Characteristics of selected irradiated material, irradiation history, methodology of experimental / analytical procedures and inventory calculations

R . Dagan[1] , M. Herm[1], A. Konobeev[2], T. Königq[1]

[1]Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal (KIT-INE), P.O. Box 3640, D-76021 Karlsruhe, Germany

[2]Karlsruhe Institute of Technology, Institute for neutron physics and Reactor technologies(KIT-INR), P.O. Box 3640, D-76021 Karlsruhe, Germany

Abstract

This study compares the characteristic of irradiated materials in form of dedicated prepared samples that were radiated in the Gösgen reactor (KKG, Switzerland)for 1226 full power days. Analytical simulations are compared to measured inventory of the cladding that was performed in the INE laboratory. Preliminary results confirm that traces of the fuel , albeit to negligible extent, are adherent to the inner surface of the cladding during the manufacturing of the samples by the so call vibrofill technique[1].

1. Characteristics and burn-up history of studied samples

The studied Zircaloy-4 plenum cladding and UO_2 fuel was sampled from a fuel rod segment which was irradiated during four cycles in the pressurize water reactor Gösgen (KKG, Switzerland) and discharged in May 1989 after 1226 effective full power days. During reactor operation, an average burn-up of 50.4 GWd/t_{HM} as well as average linear power of 260 W/cm was achieved. Initial enrichment of the fuel with U235 was 3.8%.

Characteristic data of the studied fuel rod segment are given in [2,3]. The calculation of the total inventory of the various Kr and Xe isotopes was performed with the webKORIGEN software package [4]. Based on results of a puncturing test of the fuel rod segment, the fission gas release into the plenum of the segment was determined as (8.4 ±0.9) % of the total fission gas inventory [2,3].

2. Experimental Methods and analytical procedures

Subsamples were dry cut using an IsoMet® Low Speed Saw equipped with an IsoMet® diamond wafering blade. Cutting was performed very slow to prevent overheating of the material.

The specimens were digested within 30 minutes in a dilute H_2SO_4/HF mixture at room temperature and anoxic conditions using a flask or autoclave.

Radionuclides present in the digestion liquor (and gas phase) were analyzed using various analytical and separation methods.

Fission products and minor actinides with characteristic γ-rays e.g. Cs137, Am241 and Am243 were quantified using γ-spectroscopy. Measurements in this study were performed by means of an extended range coaxial Ge detector (GX3018, Canberra Industries Inc.).

Minor actinides, activation and fission products such as Sr90 or Pu241 were measured, after separation from other radionuclides, in digestion liquor using liquid scintillation counting (LSC). LSC measurements were performed using an ultra-low-level spectrometer (Quantulus 1220, Wallac Oy, Perkin Elmer).

In addition, actinides/isotopes such as U238, Np237, Pu238, Pu239, Pu240, Pu241, Pu242, Am241, Am243, and Cm244 present in digestion liquors were also quantified using mass spectrometry. Measurements were performed using an ICP-SF-MS (inductively coupled plasma – sector field – mass spectrometer, ELEMENT XR™ ICP-MS, Thermo Scientific).

3. Referenced Benchmarking of the Gösgen sample

The first goal of task 2.3 was to establish a numerical tool, which could serve as a reference for further comparison with other codes and/or other calculation methods. As the sample to be tested was irradiated in the thermal Goesgen reactor [2] a preliminary step was to use directly a flux of a thermal reactor and run only the burn up steps according to the time steps and decay time as given in [2]. For this purpose, the webKORIGEN version [4] embedded in the NUCLEONICA [4] package was utilized. In accordance with the tested sample and the different measurements techniques discussed in section 2 the nuclide inventory within the fuel of the sample, rather than the cladding itself was analysed. Obviously, the amount of nuclides of each type is much larger in the fuel than in the cladding, which allows a better reference test case.

In table 1, representative nuclide inventory of the Gösgen sample is introduced after 1226 irradiation days and 28 years decay time. The difference between the KORIGEN burn-up with fixed flux spectrum of PWR thermal reactors and a modern Monte Carlo transport code coupled with the advanced CINDER burn-up program enables to envisage the important issues that should be looked at in the following. On general 7 of the 10 nuclides seems to be less sensitive to the type of calculation and the geometry and type of the reactor. However, other nuclides are known to be more sensitive to burn-up calculations e.g. Cm244 indeed exhibit a large deviation. Am241 is sensitive to flux fluctuation due to its pronounced resonances and its build up within the decay chain, mainly by the 14.4 years halftime life of Pu241. The big difference of the Pu239 inventory emphasize the important of accurate evaluations of the fuel inventory as it a central nuclide within the actinides generation due to fission and fission products elements. Furthermore, the nuclear data of Pu239 are continuously being improved which emphasizes the need of accurate analysis as far as the nuclear data libraries are concerned.

