Progress in NUCLEAR SCIENCE and TECHNOLOGY, Vol. 2, pp.406-410 (2011)

**ARTICLE**

# Numerical Tools for the Evaluation of Super-Compacted Radioactive Waste Residues

Stephan SCHNEIDER [1,*], Holger TIETZE-JAENSCH [1,2] and Dirk BOSBACH [1]

[1] *Forschungszentrum Jülich GmbH, Wilhelm-Johnen-Str., 52425 Jülich, Germany*
[2] *Produktkontrollstelle für Radioaktive Abfälle, Wilhelm-Johnen-Str., 52425 Jülich, Germany*

Numerical modeling and simulations of radioactive emissions from nuclear waste residues are important for the assessment of the long-term hazard potential and safety properties. The specific radioactive waste residues considered in this paper are dried and super-compacted metallic remains after sheering and dissolving spent nuclear fuel elements from commercial NPP's in the process of fuel reprocessing. They consist mainly of hulls and end pieces of the fuel assemblies and some additional technological metallic waste from the reprocessing itself. Because of activation and some small percentage of spent fuel material adhering to hulls, these rad-waste residues are classified as medium active waste. The goal of this work is to develop numerical tools to evaluate the final repository relevant properties of rad-waste residues. Simulating the gamma- and neutron-emission spectra allows numerical studying the effect of varying the concentration, activity and geometrical position of key nuclides in a heterogeneous material embedding.

***KEYWORDS: waste management, rad-waste, benchmarks***

## I. Introduction

The reprocessing of spent nuclear fuel leads to different waste streams. Most prominent is the highly active vitrified waste containing most of the fission products and minor actinides. Another stream contains the remaining metallic materials of the reprocessed fuel assemblies. These are mainly hulls, cladding and end-pieces. As some small percentage of the spent fuel remains adhering to the hulls these materials have to be considered as radioactive waste. To minimize the repository storage volume the metallic components are shredded and filled into cans, then super-compacted and stacked-up into standard waste residue containers. As for Germany there are more than 5,000 of such super-compacted waste residue containers to be disposed of.

German safety regulations require the waste residues to be inspected and their nuclear inventory declared and verified before finally transferring them to the repository. The aim of this project is to use numerical tools to calculate the expected nuclear characteristics and radiation emerging from this type of radioactive waste residue.

## II. Methods & Materials

The most crucial points about nuclear waste product control could be summarized as:

1. Inventory declaration,
2. Heat and dose rate determiniation,
3. Method evaluation.

The correct inventory declaration is the objective of all nuclear characteristics estimation. The nuclear inventory distribution determines the best choice of the surrounding material matrix for long term safety. Knowing the material composition of matrix and container properties the dose rate and exterior heat load can be calculated. Therefore product quality assurance has to deal with the individual nuclear waste product parameters but also with the production process technology.

In Germany, the evaluation of nuclear properties and the safety assessment of super-compacted rad-waste residues are expected to commence soon. Thus, the software tools are employed to provide real and hypothetical answers on all conceivable scenarios assessed.

In this paper we focus on the concept of this software environment and present benchmarks of our models.

### 1. The Concept of the Software Tools

The software aided tools are based on the results of different well known and widely accepted computer codes. The concept is divided in three parts. The first part addresses the problem of inventory declaration by using burn-up codes to calculate the nuclide distribution of spent fuel assemblies. The nuclide distribution and isotopic behavior depends on the reactor physics parameters such as: initial enrichment, burn-up, cooling time, neutron poisons and reactor design. For this part we have chosen the SCALE 6.0 software collection.[1)] Using our own models of the fuel assemblies we first perform a full-scale reactor physics simulation to generate problem specific cross-section files, which are subsequently used in ORIGEN-ARP calculations. The resulting nuclide vectors of the spent fuel nuclides distribution pattern, are stored in a hierarchical file for further processing. The variation of the reactor parameters must cover the whole parameter space of the expected spent fuel characteristics

*Corresponding author, E-mail: s.schneider@fz-juelich.de

within the reprocessing cycle. The source terms calculated from these nuclide vectors are the basis for the further analysis of the super-compacted waste residues.

The second part of the software kit deals with the problem of heat and dose rate calculations, therefore particle transport equations have to be solved. In this case we have chosen MCNP/X 2.6.0 or MCNP5 1.51 for the Monte Carlo based particle transport.

The last part of the program suite contains the user interface and auxiliary program modules. For instance, it should be possible for the user to create a MCNP model of a particular rad-waste residue just from the documentation of the waste product.

