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Title:	Delayed-Neutron and Delayed-Photon Dose-Rate Estimations for Chemical Processing of Spent Fuel Using MCNP6
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Delayed-Neutron and Delayed-Photon Dose-Rate Estimations for Chemical Processing of Spent Fuel Using MCNP6

by

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ABSTRACT

A nondestructive assay (NDA) computational study is conducted using the MCNP6 Monte Carlo radiation-transport code to calculate dose estimates caused by delayed-neutron (DN) and delayed-gamma (DG) emission for spent nuclear fuel (SNF) that is immersed in nitric acid. The SNF isotopic inventories and delayed-particle (DP) sources are prepared using a generic Westinghouse fuel assembly model for three sets of parameters: 3% and 5% ^{235}U enrichments; 20-, 30-, 40-, and 50-GWd/MTU burnups; and 3-, 5-, 10-, 20-, and 30-year cooling times. MCNP6 is used to do kcode/burnup calculations to create isotopic inventories for the irradiated fuel models. The SNF assembly is placed in a cylindrical steel vessel containing 7M nitric acid. Doses attributable to delayed-particle (DP) emissions are assessed for vessel inner diameters of 1.5 m and 0.32 m at 3.048-m intervals. Perl scripts are created and used to automate the creation of input files, execution of calculations, and data mining of output files for presentation of pertinent data in suitable format. The MCNP6 calculations are executed using message passing interface (MPI) on the Pete Linux cluster in a matter of minutes per calculation. The simulations show that the dose rates are dependent upon enrichment, burnup, and cooling. Neutron dose rates are greatest for the 0.32-m-diameter vessel containing SNF for 3% ^{235}U enrichment, 50 GWd/MTU burnup, and 3-year cooling case: ~2 rem/hr at the surface of the vessel, decreasing to ~10^{-3} rem/hr at 30.48 m from the vessel. Photon dose rates are also greatest for the 0.32-m-diameter vessel containing SNF for the case with 3% ^{235}U enrichment, 50 GWd/MTU burnup, and 3-year cooling: ~26000 rem/hr at the surface of the vessel, decreasing to ~10 rem/hr at 30.48 m from the vessel. Dose rates for the 1.5-m-diameter vessel are approximately 2 orders of magnitude smaller than the smaller vessel. Dose rates decrease with enrichment, increase with burnup, and decrease with cooling time.

KEYWORDS: MCNP6; chemical processing; delayed neutrons; delayed gammas; dose.

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Acronyms

3D	Three Dimensional
BAMF-DRT	Burnup Automation MCNPX File—Data Retrieval Tool
DG	Delayed Gamma
DN	Delayed Neutron
DP	Delayed Particle
FPAI	Fission Product and Actinide Inventory
LANL	Los Alamos National Laboratory
MPI	Message-Passing Interface
NDA	Nondestructive Assay
PC	Personal Computer
SNF	Spent Nuclear Fuel

1 INTRODUCTION

Nondestructive assay (NDA) methods are an important means by which burnup and fissile content for irradiated nuclear fuels can be quantified for fuel reprocessing and safeguards applications (Hsue et al., 1978). NDA techniques use delayed-gamma (DG) and/or delayed-neutron (DN) emissions and the relationship between fission-product yield and the number of fissions, or fuel burnup. NDA techniques are useful for laboratory and field assessment. The goal of NDA analysis is to provide a means of determining burnup and isotopic composition (fissile content) from measured DG or DN signals.

Los Alamos National Laboratory (LANL) develops and maintains the MCNP6 general-purpose radiation transport code (Goorley et al., 2012). MCNP6 accommodates intricate three-dimensional (3D) geometrical models, continuous-energy transport, criticality calculations and fuel burnup, and delayed-particle (DP) treatment. MCNP6 has a kcode/burnup feature that links Monte Carlo particle-transport and depletion capabilities. This feature enables isotopic transmutation studies for complex 3D geometries with exotic material combinations and highly anisotropic flux behavior. MCNP6 is written in Fortran 90, has been parallelized, and works on platforms including single-processor personal computers and Linux clusters. The parallel-execution capability greatly reduces execution time for simulations treating highly detailed SNF isotopic inventories and DP treatments.

The MCNP6 burnup “Tier 3” option provides the most comprehensive fission-product and actinide inventory available in MCNP6. Simulations that are executed with Tier 3 provide DG and DN sources to the fullest possible extent. The Tier 3 option was used to produce the SNF inventories in this study.

The MCNP6 DP feature treats delayed-neutron (DN) and delayed-gamma (DG) emission caused by radioactive decay of unstable fission or activation products. The MCNP6 capability pertains to reactions that have unstable residuals decay, with half-lives ranging from microseconds to thousands of years. The DG feature can provide photon emission for detailed analyses using discrete (line) data (Durkee et al., 2009a, 2009b, 2010, 2012). To our knowledge, MCNP6 is the only Monte Carlo code that can execute high-resolution DG simulations.

The following section contains a description of the fuel-assembly model used in this study.

2 SNF FUEL-ASSEMBLY MODEL

The MCNP6 burnup models for the fuel assembly used here were prepared in a previous study (Durkee et al., 2012). The SNF inventories calculated in that study are used in this study. The model and the computational processing are reviewed here.

The fuel-assembly model is based on a generic 17 × 17 Westinghouse infinitely reflected assembly (Fensin et al., 2009; OECD-NEA, 2012). The general assembly parameters are listed in Table 1. The coolant was chosen to be a 660-ppm average boron concentration at a density of 0.7245 g/cm^3 and temperature of 575 K. Figure 1 shows the MCNP6 fuel-assembly model.

Table 1. Generic Westinghouse 17 × 17 Assembly Parameters

Parameter	Data
Assembly general data	
Lattice	17 × 17
Number of fuel rods	264
Number of guide tubes	24
Number of instrument tubes	1
Fuel rod data	
Type of fuel pellet	UO_2 (10.4538 g/cm^3)
Rod pitch	1.26 cm
Clad thickness	0.065 cm (no gap between fuel and clad)
Pellet diameter	0.410 cm
Active fuel length	365.76 cm
Fuel temperature	900 K
Clad temperature	620 K
Clad material	Zircaloy-4 (5.8736 g/cm^3)
Guide and Instrument tube data	
Inner radius	0.571 cm
Outer radius	0.613 cm
Material	Zircaloy-4 (5.8736 g/cm^3)

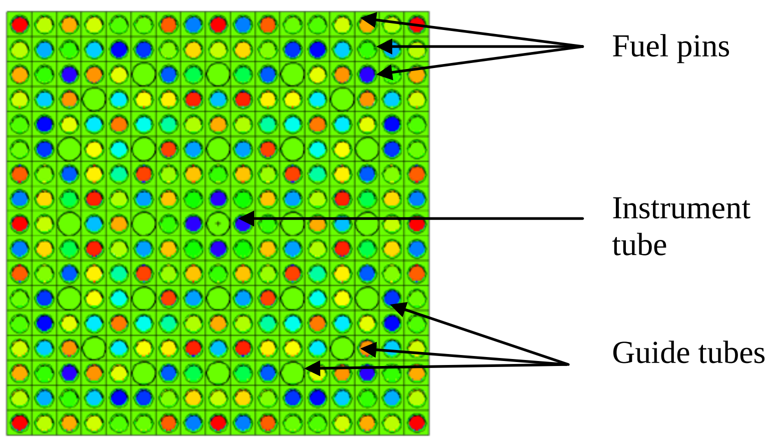

Figure 1. MCNP6 midplane fuel assembly geometry plot.

