ENVIRONMENTAL IMPACT OF A RADIOACTIVE WASTE CONTAINER FOR THE SPENT NUCLEAR FUEL

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ABSTRACT

The safety assessment for the transport and disposal of used fuel requires the calculation of radionuclide inventories in the nuclear fuel, in order to provide the source terms for radionuclide release.

In this paper, an analysis for a storage (or transport) fuel cask was performed using the previously evaluated radionuclides inventories. This analysis was made in order to contribute to a complete radioactive waste characterization, from the reactor discharging until underground final disposal.

The evaluation was performed using the SCALE 5.5 computer code system. As final results, the gamma and neutron total dose rates were obtained, at specified detector locations outside of the shipping cask, after a spent fuel cooling (decaying) time of 5 years.

1. INTRODUCTION

The activity of characterizing a spent fuel cask container consisted of:

- An evaluation of the *radiation source terms*, gamma and neutron sources strength and spectra, using SCALE5.5\SAS2H\ORIGEN code;
- A *shielding analysis* of the shipping cask, which was performed in three parts using the SCALE5.5\SAS2H\XSDRNPM-S\XSDOSE codes as follows:
    1. Cell-weighted cross sections were computed for the homogenized fuel-pin lattice representation of the assemblies of the cask, which can be either moderated or dry.
    2. Applying cross-section data for the other zone materials in the shipping cask, together with the cell-weighted fuel cross sections and zone-averaged fixed neutron and gamma source of the fuel zone, angular neutron and photon fluxes were computed for the system.
    3. A multidimensional treatment is applied to the angular leakage fluxes by XSDOSE to compute gamma and neutron dose rates at specified detector locations outside of the shipping cask.

2. PRODUCTION OF THE STRENGTH AND SPECTRA FOR THE USED FUEL NEUTRON SOURCES AND GAMMA RADIATION SOURCES

2.1. Overview of ORIGEN-S Code

ORIGEN-S (Oak Ridge Isotope GENeration) is the depletion and decay module in the SCALE code system [1]. It computes time-dependent concentrations and radiation source terms of a large number of isotopes, which are simultaneously generated or depleted through neutronic transmutation, fission, and radioactive decay. The calculations may be utilized to evaluate fuel irradiation within a nuclear reactor, or the storage, management, transportation, or subsequent chemical processing of spent fuel elements. ORIGEN-S is widely used in nuclear reactor and processing plant design studies, design studies for spent fuel transportation and storage, burnup credit evaluations, decay heat and radiation safety analyses, and environmental assessments.

2.2 Neutron Source Strengths and Spectra

The neutron source strengths and energy spectra computed by ORIGEN-S includes neutrons produced:

1) From spontaneous fission.
2) By (α, n) reactions;
3) By delayed (β–, n) neutron emission.

Only the homogeneous medium (α, n) option has been adopted. The method of computing the spontaneous fission and delayed neutron source is independent of the medium containing the fuel. However, (α, n) production varies significantly with the composition of the medium.

ORIGEN-S includes three (α, n) source options:
1) A UO2 fuel matrix,
2) A borosilicate glass matrix, and
3) An arbitrary problem-dependent matrix defined by the user input compositions.

The neutron spectra are generated in an arbitrary energy-group structure or that of a SCALE cross-section library specified by input. The total neutron source spectrum is computed as the sum of the spontaneous fission, (α, n), and delayed (β–,n) spectra.

2.3 The Gamma Source Spectrum

The ORIGEN-S decay case described applies the option for computing the gamma source spectrum of the spent fuel assembly. The spectrum is calculated for the photon energy group structure of the cross-section library specified for use in the shipping cask shielding analysis.
The Photon Data Library includes energy and photon intensity data for X-rays and gamma rays emitted during decay of 2100 nuclides taken from: the Evaluated Nuclear Data File (ENDF/B), the Evaluated Nuclear Structure Data File (ENSDF) and the Joint European File (JEF-2.2).
The code takes into account:

1) Prompt fission gamma rays from spontaneous fission of heavy nuclides;
2) Gamma rays from the fission products produced in spontaneous fission;
3) Gamma rays from (α, n) reactions for heavy-metal dioxides;
4) Gamma rays from bremsstrahlung production from beta and positron deceleration in UO2 and water using beta energy data from ENSDF.

