

Release of radioactive nuclides from spent WWER fuel

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In this paper, the instant fractional release of gaseous or volatile radionuclides from spent WWER fuel is computationally estimated. A common definition for the instant release is adopted, such that the term refers to the fraction of the inventory of radionuclides that are situated in the fuel-to-cladding gap and may be released instantaneously when the cladding fails for example in a fuel handling accident or during fuel transportation.

The analysis is mainly based on the newly amended ANS-5.4 standard by American Nuclear Society's Standards Committee. Both the nuclides with short half-lives ($6 \text{ h} < t_{1/2} < 60 \text{ d}$) and long half-lives ($t_{1/2} > 1 \text{ y}$) are considered. The relevant nuclides included to the study are krypton, xenon, iodine, caesium and tellurium.

In order to quantify the fractional release of relevant fission products in a fuel handling accident, all the rods of two WWER-440 fuel bundles are simulated with a single rod fuel performance code VTT-ENIGMA. The power histories of the two bundles originate from the design calculations of Loviisa NPP's reactor core loading, and are chosen so that the first bundle has the highest power, and the second one has the highest burnup. In addition, one more simulation with VTT-ENIGMA is conducted with a power history based on a burnup dependent linear power limit determined for the fuel used in the Loviisa NPP.

In order to calculate the release fractions with VTT-ENIGMA, the formulation presented in the ANS-5.4 standard has recently been implemented into the code at VTT.

In the analysis, the calculated release fractions of all the studied short-lived nuclides among all the simulated rods are less than 0.11%, and for the long-lived nuclides less than 5%. These results are at or below the level of experimental release fractions found in open literature.

KEYWORDS: instant release fraction, ANS-5.4, VTT-ENIGMA

1. Introduction

In this study, the fractional release of gaseous or volatile radionuclides from the fuel rod(s) whose cladding is failed is investigated. The fission products that are situated in the fuel-to-cladding gap may be released instantaneously when the cladding fails for example in a fuel handling accident or during transportation. This fraction of the inventory of radionuclides is commonly referred to as “instant release”.

The release fractions are dependent i.a. on the diffusion coefficients of the nuclides in the fuel, on linear power level during the steady-state irradiation, and on fuel burnup. Calculation of the yields of the fission products would require additional neutronics calculations, and that is out of the scope of this study.

In order to quantify the fractional release of relevant fission products in a fuel handling accident, all the rods of two WWER-440 fuel bundles are simulated with a single rod fuel performance code VTT-ENIGMA (version 5.9b with VTT applied improvements). The power histories of the two bundles originate from the design calculations of Loviisa NPP’s reactor core loading, and are chosen so that the first bundle has the highest power, and the second one has the highest burnup. In addition, one more simulation with VTT-ENIGMA is conducted with a power history based on a burnup dependent linear power limit determined for the fuel used in the Loviisa NPP.

The newly amended American National Standard ANS-5.4 [1] is considered here as the main methodological basis for determining the desired fractions of fission products. The standard is developed for determining the release of radionuclides from UO₂ fuel in an accident in which the power does not change rapidly [1]; thus the standard is well suited for depicting the releases in a fuel handling accident. The ANS-5.4 standard was originally formulated in 1982 by the Standards Committee of American Nuclear Society, and in 2011, the replacement of this standard was introduced. As new experimental data has become available, a part of the over-conservatism has been removed from the standard.

More information and experimental results related to the release of radioactive nuclides from spent fuel are found in a state-of-the-art report prepared in the European Union’s European Atomic Energy Community’s (EURATOM) 7th Framework Programme Project “FIRST Nuclides” [2].

2. Nuclides included to the study

The studied nuclides and their half-lives are presented in Table 1. The most important gaseous or volatile nuclides from the radiological point of view are krypton, xenon, iodine, caesium and tellurium. The nuclides with half-lives less than 6 hours are left outside this study as those are not relevant in fuel handling accidents. Also isotopes with very long half-lives like I-129 (15.7 million years) and Cs-135 (2.3 million years) are excluded as those are not relevant in this study but of course to be considered when analysing the final disposal of nuclear fuel.

In Table 1, the studied nuclides are divided into two groups according to their half-lives: short- and long-lived nuclides. This division is adopted from the ANS-5.4 standard in which the short-lived nuclides have half-lives between 6 hours and 60 days, and long-lived nuclides have half-lives greater than one year (in the standard, there is also a group with very short half-life $t_{1/2} < 6$ h and a method for calculating their release fractions). In the table, the short-

lived isotopes of iodine and xenon are gathered from the ANS-5.4 standard, whereas the short-lived tellurium and caesium isotopes are not explicitly named in the standard but it is only mentioned that the standard also applies to Te and Cs isotopes that have the half-life within the range $6 \text{ h} < t_{1/2} < 60 \text{ d}$. The isotopes of short-lived Te and Cs are gathered from the nuclide chart [3].