The software package itself should be modular in design to be easily modified, adapted and extended to other nuclear waste problems and not to be limited only to the super-compacted metallic waste.

### 2. Super-Compacted Metallic Waste

The waste itself is mainly composed of structural materials of the reprocessed fuel assemblies. The cladding of the fuel rods is made of Zircalloy and stainless steel. During the reprocessing the fuel elements are chopped into small pieces and afterwards bathed in boiling nitrite acid to dissolve the spent fuel from the metallic rods. The latter are then dried and filled into a can for compaction. After the compacting or pressing process the resulting discs are stacked inside another metallic container. The average density of this waste product is approx. 4.4 g/cm$^3$. In this paper we limit the discussion to waste products containing only structural materials from fuel elements. In this case it can be assumed that the radiological properties of the waste product depend only on the reactor parameters, the chemical behavior during the leaching and dissolution process and the compaction.

### 3. Assay Systems

The produced rad-waste products are scrutinized using two different measurement stations. Gamma spectroscopy is employed for the measurement of specific key-nuclides. With the estimated activities of these nuclides the fuel characteristics are evaluated in terms of burn-up and cooling-time. [2)]

A complementary second system is an active neutron interrogation measurement station. Two pulsed neutron sources are used to identify prompt and delayed neutrons emerging from the remaining fuel material inside the waste product. Emerging neutrons are counted with 249 $^3$He neutron detectors surrounding the cavity.[2)] The interpretation of the time behavior of the different signals allows the calculation of the amount of fissile materials remaining in the waste matrix.

## II. Simulation and Benchmarks

In this section we discuss the benchmarks we applied to test the quality of different calculation programs and to test our models.

### 1. Burn-up Calculation and Spent Fuel Isotopics

As mentioned in the introduction, the burn-up calculation

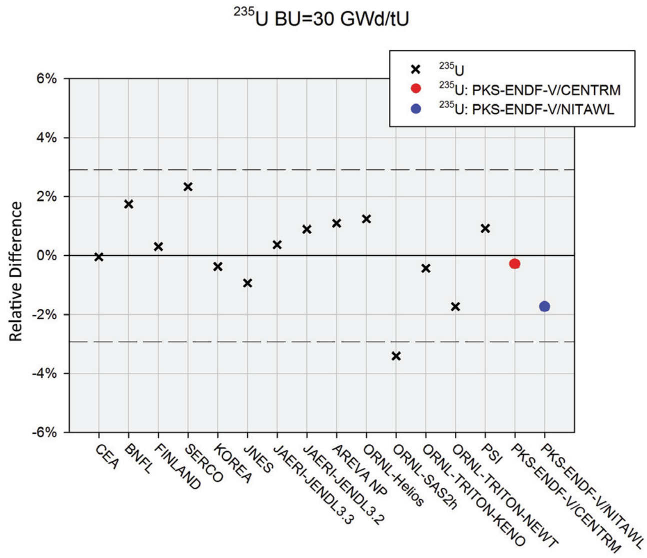


**Fig. 1** Relative difference of the $^{235}$U fraction compared with the mean value of the NEA 6227 study. The dashed lines represent the 1σ level.

is one of the basic numerical methods for the examination of the compacted waste residues. It is used to calculate the nuclide distribution of the fuel depending on the reactor physics parameters. To verify that our numerical results and our models well resemble the measured nuclide vectors we had to perform different benchmarks.

(1) NEA - Burn-up Criticality Benchmark: Phase 2D

The first benchmark performed was taken from the NEA No. 6227 report.[4)] This benchmark is used to compare different computer codes calculating the criticality depending on burn-up credit. The model was setup according to the benchmark parameters using the TRITON/NEWT sequence.[1,5)] The model consists of a single PWR 17x17 fuel element with fresh fuel composition. The initial enrichment was 4 wt.%. The SCALE functional module NEWT is a 2-D discrete-ordinates neutron transport code. NEWT calculates the forward and adjoint neutron flux solutions and eigenvalues. For this benchmark the ENDF-V library was used with a 44 group energy spectrum. **Figure 1** shows the resulting $^{235}$U fraction after 30 GWd/tU.

Our results of the calculated fraction of $^{235}$U were in good accordance with those from the other participants. Moreover, in the case of using NITAWL for the cross-section preparation our result is equal to those from ORNL who had also used TRITON/NEWT.