The photon source is determined using all nuclides in the fuel and any activated components in the problem.
The photons from X-rays and gamma rays emitted from the nucleus for all decay modes are stored in the photon library as line-energy and intensity data.

3. RADIAL SHIELDING ANALYSIS

3.1 Overview of SAS2H Code

The SAS2 (Shielding Analysis Sequence No. 2) control module is an analytic sequence that that automates the following steps [1]:

1) A depletion and decay for a specified assembly geometry and irradiation history
2) Generation of gamma and neutron sources strength and spectra (in any desired energy group format)
3) A one-dimensional radial shielding calculation, XSDRNPM-S and dose evaluation, XSDOSE for a transport storage cask. In SCALE, the primary functional tool for simulating radiation transport through shields, is represented by the XSDRNPM-S routine, which is a well established discrete ordinate program for one dimensional analysis (1-D).

3.2 Radiation Transport and Dose Evaluation

XSDRNPM-S is a highly evolved one-dimensional discrete-ordinates transport program which has a wide variety of features. XSDRNPM-S is capable of performing neutron or coupled neutron gamma calculations with the scattering anisotropy represented in any arbitrary order. The primary emphasis in the solution algorithm is on the accurate calculation of the detailed spectral variations. However, with sufficient angular quadrature and spatial mesh specification, highly precise solutions to the Boltzmann transport equation are obtained for one dimensional slab, cylindrical or spherical problems.

XDOSE is a code used in conjunction with XSDRNP to compute the neutron/gamma flux and the resulting doses at various points outside a finite cylinder or sphere. Due to the fact that here are several disadvantages to using XSDRNPM-S for calculating a dose in an external void beyond a finite shield, the model XSDOSE is used in that case. It may also be used to compute the flux and/or dose at various points due to a finite rectangular surface or circular disc.

3.3 The SAS2 Sequence and Input Description

The Shielding Analysis Sequence No. 2 (SAS2) processes fuel assembly cross sections, compute photon and neutron source spectra, and evaluate dose rates from shipping casks by one-dimensional transport shielding analysis.

The execution of SAS2 includes (see Figure 1):

- Repeated passes through BONAMI-S, NITAWL-S, XSDRNPM-S, COUPLE and ORIGEN-S for cross sections processing and fuel burnup;
- Radiation source computation
- Radial shipping cask shielding analysis applying the calculated spent-fuel composition sources;
- And the final determination of dose rates by XSDOSE from the angular flux leakage

SAS2 computes the gamma and neutron doses rates at various distances from a specified shipping cask, which contains fuel assemblies having a prescribed reactor history and cooling time.

Only basic data are required to perform the SAS2 evaluating of a shipping cask. These data, briefly, include:

1. The material zone dimensions of both the shipping cask and the unit cell representation of the fuel assembly.
2. The material densities of the fresh fuel assembly and the shipping cask
3. The material temperatures
4. The specific power exposure and shutdown time of the fuel assembly in each appropriate cycle of the reactor history
5. Various control parameters used to select libraries, nondefault options, the level of printout, or modifications to the transport computations (for instance the fineness of mesh intervals or the problem convergence criteria)
6. Other optional data such as dose detectors distances that differ from default values or light element weights per assembly.