Table 1 includes also the decay constant λ and precursor effect constant α of the nuclides. The precursor effect constant takes into account the movement (diffusion) of the precursor nuclide before it decays to the nuclide under investigation. The coefficients are reported in [1, 4]. If the coefficient is not given there, it means that the precursor effect is small and can be neglected. Then it is recommended [1] to use $\alpha=1$ when applying the ANS-5.4 standard (those cases are marked with an asterisk in Table 1). With the short-lived Te and Cs isotopes, α is not known and also then α is set to 1.

3. Method for the calculation of the release fractions

In this section, the method for determining the release fractions after the base irradiation is presented. As for the short-lived nuclides, the method is based on the ANS-5.4 standard. For the long-lived nuclides, the standard suggests a very simple method consisting of multiplying the diffusion coefficient of (or the release fraction from) a standard fission gas release (FGR) model by a constant factor. Similar methods for the coupling of the release fraction of long-lived nuclides to the FGR are studied also elsewhere [2, 5]; those are also summarized here.

As the main driving force for the nuclide movement is thermally activated diffusion [1], and as the post-irradiation temperatures in the fuel are relatively low, it can be assumed that the radioactive nuclides are immobile after the base irradiation. Thus it can be assumed that the release fractions given by the ANS-5.4 standard stay constant on that level they have had after the base irradiation.

There are some limitations in the applicability of the ANS-5.4 standard. It is intended for the steady-state conditions in which the power and temperature do not change rapidly causing burst release of fission products. In addition, fuel oxidation is not considered as it could significantly magnify the release. [1]

3.1 Nuclides with short half-lives

The derivation of the equations presented in this section is given in the ANS-5.4 background document [4] and is not repeated here. The method for calculating the release fractions for nuclides with short half-life is based on the Booth diffusion model, and it takes into account the decay of the nuclides. The model coefficients are derived based on experimental data from Halden tests and are valid in the burnup range 0-70 MWd/kgU. The release fraction, or in other words the release-to-birth ratio (R/B) of the short-lived nuclides listed in Table 1, is formulated with the help of Kr-85m ($t_{1/2}=4.48 \text{ h}$, $\lambda_{\text{Kr-85m}}=4.30\text{E-}5 \text{ s}^{-1}$, $\alpha_{\text{Kr-85m}}=1.31$) decay constant and precursor effect constant [1]:

$$\left(\frac{\text{R}}{\text{B}}\right)_{r,z} = \left(\frac{\alpha_{\text{nucl}} \lambda_{\text{Kr-85m}}}{\alpha_{\text{Kr-85m}} \lambda_{\text{nucl}}}\right)^{0.25} \left(\frac{\text{S}}{\text{V}}\right)_{r,z} \sqrt{\frac{\alpha_{\text{Kr-85m}} \text{D}_{r,z}}{\lambda_{\text{Kr-85m}}}} \quad (1)$$

Here $\text{D}_{r,z}$ is the diffusion coefficient:

$$D_{r,z} = 7.6 \cdot 10^{-7} e^{-35000/T_{r,z} [K]} + 1.41 \cdot 10^{-18} F_z^{0.5} e^{-13800/T_{r,z} [K]} + 2 \cdot 10^{-30} F_z, \quad (2)$$

where

$$F_z = 4.0 \cdot 10^{10} \text{ LHGR}_z [\text{W/cm}] (\text{Diam}_{\text{outer}}^2 [\text{cm}] - \text{Diam}_{\text{inner}}^2 [\text{cm}])^{-1}. \quad (3)$$

LHGR_z is the local linear heat generation rate and Diam is the outer and inner diameter of the pellet. In Eq. (1), the surface-to-volume ratio (S/V)_{r,z} has two values depending if the temperature is below or above the temperature in which the bubbles are interlinked on the grain boundaries:

$$\left(\frac{S}{V} \right)_{r,z} = 120 \text{ cm}^{-1} \text{ or } 650 \text{ cm}^{-1}, T_{r,z} [\text{K}] \leq T_{\text{link}} \text{ or } T_{r,z} [\text{K}] > T_{\text{link}}, \text{ respective ly.} \quad (4)$$

The threshold temperature is given by the Vitanza correlation for burnups less than 18.2 MWd/kgU and a linear correlation above that:

$$T_{\text{link}} = \frac{9800}{\ln(176 \text{ BU}_z)} + 273, \quad \text{BU}_z \leq 18.2 \text{ MWd/kgU}, \quad (5)$$

$$T_{\text{link}} = 1434 - 12.85 \text{ BU}_z + 273, \quad \text{BU}_z > 18.2 \text{ MWd/kgU}. \quad (6)$$