The conclusions of this benchmark are that our models and cross-section sets in deed produced the same results as published in the literature.[4)]

(2) JAERI-M 94-034 Report

The next step was to evaluate if our models could forecast the real nuclide distribution of the spent fuel. Therefore different reactor models were tested. For instance, we had compared the results of the Siemens PWR 14x14-16 assembly model with the real nuclide distribution published in the JAERI-M 94-034 report.[6)] The initial enrichment of the fuel was 3 wt.% of $^{235}$U. The mean burn-up was 29 GWd/tU, the fission product activities were calculated for 100 days cooling-time. For this calculation the ENDF VI library was used

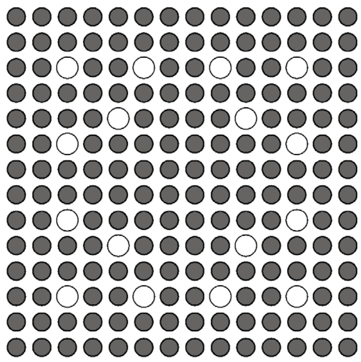

**Fig. 2** NEWT layout of the 14x14 Siemens PWR fuel element. Consisting of 180 fuel pins and 16 guide tubes.

**Table 1** Specific activity of important key-nuclides

| | Measurement (Bq/tU) | Simulation (Bq/tU) |
|---|---|---|
| $^{154}$Eu | 1.82E8±7.1E7 | 1.733E+8 |
| $^{134}$Cs | 4.52E9±1.72E9 | 4.563E+9 |
| $^{137}$Cs | 3.32E9±7.18E8 | 3.446E+9 |
| $^{154}$Eu/$^{137}$Cs | 0.0526±0.0127 | 0.0502 |
| $^{134}$Cs/$^{137}$Cs | 1.3102±0.2649 | 1.324 |

with 238 energy groups. Figure 2 shows the layout of the Siemens fuel element.

Therein, the discussion is restricted to the fission products like $^{134}$Cs, $^{137}$Cs and $^{154}$Eu, only. These radionuclides are often used to calculate the spent fuel characteristics in terms of burn-up and cooling-time.[7]

Since the simulation was performed on the whole of the fuel assembly the mean values of the key-nuclides were taken into account for each fuel assembly. The results are shown in **Table 1**.

The estimated specific activities of the different key-nuclides are within the confidence interval of the measurements. Due to the different half-life of the cesium nuclides the correlation between them is proportional to the cooling time of the spent fuel. The TRITON/NEWT module can be used to estimate the mean value of different nuclide concentrations if the reactor parameters like burn-up, cooling-time, amount of soluble neutron poison and others are well known.

## 2. Monte Carlo Benchmarks

In this section we present a selected set of our benchmarks for the neutron transport problems we are dealing with in radioactive waste characterization. The goal is to minimize systematic errors by testing models of different degrees of complexity.

(1) A Simple Neutron Scanner

The first benchmark was a comparison between a simple neutron measurement performed at our institute and the MCNP models. The detector was a single $^3$He detector surrounded by a thick layer of polyethylene to slow down fast fission neutrons emerging from a point-like $^{252}$Cf source.[8] **Figure 3** shows the experimental setup, we had varied the

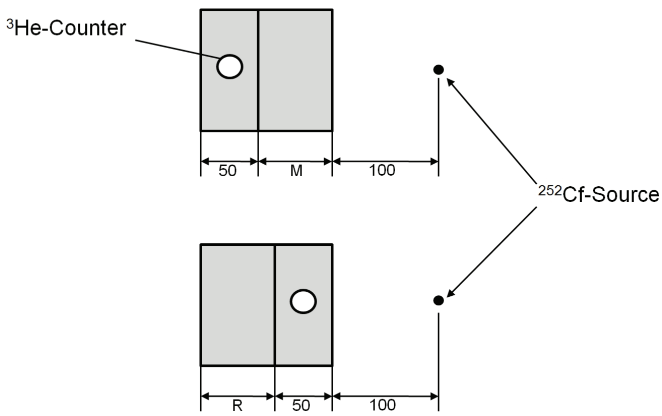


**Fig. 3** A simple neutron detector experiment for small samples. All sizes in (mm).

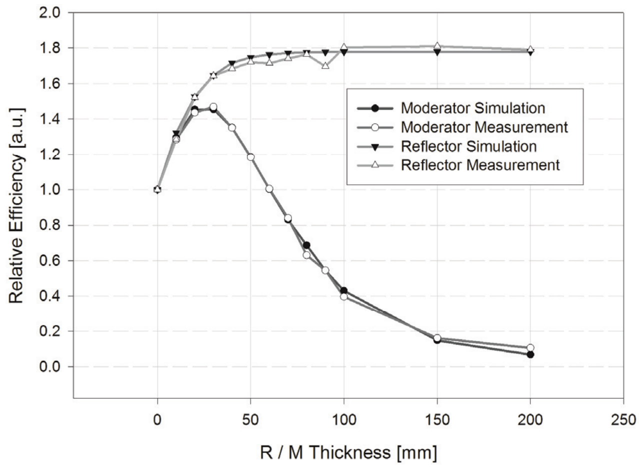


**Fig. 4** Relative detection efficiency of the 3He detector depending on the choice of moderator (M) and reflector (R) thickness

