Photon And Neutron Spectra From Spent Fuel Bundles For CANDU 6 Reactors

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ABSTRACT:

This paper describes a methodology for calculating photon (gamma) dose rate spectra from fission-product decay gammas and neutron dose rate spectra generated from a 37-element natural uranium spent-fuel bundle in air using the MCNP-4C and ORIGEN-S codes. The 37-element fuel bundles are used in all CANDU® stations, except Pickering A and B stations, which use 28-element natural uranium fuel bundles.

The gamma dose rate from spent fuel is very large at discharge and decays with time, with the gamma spectra becoming softer as short-lived, high-energy gamma-emitting fission products decay quickly with time. The neutrons are also produced from spontaneous fission and by (α,n) reactions. The neutron dose rate is insignificant with respect to the gamma dose rate, but it decays very slowly with time because of the extremely long half-lives of actinides.

The spectra of gamma and neutron radiation released from the spent fuel bundle are hardened because of the absorption of low-energy gammas in the fuel bundle, whereas the spectra of neutrons released are softened because of scatter events in the fuel bundle. The neutron spectra are much harder and few thermal neutrons are generated if the spent fuel is in air.

Gamma detection instruments such as bundle counters (e.g., one for each spent-fuel port at a CANDU 6 reactor) count the movement of spent fuel bundles between the reactor and the spent-fuel storage bays. The bundle counters are located close to the spent-fuel ports in the spent-fuel discharge room. Using Monte Carlo codes such as MCNP, the method that is used to calculate the dose rates from spent-fuel bundles can be expanded and applied to shielding modifications for the bundle counters, if desired, or it could be applied to designing a different system that detects neutrons emitted from spent fuel in air.

1. INTRODUCTION

This paper describes the calculation of gamma and neutron source, and corresponding contact dose rates and spectra from a 37-element spent fuel bundle in air as a function of decay time. The gamma rays are produced from fission-product decay and the neutrons are produced both by spontaneous fission and by (α,n) reactions. The gamma spectra are "hard" at short decay times, e.g., hours or days, but become softer as short-lived fission products decay with time. The neutron energy spectra mainly consist of fast neutrons (> 1.0 MeV); there are few thermal neutrons, since the spent-fuel is assumed to be located in air.

This methodology can be applied to new (unirradiated) fuel bundles (i.e., to verify the presence of a fresh fuel bundle) for which the gamma dose-rate spectra are much softer due to low-energy gamma rays and X-rays being emitted from heavy elements ^{235}U and ^{238}U in the new fuel bundle. The neutrons from spontaneous fission make up almost all the neutron dose rate from new fuel.

Gamma detection instruments such as bundle counters (one for each spent-fuel port at a CANDU 6® reactor) count the movement of spent fuel between the reactor and the spent-fuel storage bays. The bundle counters are located close to the spent fuel ports in the spent fuel discharge room. Using Monte Carlo codes such as MCNP, the method that is used to calculate the dose rates from the fuel bundles can be expanded and applied to shielding modifications for the bundle counters, if desired, or it can be applied to design a different system that detects neutrons emitted from spent fuel in air. Note that the spent fuel transfers occur in air when the FM ram pushes a pair of spent fuel bundles from the fuelling machine (FM), through the spent fuel port and into the elevator ladle located in the spent fuel room adjacent to the reception bay.

It should be noted that neutron source spectra have no thermal neutrons. Thus, the fast neutrons need to be thermalized by either a D_2O capsule located in front of the neutron detector or a hydrogenous material such as Lucite or polyethylene. There is also a high gamma dose rate, which needs to be suppressed so that the neutron detector can function properly.

2. ORIGEN-S CALCULATIONS

The 37-element fuel bundle contains a centre element, an inner ring (six elements), an intermediate ring (12 elements) and an outer ring (18 elements) as shown in Figure 4. This is the MCNP [1] model of the fuel bundle that is used for calculating gamma and neutron dose rates from spent fuel. The dose point is assumed to be 5.6 cm from the axis of the fuel bundle (i.e., almost on contact).

