Evaluation and Parameter Analysis of Burn up Calculations for the Assessment of Radioactive Waste – 13187

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ABSTRACT

Burn up calculations facilitate a determination of the composition and nuclear inventory of spent nuclear fuel, if operational history is known. In case this information is not available, the total nuclear inventory can be determined by means of destructive or, even on industrial scale, non-destructive measurement methods. For non-destructive measurements however only a few easy-to-measure, so-called key nuclides, are determined due to their characteristic gamma lines or neutron emission. From these measured activities the fuel burn up and cooling time are derived to facilitate the numerical inventory determination of spent fuel elements.

Most regulatory bodies require an independent assessment of nuclear waste properties and their documentation. Prominent part of this assessment is a consistency check of inventory declaration. The waste packages often contain wastes from different types of spent fuels of different history and information about the secondary reactor parameters may not be available. In this case the so-called characteristic fuel burn up and cooling time are determined. These values are obtained from a correlations involving key-nuclides with a certain bandwidth, thus with upper and lower limits. The bandwidth is strongly dependent on secondary reactor parameter such as initial enrichment, temperature and density of the fuel and moderator, hence the reactor type, fuel element geometry and plant operation history.

The purpose of our investigation is to look into the scaling and correlation limitations, to define and verify the range of validity and to scrutinize the dependencies and propagation of uncertainties that affect the waste inventory declarations and their independent verification. This is accomplished by numerical assessment and simulation of waste production using well accepted codes SCALE 6.0 and 6.1 to simulate the cooling time and burn up of a spent fuel element. The simulations are benchmarked against spent fuel from the real reactor Obrigheim in Germany for which sufficiently precise experimental reference data are available.

INTRODUCTION

The quality control of radioactive waste compounds has always been an integral part of the German safety and quality assurance concept for the disposal of radio-toxic waste. Until 1995 reprocessing of nuclear waste had been legally obligatory, then voluntary. Specific and dedicated waste conditioning methods and technologies have engaged at the reprocessing sites in France, Germany and the UK. In 2005 the German atomic act has been revised for the treatment and conditioning of high-level waste (HLW) and transport of spent fuel and reprocessing abroad has been banned. Currently, all spent fuel is temporarily stored at

the nuclear power plant (NPP) sites until further decisions are made on how to proceed and dispose of the accumulating amount of used nuclear fuel (UNF) and HLW.

The German Federal government is in charge of defining societal targets, safety standards and final disposal of German radio-toxic waste from nuclear facilities, namely the nuclear power stations, research and medical institutions and other facilities that legally deal with radioactive or fissile material. In addition, there is a vast program for the decommissioning of nuclear installations. All accumulated nuclear waste of the past has to be disposed of in a professional and safe manner. Therefore, a low-level waste (LLW) and intermediate-level waste (ILW) repository for radio-toxic waste with negligible heat generation is under construction, while an operational repository for German HLW is ways ahead from now. While some German waste is currently still reprocessed abroad and has to be repatriated into dedicated national interim storage facilities spent fuel from the more recent past and current operation of German NPPs is stacked in local on-site interim storages waiting for its further processing or final direct disposal.

The product quality control and quality assurance for reprocessed nuclear waste treatment, conditioning and packaging has never been controversial and QA-methods have gradually been developed and implemented at a very high standard. As for spent fuel accumulated since 2005 currently piling up at interim storage facilities in decay pools and transport-storages casks, only little thought has been devoted on how to check independently the actual nuclear inventory of disposable spent fuel. Mainly burn up and cooling time calculations have been employed. However the current German acceptance criteria for a virtual HLW repository require an independent verification of the crucial nuclide inventory and therefore, the waste stream properties and time dependent waste product characteristics need be established and checked [1].

Herewith, we shall present results from numerical simulation of nuclear inventory parameters and their time-dependent propagation which are affected and influenced by secondary reactor parameters. The role and amount of many declarable nuclides are usually determined from correlation laws that allow to associate these declarable nuclides with easy-to-measure key nuclides. Again, the individual fuel element history and secondary reactor parameters affect the bandwidth parameter uncertainties and validity of the applicable correlation laws. This is what we have investigated in more detail, and as a reference case, a nuclear fuel element of the German nuclear power plant Obrigheim has been scrutinized. The software, used for the burn up calculation is the worldwide accepted program SCALE version 6.0 and 6.1. All calculations include 2D-simulations of one randomly selected fuel element. The bandwidth for activities of selected nuclides is estimated by the means of parameter variation. The validation of these simulation results is based on our own benchmark analysis. This work may be regarded as a basis for the required development of nuclear inventory verification tools and can be generalized for any fuel from any reactor facility.

PROGRAM BENCHMARK

Reference data

As a first step the applied simulation programs were benchmarked against experimental data for nuclear inventory of spent fuel elements from nuclear power plant Obrigheim. The required experimental data are available in SFCOMPO-Database [2]. The data come from two independent institutions: the “Joint