To facilitate parametric studies, the MCNP6 burnup models based on the Westinghouse fuel assembly were prepared for 3% and 5% ^{235}U enrichments;[†] 20-, 30-, 40-, and 50-GWd/MTU burnups; and cooling times of 3, 5, 10, 20, and 30 years. These models were executed (in the previous study) using the MCNP6 Tier 3 burnup inventory option. The Tier 3 option provides inventories for all ENDF/B-VII.0 fission products that have yield information in the CINDER'90 transmutation code (Pelowitz, 2011). The kcode/burnup models included materials and cross

[†] Throughout the remainder of this report the enrichment terminology without specific reference to ^{235}U will be used for brevity.

sections at realistic reactor temperatures. Fuel, clad, and water were treated at 900 K, 620 K, and 575 K, respectively.

The new MCNP6 threading capability (Fensin et al., 2012) was used to execute the data-intensive kcode/burnup calculations (Durkee et al., 2012). Threading facilitates parallel execution using shared memory with reduced memory requirements. Message passing interface (MPI) execution alone enables parallel execution but requires copies of arrays—one per processor. Threading allows parallel execution with array sharing, thus reducing memory requirements. Execution was done on the LANL Pete Linux cluster. Each of the 8 kcode/burnup jobs required 4 to 7 days of execution time using 45 dedicated processors.

Once the burnup calculations were completed, the burnup input files were modified to facilitate DP calculations. To do so, the kcode/burnup (kcode) execution mode was changed to source (SDEF) execution mode. DP sources corresponding to the isotopic inventories at the various enrichment, burnup, and cooling conditions were calculated and inserted into the files. Accounting for enrichment, burnup, and cooling conditions, a set of 40 DN models and 40 DG models was created.

The DP source and the dose tally developments were done as follows. Default MCNP6 tally values are reported in units per source particle. Normalization to the source is done using the MCNP6 tally multiplier (FM) to give tally data in terms of particles/MeV-s. The FM values were calculated using DN and DG volumetric-source data produced by the Burnup Automation MCNPX File-Data Retrieval Tool (BAMF-DRT) for the previous study (Durkee et al., 2012). These DP source data are also used for the SDEF sources in the models. The volume of the active portion of the fuel assembly was used to calculate the total DN and DG source rates.

The DP input files used in the previous study were modified for use in this study as described in the following section.

3 MCNP6 MODEL PREPARATION

MCNP6 input files were prepared for 3% and 5% fuel enrichment, 20-, 30-, 40-, and 50-GWd/MTU burnup, and the 3-, 5-, 10-, 20-, and 30-year cooling times starting with the DP versions of the fuel-assembly models from the previous study (Durkee et al., 2012). Those models are modified so that:

- A 0.5-inch-thick (1.27 cm) cylindrical steel vessel encloses the fuel assembly.
- Calculations are done for two vessel inner diameters: 1.5 m and 0.32 m (this diameter allows the fuel assembly to fit inside of the vessel).
- The fuel assembly is placed in 50000 liters of 7M nitric acid (commercially available nitric acid is an azeotrope with water[†]) inside of the steel vessel.
- Cross-section libraries at 300 K are used.
- Dose-rate tallies are added.

[†] An azeotrope is mixture of two or more liquids in such a way that its components cannot be altered by simple distillation.

The analysis considers dose rates for an intact fuel assembly. No analysis has been performed to treat dissolution.

The dose-rate tallies are calculated using the MCNP6 surface-averaged flux tally ("F2"). Once these tallies are normalized to the source (see preceding Section), the normalized fluxes are converted to dose rates using the MCNP6 dose energy (DE) and dose function (DF) flux-to-dose conversion factors (and associated cards in the input files). These quantities are listed in Appendix H of the MCNP manual for neutrons and photons with units of (rem/hr)/(particle/cm^2-s).

The dose calculations in this study are calculated on 11 cylindrical surfaces beginning at the outer surface of the vessel. The cylindrical surfaces are radially spaced at 304.8-cm (10-foot) intervals. The MCNP6 tally segmentation feature is used to acquire surface-averaged doses on the cylindrical surfaces between the base of the vessel and a height of 182.88 cm (6 feet, a representative height of an adult male). The tally numbering convention for this set of tally surfaces is given in Table 2 for the two vessels.

Table 2. Tally convention for radial dose-rate calculations.

Vessel outer surface is at 76.270 cm.

Tally	Radial location (cm)	
	1.5-m model	0.32-m model
2	76.271	17.271
12	381.071	322.071
22	685.871	626.871
32	990.671	931.671
42	1295.471	1236.471
52	1600.271	1541.271
62	1905.071	1846.071
72	2209.871	2150.871
82	2514.671	2455.671
92	2819.471	2760.471
102	3124.271	3065.271

Because this study treats several sets of enrichment, burnup, cooling time, and dose-related parameters, development of the input files, their execution, and the examination of the output files is a tedious and cumbersome process that can entail appreciable error if done manually. In this study, these manipulations were automated using Perl scripts as follows:

- Perl script **makeinp.pl** was created to make the input (inp) files for each enrichment, burnup, and cooling time. First, the DP-source input files for the SNF models (Durkee et al., 2012) are copied to the DVR directory – a directory on the Pete Linux cluster that was created for this study. These files are then copied to a working directory where modifications to reflect changes to geometry (addition of the steel vessel), material (use of nitric acid), tallies (tally type and segmentation for dose estimates), number of source histories (NPS), etc., is done for the entire suite of DN or DG input (inp) files. The inp files for this study are named i3b20c3dn (3% enrichment, 20

GWd/MTU, 3-year cooling, DN), i5b50c20dg (5% enrichment, 50 GWd/MTU, 20 year cooling, DG), etc.

Execution automation on the Pete cluster was facilitated with additional scripting:

- Perl script **copyinp.pl** was created to automate the creation of execution directories and copy the DN and DG inp files to those directories on Pete. The directory naming convention follows that used for the input files.

- Perl script **makeqsub.pl** was created to make dn.qsub (DN jobs) and dg.qsub (DG jobs) files containing file name and directory labels for each inp file and to copy each qsub file to the appropriate directory on Pete. The qsub files contain commands for job submission to the Network Queueing System. Changes to the reference qsub file qsubi, such as number of nodes or processors, base directory name, and MCNP6 executable name, can easily be made. **makeqsub.pl** then generates dn.qsub or dg.qsub files and places them on the subdirectories.

- Perl script **rundp.pl** was created to execute the MCNP6 jobs. Script **rundp.pl** submits the qsub files on each subdirectory for all jobs.

- Perl script **moveomr.pl** was created to rename the output (outp), tally (mctal), and run tape (or runtpe) files, giving unique identifiers with enrichment, burnup, and cooling time. The filenames correspond to the inp files, with suffixes "o", "m", and "r" affixed (instead of the "i" suffix for the input files).

- Perl script **extractt.pl** was created to (1) parse the outp files; (2) locate the bins and tally data for the dose rates (energy-dependent and total); and (3) write the dose-rate data files whose formats are suitable for plot creation using gnuplot (Williams and Kelley, 2007). Naming conventions adhere to those for the DN and DG outp files with additional suffixes denoting dose rates for gnuplot plots. For the energy-dependent dose-rate data, the suffixes include the tally number (which corresponds to the radial distance of each tally surface outside the vessel) and "g" to denote gnuplot. For the energy-integrated (total) dose rates, the file suffixes are "Tg". Filenames for energy-dependent dose rates are thus of the form i3b20c3dgt2g (3% enrichment, 20 GWd/MTU burnup, 3-year cooling, tally 2). For energy-integrated files, the convention is of the form i3b20c3dgTg (3% enrichment, 20 GWd/MTU burnup, 3-year cooling, total). The gnuplot files **gnuptalT.plt** and **gnuptalE.plt** were created to provide plots of total (energy-integrated) and energy-dependent dose rates for DN and DG radiation.