Finally for the whole rod, the R/B ratio is obtained by summing and normalizing the local R/B ratios:

$$\left(\frac{R}{B} \right)_{\text{rod}} = \sum_{z=1}^Z \frac{P_z [\text{W/cm}] V_z [\text{cm}^3]}{P^{\text{ave}} [\text{W/cm}] V^{\text{rod}} [\text{cm}^3]} * \sum_{r=1}^R \frac{P_{r,z} [\text{W/cm}] V_{r,z} [\text{cm}^3]}{P_z [\text{W/cm}] V_z [\text{cm}^3]} \left(\frac{R}{B} \right)_{r,z}. \quad (7)$$

The standard is applicable when the power has been constant over three half-lives of the nuclide in question. If this constraint is not met, the highest power and temperature during the three half-life period is used in a conservative fashion. [1]

The formulation is used for iodine and xenon nuclides presented in Table 1 as such. Due to lack of experimental data in case of short-lived tellurium and caesium nuclides, however, conservative multipliers are suggested [1] in front of the diffusion coefficient, Eq. (2). The coefficients are 4 and 2 for tellurium and caesium, respectively, and those are applied with the calculations presented in this paper.

In order to calculate the release fractions with VTT-ENIGMA, Eq. (1) – (7) have been coded into the code's output routine within another project [6].

3.2 Nuclides with long half-lives

According to the ANS-5.4 standard, the release fraction of Kr-85 can be stated to be equal to the FGR calculated with a fuel performance code. For Cs-134 and -137, the fractions are

conservatively estimated to be $\sqrt{2}$ times the calculated FGR, or in another fashion, the diffusion coefficient of the code's FGR model is multiplied by two. Another reference suggests that the caesium release is the FGR divided by 3 [2, 5]. However, there is disagreement between the references [2], [5] and [7] whether the ratio 1:3 is between Cs release and FGR or between the diffusion coefficients, in which case the ratio would be $1:\sqrt{3}$ for the release fractions. As the original reference [7] suggests that it is the ratio between the diffusion coefficients, the ratio $1:\sqrt{3}$ is applied here as the ratio between Cs release and FGR. In [5], new experiments are conducted in order to confirm the ratio given in [7], and the results were found to have good correspondence with the old results. The new tests were performed in aqueous environment and the burnup range was 58-75 MWd/kgU.

If the defected fuel rod is in water environment, Cs and I are among the elements that dissolve in water and move away with the water from the gas gap [8]. In aqueous environment, yet another correlation between the caesium release and FGR is fitted based on a number of experimental results from various sources [2]:

$$\text{Cs}[\text{release} - \%] = (0.19 \pm 0.02) \text{FGR}[\%] + (0.74 \pm 0.19) \quad (8)$$

The data used for fitting this correlation is presented in Fig 1, with the exception that the tests in which only Cs release or FGR is measured are excluded.

4. Results

Average power histories, fuel maximum temperatures, and the calculated FGRs are presented in Fig 2 for the fuel rods of the two analysed bundles. There are total of 126 rods in one bundle, and six of them are Gd-doped; the fabrication specifications used as code input are found in open literature [9]. The release fractions of the short-lived nuclides explicitly mentioned in ANS-5.4 standard (iodine, xenon) as a function of time are presented in Fig 3 for the fuel rods of the highest power bundle. The results of the single rod with the power corresponding to the burnup dependent linear power limit are presented in Fig 4. The main results are gathered to Table 2 and 3. As for the long-lived Cs nuclides, the release fractions are calculated with three different ways as explained in Section 3.2.

As mentioned in Section 3.1, the ANS-5.4 standard is applicable when the power has been constant over three half-lives of the nuclide in question, and if this constraint is not met, the highest power and temperature during the three half-life period are used. With the present calculations concerning the two fuel bundles, the duration of the constant power period at the end of the irradiation is 11 days. This period is shorter than three times the half-life of some nuclides given in Table 1, and therefore with these nuclides, the R/B values calculated before the final constant power period are reported in Table 2 (marked with an asterisk) instead of the values at the very end of the simulation (the final power before the shutdown is at lower level than the preceding power, see the power plots in Fig 2).

The release fractions of the short-lived nuclides (average values, see Table 2) are found to be slightly higher in the highest power bundle compared to those of the highest burnup bundle but the difference is not very large. FGR, on the other hand, is higher with the highest burnup bundle. However, the single rod calculated with the power corresponding to the linear power limit has the highest release fractions and FGR among all the simulated rods. Therefore it can be concluded that it definitely is the worst case scenario with respect to the release of

radionuclides. The average release fractions in case of the two bundles represent the best-estimate predictions whereas the maximum values give a conservative estimation.