Table I. Calculation vector from SFCOMPO data base

Variable	Unit	Variable	Unit	Variable	Unit
1. Cs-134/Cs-137[1]	Bq/Bq	18. Total Pu/Total U	kg/kg	35. Cs-134[1]	kg
2. Cs-137/U-238[1]	mol/mol	19. U-235/Total U	kg/kg	36. Cs-137[1]	kg
3. Eu-154/Cs-137[1]	Bq/Bq	20. (U-235/Total U)/ (U-235/Total U init)	kg/kg	37. Eu-154[1]	kg
4. Kr-83/Kr-86[2]	mol/mol	21. U-235/U-238	mol/mol	38. Pu-238	kg
5. Kr-84/Kr-86[2]	mol/mol	22. U-236/Total U	kg/kg	39. Pu-239	kg
6. Kr-85/Kr-86[2]	mol/mol	23. U-236/U-238	mol/mol	40. Pu-240	kg
7. Nd-148/U-238	mol/mol	24. U-238/Total U	kg/kg	41. Pu-241	kg
8. Pu-238/Total Pu	kg/kg	25. Xe-131/Xe-134[2]	mol/mol	42. Pu-242	kg
9. Pu-239/Total Pu	kg/kg	26. Xe-132/Xe-134[2]	mol/mol	43. Total Pu	kg
10. Pu-239/U-238	mol/mol	27. Xe-136/Xe-134[2]	mol/mol	44. Total Pu, U	kg
11. Pu-240/Pu-239	mol/mol	28. Burn up (by Cs-137 Destructive method)[1]	GWd/MTU	45. Total U	kg
12. Pu-240/Total Pu	kg/kg	29. Burn up (by Cs-137 Non-destructive method)[1]	GWd/MTU	46. U-235	kg
13. Pu-241/Pu-239	mol/mol	30. Burn up (by Nd-148 method)	GWd/MTU	47. U-235 Depletion	kg
14. Pu-241/Total Pu	kg/kg	31. Burn up (by Theoretical)	GWd/MTU	48. U-236 Build-up	kg
15. Pu-242/Pu-239	mol/mol	32. Am-241[1]	kg	49. U-238	kg
16. Pu-242/Total Pu	kg/kg	33. Cm-242	kg	50. U-235 Depletion	kg
17. Total Pu/Total U	mol/mol	34. Cm-244	kg		

[1] Only from “Joint Research Center” (Ispra)

[2] Only from Karlsruhe Institute of Technology (Karlsruhe)

Research Center" in Ispra and Nuclear Research Center Karlsruhe (today Karlsruhe Institute of Technology).

Altogether 50 variables were measured or calculated by both the institutions. These values are summarized in Table I. There is no detailed description of the individual measurement methods in SFCOMPO data base. It is however mentioned, that decay corrections have been performed for all the results and the listed values correspond to the cooling time equal to zero. This time corresponds to the facility shutdown to change or remove the fuel and marks the end of irradiation cycle for the corresponding used fuel element.

Due to differences in measurement methods, different units are used for different experimental values. All values are normalized to the metric tonne of the heavy metal of an initial material (MTU initial).

Nuclear power plant Obrigheim – Layout of reactor and fuel element

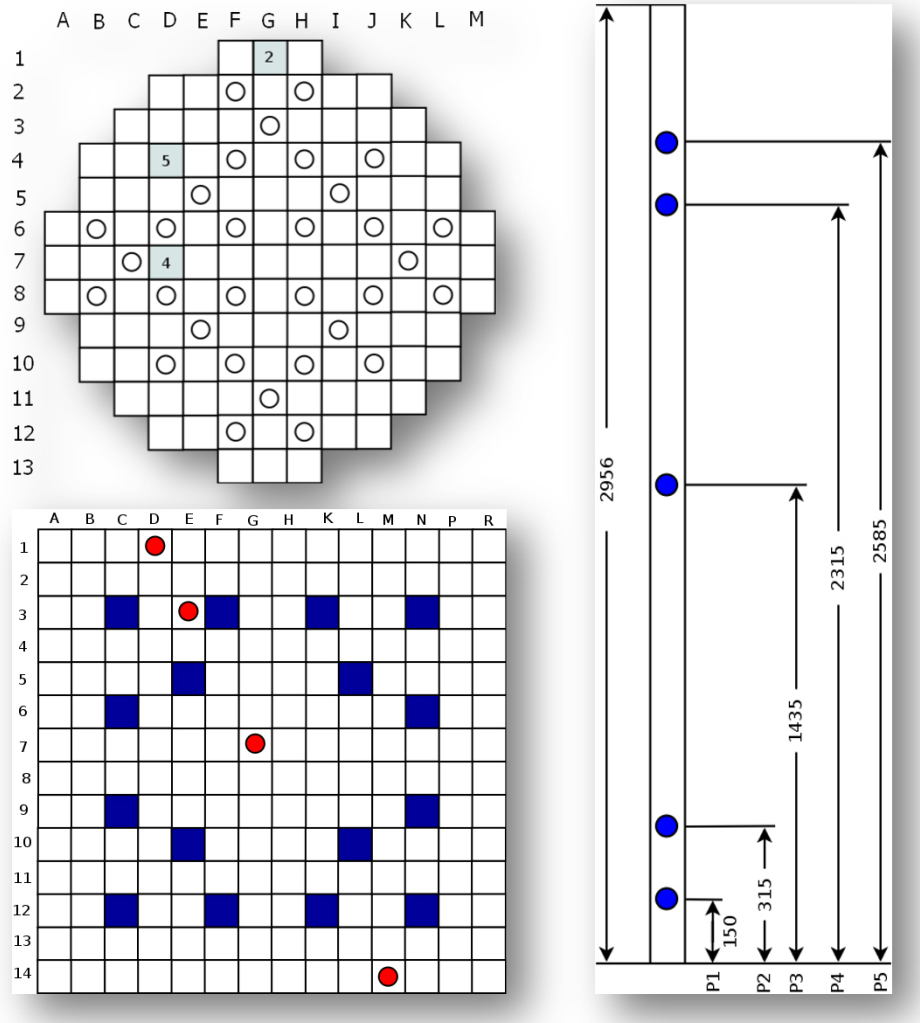

Fig. 1. Layout of the Obrigheim reactor, fuel element and fuel rod [2]

The online database SFCOMPO contains results of destructive measurements performed on spent fuel elements from a pressurized-water reactor (PWR) of the nuclear power plant Obrigheim in Germany. The measurements were performed on several fuel elements with different individual operation histories. For this work the data for fuel element BE 124 (according to SFCOMPO notations) were used.