thickness of the reflector and the moderator to examine different influences on the detector signal and determined the optimal thickness for both the moderator and reflector material.

The MCNP model was setup using an isotropic $^{252}$Cf point source with a Watt-fission spectrum. The variations of the tallies depending on the thicknesses of the different layers are shown in **Fig. 4**. The estimated detector efficiencies were normalized to the case of a single $^3$He tube inside a polyethylene block with the dimensions of (50x100x400) mm. MCNP 5 1.51 and the ENDF VII.0 library was used for these calculations. The problem was run until the relative uncertainty was smaller than 5% and the ten statistical tests of MCNP passed. The calculated efficiencies are in good agreement with the measured values. The variation of the reflector thickness has a larger impact on the total detection efficiency. This is due to the increased backscattering probability. The increase of the moderator thickness results in larger detection efficiency because the neutrons are slowed down before reaching the $^3$He tube. A further increase of the moderator thickness is counterproductive because the distance between the sample and the detector is increased and therefore the detection efficiency is rather reduced due to

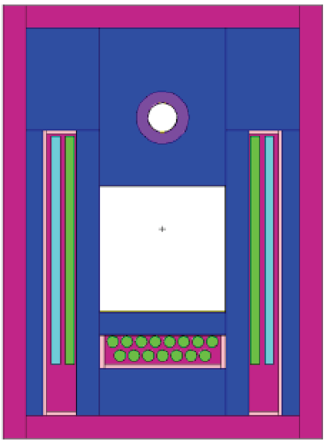

**Fig. 5** MCNP5 model of the PROMETHEE 6 detection system. The green and light blue areas are the $^{3}He$ detector banks.

geometrical reason. The optimum values for this detector assembly were found as M=3 cm and R=10 cm, which yields a factor 2.4 times higher detection efficiency.

(2) PROMETHEE 6

To increase the level of complexity we chose a different sophisticated neutron scanner for the next benchmark. The PROMETHEE 6 (Prompt Epithermal and Thermal Interrogation Experiment) was developed by the French Atomic Commission (CEA) and COGEMA as an alpha low-level waste assay system and is therefore a valuable test object for the modeling of large scale detection systems.[9)] The geometrical set-up of the system was taken from various published sources and the model was created using MCNP5.[9,10)] The main feature of the PROMETHEE 6 is the use of a pulsed neutron source (D-T tube) and a neutron detector system of $^{3}He$ proportional counters embedded into various combined layers of moderator, reflector and absorption/neutron energy cut-off materials. **Figure 5** shows the layout of the MCNP5 model. The cavity was designed for 118 l waste drums. The dimension of the empty cell is (55x55x90) cm. The walls of the cavity are covered by 2 mm thick aluminum plates. The neutron generator GENIE 26 can produce 15 µs pulses at 125 Hz repetition rate. The achievable neutron fluxes are between $3.7 \times 10^{7}$ to $1.8 \times 10^{8}$ n/s emitting in $4\pi$.[10)]

The real experiment detection efficiency of the empty cell is 25.7% ± 0.3%.[5,8)] With our Monte Carlo methods we calculated the detection efficiency at 28% ± 0.1%. So the efficiency is slightly overestimated in our simulation. This could be so because of some geometrical inaccuracies in the model or due to the neglect of the wall effect on the $^{3}He$ tubes.[11)] In this case only the probability for the (n,p) reaction was tallied for each starting particle.

(3) Conclusion Benchmarks & Simulation

The computer codes MCNP5/MCNPX as well as SCALE 6.0 are well known and worldwide accepted codes for simulations in the field of nuclear engineering. The extensive numbers of benchmarks were used to become familiarized with different methods and applications and to minimize systematic errors because of incorrect use or misinterpreting of different computer codes. The results of the simulations shown here are all in good accordance with real experimental results or published benchmarks. At the moment we are compiling some additional measurements for testing the capabilities of our codes in use to estimate the correct detector efficiencies in combined neutron-photon experiments.