In Table 3, the maximum release fractions of the short-lived nuclides during the whole base irradiation are reported (thus not the end-of-life values). These values can be considered as conservative upper limits for a situation in which the fuel is unloaded from the reactor before the end of its planned operation time. Again, when comparing the two bundles, the highest release fractions are found in the highest power bundle (it should be noted that the rod with the power corresponding to the linear power limit has the highest values of all).

5. Discussion and Conclusions

In ANS-5.4 standard it is stated that the best-estimate release of I-131 (this nuclide gives the highest equivalent dose after the reactor shutdown) in a typical commercial plant operation is less than 2%. In order to have a bounding prediction at a 95/95 tolerance level, the release fraction is multiplied by a constant factor of 5 (considered as a very conservative multiplier for I-131 as it is derived for Kr-85m) [1]. This means that the bounding prediction at the tolerance level of 95/95 for I-131 is less than 10%. The calculated releases of I-131 presented in Table 2 and 3 are less than 0.031% for the two bundles, and less than 0.07% for the rod with the power corresponding to the linear power limit. Thus the calculated values are far less than 2 or 10%. The highest value (0.10897%) for the release fraction among all the studied short-lived nuclides is calculated for Te-125m (Table 3).

According to the state-of-the-art report [2], Cs release is below 5 % in almost all experiments made for PWR and BWR fuel samples, see Fig 1a. When compared to the calculation results, closest to this value reaches the result (4.91%) from the rod with the power corresponding to the linear power limit when the FGR is multiplied by $\sqrt{2}$. Also the calculated FGRs and that way determined release fractions of Kr-85 are within the same low range as in most of the experiments, see Fig 1b.

It should be noted that the scatter in Fig 1a-b is very large, and also that the set-up of the experiments from which the data originates was different (for example, the samples were from bare fuel, pellets, fragments and powder, the aqueous solutions were different etc.). Due to the pellet central hole in WWER-440 fuel, the fuel temperatures stay moderate and the thermal diffusion of gaseous fission products remains at lower level than in western PWRs and BWRs (cp. Cs release and FGR in Table 2 to those presented in Fig 1a-b).

For the airborne release fractions of Cs and Kr-85, values are suggested in an analysis document considering an accident at the Yucca Mountain repository (Table 5 in [10]). The release fractions are agglomerated from NRC regulatory guides and NUREG reports, and are originally based on burst rupture tests. Burst rupture tests are suggested to simulate the instant release from a fuel rod with a cladding that fails for any reason [10]. In the report, the release of Kr-85 (and thus the FGR) is reported as 30%, and Cs-134, -137 as 0.02%. The release fraction of caesium is based on limited experimental data. When comparing the Cs value with the results presented in Fig 1a, it appears that the Cs release is at much lower level in air than in aqueous phase, exactly as suggested in [8].

It should be recognized that the methods for determining the release fractions are developed and fitted based on a relatively scarce set of experimental data, and therefore multiple conservative assumptions have been made when developing these methods. Again, sometimes

even the conservative assumptions are not enough to cover the lack of data. When the premises of the analysis are changed, for example the steady-state power level, discharge burnup, environment (gas/aqueous), whether there is oxidation or not, etc., the release fractions may be significantly affected.

6. Summary

In this paper, an estimation of the instant release fractions of gaseous or volatile radionuclides from WWER-440 fuel is determined. The analysis is mainly based on the newly amended ANS-5.4 standard by American Nuclear Society's Standards Committee. Both the nuclides with short half-lives ($6 \text{ h} < t_{1/2} < 60 \text{ d}$) and long half-lives ($t_{1/2} > 1 \text{ y}$) are considered. The relevant nuclides included to the study are krypton, xenon, iodine, caesium and tellurium.

All the rods from two fuel bundles (with the highest power and the highest burnup) of Loviisa NPP were simulated with VTT-ENIGMA fuel performance code, and an additional case considering a rod with the power history corresponding to the linear power limit determined for the fuel used at Loviisa NPP. As a sum-up of the analysis, the calculated release fractions of all the studied short-lived nuclides were less than 0.11% in all cases, and for the long-lived nuclides less than 5%. Despite the multiple conservative assumptions included in the method used for determining the release fractions, the obtained results are at or below the level of experimental release fractions found in open literature.