The fuel element layout and the operating history were taken from the Ispra report [3], the SFCOMPO data base [2] and "International Journal for Nuclear Power" [4]. The upper left frame of Figure 1 displays the transversal configuration of the reactor [2]. The fields marked with grey represent the position of considered fuel element during different operational cycles.

The modeled fuel element BE 124 encloses a right-angled grid with 14 × 14 rod positions (Figure 1, lower left frame). The blue cells indicate the control rod guide tubes. The measurements were carried out on fuel rods (red points) on different depths, hence, there are several different measurement data of the same fuel rod. The right frame of Figure 1 shows the position of the measurement points in a fuel rod. Besides, the various measuring depths are given in mm and are the same for every fuel rod. As a reference Rod G7 (Figure 1, lower left frame), measurement position p3 was used (further "RodG7p3").

First of all the comparison was performed between two measurement sets. The Figure 2 displays the ratio between the measurements performed by two institutions. Most of the results agree within 5%. Some variables show a larger deviation, for example over 45% for Cm-242. However, there are no measurement uncertainties listed in SFCOMPO database and it is therefore difficult to conclude on the agreement or disagreement of these two measurements.

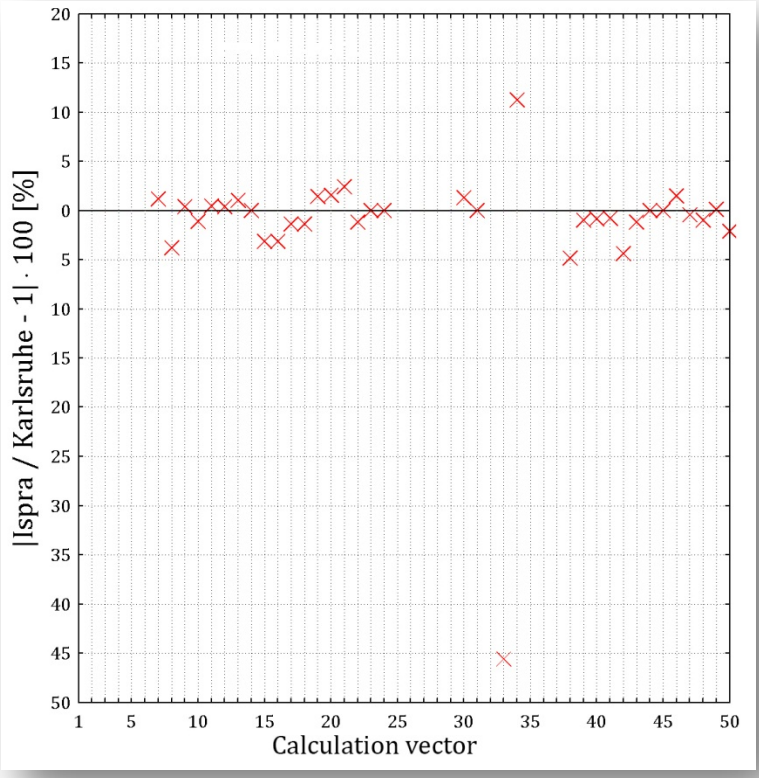

Fig. 2. Relative deviation of two measurement sets for a fuel element (BE 124) from the NPP Obrigheim in Germany. Measurement position RodG7p3, BU = 31.5 GWd/MTU

The Table II contains main modeling parameters of the Obrigheim reactor and its fuel element.

Table II. Summary of parameters for the NPP Obrigheim reactor and fuel element BE 124

Considered operating time	1970 – 1974
Type of fuel element	Siemens 14 × 14
Fuel	UO_2
Initial enrichment of U-235	3.0 %
Medium burn up	29 GWd/MTU
Fuel density	10.408 g/cm^3
Moderator density (H_2O)	0.7283 g/cm^3
Fuel tempeature	900 K
Moderator tempeature	570 K

Benchmark

For the simulation of experimental data two models were used. The first model ("arp") represents a simplified approach. For the calculation of the nuclear vector only the most important parameters, such as fuel element type, initial enrichment, burn up and moderator density, were taken into account. Such approach allows the user to create an input file and perform swift and fast calculations. The second model ("newt") is more complicated, but also more precise. The "newt"-model calculation of nuclear vectors requires additional parameters, those of the "arp"-model and hitherto more detailed information about the fuel, moderator and operation history. In this case the calculation may take up to several days. The comparison of two models is performed in order to find optimal ratio between time, costs and required precision.

The calculation was performed with two versions of SCALE software 6.0 and 6.1. For each version two models were applied. Table III summarizes main parameters of both models.

Table III. Properties of the two selected models

Name	arp	newt
Measurement point	RodG7p3	RodG7p3
Burn up	29 GWd/MTU	31.5 GWd/MTU
Solver	Arp	Newt
Library	ENDF/B-V	ENDF/B-VI (SCALE 6.0) ENDF/B-VII (SCALE 6.0 and 6.1)
Number of the energy groups	44	238
Grid	–	4 x 4 per Cell

The modeling of the operation history is based on SFCOMPO data. Comparing absolute values of test case “arp” with experimental data gives an overview on the order of magnitude for differently determined values (Figure 3).

The experimental results are marked in black points for Ispra and red triangles for Karlsruhe, respectively. The blue symbols show the simulation results of the same “arp” case, “×” for SCALE 6.0 and “+” for SCALE 6.1 calculations, respectively.