Each MCNP6 dose-rate calculation was executed using 10^6 source particles (neutrons or photons). The calculations were performed using an Intel MPI MCNP6 executable on the Pete Linux cluster using 48 processors. Execution of the respective DN and DG simulations was done using either neutron or photon transport. No coupled neutron-photon simulations were executed. The dose-rate estimates are thus somewhat underestimated because neutrons and photons created fission and activations reactions, including activation of the steel vessel, are neglected.
The following sections contain the results for the two vessel models.

4 MCNP6 DOSE ESTIMATES FOR THE 1.5-M MODEL

The MCNP6 models consist of the fuel assembly immersed in nitric acid inside of a cylindrical steel vessel. MCNP6 geometry plots for the 1.5-m-diameter model are shown in Figure 2.

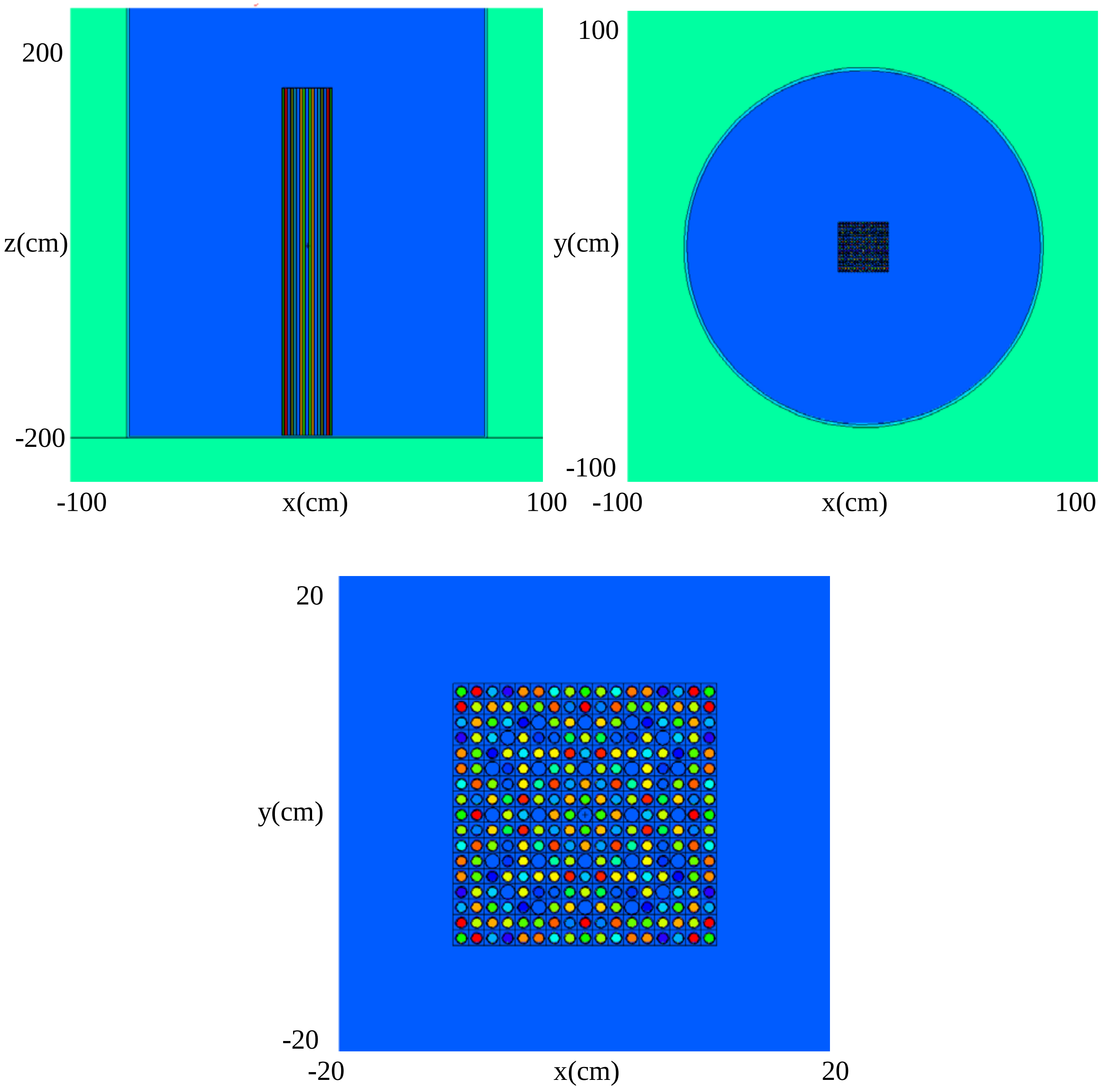

Figure 2. MCNP6 geometry plots of the 1.5-m-diameter chemical processing model showing fuel assembly, nitric acid (blue), and air (green).

Figure 3 contains plots of the neutron dose rate as a function of radial distance for 3% ^{235}U enrichment. The tally data are reported at locations listed in Table 2. The outer surface of the vessel is located at 76.27 cm. At a given position, the dose rate increases as the burnup increases because of the increase in fission-product inventory with increased burnup. Error bars are included in the plots – the relative uncertainties for the energy-integrated dose rates are on the order of 0.01 at the surface of the vessel and 0.03 at 100 feet. These values are statistically reliable. Each of the 10 statistical checks* used by MCNP6 (Brown, 2003, pp. 123–127) were passed, suggesting the acceptability of the dose-rate tallies.

As a check, the neutron dose rate can be estimated as follows. The DN emission is caused to a large extent by emission from ^{242}Cm and ^{244}Cm. The masses of these isotopes are available MCNP6 output files (print table 220). For 3% enrichment, 20 GWd/MTU burnup, and 3 year cooling, the masses of these isotopes are 1.545e-2 and 1.437 g, respectively. The spontaneous fission yield of these isotopes is 2.10e7 and 1.08e8 n/s-g, respectively (Table 11-1 of Reilly et al., 1991). Assuming that the entire mass of these nuclides can be represented as a point source, then the source strength is 1.58e7 n/s. The corresponding flux at the surface of a voided sphere 10 cm in diameter (the fuel assembly is 11.7 cm wide), the neutron flux is 5.03e4 n/cm^2-s. According to experimental work (Batchelor and Hyder, 1956), the mean energy of delayed neutrons is ~0.45 MeV. From Table H.1 of the MCNP manual, the flux-to-dose conversion factor for 0.5-MeV neutrons is 1.32e-04 (rem/hr)/(n/cm^2-s).† The corresponding dose rate is 4.65 rem/hr. The MCNP6 dose estimate for the surface of the fuel assembly is 0.18 rem/hr. The analytic result should be greater than the MCNP6 result because the analytic result is estimated for a void. The analytic and MCNP6 results are of the same order.

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*MCNP6 uses 10 statistical checks to form statistically valid confidence intervals for each tally bin.

† The tabulated values vary from 3.7e-6 at E = 2.5e-8 to 2.27e-4 at E = 20 MeV. Data are listed for 0.1, 0.5, and 1 MeV. For the approximation purposes here, the flux-to-dose conversion value at 0.5 MeV is used.

