NENE 2019
September 9–12
PORTOROŽ
SLOVENIA
28th International Conference Nuclear Energy for New Europe

Impact of Fuel Type and Discharge Burnup on Source Term

Pauli Juutilainen
VTT Technical Research Centre of Finland
P.O. Box 1000
02044 VTT, Finland
pauli.juutilainen@vtt.fi

Silja Häkkinen
VTT Technical Research Centre of Finland
P.O. Box 1000
02044 VTT, Finland
silja.hakkinen@vtt.fi

ABSTRACT

Knowledge of spent nuclear fuel (SNF) source term (decay heat, reactivity, nuclide inventory, other relevant properties of SNF) is essential in the safe handling and final disposal of SNF. For example, decay heat power determines how densely the fuel canisters can be packed in the final disposal tunnels. The fuel type and burnup affect the nuclide inventory of the SNF and therefore have an essential impact on the source term. Fuel discharge burnup has increased over time and new types of fuel typically enable higher burnups than before.

In Finland, two different reactor types have been operated (VVER-440 and BWR). Additionally, an EPR is expected to start operating in the near future and preparations to construct a VVER-1200 unit during the 2020s are underway. The fuel assembly design used in these types of reactors varies in many parameters such as e.g. geometric shape, axial enrichment zoning and nuclide content (VVER-1200). Also, several different fuel assembly types can be used in one type of a reactor. The possible differences include e.g. average enrichment, enrichment zoning, number of gadolinium absorber rods and geometric parameters such as e.g. fuel pellet dimensions and cladding thickness. The operating parameters such as power and boron concentration also depend on the reactor and fuel assembly types and have an effect on the SNF source term.

In this work, fuel assemblies representing each of the above-mentioned reactor types were calculated using the Monte Carlo particle transport code Serpent 2. The effect of discharge burnup and fuel type on different components of the source term such as e.g. decay heat, photon emission rates and the nuclide inventory was examined.

1 INTRODUCTION

The spent nuclear fuel (SNF) characteristics such as decay heat and reactivity determine the number of assemblies that can be loaded in a final disposal canister and hence have a great impact on the volume needed in the underground facility. Also other components of the source term such as e.g. nuclide inventory determining the radioactivity of SNF, are essential in the

safe handling and disposal of the SNF. However, the computational characterization of spent fuel assemblies involve several sources of uncertainty such as e.g. uncertainties in nuclear data, impurities in fuel and structural materials, choice of calculation parameters, uncertainties in operation history etc. In the present study, we focus on the impact caused by different kind of fuel assemblies and different levels of discharge burnup while keeping the other factors constant.

2 METHODOLOGY

2.1 Studied assemblies

Several different types of fuel assemblies have been used in the Finnish power reactors, VVER-440 and BWR, during the history of their operation. For the calculations of this work, a GE14-type assembly [1],[2] was chosen to represent BWR fuel and TVEL 2$^{\mathrm{nd}}$ generation [3] [4] [5] VVER-440 fuel. An assembly was also modelled for the EPR fuel [6] [7] and three assembly variations for VVER-1200 fuel [5] [8]. The VVER-1200 assemblies differ from each other with respect to uranium enrichment and number of gadolinium rods. A selection of key parameters of the studied assemblies are listed in Table 1. As the table suggests, the irradiation conditions, such as power density, boron concentration etc., were approximated with constant values for the whole operating history. All calculations were performed in two-dimensional geometry.

The calculated VVER-1200 assemblies differ from the others such that the fuel was based on recycled uranium, causing some U-236 impurity into the fresh fuel. A weight fraction of 0.7 wt-% was assumed for U-236 concentration. All other studied assemblies used UOX fuel based on natural uranium. Additionally, all studied assemblies contained a various number of rods that were doped with burnable absorber gadolinium. Gadolinium (Gd) was mixed in the form of Gd_2O_3 whose fraction varied between 3 and 9 wt-% whilst the rest of the fuel mass was still UOX. The uranium in Gd-rods was based on fresh uranium also in the VVER-1200 fuel.