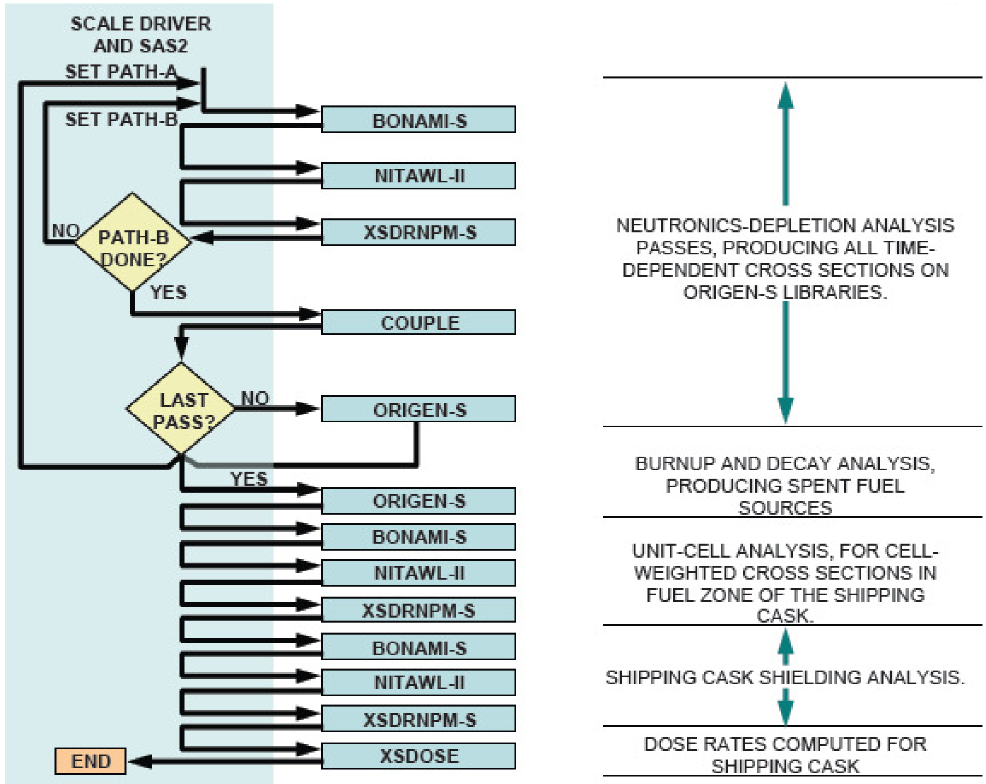


Figure 1 Computational flow chart for the SAS2 procedure [2], [3]

4. DESCRIPTION AND EVALUATION OF THE BWR CASK

The BWR cask case is an example pertaining to an analysis of an actual storage cask designed to contain 52 BWR spent fuel assemblies. Gamma and neutron dose rates have been measured at various positions on the surface of the cask.

4.1 BWR Assembly Description Data

In generally, the assembly and moderator specifications used in this case are those of References [4] and [5]. The 189-kg U assembly and fuel rod data are listed in Table 1. The weighted average U-235 enrichment is 2.40 wt % U-235 and other uranium isotopic contents are 0.021 wt % U-234, 0.011 wt % U-236, and 97.568 wt % U-238.

The SAS2 model for the assembly locates a gadolinium poison rod at the center of the larger unit cell. Four gadolinium poison rods (type 5 rods listed in Table 1) are located in the assembly, as shown in Figure 2.

Table 1 BWR assembly description [4]

Assembly general data:	
Lattice, designer	8 × 8, General Electric
Type	Burnable poison
Water density, vol-av, g-cm-3	0.392
Water temperature, av K	558
Number of poison rods	4
Number of holes	1
Channel material	Zircaloy-4
Channel water density, av, g-cm-3	0.743
Fuel rod data:	
Pellet diameter, in.	0.4160
Gap (diametrical), in.	0.0090
Rod OD, in.	0.493
Fuel rod pitch, in.	0.640
Pellet stack density, % TD	94
Clad material	Zircaloy-2
Active fuel length, in.	148
Plenum (fuel rod) length, in.	10
Fuel temperature, K	840
Clad temperature, K	620
Type 1: 31 rods, wt % 235U	2.64
Type 2: 16 rods, wt % 235U	2.30
Type 3: 8 rods, wt % 235U	2.12
Type 4: 4 rods, wt % 235U	1.65
Type 5: 4 rods, wt % 235U	2.30, with 2.0 wt % Gd2O3