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Table 1 shows the gamma source associated with a 37-element fuel bundle as a function of energy, 15 minutes after discharge. The gamma sources are calculated using the ORIGEN-S code [2] for an average-power bundle (473 kW) and a high-power bundle (650 kW), both irradiated to the same exit burnup of 7800 MW.d/Mg(U), i.e., 314- and 229-day irradiations respectively, for a CANDU 6 reactor. The gamma source is generally higher with the higher-power bundle, which is to be expected at discharge.

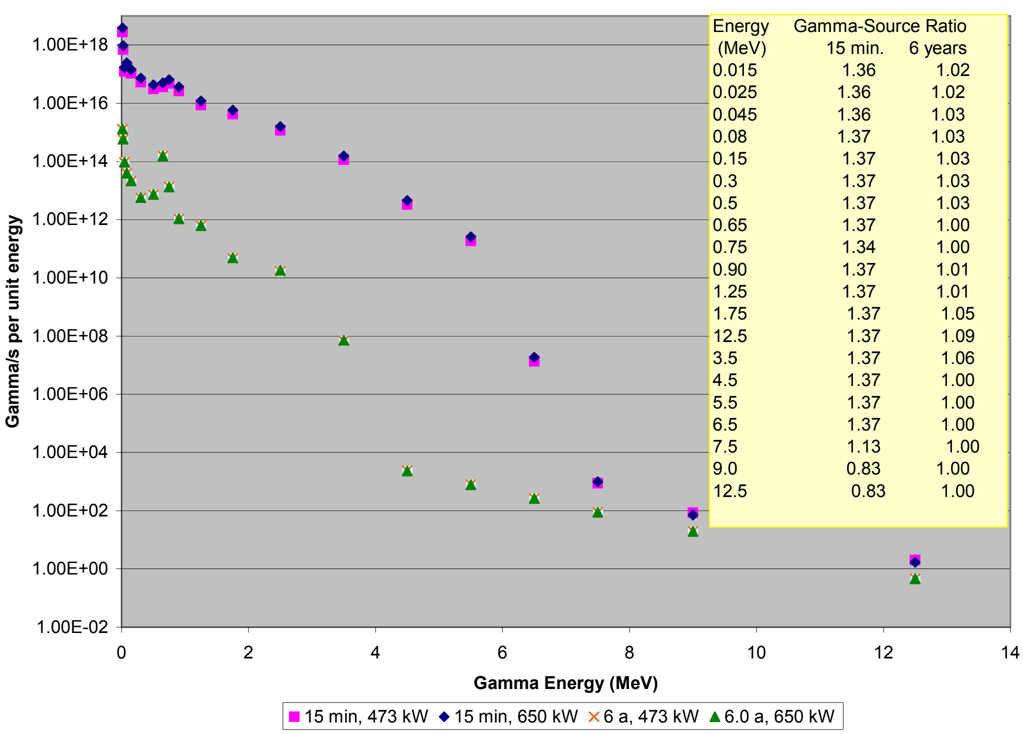
Table 1
Gamma source in spent Fuel after 15-minute decay

