On the Influence of the Power Plant Operational History on the Inventory and the Uncertainties of Radionuclides Relevant for the Final Disposal of PWR Spent Fuel – 15149

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ABSTRACT

A mandatory requisite for official granting safe final disposal approval of SNF is the comprehensive specification and declaration of the nuclear inventory in SNF by the NPP-operators. The whole radiochemical characterization of each spent fuel assembly hides many difficulties such as very high radiation burden, destruction method, the inhomogeneous burn-up of fuel assembly, very high cost. Thus, non-destructive characterization is usually applied on industrial scale. In this case only several nuclides can be measured, and the rest is calculated using simulations and typical irradiation history. Like any other German rad-waste acceptance requirements all these declarations will have to be checked and verified by an independent inspector. In this verification process not only values of radionuclide (RN) activities are important, but also their uncertainties. The simple simulation using burn-up calculations based on typical and generic reactor operational parameters do not answer the question about possible uncertainties observed in real reactor operations. At the same time, the details of irradiation history are often unknown, which complicates the assessment of declared RN inventories. Therefore, the development of dedicated generic proof tools, which can be applied to different types of waste, often of unknown origin and history is worth a serious consideration.

Here, we have compiled a set of burn-up calculations considering the operational history of 339 published or anonymized real PWR fuel assemblies. This gives us the possibility to calculate the realistic variation or spectrum of RN inventories. These histories were used as a basis for “SRP analysis”, to provide information about the range of the values of the associated secondary reactor parameters (SRP’s), i.e. initial enrichment (IE), specific power (SP), irradiation time (IT), downtime (DT), boric acid concentration (BA), moderator and fuel density (MD and FD) and temperatures (MT and MT) used as input parameters for the simulation models.

Then, burn-up calculation codes provide the anticipated results for the entire nuclear inventory such as characteristic activity values and the corresponding minimum and maximum limits as a function of the fuel burn-up.

These results are being validated with the experimental data from various publications and the online database – SFCOMPO [1] and subsequently transferred and summarized into our own database. Ultimately, this database will provide the basis of proof tools for the operator independent verification of the declared nuclear inventories for the quality and safety assessment of a radioactive waste compound and bounding values for certain RN as a function of burn-up. The RN inventory can hence be verified without taking credit of the whole of the operational history of the SNF.

INTRODUCTION

Since 2005 spent fuel assemblies from German commercial nuclear power plants must no longer be submitted to reprocessing but stored on the NPP sites in local interim storage facilities. According to a reference concept for direct disposal spent fuel is first wet cooled before it is transferred to subsequent dry interim storage in appropriate casks and waiting for finally being disposed of. Of course, local waste acceptance criteria for the on-site storage facilities are followed; yet, for a long-term disposal scheme independent national product quality control measures will have to be implemented similarly as to other

HLW, e.g. vitrified and super-compacted waste residues. Moreover, as for the lack of a HLW repository in Germany, the independent expert working group for HLW has identified a set of repository relevant criteria for the dominant waste streams from reprocessing [2].

Obviously a similar approach will be followed for the direct disposal path of SNF. Despite the acceptance criteria for temporary storage being clearly defined, the long-term safety of a deep geological repository depends on the geochemical behavior of radionuclides (RN) that are not necessarily relevant for interim storage. The complexities of the chemical composition of SNF results from fission and n-capture reactions during the reactor operation time and are, thus, highly correlated with the individual burn-up history of the SNF. Clearly, the formation of direct fission products, like Cs-137, is straight forward. However, the formation and abundance of other radionuclides, such as Tc-99, Nb-95, I-129, C-14 and a few others result from much more complex processes. Yet, the mechanism for the TRU is different and we have found that for instance the Am-241 inventory may vary by 400% depending on the individual burn-up history and secondary reactor parameters.

SRP ANALYSIS

Analyzing the secondary reactor parameter or (SRP analysis) provides realistic ranges and typical values for each SRP in dependence of burn-up. The particularity of this work is that the SRP analysis is based on the data of a real PWR operational history of the individual fuel assemblies, which is summarized in own FA database. Thus, SRP analysis includes information of the operational history of 339 fuel assemblies (FA), thereby 270 FA data come from industrial company Wissenschaftlich Technische Ingenieurberatung GmbH (WTI) and 69 FA data from several publications.