In most of the cases the numerical simulation agrees pretty well with the experimental data. However, several values show large deviation, as for Cs-137/U-238, Cm-242, Cm-244 and above all Am-241 with the largest relative deviation.

The Figure 4 displays the deviations between measured data and the “arp”-model (left frame) and the “newt”-model (right frame).

The comparison between numerical and experimental data is shown: Ispra in black with SCALE 6.0 simulation marked with “O” and SCALE 6.1 results with “●”. The red symbols compare the Karlsruhe experimental data with SCALE 6.0 “×” and SCALE 6.1 “+” simulations.

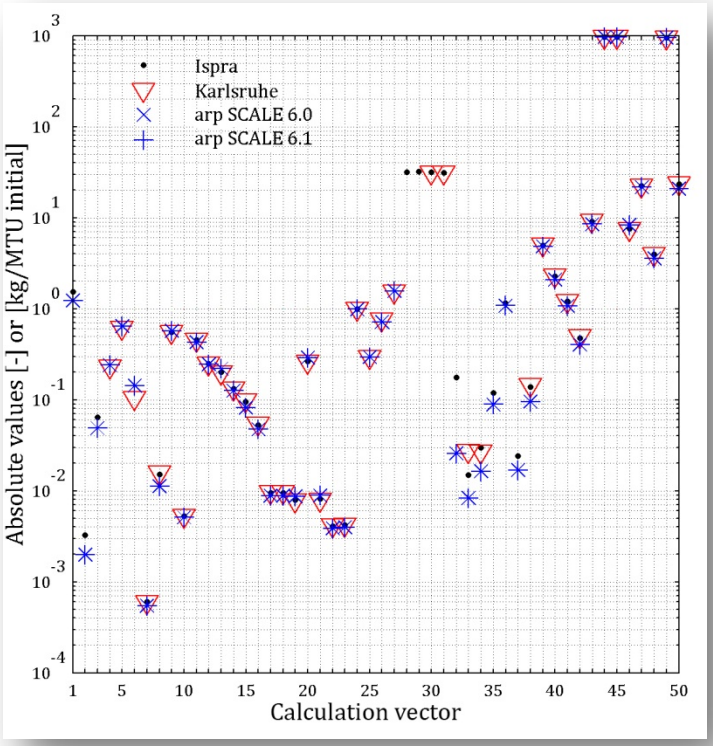

Fig. 3. Comparison between measured and simulated (arp model) data for a fuel element (BE 124) of the NPP Obrigheim. Measurement position RodG7p3, BU = 31.5 GWd/MTU

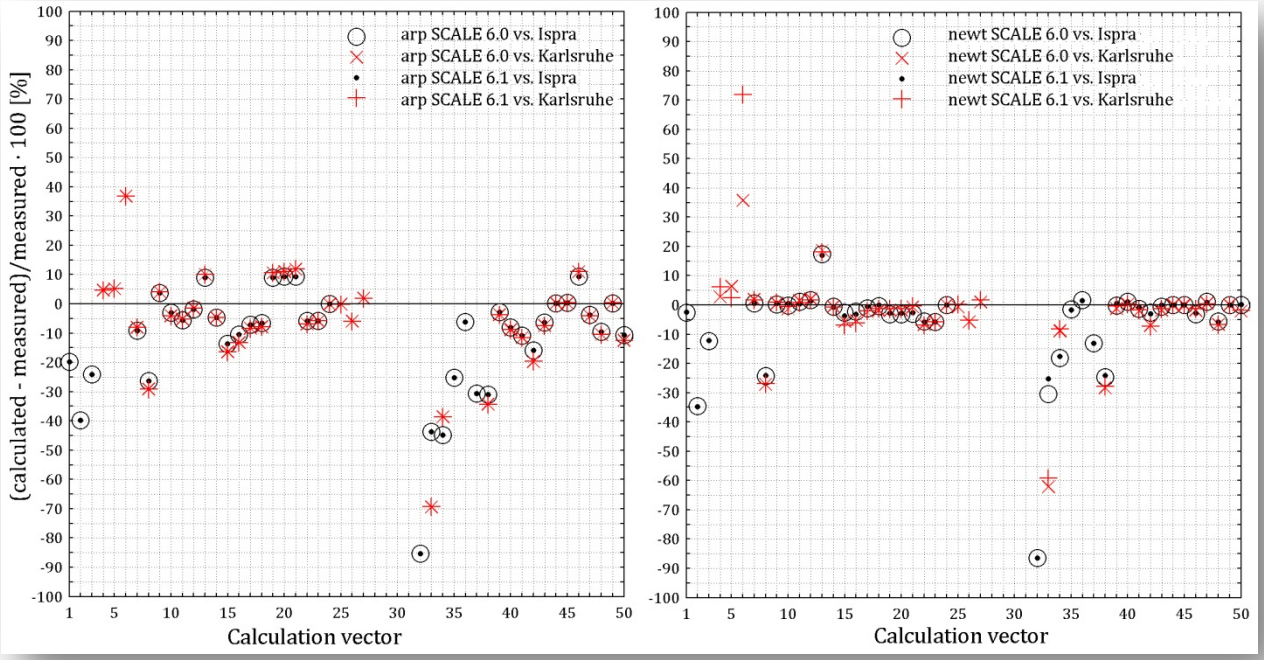

Fig. 4. Deviation between measured and simulated (left frame for arp model and right frame for newt model) data for the fuel element (BE 124) of the NPP Obrigheim. Measurement position RodG7p3, BU = 31.5 GWd/MTU

In case of the „arp"-model the both SCALE versions produce the same results, because the modeling does not differ on this level and the both SCALE versions use the same libraries. In most of the cases deviations between numerical simulations and experimental reference data are within 20% accuracy. This is a good result for a simple model without complex specification. However, several values differ obviously (up to 85% for Am-241). As one can see in the left frame of Figure 4 the deviations for Cm-242 are 70% and 45% for Karlsruhe and Ispra respectively. It is difficult to draw solid conclusions about such deviations without knowing the uncertainties of these measurements.