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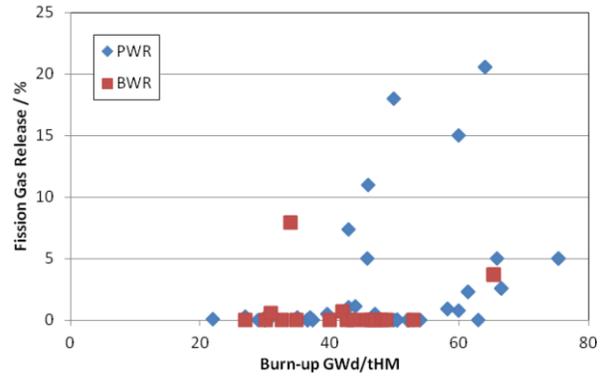
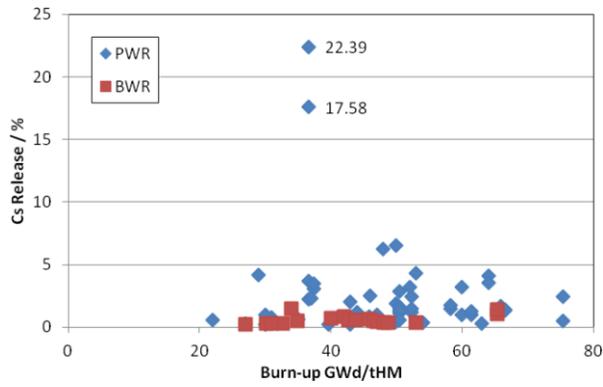


Figure 1a-b. Caesium release and FGR in fuel experiments made in aqueous phase. [2]

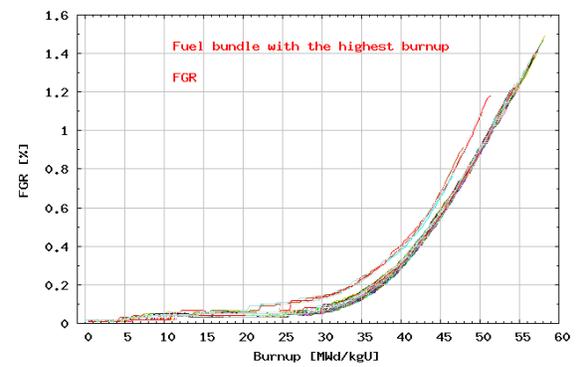
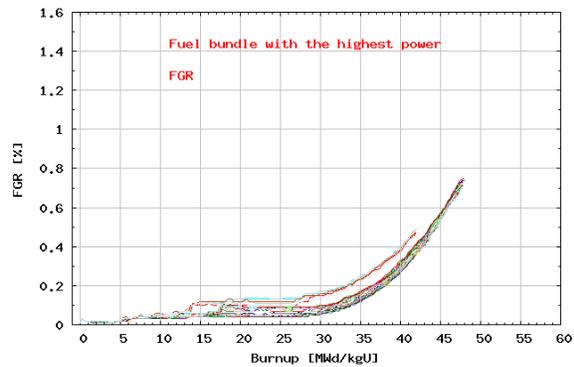
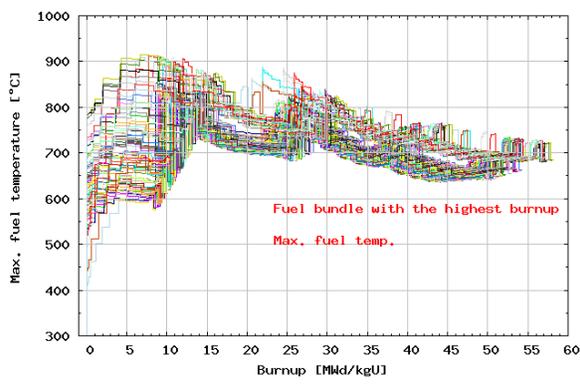
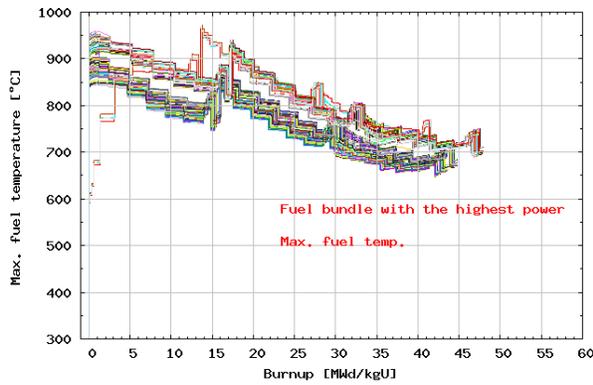
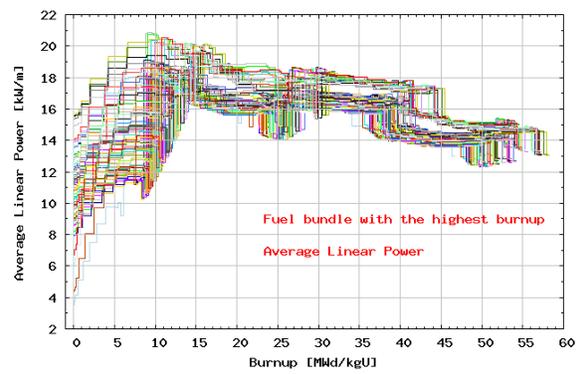
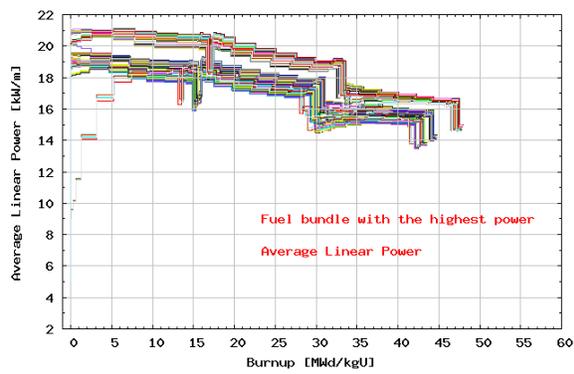