Regardless of the reactor or assembly type, the fresh fuel is generally known to contain trace amounts of N-14 and Cl-35 that yield C-14 and Cl-36 nuclides, respectively, during the irradiation. These nuclides, in conjunction with Mo-93, Ag-108m and I-129, have been assessed to be the most likely radionuclides to get released to biosphere from the final disposal repository [9]. Therefore, 10 ppm of both N-14 and Cl-35 were added to the fuel material for these calculations in order to get an estimate of the C-14 and Cl-36 concentration in the spent fuel.

Table 1: Some specifications of the studied fuel asssemblies

	VVER-440	**VVER-1200**			**BWR**	**EPR**
Parameter	**TVEL 2nd gen.**	**Assembly 1**	**Assembly 2**	**Assembly 3**	**GE14**	
U mass (kg/cm)	0.520	1.29	1.29	1.29	0.505	1.21
Average U-235 (wt-%)	4.37	4.92	3.76	4.90	4.23	3.60
Normal rods	120	306	300	300	74	253
Gd-rods	6	6	12	12	18	12
Gd_2O_3 in Gd-rods (wt-%)	3.35	5	5	5	3 or 8	9
Boron (ppm)	500	600	600	600	0	600

2.2 Computational model and methods

All calculations were performed with the continuous-energy Monte Carlo code Serpent [10] that has been developed at VTT since 2004 for various reactor physics and radiation shielding applications. These calculations utilized the neutron transport and burnup calculation functionalities of the code to provide source term information needed in the spent fuel management. The version 2.1.31 of the code was used. The nuclear data was obtained from JEFF-3.2 cross-section library for neutron transport and from JEFF-3.1.1 fission yield and decay libraries for the burnup section of calculations.

The studied fuel assemblies were irradiated up to 80 MWd/kgU discharge burnup, even though the realistic maximum discharge burnup levels have been much lower. However, the nuclide compositions were recorded at every burnup step allowing the spent fuel analyses to be performed flexibly for almost any discharge burnup. In this study, we used 50 MWd/kgU as a reference discharge burnup, even though it is historically still somewhat higher than the realised averages for all types.

The length of the burnup steps in the calculations was varied based on various needs over the irradiation period. First, before anticipated xenon equilibrium, 0.1. . .0.3-MWd/kgU-long steps were used. At the next phase, up to 25 – 30 MWd/kgU, 0.5 MWd/kgU was used to ensure that the burnable absorbers deplete completely and finally, the rest of the irradiation was calculated with 2.5 MWd/kgU long steps. Some minor differences in the step lengths exist between the modelled assemblies, but they should not be significant. The substep method [11] with linear interpolation in the predictor and quadratic extrapolation in the corrector with 10 substep in both predictor and corrector was used.

Every pin in the fuel assemblies was handled as a separate depletion zone in the burnup calculations. The Gd pins were additionally divided into 10 equally large depletion zones. These were perfomed with the automatic zone division tool of Serpent.

The neutron transport part of the calculations were based on 10 million neutron histories at every burnup step. As a result, the statistical uncertainty in k_{eff} was mainly 15. . .20 pcm. However, the uncertainty indicated growing trend up to roughly 25 pcm along with increasing burnup. The calculations were performed in 2D geometry with either periodic (VVERs) or reflective (BWR, EPR) boundary conditions.

3 RESULTS

3.1 Decay heat

The decay heat production is one of the most important factors in the design of storage configurations, since it practically sets the limits for the positioning density of the spent fuel canisters. In order to provide a general view about the nature of decay heat production of the spent fuel over the first 100,000 years after being discharged from the reactor, the total decay heat production of the studied fuel assemblies is illustrated in Figure 1. Fig. 1a shows the heat production per unit mass for the assemblies with 50 MWd/kgU discharge burnup. Since the logarithmic scale somewhat hides the differences between the assembly types, the relative difference of the heat production between the most and least heat-producing assemblies — VVER-1200 assembly 2 and BWR, respectively — for various burnups at each calculated time is depicted in Fig. 1b. The latter figure clearly suggests that the relative difference may be rather large and is affected by the discharge burnup.