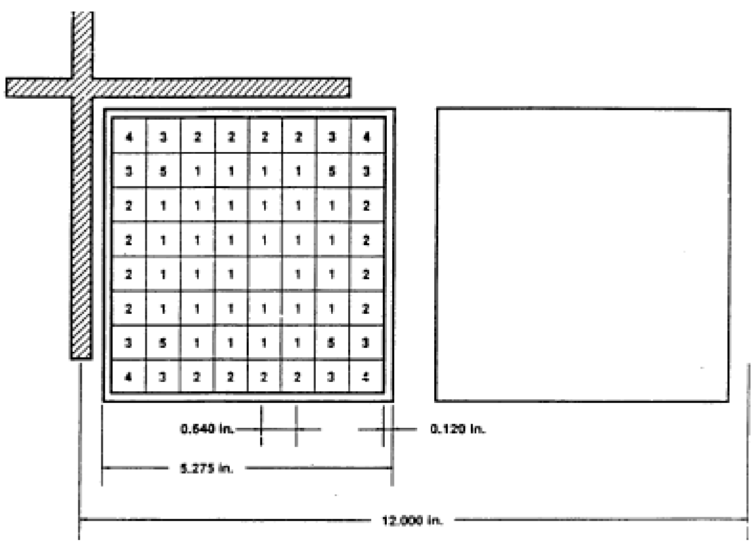


Figure 2 BWR assembly schematic [5]

Table 2 BWR assembly operating history [4]

Cycle	Accumulated time, d	Time difference, d	Burnup, GWd/MTU	Cycle burnup, GWd/assm.	Power, MW/assm.
1	807	807	10.651	2.013	2.494
	866	59	10.651	0	0
2	1367	501	21.430	2.037	4.066
	2166	799	21.430	0	0
3	2878	712	25.383	0.747	1.049

Given SAS2 parameter selections:

- Reactor-fuel-depletion-case library: 44(n) groups;
- Shielding-analysis-case library: 27(n)-18(g) groups;
- Number of libraries produced/cycle = 1;
- Skip cask cell-weight case, or PARM='SKIPCELLWT';
- Detector locations (at midplane, only): 0, 2.54, 100, 200, 500 cm from cask.

Given storage cask description data:

- Type of fuel zone coolant: none, dry fuel;
- Temperature of cask, K = 380;
- Number of zones = 9;
- Materials by zone radii:
  1: 75.32 cm; 52 fuel assemblies, SS304, boral, Cu, Zircaloy;
  2: 75.70 cm; Zircaloy;
  3: 75.93 cm; boral;
  4: 76.57 cm; Cu;
  5: 79.29 cm; SS304;
  6: 90.09 cm; Pb;
  7: 95.17 cm; SS304;
  8: 110.41 cm; ethylene glycol water solution;
  9: 111.05 cm; SS304;

4.2 Dose rates results

Fluxes and Dose rates at 0, 1, 2 and 5 meters radial position from the cask are shown in Table 3 below: (BWR Fuel Elements, Burnup = 25 300 MWd/t, after a cooling time of 5 years)

Table 3 Total fluxes and dose rates

Detector	**Fluxes**		**Doses**		**Total Dose Rate**	
Position	neutrons	gamma	neutrons	gamma		
	n/cm^2*s	photon/cm^2*s	mrem/h	mrem/h	mrem/h	mSv/h
Surface	8.32E+01	1.05E+04	1.16	17.3	18.46	0.18
1 m	3.06E+01	4.32E+03	0.45	7.63	8.08	0.08
2 m	1.66E+01	2.50E+03	0.25	4.57	4.82	0.05
5 m	4.56E+00	7.67E+02	0.08	1.47	1.55	0.02

The following charts chart in Figure 3 and Figure 4, illustrate the data from Table 3.