The right frame of Figure 4 shows the deviations between experimental results and numerical calculations with the "newt"-model. Since this model is more detailed, many values were significantly improved and are within ± 10% limit. The problematic results (Am-241 etc.) remain unchanged. In additional a new question mark arises for the Kr-85/Kr-86 values, because the numerical results are different for the two SCALE versions. A possible explanation is the use of different libraries.

VARIATIONAL ANALYSIS OF THE CORRELATION BANDWIDTH

Correlations are often used for calculations of nuclear vector composition in case destructive assay methods are not applicable. They may also be used for estimations, when fuel operational history is unknown or other important information is missing.

Secondary reactor parameters

Primary reactor parameters have immediate impact on the composition and variation of the nuclide vector. These are the cross sections and neutron flux. The secondary parameters are those factors affecting the nuclide vector indirectly through the change of the primary reactor parameters. The secondary reactor parameters are:

- Fuel properties: initial enrichment, density, temperature;
- Moderator properties: density, temperature.

The influence of the secondary reactor parameters on the selected key nuclides

Non-destructive waste characterization methods are often used. In this case only the so-called key nuclides are determined experimentally.

The key nuclide is such a radionuclide that can be detected easily through its characteristic gamma rays by means of non-destructive metrology [5]. The activities of nuclides, which are difficult or impossible to measure, can be determined numerically through correlations with appropriate key nuclides [5]. If the operational history for a fuel element is unknown or if the waste package contains wastes from different fuel elements with different characteristics it is possible to introduce the mean or characteristic variables: burn up and cooling time. The following radionuclides are often used as the key nuclides for the determination of the burn up and the cooling time: Cs-134, Cs-137, Eu-154 [6]. However, in case of limited information about spent fuel such correlations may have large uncertainties. The aim of this work is to estimate the influence of secondary reactor parameters on correlation uncertainties.

The influence of the fuel and moderator properties on the nuclide composition is investigated for burn ups of up to 60 GWd/MTU. The properties of a fuel element, such as geometry, composition of the fuel and moderator parameters correspond to the properties of a fuel element of the Obrigheim reactor (Table II). The operation history comprises three cycles and covers the period of four years in total. There are 24 models defined for the parameter analysis. The variation was performed for the following secondary reactor parameters:

- Initial enrichment: 1.5%,3%, 4.5%
- Fuel temperature: 900 K, 1800 K
- Moderator temperature: 556 K, 586 K
- Fuel density: 90%, 100% respective of the physical fuel density (Table I)
- Moderator density: 90%, 110% respective of the physical moderator density (Table I)

The Figure 5 presents the results of the parameter analysis for Eu-154. The bandwidth for Eu-154 shows, that its build up is very sensitive to the parameter change in a section with lower burn up. The bandwidth meets its minimum at roughly 50 GWd/MTU. The average bandwidth for the whole burn up range is about 39%. However, the intermediate and high burn ups are in the practice more relevant and that is why the average bandwidth for Eu-154 can be limited to 26%, if any burn up less than 20 GWd/MTU is neglected. For low and intermediate burn ups up to approximately 35 GWd/MTU the bandwidth is dominated by uncertainty of the initial enrichment, while at higher burn ups the influence of moderator density is the dominant factor. The influence of other parameters is weaker. It is obvious from Figure 5 that the activity of Eu-154 is not a linear function of the burn up. As Eu-154 is not a direct fission product and as it is screened by stable Sm-154 its build-up is therefore somewhat delayed. Thus, Eu-154 is bred

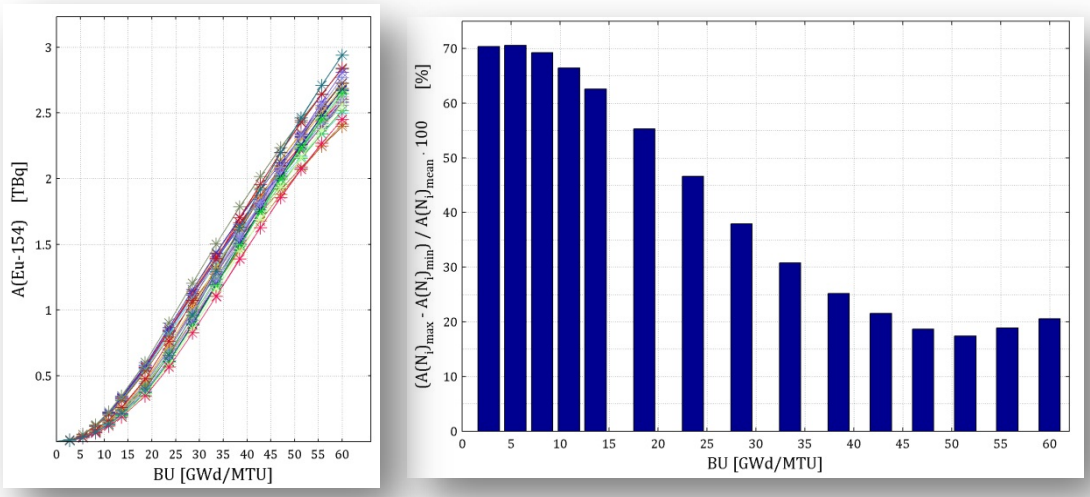

Fig. 5. Build-up of Eu-154 as a function of burn up (left frame) and corresponding bandwidth (right frame)

mainly by means of the neutron capture by Eu-153. The maximum production rate of Eu-154 is achieved at burn up of ~50 GWd/MTU, thereafter it is somewhat reduced. The reason is neutron capture by Eu-154, with a cross section of 1500 barn [7]. This behaviour can explain the large bandwidth at the beginning and the gradient of the bandwidth after the minimum, and this interpretation is supported by the parameter variation of a number of models.