However, an earlier study [12] suggests that when the spent fuel is deposited into its final repository, the surrounding bedrock achieves its peak temperature 50 – 100 years after the

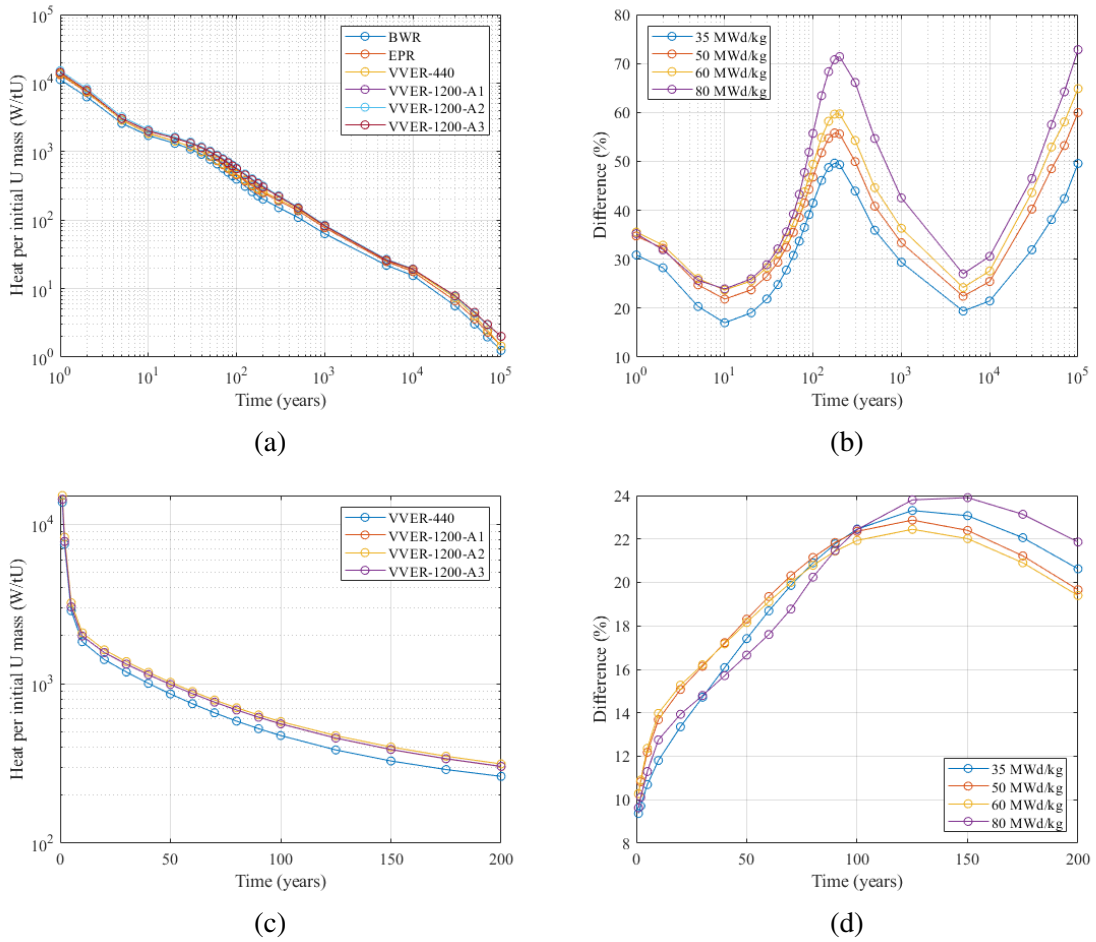
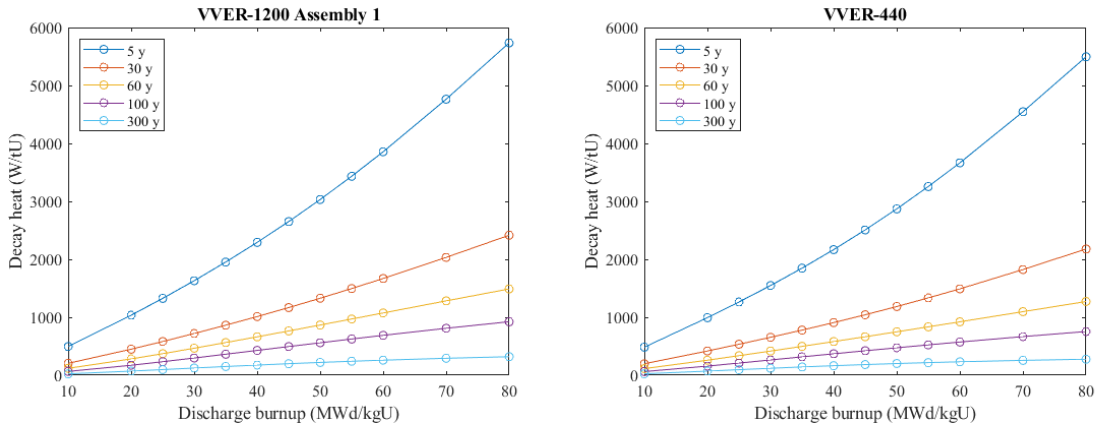