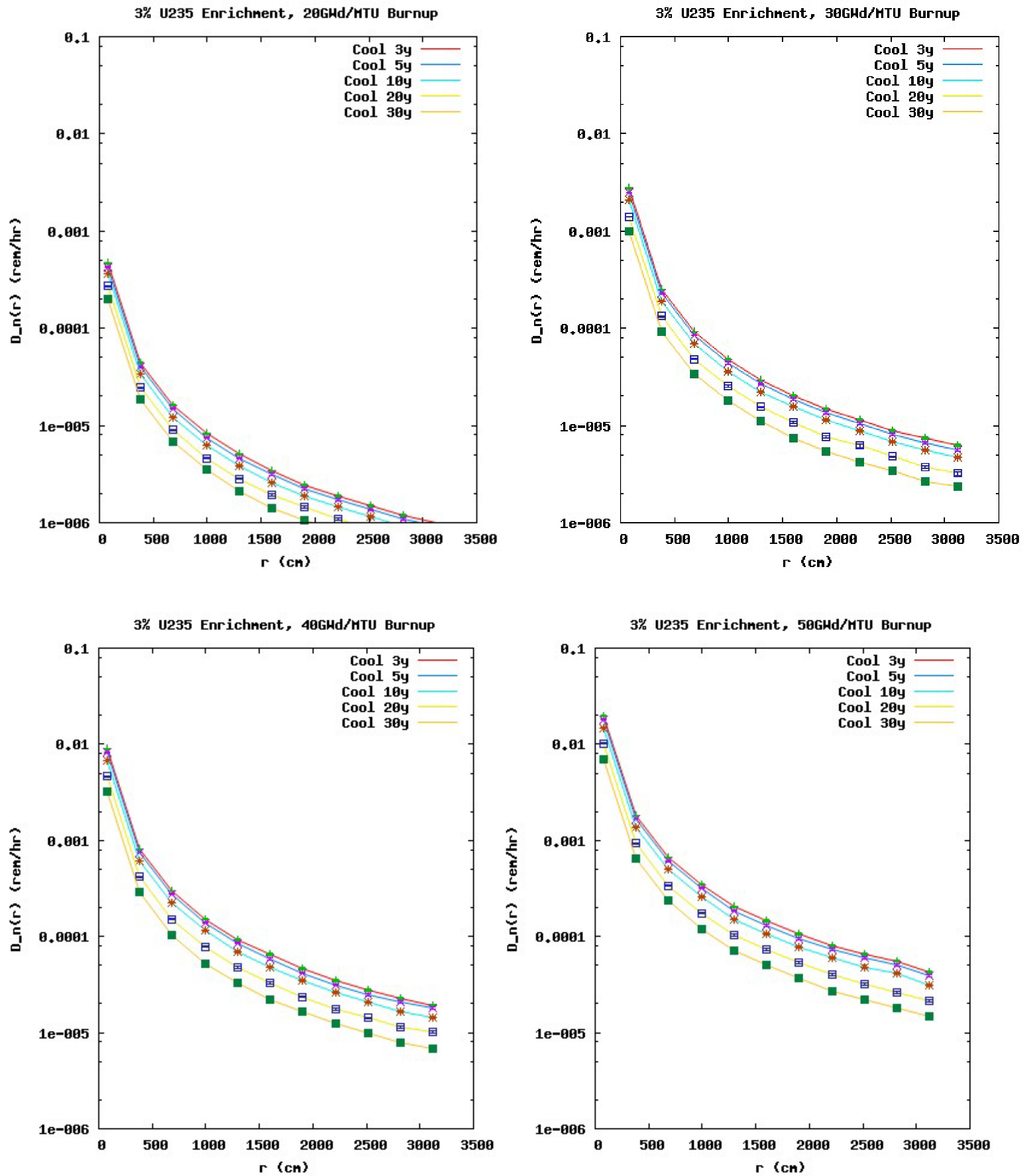

Figure 3. Neutron dose rate as a function of distance for 3% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

Figure 4 contains plots of the neutron dose rate as a function of radial distance for 5% ^{235}U enrichment. The dose rates are lower than those for the corresponding conditions at 3% enrichment because constant-power irradiation for 5% enrichment occurs at a lower flux than for 3% enrichment. Consequently, the buildup of fission products for 5% enrichment is lower than that for 3% enrichment and the corresponding neutron emission and dose rates are lower at 5% enrichment than 3% enrichment.

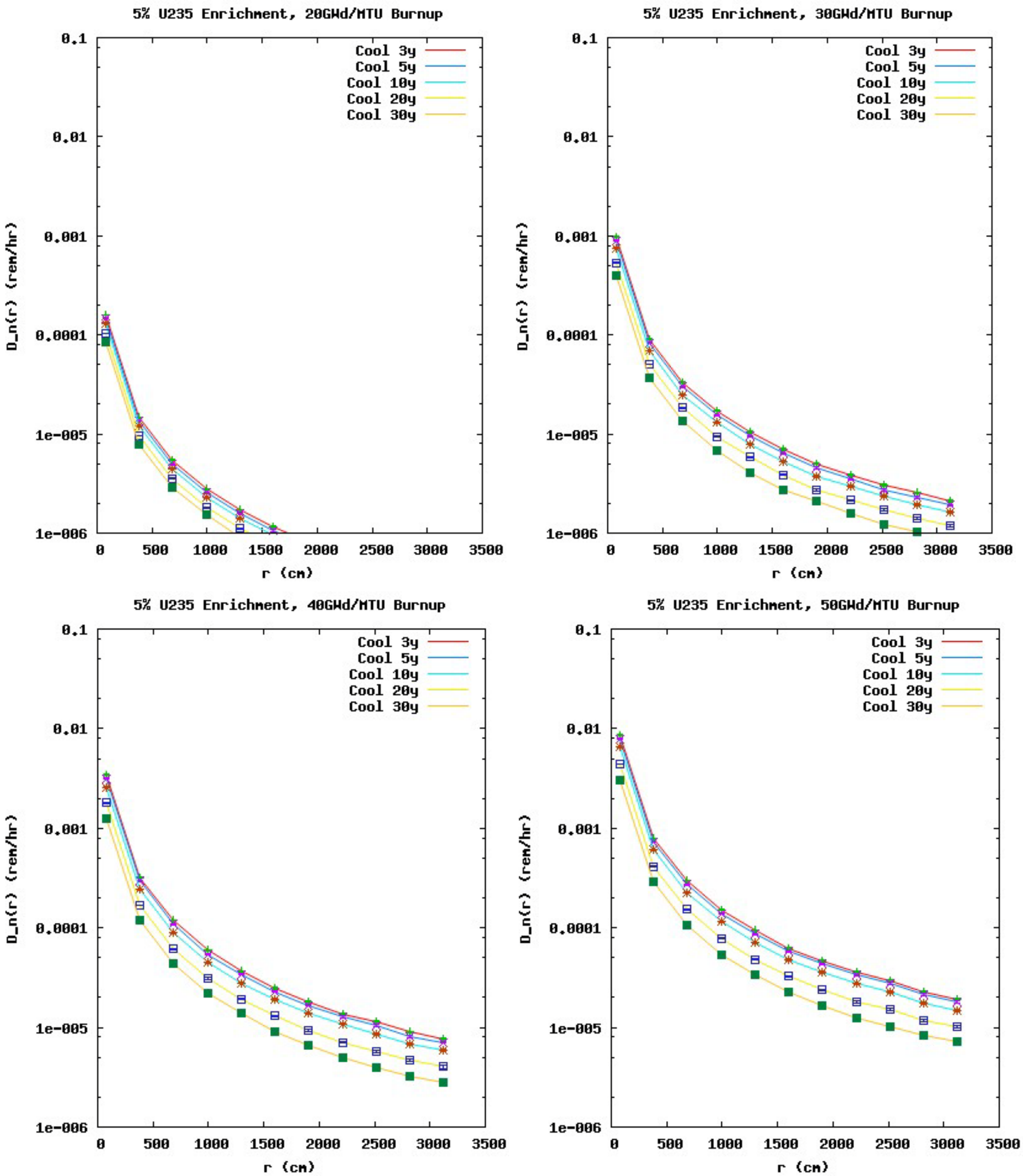

Figure 4. Neutron dose rate as a function of distance for 5% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

Figure 5 contains plots of the photon dose rate as a function of radial distance for 3% ^{235}U enrichment. The relative uncertainties for the energy-integrated dose rates are on the order of 0.01 at the surface of the vessel and 0.05 at 100 feet. In general, the gamma dose rate should dominate the neutron dose rate because the neutron production by spontaneous fission of (primarily) ^{242}Cm and ^{244}Cm occurs only a small fraction of the times that decay occurs. The MCNP6 photon dose-rates dominate their neutron counterparts.