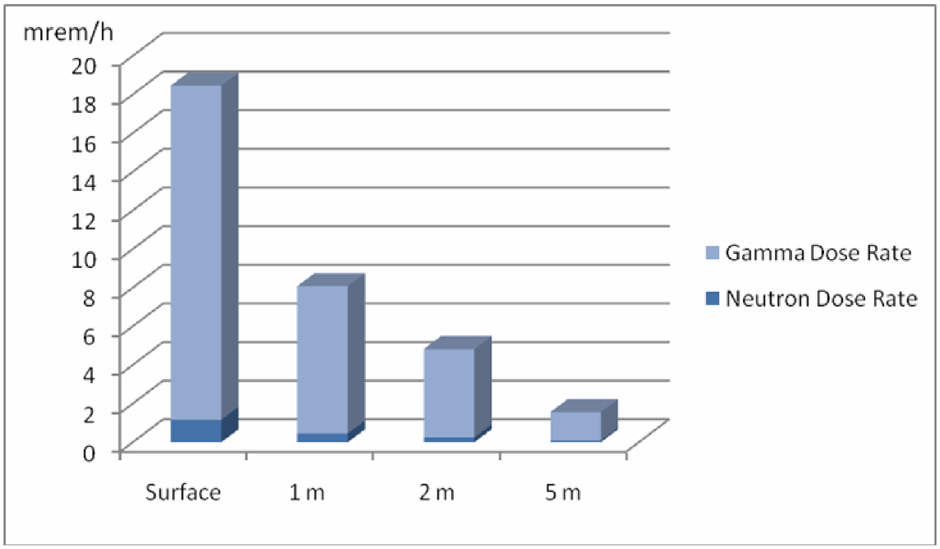


Figure 3 Total dose rates at the surface, 1, 2 and 5 meters from BWR cask

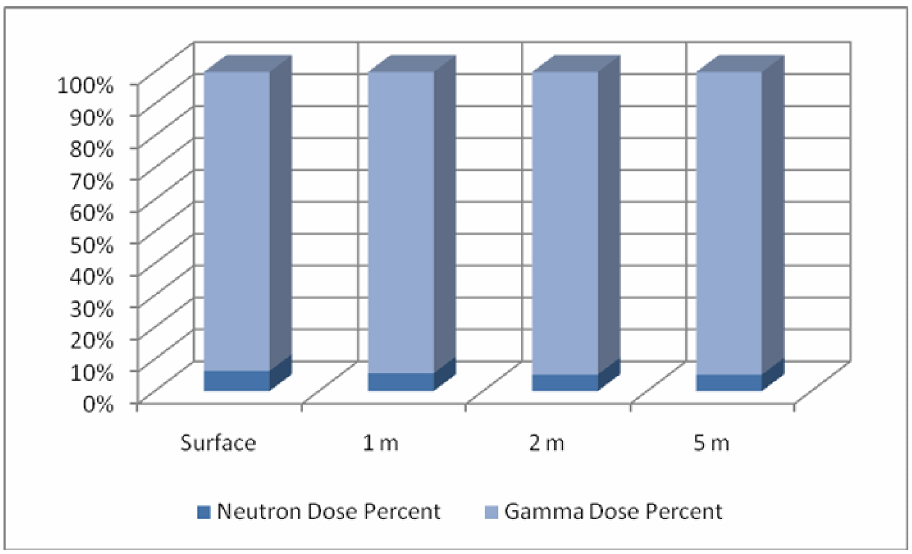


Figure 4 Total dose rates percent proportions at the surface, 1, 2 and 5 meters from the BWR cask.

4.3 Comparison of the calculated and measured doses

A profile of measured gamma dose rates was obtained along the axial elevation of the cask surface. The thermoluminescent dosimeters (TLDs) used to obtain the profile of dose rates required a calibration to the gamma spectrum. The average gamma dose rate derived by height-segment weighting was 12.5 mrem/h. The gamma dose at the cask midplane (peak) was 14.4 mrem/h. [5]
The final average neutron dose rate, after applying the average calibration factor, was 1.24 mrem/h. The midplane (peak) neutron dose rate was 1.77 mrem/h after calibration. [5]