Figure 2. Average linear power, maximum fuel temperature, and FGR from VTT-ENIGMA simulations for the rods in the highest power (on left hand side) and highest burnup (on right hand side) bundle.

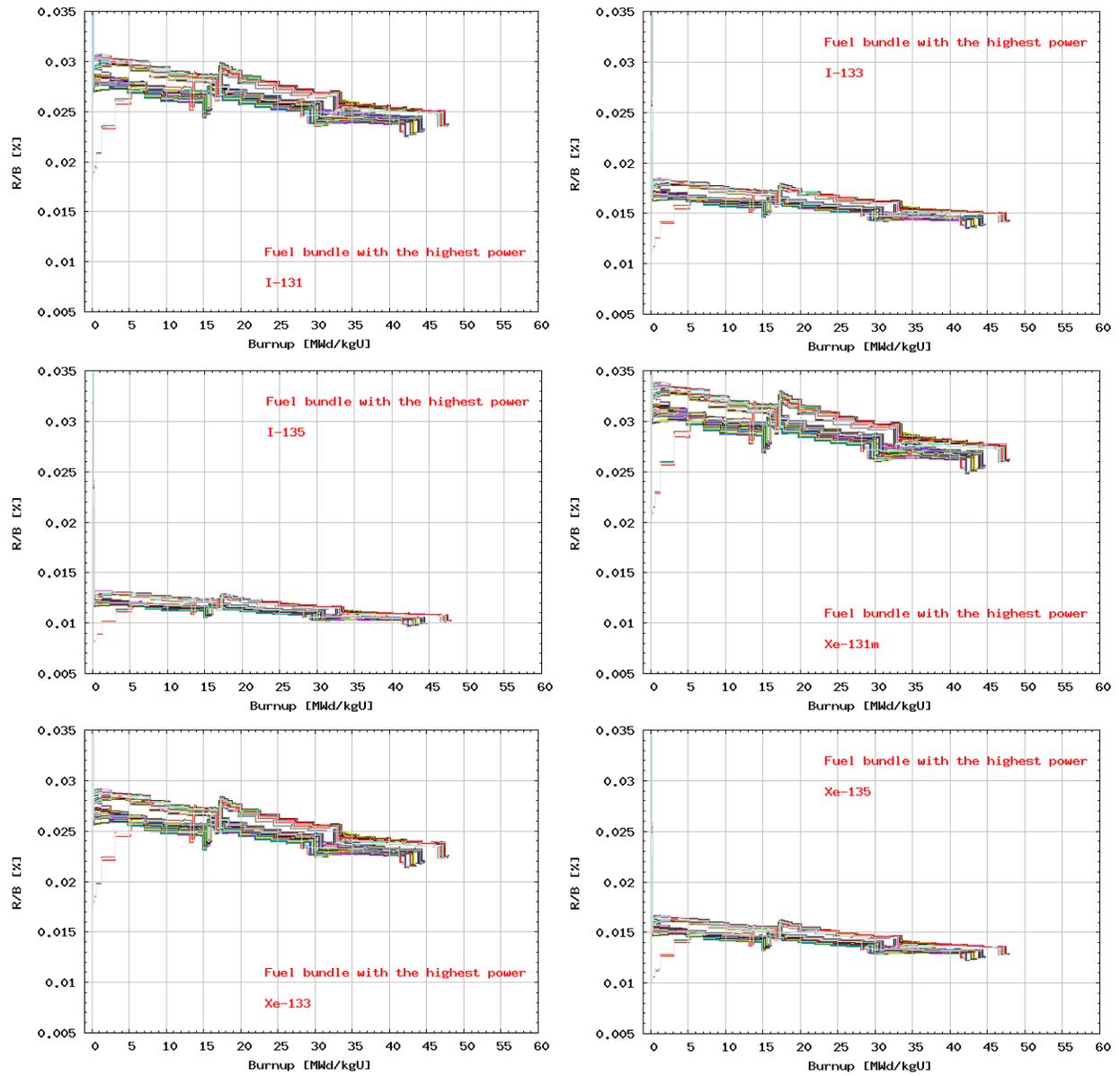


Figure 3. The percentages of iodine (I-131, -133, -135) and xenon (Xe-131m, -133, -135) isotopes released to the gas gap from the highest power bundle according to the ANS-5.4 standard [1] implemented into VTT-ENIGMA.