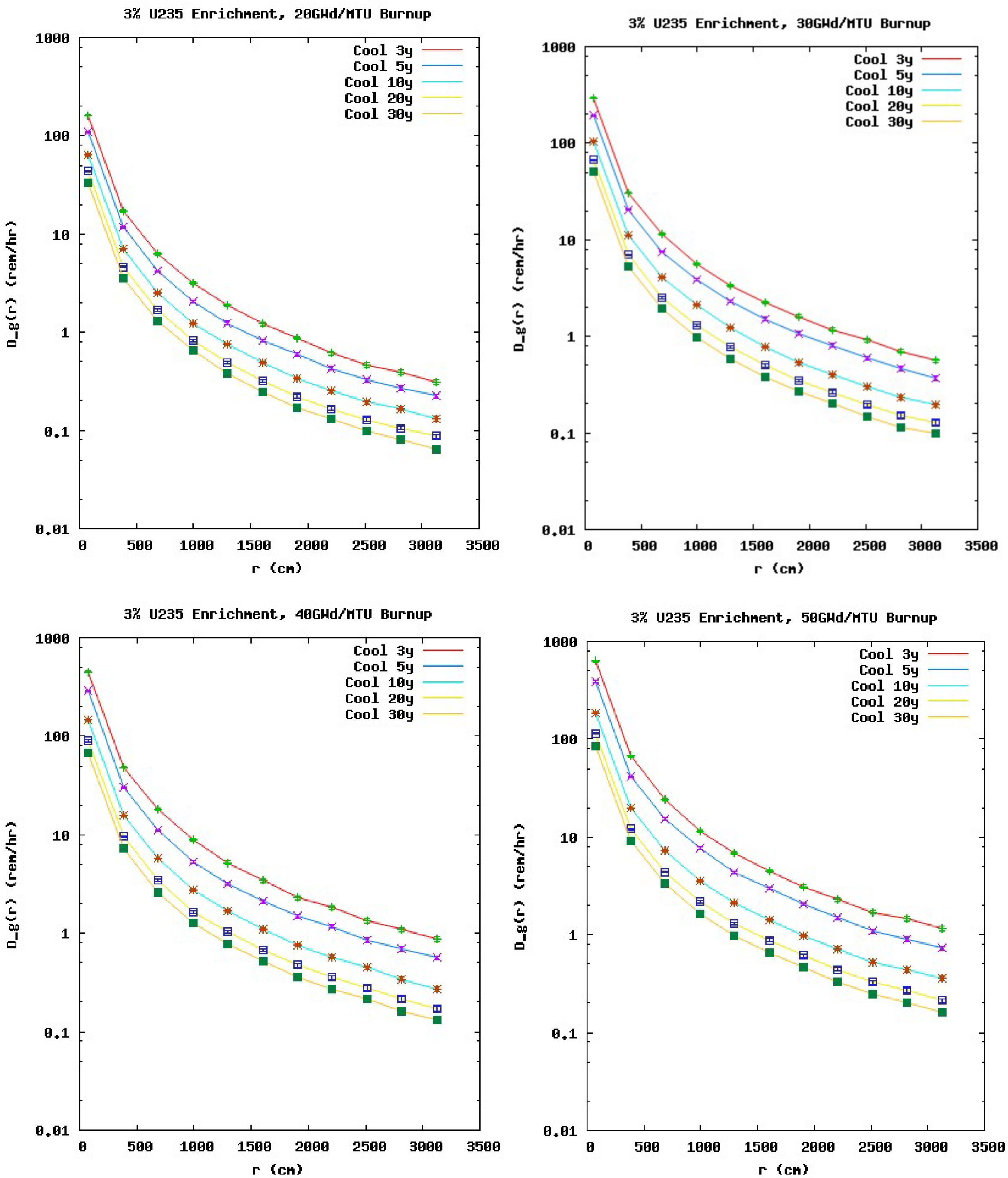

Figure 5. Photon dose rate as a function of distance for 3% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

Figure 6 shows the photon dose rates for 5% enrichment. The corresponding photon emission and dose rates are lower at 5% enrichment than 3% enrichment for the reason discussed for the neutron dose rates.

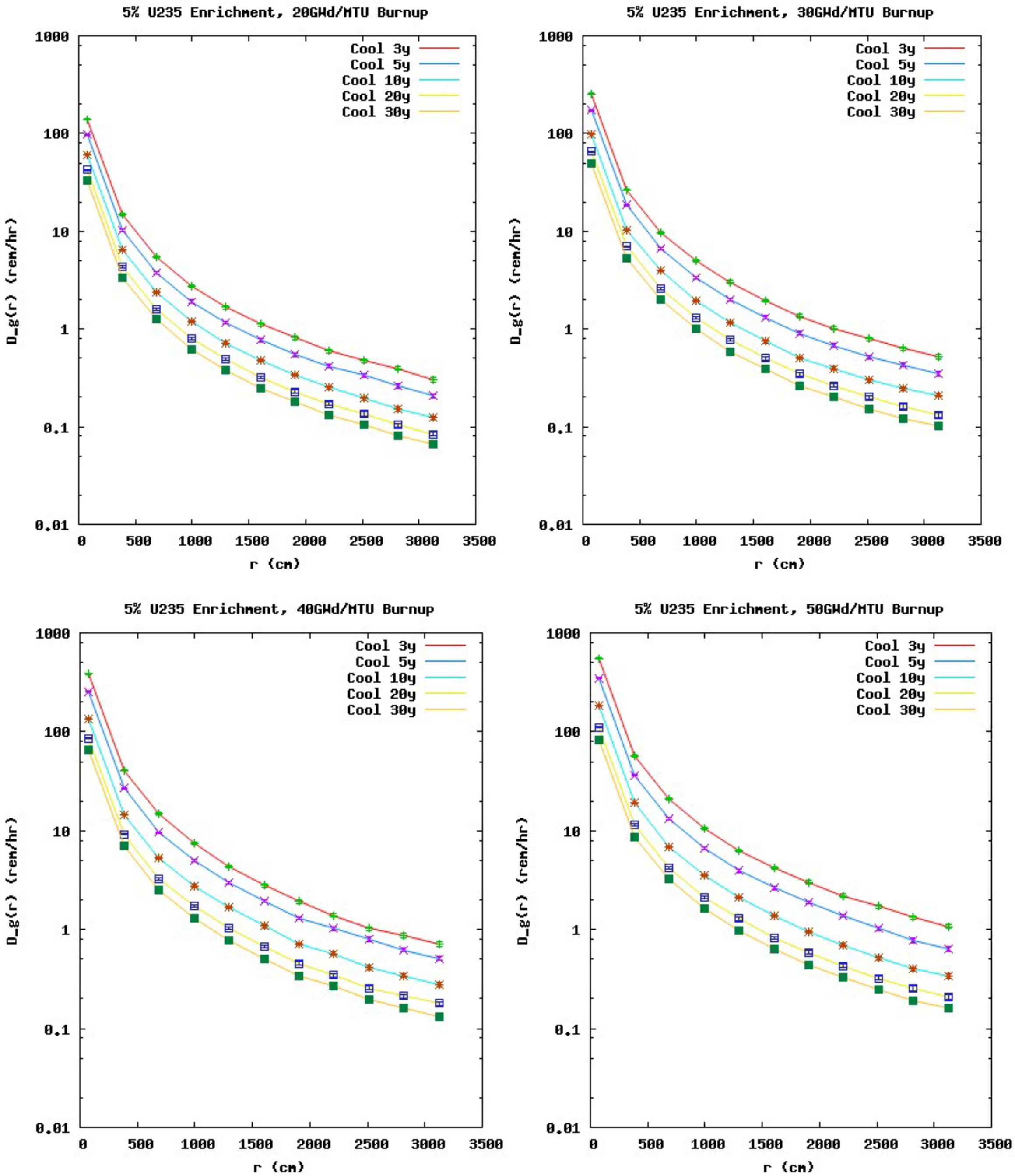

Figure 6. Photon dose rate as a function of distance for 5% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

5 MCNP6 DOSE ESTIMATES FOR THE 0.3-M MODEL

MCNP6 geometry plots for the 0.32-m-diameter model are shown in Figure 7. This diameter was selected (rather than 0.3 m) so that the fuel assembly would just fit inside of the vessel.