Figure 6 shows the result of the parameter analysis for the build-up the key nuclides Cs-134 and Cs-137. The bandwidth for Cs-134 is again dominated by the uncertainty of the initial enrichment and moderator density, likewise Eu-154. The difference here is that the initial enrichment plays a role for all burn ups. The contribution to the bandwidth by any other parameter is weaker. The bandwidth for Cs-137 is basically insensitive to any parameter variations, because the production of this nuclide depends on the burn up, only.

The build-up of Cs-134 at the beginning follows the same pattern as for Eu-154. The nuclide Cs-134 is screened off by the stable nuclide Xe-134. However, the thermal neutron capture cross section of 140 barn for Cs-134 is rather low [7]. Thus, the build-up rate of Cs-134 is not reduced at higher burn ups.

In contrast to Eu-154 and Cs-134, the nuclide Cs-137 is a direct fission product. The build-up of Cs-137 is linear therefore with the burn up and does not depend at all on any secondary reactor parameter, as can be concluded from the extremely narrow bandwidth. Due to low capture cross-section of 0.2 barn there is no reduction in build-up rate. The precursor nuclide Cs-136 has also a very low capture cross section (1.3 barn), and that excludes the build-up of Cs-137 through neutron capture [7].

The build-up curves presented here qualitatively agree with calculations performed for other light water reactors [8].

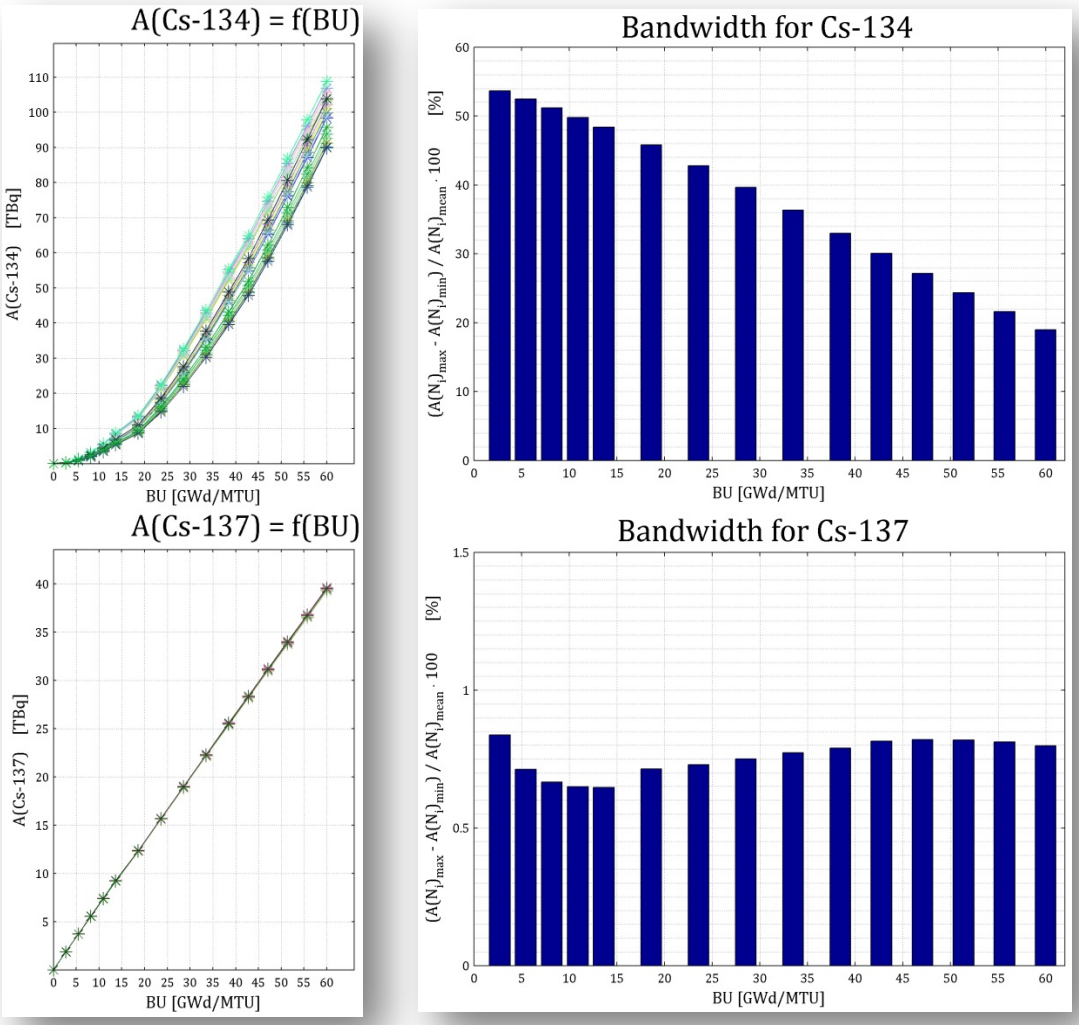

Fig. 6. Build-up of Cs-134 (upper frame) and Cs-137 (lower frame) as functions of burn up and corresponding bandwidths

Burn up correlation

In common practice different burn up correlations are used. One of the most common is the Eu-154/Cs-137 ratio [8]. Usually the dependence of the Eu-154 activity on burn up is described by a parabolic square function and the dependence of the Cs-137 activity by a linear function. Therefore burn up is commonly considered being a linear function of Eu-154/Cs-137 ratio.