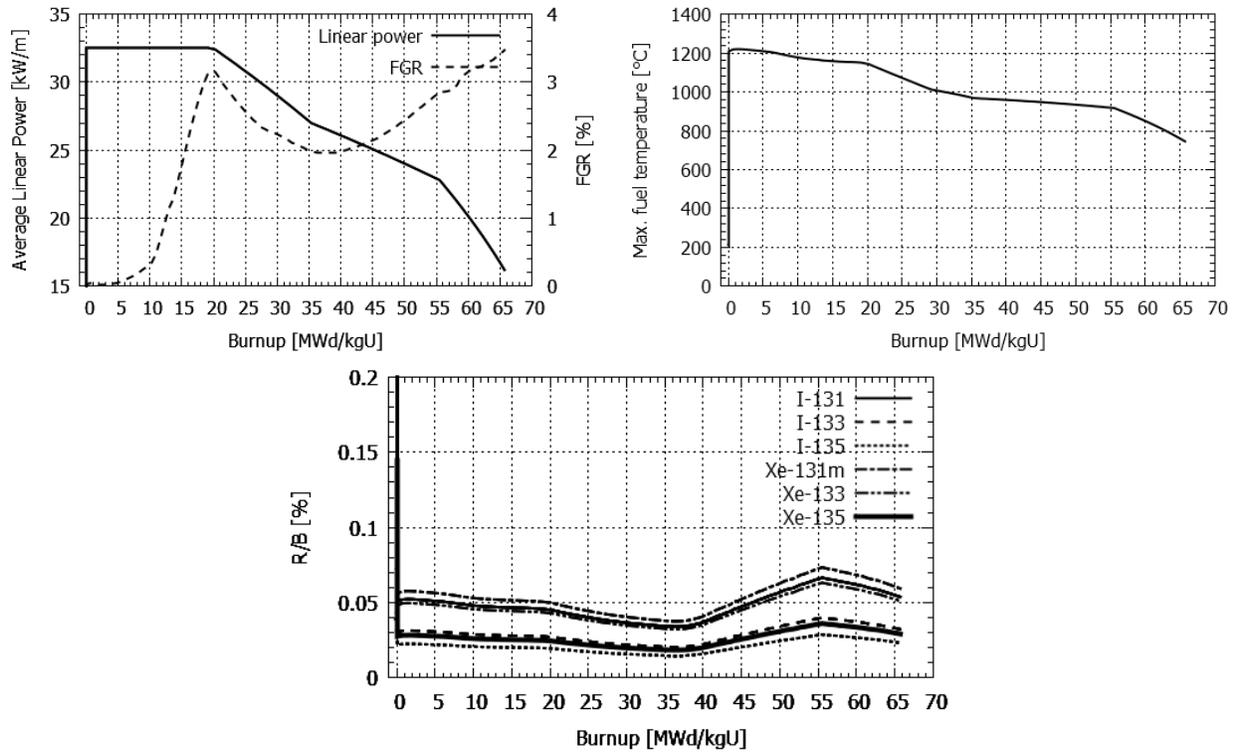


Figure 4. Average linear power and FGR (top left), and maximum fuel temperature (top right) from VTT-ENIGMA for the rod with the power corresponding to the linear power limit. The percentages of short-lived nuclides (bottom) released to the gas gap according to the ANS-5.4 standard [1] implemented into VTT-ENIGMA.

Table 1. Nuclides included to the study. [1, 3]

	Nuclide	$t_{1/2}$ [d, h or y]	λ [s^{-1}]	α
Short half-life $6 \text{ h} < t_{1/2} < 60 \text{ d}$	I-131	8.04 d	9.98E-7	1.0
	I-133	20.8 h	9.26E-6	1.21
	I-135	6.72 h	2.86E-5	1.0 (?)*
	Xe-131m	11.9 d	6.73E-7	1.0 (?)*
	Xe-133	5.243 d	1.53E-6	1.25
	Xe-135	9.10 h	2.12E-5	1.85
	Te-118	6.00 d	1.34E-06	1.0 (?)*
	Te-119	16.05 h	1.20E-05	
	Te-119m	4.70 d	1.71E-06	
	Te-121	19.17 d	4.18E-07	
	Te-125m	57.40 d	1.40E-07	
	Te-127	9.35 h	2.06E-05	
	Te-129m	33.6 d	2.39E-07	
	Te-131m	33.25 h	5.79E-06	
	Te-132	3.20 d	2.50E-06	
	Cs-127	6.25 h	3.08E-05	
	Cs-129	32.06 h	6.01E-06	
	Cs-131	9.689 d	8.28E-07	
Cs-132	6.480 d	1.24E-06		
Cs-136	13.16 d	6.10E-07		
Long half-life $t_{1/2} > 1 \text{ y}$	Cs-134	2.1 y		
	Cs-137	30.2 y		
	Kr-85	10.7 y		

Table 2. Release of the radionuclides. Comparison of the average release values between the highest power bundle and the highest burnup bundle is made, and the higher value of those two is marked in grey.

