}Tc	47	^{94}Nb	42	^{93}Zr	48
	^{92}Nb	48	$^{14}C_{org.}$	49		

Tab. 2. *Percentage differences in release rates between two migration cases.*

	δ [%]		δ [%]		δ [%]
^{240}Pu	15	^{235}U	-6	^{107}Pd	-2
^{236}U	22	^{231}Pa	-31	^{126}Sn	49
^{237}Np	0.03	^{227}Ac	-31	^{135}Cs	30
^{242}Pu	86	^{92}Nb	48	^{129}I	42
^{238}U	0.72	^{93m}Nb	29	^{36}Cl	48
^{234}U	0.75	^{94}Nb	42	$^{14}C_{inorg.}$	50
^{230}Th	92	^{93}Zr	29	$^{14}C_{org.}$	49
^{226}Ra	0	^{79}Se	-3		
^{210}Pb	94	^{99}Tc	1		

5. Conclusion

Release rates of nuclides which exceed their solubility limits are effectively lowered. On the other hand, if a solubility limit is higher and a radionuclide does not reach this limit, release rates from EBS are relatively proportional to a change in concentration. In consideration of only one disposal container, changes in release rates are negligible but it could be a possible source of uncertainty if a whole source term of repository is modelled. It is also necessary to model and analyse processes occurring in the far field and perform a sensitivity analysis of parameters that have a crucial influence on radionuclide release.

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