Figure 1: The decay heat per uranium mass with 50 MWd/kgU discharge burnup is depicted for all studied assemblies in (a) over 1 – 100,000 years after discharge and for the studied VVER assemblies in (c) over 1 – 200 years after discharge. The relative difference between the most and least heat producing assemblies out of all studied assemblies and VVER assemblies are presented in (b) and (d), respectively.

disposal. Therefore, and combined with the fact that the heat production decreases continuously, the following decay heat considerations are restricted to the rather short period with respect to the scale of the final disposal in general, that is, up to 200 – 300 years.

Figures 1c and 1d describe the heat production of the studied VVER assemblies similarly to Figs. 1a and 1b, but the plotted period is cut to 200 years in order to make the differences between the assemblies become better visible. However, the three VVER-1200 assemblies can still hardly be seen to differ much from each other. The difference between the three and the VVER-440 can be seen, and the difference between the hottest and coolest assembly peaks at above 20 %. When only the VVER-1200 assemblies are compared, the difference is kept below 7 % with the discharge burnup higher than 40 MWd/kgU. For lower burnups, the maximum difference can be much larger.

The dependency of decay heat on discharge burnup is illustrated in Figure 2, where the heat production of spent VVER-1200 and VVER-440 fuel assemblies is depicted for various cooling times after the discharge. It can be seen that the additional heat production caused by higher burnup is much more prominent with short cooling times, but the difference between these assembly types is relatively small.



(a) (b)

Figure 2: The decay heat per unit mass as a function of discharge burnup at various cooling times in (a) for VVER-1200 assembly 1 and in (b) for VVER-440 assembly.

The heat production is largely dominated by a rather small number of the most important heat producers, even though the share of the top heaters drops temporarily soon after the discharge. Figure 3 illustrates as an example, how various numbers of top heat producers contribute to the total heat production, when a VVER-1200 assembly is considered. It can be noticed that a few decades after the discharge, the top-10 heaters are practically responsible for the whole decay heat, and even the top-5 nuclides explain more than 80 % of the heat production. As an example, Table 2 presents the top-5 heat producers of spent VVER-1200 fuel assembly at a few selected points after discharge. Generally, when the top-5 lists are compared between fuel types, the same nuclides are mostly present, but partly in different order. Additionally, when the same studies as shown in Fig. 3 are performed for other fuel types and various discharge burnups, relatively similar — but not identical — behaviour can be observed.