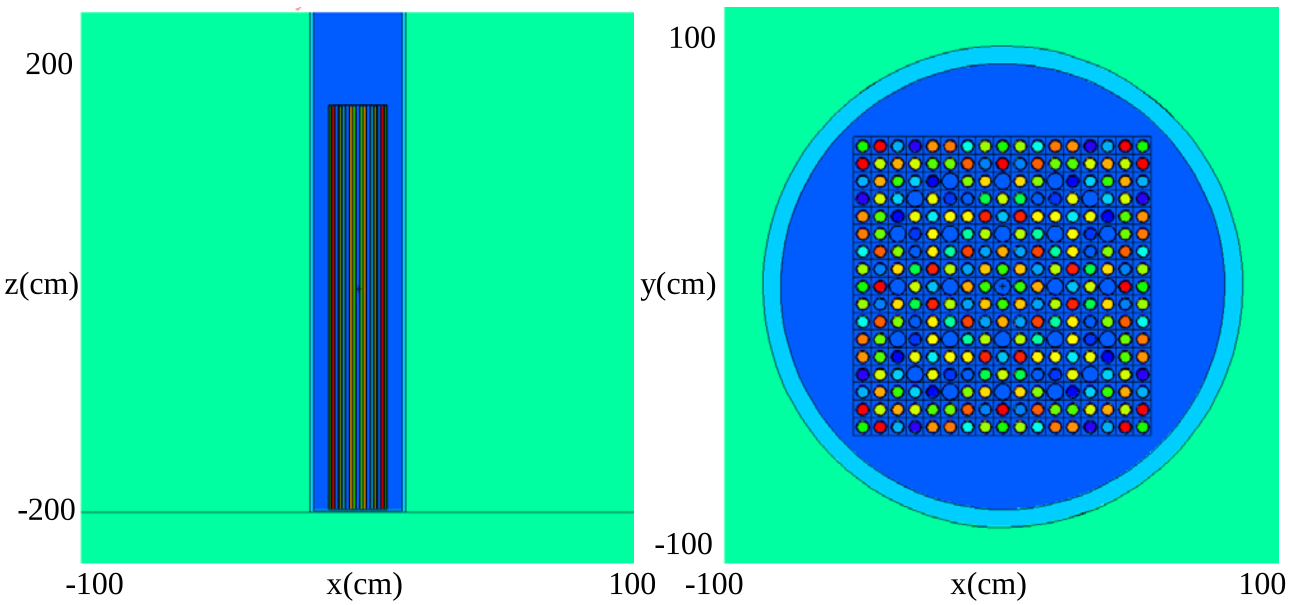

Figure 7. MCNP6 geometry plots of the 0.32-m-diameter chemical processing model showing fuel assembly, nitric acid (blue), and air (green).

Figure 8 contains plots of the neutron dose rate as a function of radial distance for 3% ^{235}U enrichment. Relative uncertainties for the dose rates are < 0.01. These dose rates are approximately 100 times greater than those for the 1.5-m model (Fig. 3). Other behavior resembles that for the 1.5-m model.

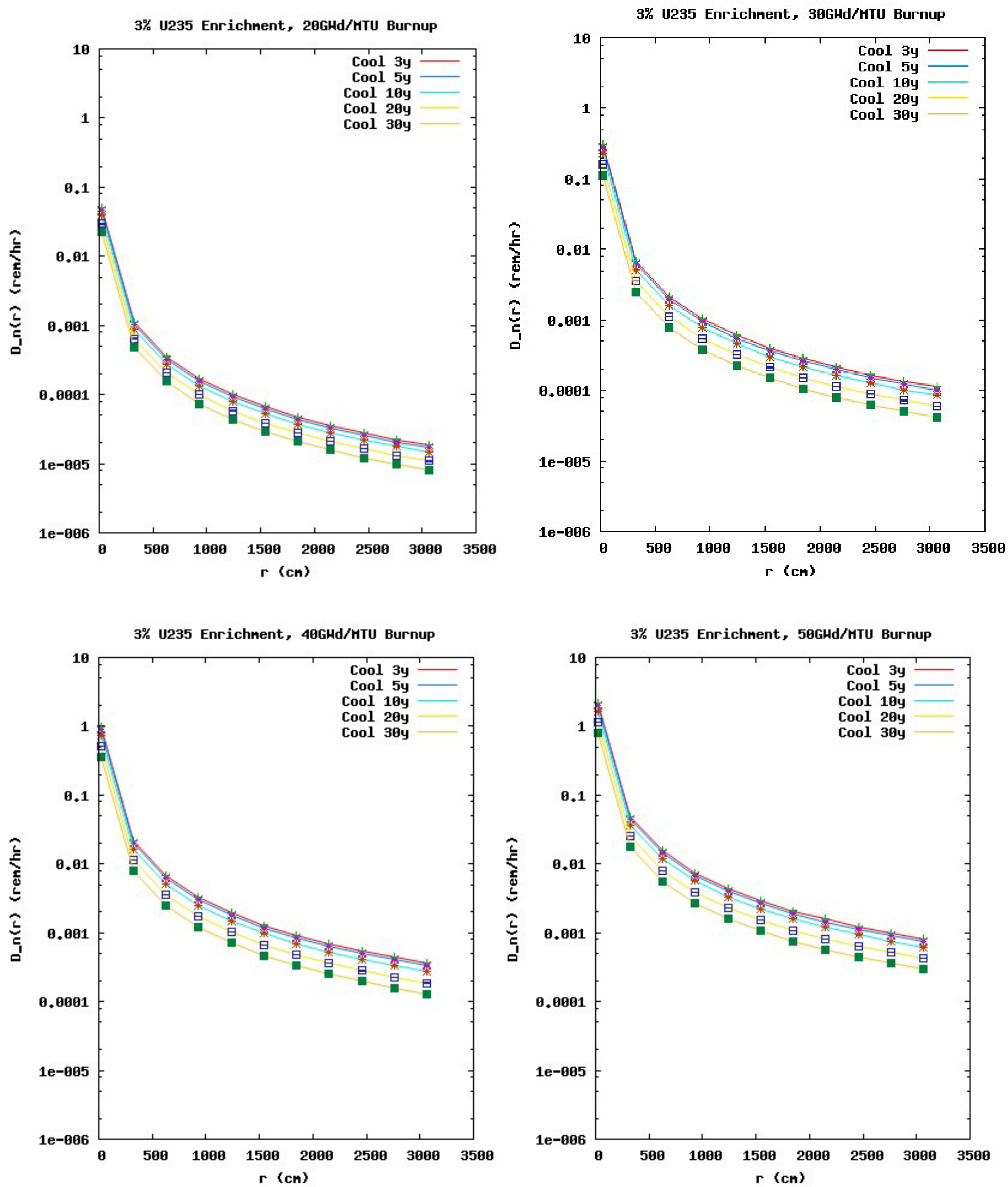

Figure 8. Neutron dose rate as a function of distance for 3% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

Figure 9 contains plots of the neutron dose rate as a function of radial distance for 5% ^{235}U enrichment. These dose rates are approximately 100 times greater than those for the 1.5-m model (Fig. 4). Other behavior resembles that for the 1.5-m model.