We started our SRP analysis from the definition of the individual parameters of the PWR operational history and possible interdependencies. There are nine SRP’s: specific power (SP), irradiation time (IT), downtime (DT), initial enrichment (IE), fuel density and temperature (FD and FT), boric acid concentration (BA) and moderator density and temperature (MD and MT). In our burn-up calculations model all SRP’s are varied independently from one another. Therefore, the absence of dependencies had to be ensured. The possible interdependencies were identified both empirically (based on the statistics of 339 FA data) and theoretically (if applicable).

The product of the specific power parameter with the irradiation time provides the burn-up. Since burn-up steps were fixed, the range and typical value of the specific power were determined from the FA database and the irradiation time is calculated from burn-up and specific power.

For the MD and MT a theoretical correlation exists (thermo-hydraulic law) [3]. The MD and MT data from the FA database are in good agreement with theoretical data from the NIST database (see Fig. 1). Thus, one of two either the range of MD values or the range of MT values can be determined from the FA database while the other SRP is then determined and fixed through-by the NIST correlation law. In this work we determine range of MT values from the FA database.

The analysis of all other, remaining SRP’s from the FA database yields no interdependencies. As a result, there are seven independent SRP’s (IE, SP, FD, FT, BA, MT and DT) and two SRP’s biased by correlations (IT and MD) which should be varied for burn-up calculations.

A very special SRP is the downtime DT (i.e. the time between two operational cycles) because this is not a physical parameter but it is merely given by the operational conditions of the NPP, and it is determined by time needed to make changes and service the FA layout. However, DT can have a significant influence on the RN inventory. The data of FA database reveal that DT can exceed 1000 days, however this is rather an exception. The typical time of DT is either about 30 days, which corresponds to the typical change time of FA of one month, or about 400 days. This corresponds to one reactor operation (irradiation) cycle, when the FA was discharged from core into the cooling pool, and two short change time of FA. Thus, the purpose is to investigate the impact of DT on the RN inventory and to ensure that the DT can be eliminated from other SRP analysis.

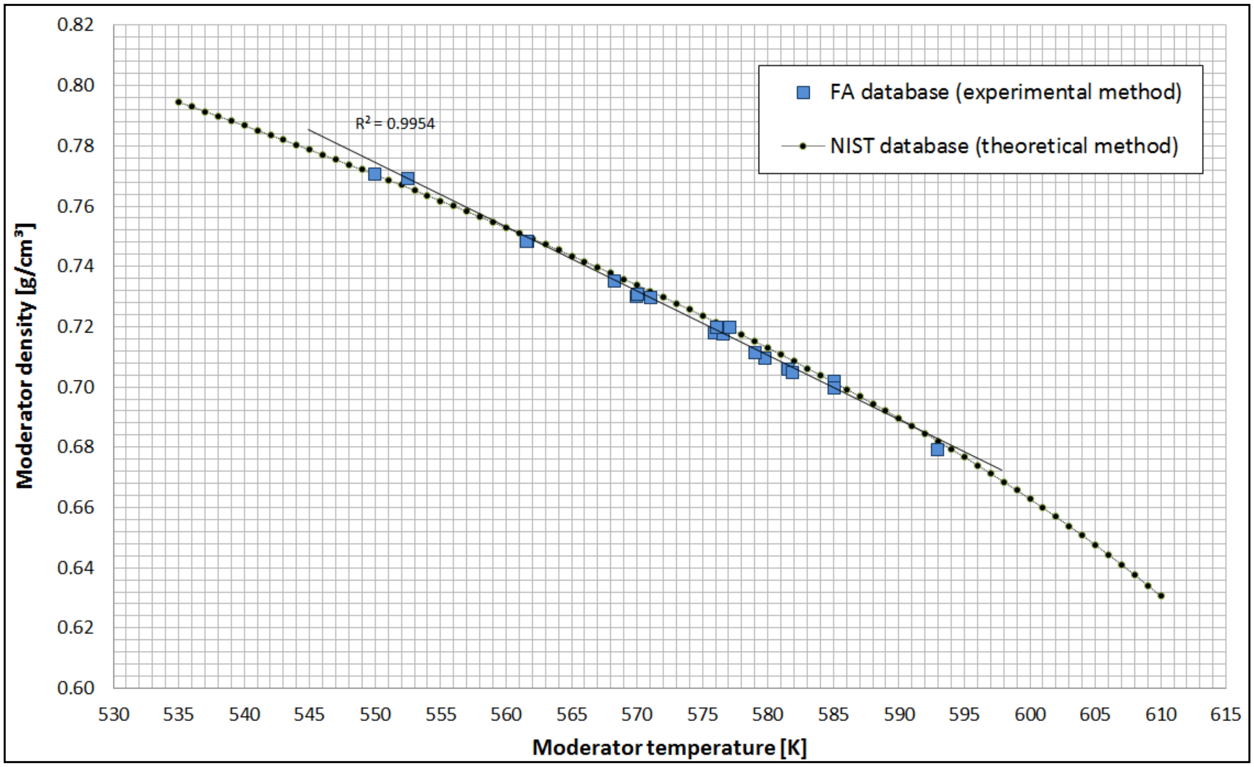

Fig. 1. Theoretical and experimental data of MD and MT.