	Release [%]	Highest power bundle			Highest burnup bundle			Rod with the power corresponding to the linear power limit
		min.	max.	ave.	min.	max.	ave.	
short half-life	I-131	*0.02366	*0.02517	*0.02430	*0.02269	*0.02862	*0.02372	*0.05561
	I-133	0.01348	0.01431	0.01383	0.01293	0.01393	0.01328	0.03238
	I-135	0.00969	0.01029	0.00995	0.00930	0.01001	0.00955	0.02328
	Xe-131m	*0.02610	*0.02778	*0.02682	*0.02504	*0.03159	*0.02617	*0.06137
	Xe-133	*0.02248	*0.02392	*0.02309	*0.02157	*0.02720	*0.02254	*0.05285
	Xe-135	0.01218	0.01294	0.01251	0.01169	0.01259	0.01200	0.02927
	Te-118	*0.02199	*0.02340	*0.02259	*0.02109	*0.02661	*0.02205	*0.05169
	Te-119	0.01205	0.01279	0.01236	0.01155	0.01245	0.01187	0.02894
	Te-119m	*0.02069	*0.02201	*0.02125	*0.01985	*0.02503	*0.02074	*0.04863
	Te-121	*0.02940	*0.03128	*0.03020	*0.02820	*0.03557	*0.02947	*0.06911
	Te-125m	*0.03867	*0.04115	*0.03973	*0.03710	*0.04679	*0.03877	*0.09091
	Te-127	0.01052	0.01117	0.01080	0.01009	0.01087	0.01037	0.02528
	Te-129m	*0.03382	*0.03600	*0.03475	*0.03245	*0.04093	*0.03391	*0.07952
	Te-131m	0.01445	0.01535	0.01483	0.01386	0.01493	0.01423	0.03471
	Te-132	0.01782	0.01892	0.01829	0.01709	0.01842	0.01755	0.04281
	Cs-127	0.00952	0.01010	0.00977	0.00913	0.00983	0.00937	0.02286
	Cs-129	0.01432	0.01521	0.01470	0.01374	0.01480	0.01411	0.03440
	Cs-131	*0.02479	*0.02638	*0.02546	*0.02378	*0.02999	*0.02485	*0.05827
Cs-132	*0.02241	*0.02385	*0.02303	*0.02150	*0.02712	*0.02247	*0.05270	
Cs-136	*0.02676	*0.02848	*0.02749	*0.02567	*0.03238	*0.02683	*0.06291	
long half-life	FGR (Kr-85)	0.38	0.75	0.51	0.77	1.49	1.03	3.47
	FGR * $\sqrt{2}$	0.54	1.06	0.73	1.09	2.11	1.46	4.91
	FGR / $\sqrt{3}$	0.22	0.43	0.30	0.44	0.86	0.60	2.00
	=0.19*FGR +0.74	0.81	0.88	0.84	0.89	1.02	0.94	1.40

Table 3. Maximum release of radionuclides during the base irradiation. Comparison of the maximum release values between the highest power bundle and the highest burnup bundle is made, and the higher value of those two is marked in grey.

	Release [%]	Highest power bundle	Highest burnup bundle	Rod with the power corresponding to the linear power limit
		max.	max.	max.
short half-life	I-131	0.03075	0.02958	0.06664
	I-133	0.01848	0.01778	0.04005
	I-135	0.01329	0.01278	0.02879
	Xe-131m	0.03393	0.03264	0.07354
	Xe-133	0.02922	0.02811	0.06333
	Xe-135	0.01671	0.01607	0.03620
	Te-118	0.02859	0.02750	0.06196
	Te-119	0.01652	0.01589	0.03580
	Te-119m	0.02690	0.02587	0.05829
	Te-121	0.03822	0.03676	0.08284
	Te-125m	0.05028	0.04836	0.10897
	Te-127	0.01443	0.01388	0.03128
	Te-129m	0.04398	0.04230	0.09531
	Te-131m	0.01982	0.01906	0.04295
	Te-132	0.02444	0.02351	0.05296
	Cs-127	0.01305	0.01255	0.02828
	Cs-129	0.01964	0.01889	0.04255
	Cs-131	0.03222	0.03100	0.06983
	Cs-132	0.02914	0.02803	0.06315
Cs-136	0.03479	0.03346	0.07539	