The simulation results (Figure 7) show, that the linearity of Cs-137 activity is reproduced very well. Whereas the quadratic behaviour of the Eu-154 nuclide complies with reality only for intermediate burn up of < 30 GWd/MTU (Fig. 7 or [8]).

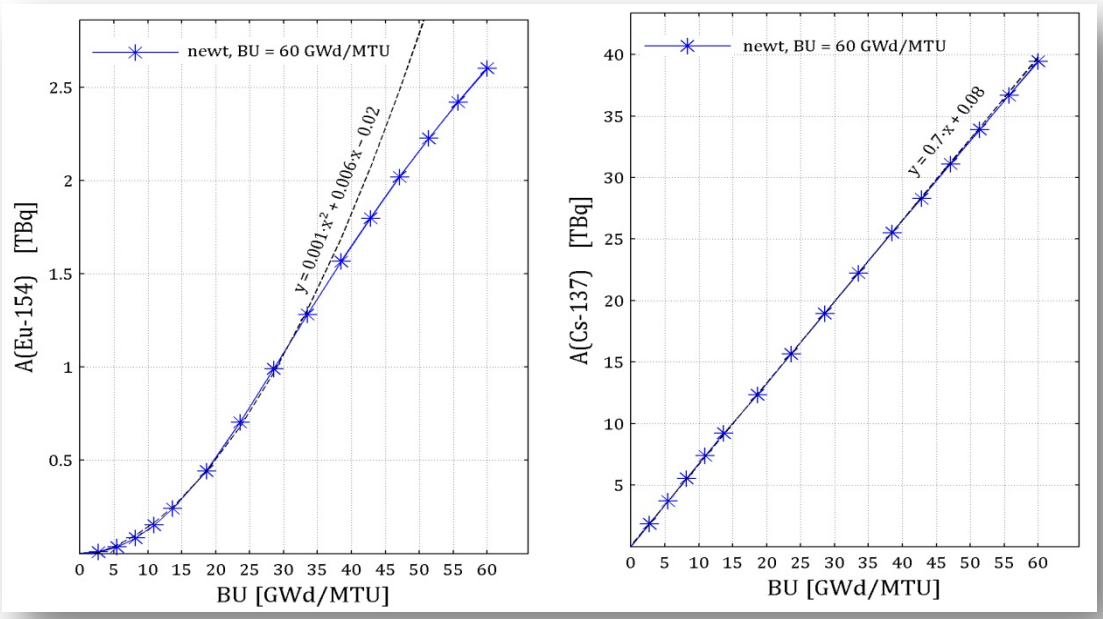

Fig. 7. Build-up of Eu-154 (left frame) and Cs-137 (right frame) as a function of burn up. Calculations are performed with SCALE 6.1 using newt model for burn ups < 60 GWd/MTU

The strong dependence of Eu-154 build-up on secondary reactor parameters leads to a large bandwidth of the associated burn up correlation (Figure 8). Moreover, one can see that correlation is valid only for burn ups of up to 45 GWd/MTU. For higher burn ups different methods should be considered. The uncertainty of the burn up correlation due to secondary reactor parameters variation is estimated to be between 60 - 85%. The largest contribution at low and intermediate burn ups of up to 35 GWd/MTU is due to influence of an unknown initial enrichment on Eu-154 production (s. Figure 5 and Figure 6 lower frames). For higher burn ups the production of Eu-154 does not obey the quadratic law and the correlation is not very stringent (s. Figure 7). This leads to an increased bandwidth. This is quite a large uncertainty and in many cases higher precision is required. Therefore, it is recommended to use information of the initial fuel enrichment, if possible.

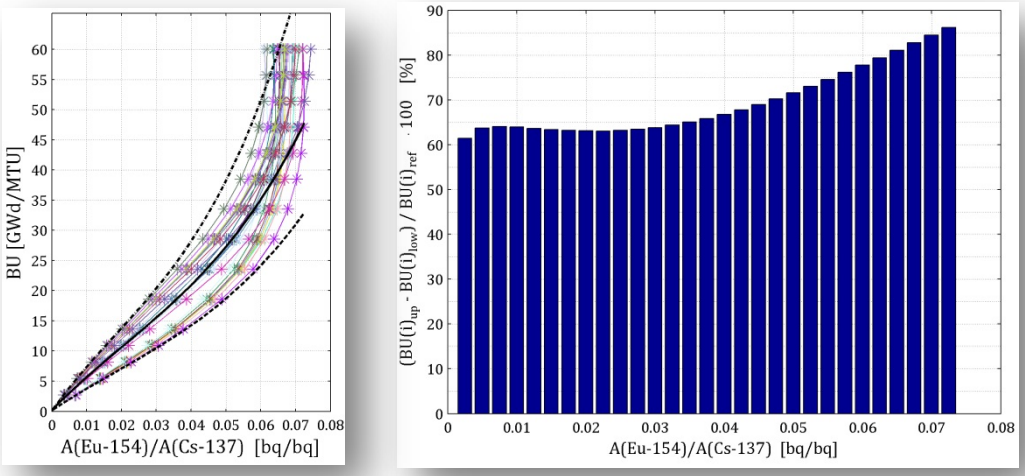

Fig. 8. Burn up as a function of the ratio Eu-154/Cs-137 (left frame) and the corresponding bandwidth (right frame). Calculations are performed with SCALE 6.1 using newt model for burn ups up to 60 GWd/MTU