Therefore, we have calculated the expected burn-up for several DT-parameter values between 0 and 400 days, thereby the values of residual SRP's included either as typical or mean values. The investigated burn-up (BU) is equal to 65 GWd/MTU. The results of DT analysis show that for the most fission products the operational history without DT is either independent (Ag-108m, C-14, Cs-135, I-129, etc.) or conservative (Ag-110m, Ce-144, Cs-134, Cs-137, Eu-154, etc.). In contrast the activities of some actinides are underestimated in the case of operational history without DT (Am-241, Ac-227, Am-242m, Cf-249, etc.), though some are independent of DT (e.g. U-235, U-238, Pu-239, Np-237, Cm-244, etc.). It has to be taken into account, that for the long-term scale of final disposal long cooling times (CT) are of consideration under which the influence of DT becomes in fact negligible.

Thus, the downtime analysis provides the possibility for burn-up calculations without DT, i.e. DT is defined to zero.

So there are now six SRP's (IE, SP, FD, FT, BA and MT), for which the range values or limits have to be determined as a function of burn-up. For example, Fig. 2 shows the limits and typical values for the initial enrichment (IE) for each of the mentioned burn-up steps. Here, we divide possible burn-up range into four steps: 15, 31, 49 and 71 GWd/MTU. The colored points in the Fig. 2 correspond to real PWR fuel assemblies [4]. Each colored point represents between < 10 (green points) and > 500 (dark gray points) fuel assemblies. The black points on the Fig. 2 correspond to 339 fuel assemblies from our FA database; thereby each point represents one fuel assembly. Thus, our FA database covers, to a large extend, all parameters sets of fuel elements that are to be expected, e.g. in Germany. For each of the four BU steps the corresponding typical values of IE as well as lower and upper limits have been identified. A similar procedure was applied to determine the range of other SRP's.