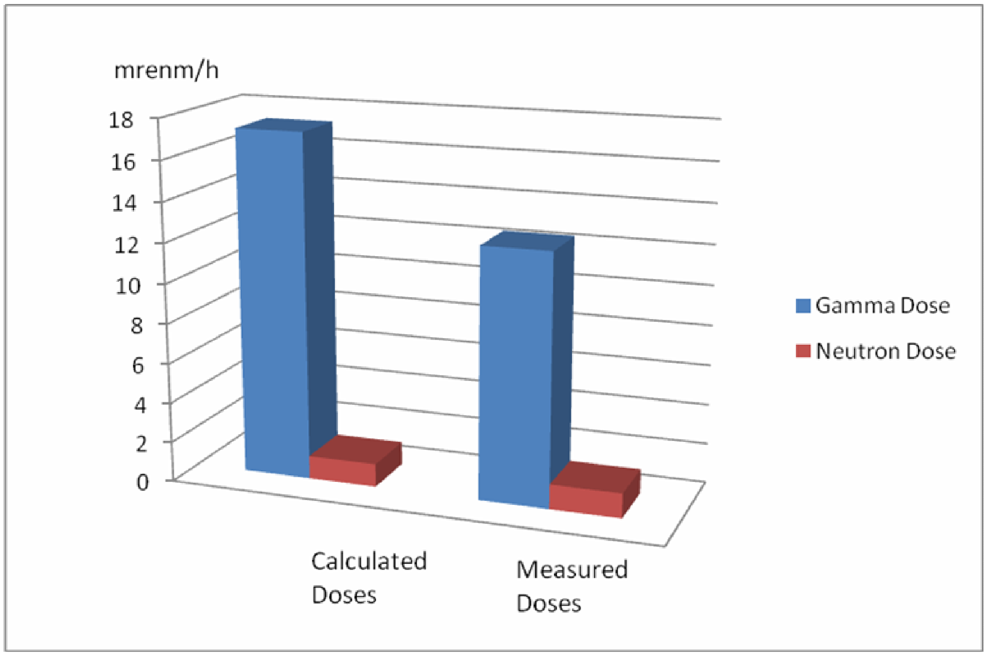
The Table 4 compares the measured doses located at 26.5° on cask surface with the calculated doses obtained from the SAS2 sequence. The calculated doses are lower compared with the measured neutron dose and higher compared with the measured gamma dose.

Table 4 Computed dose rates and measurements in mrem/h at the cask surface

Radiation type	Calculated by SAS2 [mrem/h]	Average dose measurement [5] [mrem/h]	Peak dose measurement [mrem/h]	Discrepancy (Calc.-Meas.)/Meas*100 [%]
Gamma dose rate	17.3	12.5	14.4	34..8%
Neutron dose rate	1.16	1.24	1.77	-6.45%

The previous table is graphically illustrated in Figure 5 bellow.

Figure 5 Comparison between calculated and measured gamma and neutron doses



The differences between measured and SAS2-computed values may result from several reasons:

- The depletion analysis used a different BWR assembly design;
- Uncertainty in the spent fuel depletion model;
- The shielding analysis assumed the cask to be fully loaded with assemblies having the characteristics of the one assembly nearest the detector location;
- The shielding analysis model homogenizes the fuel over the entire cask cavity;
- General limitations of a 1-D computational model;
- Uncertainty in the cross-section data.

5. CONCLUSIONS

The activity described in the paper represents a study of the transport and interim storage container casks for a BWR type used fuel. The cask shielding analysis produced the neutron and photon fluxes and corresponding doses were calculated using the SCALE5.5\SAS2 codes system.

The BWR fuel type was primarily chosen for the analysis due to the availability of the experimental data for providing a validation of the calculation procedure. A similar evaluation for the NPP Cernavoda CANDU spent fuel is currently under development for a CASTOR/POLUX type container cask.

The ANSI standard neutron flux-to-dose-rate factors and ANSI standard gamma-ray flux-to-dose-rate factors were used to estimate the neutron and gamma doses. [6]

The doses were calculated, in both analyzed cases, at 4 detector positions, at 0, 1, 2, and 5 meters radial position from the container, after a cooling time of 5 year. As expected the doses are decreasing as the distance from the container increases. The dose coming from the gamma rays is constantly higher than the dose generated by the neutrons.

A comparison between the measured doses located on cask surface and the calculated doses was made. The calculated doses are lower than the measured neutron dose and higher than the measured gamma dose. The discrepancy between the calculated and measured neutron dose is 6.45% resulting a good agreement. The discrepancy between the calculated and measured gamma dose is 34.8%. The difference is within the measurement uncertainty.
This comparison could represent a validation of the dose evaluation methodology presented in the paper.

To completely characterize the nuclear spent fuel at the end of its cycle, a future study would be performed. This will involve a complete analysis for the final disposal of the spent fuel including the criticality safety analysis of the disposal configuration using the SCALE5\CSAS sequence (KENO Monte Carlo module).

6. BIBLIOGRAPHY

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