Nuclides	**KORIGEN** mol/g_{UO2}	**MCNP-CINDER** mol/g_{UO2}	**Ratio MCNP/KORIGEN**
Kr total	4,97E-06	5,08E-6	1,02E+00
Xe total	5,17E-5	4,83E-05	9,34E-01
Sr90	3,79E-06	3,78E-06	9,96E-01
Cs137	6,17E-06	6,25E-06	9,74E-01
U238	3,44E-03	3,57E-03	1,04E+00
Np237	2,82E-06	2,98E-06	1,06E+00
Pu239	2,12E-05	2,93E-05	1,38E+00
Am241	4,67E-06	6,17E-06	1,32E+00
Am243	8,37E-07	8,47E-07	1,01E+00
Cm244	1,23E-07	1,64E-07	1,24E+00

Table 1 – Inventory of Gösgen fuel after 1226 irradiation days and 28 years of decay.

In summary, the planned benchmark within sub task 2.3 should concentrate on code to code validation on different levels based also on the results of sub task 2.1. This will enable to deal in a better manner with the more complicated and yet important inventory of the cladding of the irradiated sample of Goesgen.

4. Preliminary analysis of the plenum cladding sample

The plenum cladding material under investigation includes traces of fuel, which were adhered onto the cladding inner surface during the insertion process of the pellets into the fuel rod. As part of the cladding analysis, the amount of several anticipated nuclides based on the initial material composition of the cladding was calculated and compared to experimental measurements. In Particular several actinides on one side and some fission products on the other side were analysed in accordance with the available measuring techniques.

The numerical calculation was performed with the MCNP CINDER code system, mentioned in the previous section.

Table 2 presents the experimentally obtained results as well as the calculated ones for several minor actinides. The sliding friction effect of the pellets on the inner surface is obvious. The calculation is based on the material composition data of the cladding and it can be seen that the ration C/E is between 1.5 to 2.2% with the exception of Pu240 that amount only to 1.07% of the observable Pu240.

Nuclides	exp. inventory Bq/g	MCNP-CINDER Bq/g	Ratio C/E
Pu238	7,81E+04	1,19E+03	1,52E-02
Pu239	1,20E+04	2,50E+02	2,07E-02
Pu240	2,04E+04	2,19E+02	1,07E-02
Pu241	2,06E+06	4,294E+04	2,08E-02
Pu242	2,19E+02	3,93E+00	1,80E-02
Am241	1,80E+05	3,83E+3	2.127E-2
Am243	3,71E+03	5,766E+01	1,55E-02
Cm244	3,13E+5	5.47E+3	1,747E-02

Table 2 – Actinide inventory of Gösgen cladding sample after 1226 irradiation days and 26 years of decay.

Altogether, those preliminary results indicate that the amount of additional fuel on the inner cladding surface increases the initial fuel inventory by up to one order of magnitude. The accurate evaluation is however still a challenge as this amount of fuel traces on the inner surface cladding is still quite small which makes it more sensitive to the quality of the numerical tools, in particular Monte Carlo ones.

It should be noticed that the experimental data are based on averaging different measuring techniques like mass-spectrometry or gamma spectroscopy. Apparently, it introduces some uncertainties, which are to be further explored in the ongoing work.

5. Summary and future plans

The new coming measuring campaign together with additional calculating tools of different participants will enable a broadened sight on nuclear inventory including, beside the minor actinides, also additional fission and activation products. As far as direct human safety is concerned volatile nuclides like Cs137 and C14, I129, Cl36 and Tc99 should be also handled and analysed within the framework of the subtask.

On the experimental side updated separation methods for Pu isotopes are being currently tested. Further on, nuclide dependent measurement techniques are being exploited to confirm the applicability range of alpha-, beta-, gamma-spectroscopy and mass spectrometry.

Acknowledgement

This document is a report of the European Joint Programme on Radioactive Waste Management (EURAD). EURAD has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 847593.

References

1. STRATTON, R. W., BOTTA, F., HOFER, R., LEDERGERBER, G., INGOLD, F., OTT, C., REINDL, J.,
   ZWICKY, H. U., BODMER, R., AND SCHLEMMER, F. 1991. A comparative irradiation test of UO2 sphere-pac and pellet fuel in the Goesgen PWR. Proceedings of Avignon, France.
   Int. Topical Meeting on LWR Fuel Performance "Fuel for the 90's

2. González-Robles, E., Wegen, D. H., Bohnert, E., Papaioannou, D., Müller, N., Nasyrow, R., Kienzler, B., Metz, V. Physico-chemical characterization of a spent UO2 fuel with respect to its stability under final disposal conditions. Mater. Res. Soc. Symp. Proc., 1665 (2014) pp. 283-289.

3. Gonzalez-Robles , E., Metz, V., Wegen, D.H., Herm, M., Papaioannou, D., Bohnert E., Gretter, R., Müller, N., Nasyrow, R., de Weerd, W., Wiss, T., Kienzler, B. Determination of fission gas release of spent nuclear fuel inpuncturing test and in leaching experiments under anoxic conditions. Journal of Nuclear Materials 479 (2016) 67-75.

4. Nucleonica GmbH (2011) Nucleonica Nuclear Science Portal (www.nucleonica.com), Version 3.0.11. KORIGEN code. http://www.nucleonica.net/Application/Korigen.aspx