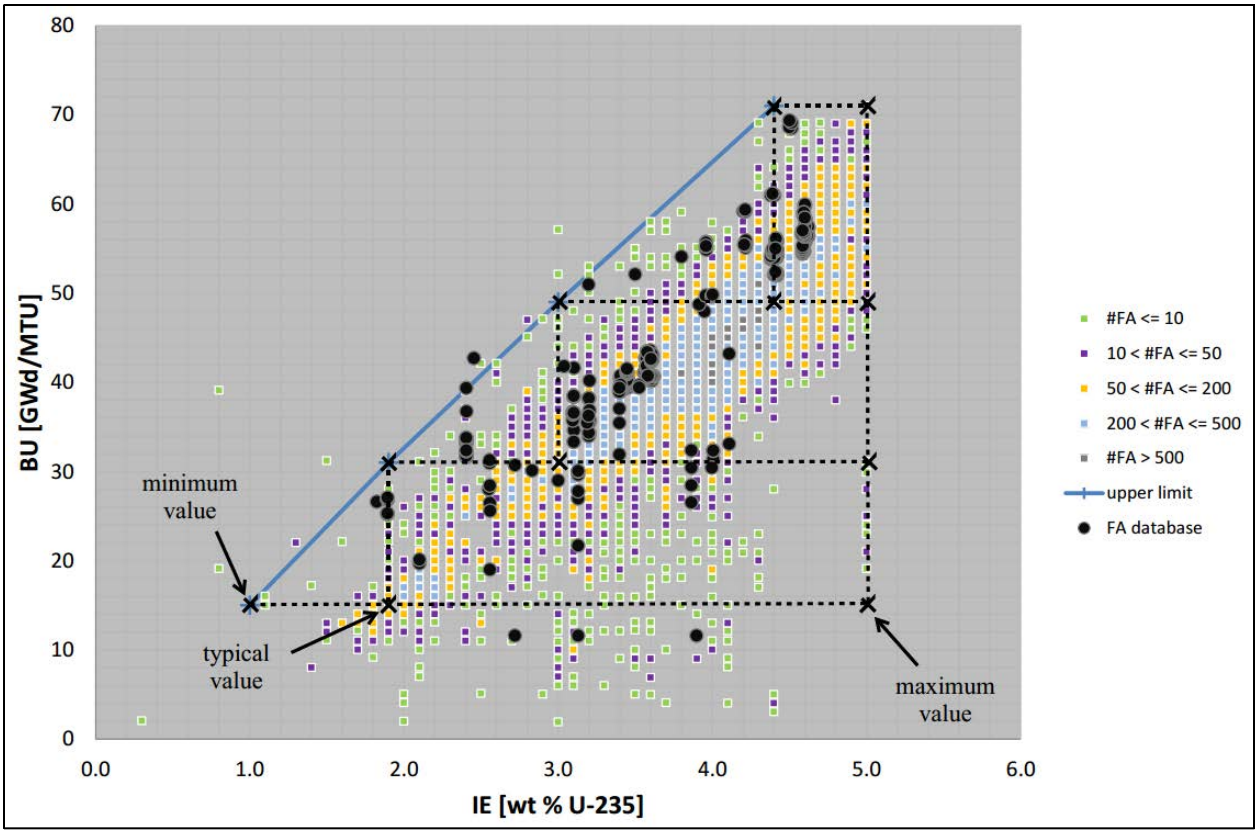

Fig. 2. Burn-up (BU) vs. Initial Enrichment (IE).

Using the information about all SRP's with lower, upper limits and typical values numerous burn-up calculations were performed with SCALE 6.1 software. This covered all possible variations of SPR's. As a result, corresponding realistic variations of RN inventories were determined.

RADIONUCLIDE INVENTORY: THEORETICAL BANDWIDTH AND MEASURED DATA

Burn-up calculations provide the anticipated results for the entire RN inventory such as characteristic activity values and the corresponding minimum and maximum limits as a function of the fuel burn-up, the so-called theoretical bandwidth.