Cooling time correlation

As in the case of burn up, many different correlations can be used for the cooling time determination. The usual approach is to use two easy measurable nuclides with different half-lives. The ratio of their activities is usually independent on burn up and changes only with cooling time. Possible candidates are: Pr-144/Cs-137, Rh-106/Cs-137, Rh-106/Pr-144 and Eu-154/Cs-134 [6]. Each one of them has some advantages and disadvantages and the most appropriate one has to be selected upon the application considerations. In our case the important quality is a validity range, which is limited by nuclide half-lives. Here the validity range of correlations involving Pr-144 or Rh-106 is about 10 years, whereas the one with Cs-134 may by applied for approximately 20 years. Therefore it has been selected for our analysis.

The validity range of this correlation is approximately 20 years which is limited by the short-lived nuclide Cs-134 of 2.1 years half-life and which is almost completely degraded after about 10 half-lives.

The dependence of the cooling time correlation on parameter variations with the respective lower and upper boundary is shown in the Figure 9. This diagram reflects the whole parameter spectrum.

In this case the bandwidth remains constant (the range between two boundaries in the y-direction) with increasing nuclides ratio value. It amounts to almost 400 days by the observation of the whole parameter range. The aforementioned unknown initial enrichment and moderator density is mainly responsible for the uncertainty. If however, the initial enrichment is well known, the bandwidth is limited to 200 days.

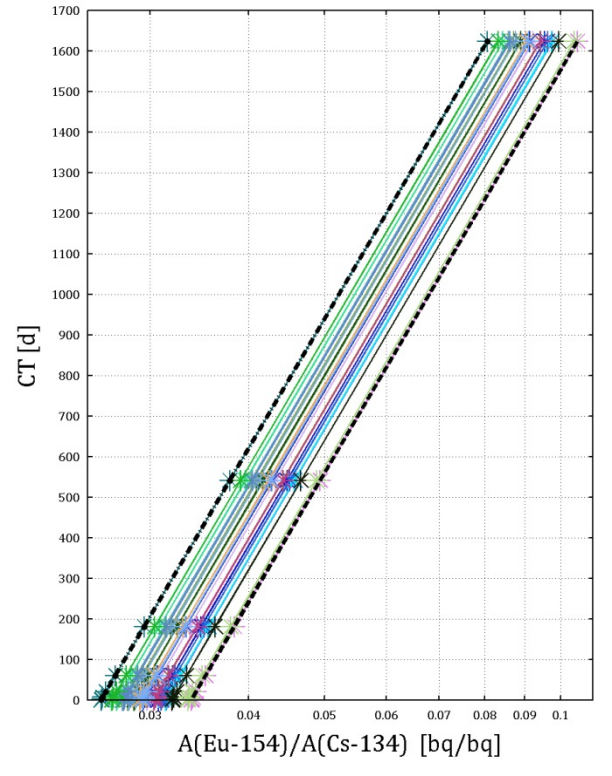

Fig. 9. Cooling time is as a function of the Eu-154/Cs-134 ratio. Calculations are performed with SCALE 6.1 using newt model for burn ups up to 60 GWd/MTU

SUMMARY

Correlations between easy-to-measure key nuclides and other declarable nuclides are not as solid as one would wish. They must be justified and verifiable for each individual waste stream, and their uncertainties are subject of specific considerations. Here, we have scrutinized commonly used correlations to assess the cooling time and burn up of spent reactor fuel and have found the range of validity being limited and the uncertainties being influenced by a number of secondary reactor parameters. This is not so surprising per se, but these limitations of commonly used correlations should be implemented into the proof tools to check and verify waste inventory declarations. And uncertainty propagation must be considered for the sake of waste property quality assessment.

To ensure that our numerical assessment of correlation dependencies on primary fuel information, secondary reactor parameters and their uncertainties produces trustworthy results, we have successfully benchmarked our programs using both, SCALE 6.0 and 6.1, against an experimental data for real spent fuel element of the German NPP Obrigheim from SFCOMPO database. Most of calculated values agree with measured ones within 5% of accuracy. Some of the calculated values for actinides deviate significantly from the experimental values, up to 85% (Am-241). There is also a large discrepancy between the different SCALE versions in case of the Kr-85/Kr-86 activity ratio. One difficulty of the benchmark is insufficient information on the reference data, namely the accurate operating history or the dates of the measurements. These data have direct influence on simulation accuracy.

The parameter analysis and the determination of the bandwidth for burn up dependent build-up of individual nuclides show clearly a very strong sensitivity to the variation of secondary reactor parameters. This seems so in particular for the studied key nuclides Eu-154, Cs-134. Apparently, the bandwidth is most sensitive to the variation of the initial fuel enrichment and the reactor's moderator density. The build-up of Cs-137 is strictly linear with the burn up and insensitive to any other parameter variations.

The analysis of the burn up and cooling time correlations serves as very useful tool to estimate the nuclide vector composition and its uncertainties. However, these two commonly used correlations are applicable only in a limited range, because of the specifics of the Eu-154 build-up for the burn up correlation and the short-lived nuclide Cs-134 for the cooling time correlation.

The knowledge of the secondary reactor parameters, especially initial enrichment and moderator density, is advisable for the optimization of the bandwidth and uncertainties. In order to make a method more general it is necessary to extend and complement it with calculations for other reactor types.

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