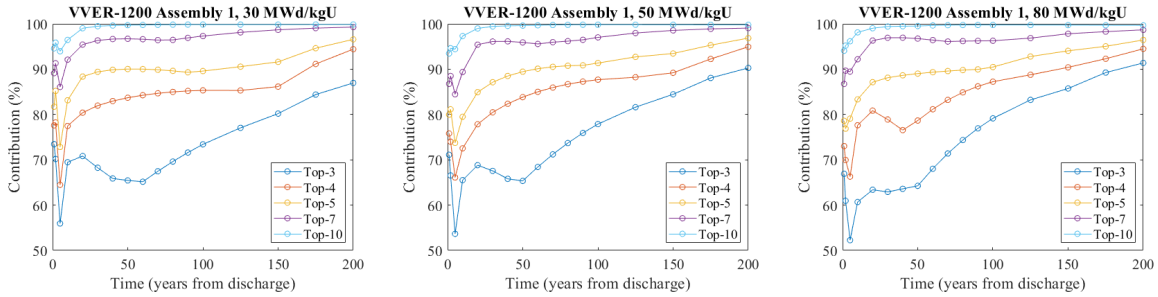
Table 2: Top-5 heat producers at various times after discharge (VVER-1200 assembly 1, 50 MWd/kgU)

5 years	**20 years**	**50 years**	**100 years**	**150 years**
Cs-134	Y-90	Pu-238	Am-241	Am-241
Y-90	Ba-137m	Am-241	Pu-238	Pu-238
Ba-137m	Pu-238	Ba-137m	Ba-137m	Pu-240
Pu-238	Am-241	Y-90	Y-90	Ba-137m
Rh-106	Cs-137	Cs-137	P-u240	Y-90

3.2 Photon emission rates

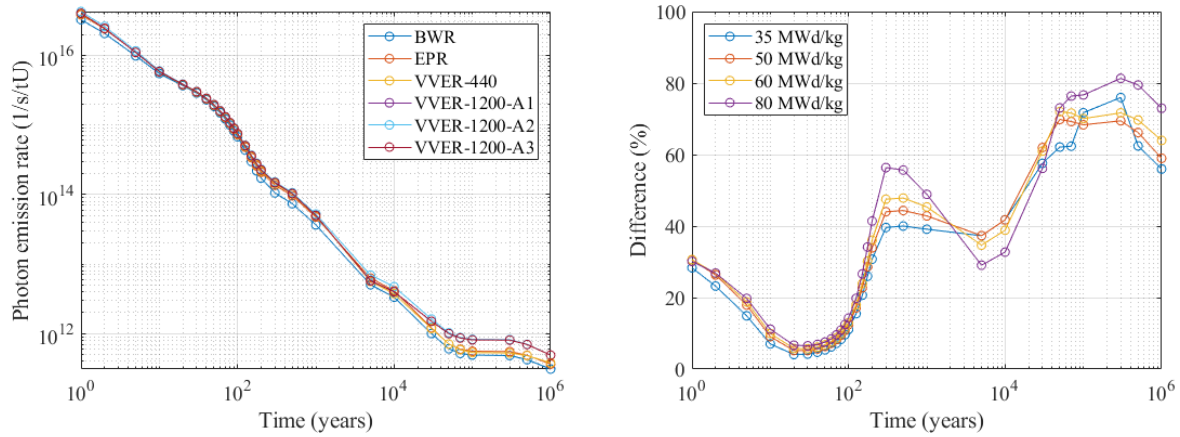
The photon emission rates per unit mass of initial uranium is depicted in Figure 4. Analogically to Figs. 1a and 1b, the total emission rate is presented for all studied fuel types with the discharge burnup of 50 MWd/kgU (in Fig. 4a) and the relative difference of the emission rate between the most and least active bundles at each calculated time step is illustrated in Fig. 4b. Somewhat large relative differences begin to appear after the first post-discharge centuries.