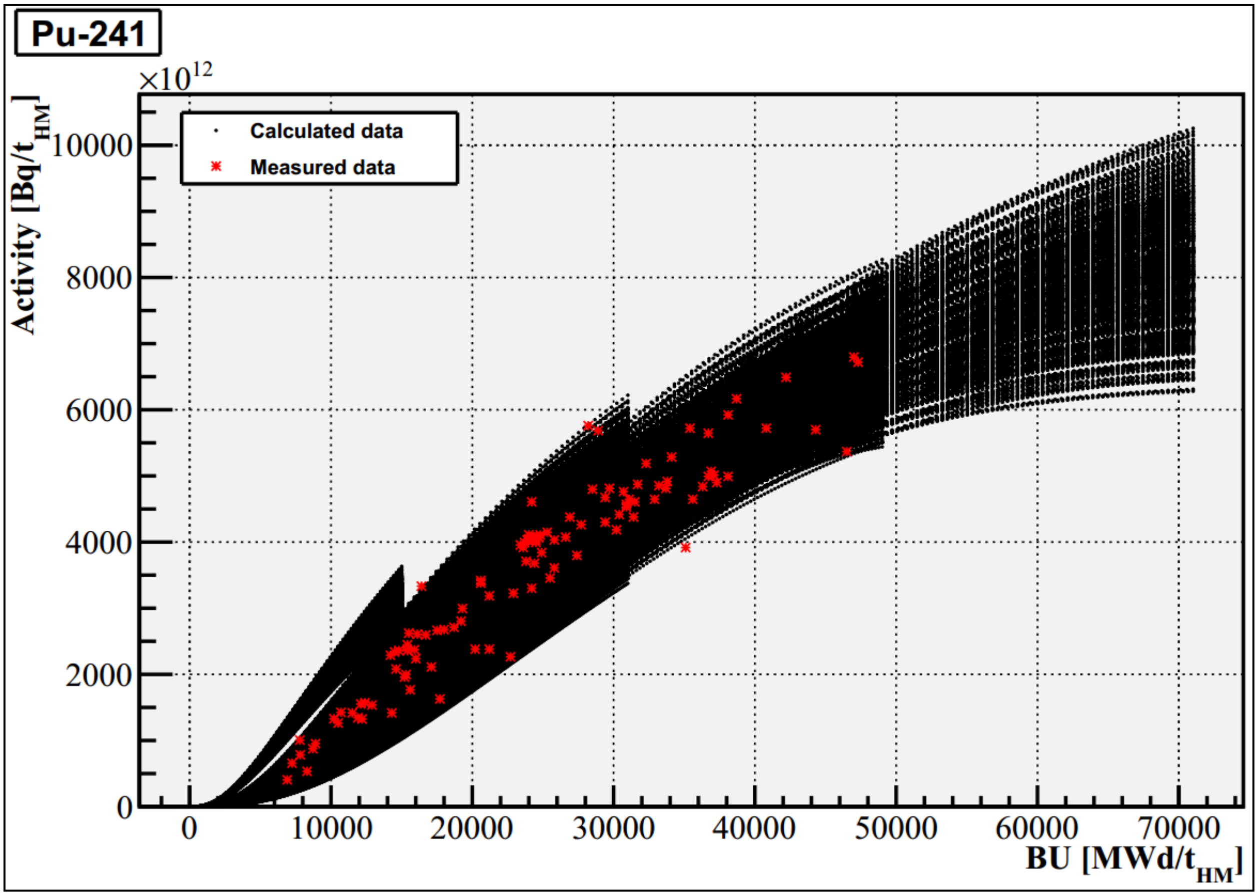

Fig. 3. Calculated and measured data of Pu-241 with CT correction.

We performed these calculations for more than 60 final disposal relevant RN. This corresponds to the typical list of RN, which are relevant for the long-term safety assessment of rad-waste repositories [5]. The results for about 30 of these RN can be benchmarked with the SFCOMPO database [1]. The Fig. 3 shows calculated (black) and measured (red points) data of the built-up Pu-241 activity vs. burn-up. The calculated data represent the bandwidth of the theoretical activity of Pu-241 based of realistic SRP's values; therefore all SRP's have been varied accordingly. The behavior of Pu-241 on varying all SRP's shows the large influence of each SRP. The measured data represent measurements of several samples from different PWR with different histories as extracted from the online database - SFCOMPO. It has to be mentioned here, that in our calculations we considered no post-irradiation cooling time (CT=0), while in SFCOMPO database the measured data correspond to different cooling times. In order to compare our calculations with measured data from SFCOMPO database we had to correct the latter for corresponding cooling times, where it was necessary.

There are RN's, for which the CT correction is not enough for the perfectly agreement between theoretical and measured data (Am-241, Cm-242, Pu-238, Sb-125, Sr-90, U-232, U-234). Still, there is one interesting observation. For example for Cm-242 (Fig. 4) there are a few measurements from the same samples published from two different institutions. E.g. one such sample (Fig. 4 blue marked measurement points) corresponds to the FA of– "Obrigheim PWR BE210 G14 1328" with a burn-up of ~ 38 GWd/MTU and CT = 0 [1]. The different measurements were performed at the "Joint Research Center" in Ispra and the

"Kernforschungszentrum" Karlsruhe. As can be seen from Figure 4 the data point from Ispra is within our calculated bandwidth, whereas the measured Karlsruhe data is overshooting. A reason for that obvious discrepancy still needs to be established; more information about measurements methods, conditions and uncertainties may well be required.