**Table 2** Assumed dominant spent fuel parameters

| | min | max |
|---|---|---|
| Initial Enrichment (%) | 1.5 | 4.5 |
| Burn-up (GWd/tU) | 15 | 50 |
| Cooling-time (y) | 2 | 30 |

## III. Application to Super-Compacted Waste

After completing the benchmarking we started with the evaluation of real super-compacted waste residues. **Table 2** shows the assumed parameters for the spent fuel characteristics of the hypothetical super-compacted rad-waste.

At the first stage we calculated different ORIGEN-ARP cross-section files covering the whole parameter range of the assumed fuel characteristics from our SCALE/NEWT models of the different fuel element layouts of the German nuclear power plants,. The nuclide distributions of the fuels were calculated in subsequent runs and stored in a hierarchical data file for further processing. From these the relevant key nuclides information was transferred to the MCNP5 model. The coupling between these two computer codes is necessary to correctly calculate the detector responses depending on the spent fuel parameters.

The next step of the evaluation is to set-up the models of the real detection systems used to characterize the waste residues.[2)]

## IV. Conclusion & Outlook

In this paper we presented our work combining different well-known computer codes to assist / support nuclear experts with the evaluation or assessment of a specific rad-waste product. The results presented here are limited to a comprehensive collection of benchmarks performed to ensure the quality of the simulation packages and their outputs. The next steps will be the evaluation of the waste product with the variation of different product specific model parameters. The burn-up calculations were limited to the PWR fuel type, which is the most dominant part of the expected waste stream. For a more general evaluation of the waste packages we have to evaluate the BWR fuel as well. We are convinced that this approach of simulating a variety of different aspects of nuclear waste residues and their properties will help to achieve a deeper understanding of the waste product characteristics themselves. With the combination of validated and benchmarked burn-up calculations and neutron/photon transport calculations of the rad-waste containers, the inventory declaration and derived parameters like dose-rates and thermal powers could be independently verified.

## References

1) S. M. Bowman, "Overview of the SCALE Code System," *Trans. Am. Nucl. Soc.*, **97**, 589–591 (2007).
2) A. C. Raoux, A. Lyoussi, C. Passard, C. Denis, J. Loridon, "Transuranic waste assay by neutron interrogation and online prompt and delayed neutron measurement," *Nucl. Instr. Meth.Phys. Res.*, **B207**, 186-194 (2003).
3) H. Toubon, T. Gain, B. Perot, A. Lyoussi, A. C. Raoux, G. Mehlman, X. de Brimount, P. A. Tulle, "Method for ascertaining the characteristics of the radiological content of canister of compacted hull and nozzle waste resulting from reprocessing at the La Hague of irradiated fuel assemblies of light water reactors," *Proc. Safewaste 2000* (2000).
4) A. Barreau, *Burn-up credit criticality benchmark: Phase II-D*, NEA No. 6227, (2006).
5) D. F. Hollenbach, L. M. Petrie, "Comparison of the CENTRM Resonance Processor to the NITAWL Resonance Processor in SCALE," *Trans. Am. Nucl. Soc.*, **78**, 158 (1998).
6) Y. Naito, M. Kurosawa, *T. Kaneko Data book of the isotopic composition of spent fuel in light water reactors*, JAERI-M 94-034, Japan Atomic Energy Research Institute (JAERI) (1994)
7) A. Hermann, R. Berndt, "New Possibilities in Isotope Correlation Analysis of Spent Nuclear Fuel," *J. Radioanal. Nucl. Chem.*, **80**, 189-198 (1983).
8) G. Caspary, *Bestimmung von spaltbaren Stoffen in Abfallproben durch Neutroneninterrogation*, Juel-3498, Forschungszentrum Juelich (1998).
9) A. C. Raoux, A. Lyoussi, C. Passard, J. Romeyer-Dherbey, C. Denis, J. Loridon, J. Misraki, P. Chany, "Nondestructive radioactive waste assay by using fast and thermal neutron interrogation," *Proc. Safewaste 2000*, (2000).
10) A. Mariani, C. Passard, F. Jallu, H. Toubon, "The help of simulation codes in designing waste assay systems using neutron measurement methods: Application to the alpha low level waste assay system PROMETHEE 6," *Nucl. Instr. Meth. Phys. Res.*, **B211**, 389-400 (2003).
11) K. H. Beckurts, K. Wirtz, *Neutron Physics*, Springer-Verlag, Berlin Goettingen New York, (1964).