Determination of minor actinides in irradiated fuel rod components

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Abstract

This study is sequential to the work presented in the AMNT meeting in 2016 concerning the measurement and simulation of several isotopes, in particular C-14 and Cs-137 in components of an irradiated PWR fuel rod segment. The current research deals with minor actinides (MA) inventory within the Zry-4 cladding sampled from the of the fuel rod segment. The relevance of MA for the fuel waste is evident; being important long lived radio-toxic contributors besides long lived fission and activation products such as I-129 and C-14. The current study shows that the amount of minor actinides is only partially due to traces of Uranium entering, during the manufacturing process, to the cladding. The major concentration of MA is found on the inner surface of the cladding and is corollary to friction between pellets and the inner surface of the cladding during the fabrication of fuel rods.
Moreover, the comparison between numerical simulations and measurements lead in addition to identify the mobility strength of some isotopes like Cs-137 based on the knowledge gained from the MAs. In particular, it was shown that more than half of the Cs-137 which was found within the Zry-4 comes, most probably, from the adjacent pellets. This fact emphasizes the likelihood of Cs-137 to be released, also to the environment, under unforeseen incidents.

Introduction

The identification of MA vector in spent nuclear fuel assemblies is of highly importance not only as far as the disposal of the waste is concerned, but rather also as measurement of the quality of the simulation for the burning cycles during the operation of the reactor.
The depiction of the MA inventory in the cladding is more cumbersome, as the amount of uranium traces within the cladding is small and demands thereafter very accurate calculations. Moreover, the manufacturing of fuel rods evolves abrasion during the pressing of the fuel pellets into the cladding. The contamination of Uranium on the inner surface of the cladding can reach a depth of 25 µm. A combined analysis based on calculations and measurements can allow for estimation of the amount of MA and in particular the origin of it.
The performed measurements contain surface sensitive techniques and digestion of cladding specimens for (radio) chemical analysis which are compared against numerical calculations to assess the radionuclide inventory of the Zry-4 cladding. Complementary (radio) chemical analysis methods are used e.g. alpha- and gamma-spectrometry.
On the computational side two independent Monte Carlo based burn-up simulation methods are used. The first code MCNPX-CINDER [1], with the ENDF-7 [2] reference data based library, is compared against the MURE (MCNP utility for reactor evolution) package. The latter code system is highly configurable and was modified to allow for the use of a complete set of burn-up and activation data. In

particular, it includes the use of TENDL-2014 as backup data for nuclei that are not covered by the ENDF/B evaluation.

Numerical Simulation of the irradiated fuel rod segment in the PWR Gösgen

The irradiated fuel segment of which the Zry-4 cladding was investigated was part of 5 rod pellet segments put on top of each other for experimental purposes. On top of each fuel segment was a spring covered by the Zr cladding as can be seen in Figure 1. The analyzed Zry-4 was taken from the top of the 50 cm long segment, namely from the upper part of the sample which covered the spring. The experimental rod was irradiated in the middle of a subassembly between 1985 and 1989 in the Goesgen PWR.

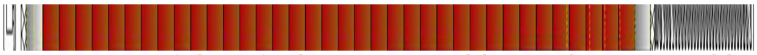

Figure 1: One of the 5 fuel segments with a spring covered by Zr cladding at the top

The Goesgen subassembly is a 15 X 15 fuel rod configuration with 20 water locations as can be seen in Figure 2. The burn-up measured segment was irradiated for 1226 EFPD (Effective Full Power Days). The average burn-up was 50.4 GWd/t_{HM}. The average linear power of the segment was 260 W/cm [3].
The equivalent subassembly that was simulated is depicted in Figure 2. The underlying numerical concept was to meet the conditions as shown above to which the fuel segment is exposed to. In order to avoid statistical error and without violating the above mentioned constraints, 4 equivalent fuel segments were simulated in the center of the subassembly as can be seen in Figure 2.

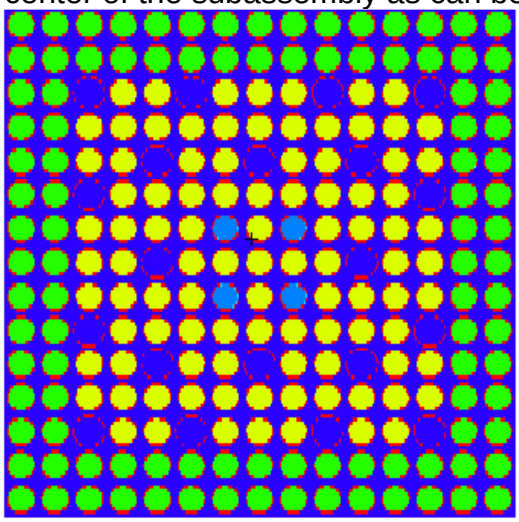

Figure 2: The Goesgen Subassembly. The 4 bright blue fuel elements in the middle depict equivalent positions of the investigated fuel segment. The water holes are seen as to some extent enlarged red cycles within the dark blue color of the simulated water. The green and yellow pins are the rest fuel pins of the subassembly.