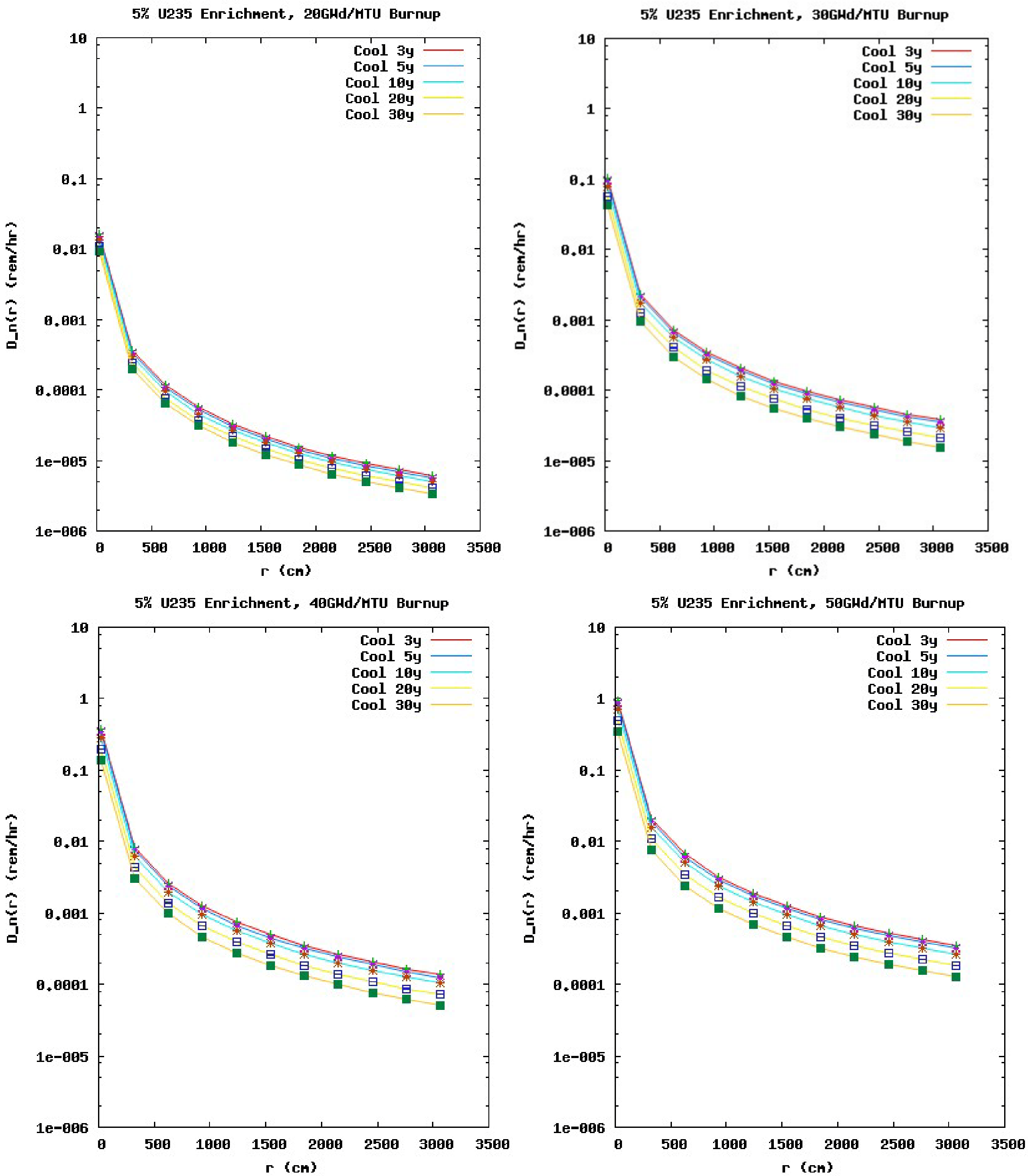

Figure 9. Neutron dose rate as a function of distance for 5% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

Figure 10 contains plots of the photon dose rate as a function of radial distance for 3% ^{235}U enrichment. The relative uncertainties for the energy-integrated dose rates are < 0.01 at the surface of the vessel and 0.02 at 100 feet. These dose rates are approximately 100 times greater than those for the 1.5-m model (Fig. 5). Other behavior resembles that for the 1.5-m model.

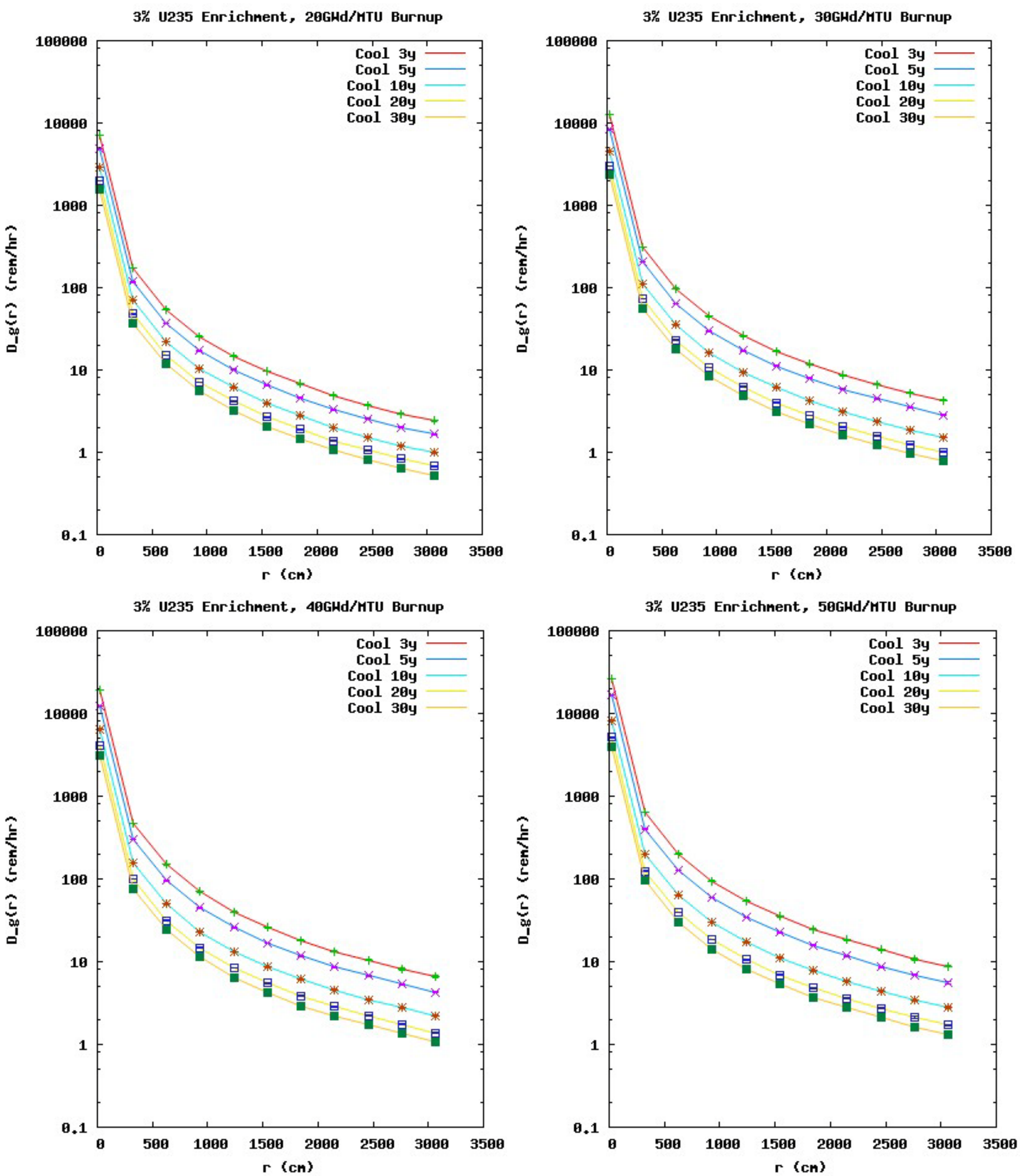

Figure 10. Photon dose rate as a function of distance for 3% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

Figure 11 shows the photon dose rates for 5% enrichment. These dose rates are approximately 100 times greater than those for the 1.5-m model (Fig. 6). Other behavior resembles that for the 1.5-m model.