When the contribution of the top photon emitters is considered, the photon emission is much more concentrated to only a few nuclides than heat production, at least in the scope of some tens to a few hundred years after discharge. The three most photon-emitting nuclides are



(a) (b) (c)

Figure 3: The contribution of a few selected number of top heat producers of the total heat production at each calculated time, when a VVER-1200 Assembly 1 is considered with various discharge burnups.



(a) (b)

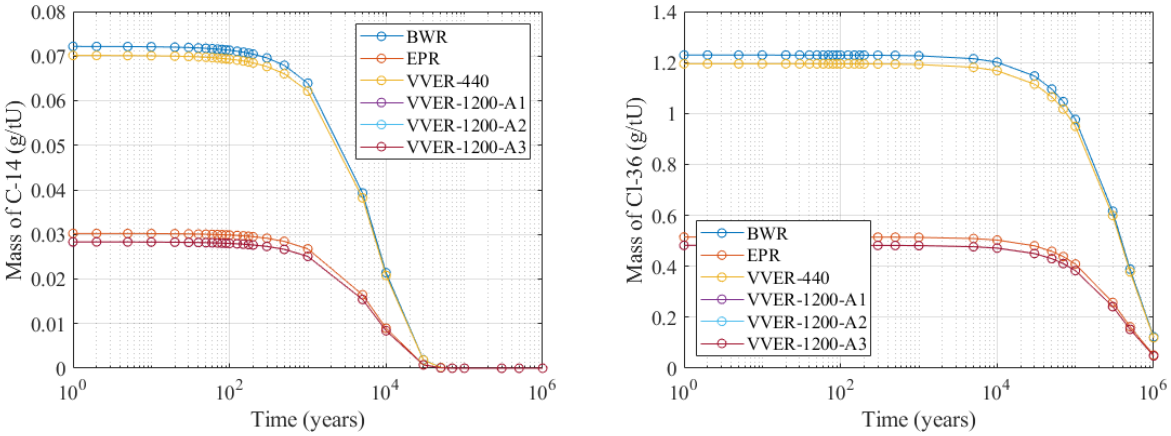
Figure 4: In (a): photon emission rates of the studied assemblies with 50 MWd/kgU discharge burnup. In (b): the relative difference of the emission rates between the most and least active bundle with various discharge burnups.

almost completely responsible for photon emissions over that period, even though the nuclides in the top-3 slightly vary over time. Generally, Ba-137m, Am-241 and Pu-238 are typical nuclides on the list, when all studied fuel types with 50 MWd/kgU discharge burnup is considered. In contrast to the heat producers, the contribution of top photon emitters becomes more evenly distributed at later phases.

3.3 Radionuclides

The most probable radionuclides to propagate out of the final disposal repository are C-14, Cl-36, Mo-93, Ag-108m and I-129, so the contents of these nuclides are of particular interest when the propagation barriers are analysed. As mentioned in Section 2.1, the presence of C-14 and Cl-36 is due to impurities whose actual concentration in the fresh fuel is an assessment. However, the same concentration was used in all assemblies, so the differences between them can be compared, which is done in Figure 5 for discharge burnup of 50 MWd/kgU.

The behaviour of the other listed nuclides is similar, at least in the sense that the studied VVER-440 and BWR fuel types contain them roughly double to the contents in other studied assemblies. When the effect of discharge burnup is considered, increasing burnup adds



(a) (b)

Figure 5: Contents of C-14 (a) and Cl-36 (b) in the spent fuel as a function of time after discharge.

the amount of these nuclides, but the relative differences between the fuel assemblies remain somewhat constant.