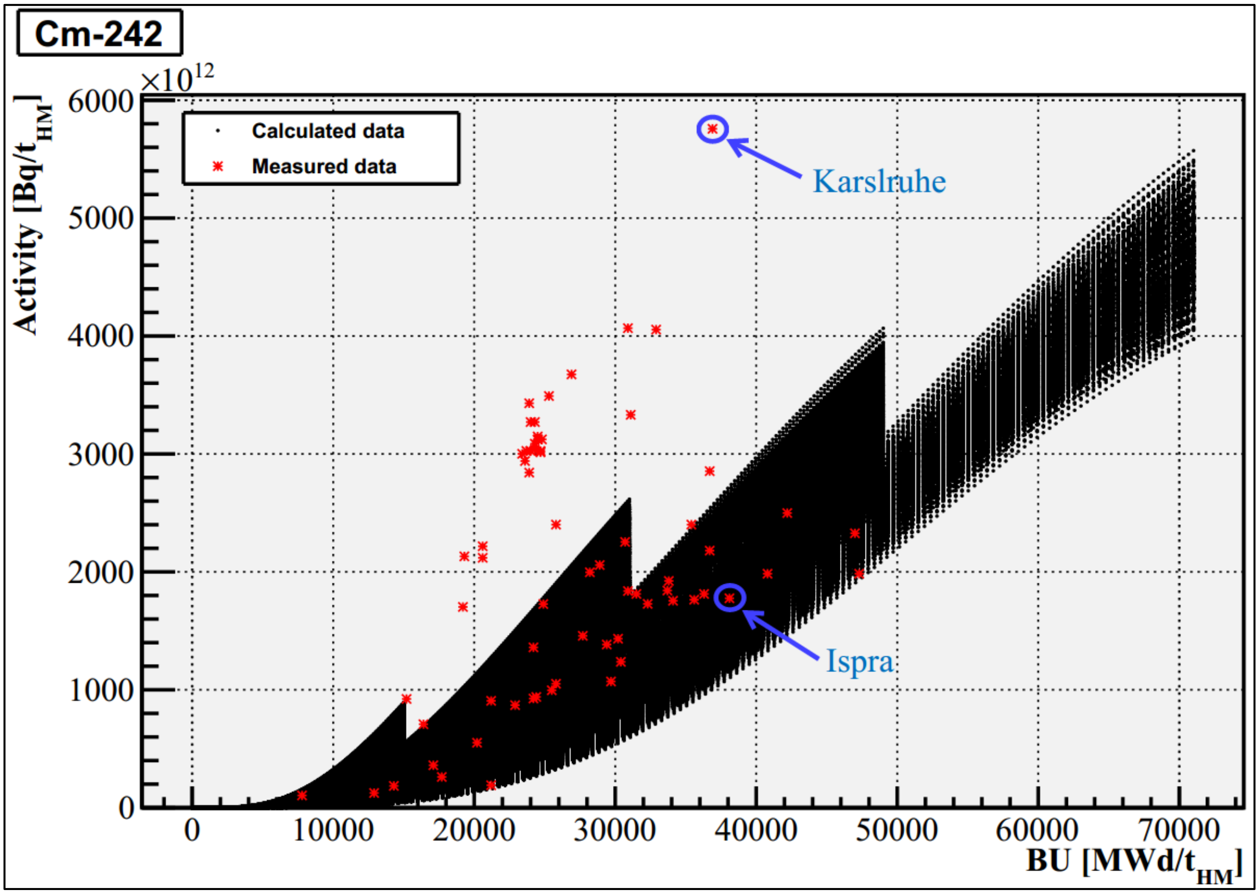

Fig. 4. Calculated and measured data of Cm-242 with CT correction.

Still we can utilize our simulation results for assessing the remaining nuclides, for which only a limited amount of measurement data are available, if at all. The calculated bandwidth of the RN inventory provides the basis of proof tools for an operator-independent verification of the declared nuclear inventories and can be used for the quality and safety assessment of radioactive waste content and evaluating the bounding values for certain RN as a function of burn-up. The RN inventory can hence be verified without detailed knowledge of the complete operational history of the particular SNF.

CONCLUSION AND FUTURE WORK

In this work only the bare fuel composition has been investigated, i.e. fission products and actinides, in total about 60 RN. Activation products have not been considered here. The SFCOMPO database contains data for 30 fission products and actinides that can be used for benchmarking.

For each of these RN relevant for final disposal the theoretical bandwidth like the one for Pu-241 (Fig. 3) has been calculated. All nuclides can be grouped, depending on their response to varying SRP's:

- Independent of all SRP: Cs-137, Se-79 and Tc-99
- Dependent of all SRP: Am-241, Am-242, Pu-239, Pu-241 etc.
- Strongly dependent of IE: U-235, Am-243, Eu-155, I-129 etc.
- Strongly dependent of IE and SP: Ac-227, Ce-144, Zr-95 etc.

The validation using the SFCOMPO database shows that for some RN our calculations do not cover all possible variations. Moreover, the deviations between calculated and measured data for several RN, such as Am-241, Cm-242, Np-237, Pu-238, Sb-125, Se-79, Sn-126, Sr-90, Tc-99, U-232, U-234 need to be explained. Furthermore, the SFCOMPO database does not contain any measured data for BU > 50 GWd/MTU. Whenever these data become available (e.g. from the FIRST-Nuclides project [6]), they must be integrated into the theoretical considerations and calculations such as ours that is introduced here. It is also important to make a clear statement on those RN that lack measured data.

Another perspective is to analyze different scenarios and methods for those cases of incomplete or insufficient information about SRP's. Thereby the question is: what exactly is the minimum required information about the SRP's for successful characterization of the comprehensive RN inventory and to provide an estimate of the expected uncertainty bandwidth.

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