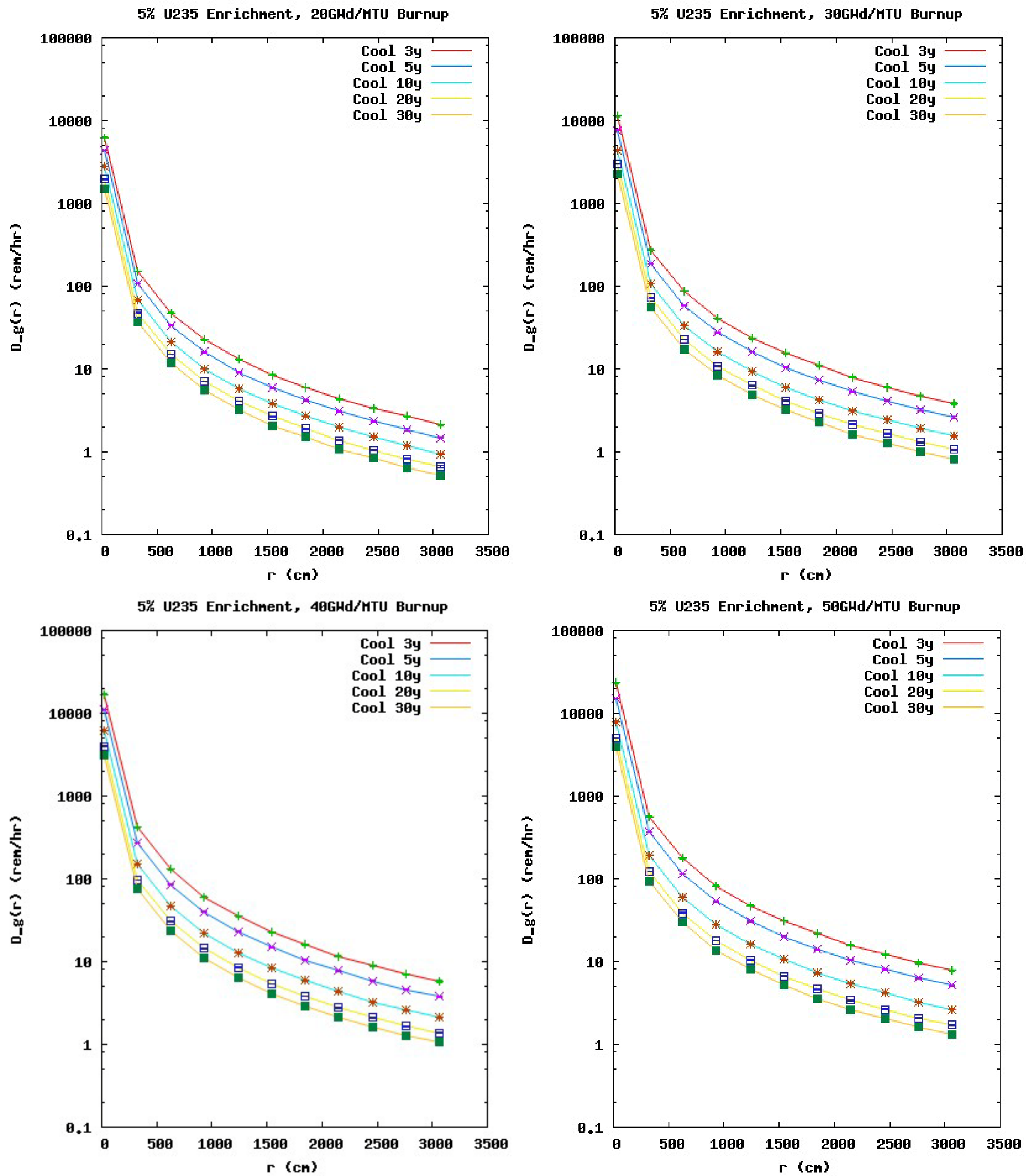

Figure 11. Photon dose rate as a function of distance for 5% ^{235}U enrichment; 20, 30, 40, and 50 GWd/MTU burnup; and 3, 5, 10, 20, and 30 year cooling following irradiation.

6 SUMMARY AND CONCLUSIONS

The MCNP6 Monte Carlo radiation-transport code was used to perform radiation-transport simulations supportive of an NDA study of dose estimates caused by DN and DG emission from an SNF assembly that is placed in a cylindrical steel vessel and immersed in 7M nitric acid. The SNF isotopic inventories and the DP sources were prepared using a generic Westinghouse fuel assembly model. To facilitate parametric analysis, the simulations were executed using three sets of parameters for the SNF: 3% and 5% ^{235}U enrichments; 20-, 30-, 40-, and 50-GWd/MTU burnups; and 3-, 5-, 10-, 20-, and 30-year cooling times. In addition, 2 vessel diameters were treated: 1.5 and 0.32 m. The dose rates were assessed for an intact assembly. No modeling was performed to treat the assembly in a state of dissolution.

The SNF inventories were produced in a related pyroprocessing study using MCNP6 kcode/burnup calculations to give isotopic inventories for the irradiated fuel models. The MCNP6 Tier 3 actinide and nonactinide option was used to provide the most comprehensive inventory available. Those inventories were used to the provide DN and DG sources and tally normalizations in this study.

The MCNP6 radiation-transport was executed using either neutron or photon modes. Coupled neutron-photon execution was not done. Gamma emission caused by neutron-induced fission or activation was therefore not treated, which causes an underestimate of the photon dose rate.

Perl scripts were created and used to automate the creation of input files, execution of calculations, and data mining of output files for presentation of pertinent data in suitable format. The MCNP6 calculations were executed using message passing interface (MPI) on the Pete Linux cluster in a matter of minutes per calculation.

The calculations show dose rate dependence on enrichment, burnup, and cooling time. Neutron dose rates are greatest for the 0.32-m-diameter vessel containing SNF for the case 3% ^{235}U enrichment, 50 GWd/MTU burnup, 3-year cooling: ~2 rem/hr at the surface of the vessel, decreasing to ~10^{-3} rem/hr at 30.48 m from the vessel. The prominent DN emitters ^{242}Cm and ^{244}Cm likely contribute much of the dose rates. Photon dose rates are also greatest for the 0.32-m-diameter vessel containing SNF for the case with 3% ^{235}U enrichment, 50 GWd/MTU burnup, and 3-year cooling: ~26000 rem/hr at the surface of the vessel, decreasing to ~10 rem/hr at 30.48 m from the vessel. Dose rates for the 1.5-m-diameter vessel are approximately 2 orders of magnitude smaller than the smaller vessel. Dose rates decrease with enrichment [reduced fission-product and actinide inventories FPAIs], increase with burnup (increased FPAIs), and decrease with cooling time (radioactive decay of FPAIs).

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