4 CONCLUSIONS

Monte Carlo burnup calculations were performed with Serpent 2 for a selection of fuel assemblies that have been or are likely to be used in Finnish NPPs and some of the results are reported in the present study. The purpose of the study was to identify the differences in the spent fuel characteristics of the studied fuel assemblies. In the wider scope, the calculations were part of the process to formulate standardised practicalities to burnup calculation for waste management puproses with VTT's computational tools.

The burnup calculation mode of Serpent provides plenty of information but only decay heat production, photon emission rates and the concentration of a few important radionuclides could be discussed in the present study. These illustrated various differences between the fuel assemblies, but how the observed differences practically affect the waste management, is a question to be answered in the studies utilizing the source term, such as heat transfer and radiation shielding analyses.

The calculations were performed in 2D geometry with periodic or reflective boundary conditions, which is a potential source for uncertainties, since the neighbouring assemblies usually are at different phase of irradiation. As another approximation, the neutronics conditions were assumed constant over the whole irradiation period, which obviously is not true particularly with the boron concentration. More accurate operating data would be useful for future calculations, but not necessarily available.

ACKNOWLEDGMENTS

The work has been funded by the Finnish Research Programme on Nuclear Waste Management KYT2022.

REFERENCES

[1] B. Ade, S. Bowman, I. Gauld and G. Ilas, "Technical Basis for Peak Reactivity Burnup Credit for BWR Spent Nuclear Fuel in Storage and Transportation Systems", NUREG/CR-7194, ORNL/TM-2014/240, 2015.

[2] I. Gaud and U. Mertyurek, "Margins for Uncertainty in the Predicted Spent Fuel Isotopic Inventories for BWR Burnup Credit", NUREG/CR-7251, ORNL/TM-2018/782, 2018.

[3] T. Lahtinen, M. Antila and S. Saarinen, "Review of core design and operating experience in Loviisa", 20th Symposium of Atomic Energy Research, Hungary, 2010.

[4] M. Antila and T. Lahtinen, "Recent core design and operating experience in Loviisa NPP", in Proceedings of the 17th Symposium of AER, Ukraine, 2007.

[5] TVEL, "VVER Nuclear Fuel," [Online]. Available at https://www.tvel.ru/wps/wcm/connect/tvel/tvelsite.eng/resources/9ef3688045a250ee976cf73487cca6b0/NuclearFuel_ENG.pdf. [Accessed February 26, 2019].

[6] R. Boehm, G. Schmidtlein, E. Riedl and P. Pulkus, "Simplicity, Flexibility and Margins - the First Core of Olkiluoto 3", in Top_Fuel 2006 Transactions, Salamanca, Spain, October 22 – 26, 2006.

[7] G. Sengler, F. Forêt, G. Schlosser, R. Lisdat and S. Stelletta, "EPR Core Design", Nuclear Engineering and Design, vol. 187, no. 1, pp. 79 – 119, 1999.

[8] V. Molchanov, "Nuclear Fuel for VVER Reactors. Current Status and Prospects", in 6th International Conference on WWER Fuel Performance, Modelling and Experimental Support, Bulgaria, September 19–23, 2005.

[9] Posiva Oy, "Safety Case for the Disposal of Spent Nuclear Fuel at Olkiluoto —- Synthesis 2012. POSIVA 2012 – 12. Posiva Oy, 2012.

[10] J. Leppänen, M. Pusa, T. Viitanen, V. Valtavirta and T. Kaltiaisenaho, "The Serpent Monte Carlo code: Status, development and applications in 2013", Annals of Nuclear Energy, vol. 82, pp. 142 – 150, 2015.

[11] A. Isotalo, P. Aarnio, "Substep methods for burnup calculations with Bateman solutions", Ann. Nucl. Energy Vol. 38, pp. 2509 – 2514, 2011.

[12] K. Ikonen, J. Kuutti and H. Raiko, "Thermal Dimensioning for the Olkiluoto Repository - 2018 Update", Working Report 2018-26, 2018.