In figure 2 the chosen option is shown, namely the yellow pins were considered as generating the power which exhibits the flux and the power defined for the fuel segments under investigation. The green pins in Figure 2 are also fuel pins, however they were only considered as far as their neutron generation was concerned. This kind of simulation leads to the requested linear power and total burn-up level for the 1226 EFPD irradiation period.

The flux solver MCNPX –CINDER was used for the burn-up calculation. In this way the reaction rate data are transferred directly from the transport solver to the burn-up module.
The burn up simulation of the subassembly, shown in figure 2, was optimized to 5 steps. At the end of the 1226 EFPD a decay period of 9600 days was considered. This is the period between the end of the irradiation at 1989 and the measurements of the sample which were recently performed.

The radiochemical measurement technique is generally based on alpha spectrometry. α- particles, emitted from the analyzed radionuclide, transfer their energy to a semiconductor, thus creating electron-hole pairs. The electron can be then detected to account for the existence of the specific radioactive nuclei under investigation. The measurements in this work were performed using the Canberra S100 (Canberra Industries Inc.) detector.
For the MA isotopes with characteristic γ-rays e.g., Am-241, the quantification was performed in acidic digestion liquors obtained from dissolution experiments performed with irradiated Zry-4 using γ-spectroscopy. The measurements were performed by means of an extended range coaxial Ge detector (GX3018, Canberra Industries Inc.).
Table 1 presents the reference results of the current study. It shows the comparison between the calculated nuclides and combined measured isotopes which have close (α spectrometry based) peaks. The ratio of those results are considered to be a reference as they exhibit the less statistical error which is considered to be below 5% for each α or γ type measurements. In particular the ratio of the second and third column is more reliable as the signal of counting is higher. Based on table 1 one can see that the comparison of calculated and measured ratios between the pairs “2” and “3” is very good. Using the ratio of “1” leads to difference of about 25%. Further, the absolute counting shows a factor 50 for columns 2 and 3 and factor 65 for column 1.

Table 1: Comparison between the measured and the calculated activities of 3 pairs of MA isotopes within the Zircaloy of the fuel segment under investigation in unit Bq/(g Zry-4)

MAs	**1)Pu-239 + Pu-240**	**2)Pu-238 + Am-241**	**3) Cm-243 + Cm-244**	**Ratio 2/3**
Experimental	3.06×10^4	2.29×10^5	2.61×10^5	0.877
Calculated	4.57×10^2	4.52×10^3	5.26×10^3	0.859

In order to extract more information further measurements were performed based on alpha spectrometry for several Pu isotopes and γ-spectroscopy for Am-241 as mentioned above. Table 2 shows the obtained results.
Analyzing the results in tables 1 and 2 and accounting for the statistical errors it can be seen that except for the Measurement of Pu-240 all results are consistent in a sense that the contribution of the inner surface of the cladding –which could not be accounted for by the calculations- is by about factor 50 higher than the traces within the material. The larger deviation due to the Pu-240 is consistent in both tables, suggesting that measurement of Pu-240 was counting signals which did not come from Pu-240 by about 40%.

Table 2: Comparison between the measured and calculated activities of isotopes, using alpha spectrometry for several Pu isotopes and γ-spectroscopy for Am241 within the Zircaloy of the fuel segment under investigation in unit Bq/(g Zry-4)

MAs	Measured	calculated	Ratio measured/calculated
Pu-239	1.2×10^4	2.38×10^2	50.4
Pu-240	2.04×10^4	2.19×10^2	93
Pu-241	2.06×10^6	4.4×10^4	47
Pu-242	2.19×10^2	4.1×10^0	53
Am-241	1.72×10^5	3.76×10^3	46

Conclusions

A burn-up simulation of a fuel rod sample, which was irradiated in the PWR Goesgen for 1226 Effective Full Power Days, was performed. In this study the activity of several MA in the Zry-4 cladding, covering the spring on the tops of the sample, was measured and compared to burn up calculations. Analyzing the overall trend of the results it was shown that for almost all isotopes the measured activity of the investigated MAs was higher by about a factor 50 than the calculated activity. Taking into account that the inner surface of the cladding was "contaminated" by Uranium due to the abrasion during the pressing of the fuel pellets, strengthen the assumption that the higher activity of the cladding is due to the mechanical production process of the fuel rod.

Further on, the shown results give also an indication for the large deviation between the measured and calculated Cs-137, presented in [4]. In this former study the measured amount of Cs-137 was larger by more than a factor 100 in comparison to the calculation. However based on the current study, one would expect again an increase of a factor 50, similar to the measured/calculated ratio of the MAs. However, the fact that in the former study the measured amount of Cs was much higher indicates that not all the measured Cs137 should be attributed to the fission process directly within the inner of the Zry-4 surface. Most probably, more than half of the activity of Cs-137 is due to migration of Cs-137, from the adjacent pellet within the sample, which was adhered to the cladding.

References

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