Gamma Group	Upper Energy Limit (MeV)	Mean Energy (MeV)	Source (γ/s.bundle) [473 kW bundle]	Source (γ/s.bundle) [650 kW bundle]
1	14	12.5	8.03E+00	6.68E+00
2	10	9	1.69E+02	1.41E+02
3	8	7.5	8.73E+02	9.89E+02
4	7	6.5	1.36E+07	1.86E+07
5	6	5.5	1.87E+11	2.57E+11
6	5	4.5	3.32E+12	4.56E+12
7	4	3.5	1.14E+14	1.56E+14
8	3	2.5	1.16E+15	1.60E+15
9	2	1.75	2.09E+15	2.88E+15
10	1.5	1.25	4.36E+15	5.99E+15
11	1	0.90	5.27E+15	7.24E+15
12	0.8	0.75	4.81E+15	6.46E+15
13	0.7	0.65	3.66E+15	5.01E+15
14	0.6	0.50	6.28E+15	8.58E+15
15	0.4	0.30	1.07E+16	1.46E+16
16	0.2	0.15	1.07E+16	1.46E+16
17	0.1	0.08	7.20E+15	9.86E+15
18	0.06	0.045	3.70E+15	5.02E+15
19	0.03	0.025	6.99E+15	9.51E+15
20	0.02*	0.015	2.81E+16	3.83E+16

* Lower limit is 0.01 MeV.

Gamma sources from spent fuel bundles are proportional to operating power for short decay times, e.g., from a few hours to a few months, and to burnup for longer decay times, e.g., years. This means that, if the burnup were to be the same between two bundles with different operating powers (e.g., 473 and 650 kW), then the gamma source spectra (and thus, the corresponding dose-rate spectra) would be slightly different (a factor of 650/473 = 1.37) for short decay times, and about the same for longer decay times. This is shown in Figure 1.

Figure 1 also includes the ratios of the gamma source spectra as a function of energy between the 650 and 473 kW spent fuel bundles. At 15 minutes after discharge, these ratios are around 1.37 except for Groups 1 and 2 (i.e., > 8 MeV), but noting that the corresponding gamma source is also small (~175 γ/s). This was attributed to the presence of extremely short-lived, but also long-lived fission products, which are affecting the gamma source for the longer irradiation case, i.e., 473 kW bundle. The corresponding ratios are generally around 1.0 for the 6-year decay time. It is realized that the gamma source is a complicated function of operating and decay times.



Energy (MeV)	Gamma-Source Ratio 15 min.	6 years
0.015	1.36	1.02
0.025	1.36	1.02
0.045	1.36	1.03
0.08	1.37	1.03
0.15	1.37	1.03
0.3	1.37	1.03
0.5	1.37	1.03
0.65	1.37	1.00
0.75	1.34	1.00
0.90	1.37	1.01
1.25	1.37	1.01
1.75	1.37	1.05
12.5	1.37	1.09
3.5	1.37	1.06
4.5	1.37	1.00
5.5	1.37	1.00
6.5	1.37	1.00
7.5	1.13	1.00
9.0	0.83	1.00
12.5	0.83	1.00

Figure 1 Gamma-source Spectra after discharge from 473 kW and 650 kW fuel bundles irradiated to the same burnup of 7800 MW.d/MgU.

The corresponding neutron source 15 minutes after discharge for the average power bundle, irradiated for 314 days to a burnup of 7800 MW.d/MgU is 3.0E5 n/s. ORIGEN-S indicates that 85% of the neutrons originate from spontaneous fission and 15% from (α,n) neutrons. The total neutron source was normalized to the U-235 fission spectra to obtain the neutron source spectra in the bundle. The resulting source is shown in Table 2. ORIGEN-S was also run for a high power bundle (650 kW) irradiated to the same burnup. The total neutron source after discharge for this bundle was lower, i.e., 2.5E5 n/s. This result is not unexpected, because of the shorter irradiation time (229 days) the fraction of actinides available at discharge to produce spontaneous fissions, and the inventory of alpha emitters at discharge are reduced with the latter case. The spontaneous fission neutron source was lower by about 20%, while the (α,n) neutrons were lower by about 25%. At longer decay times, e.g., 6 years, the neutron sources become comparable. This behaviour is shown in Figure 2.

Table 2
Neutron source in spent fuel as a function of neutron energy, 15 minutes after discharge (P=473 kW, 650 kW; burn-up=7,800 MW.d/MgU).

Neutron Group	Upper Energy Limit (MeV)	Mean Energy (MeV)	Neutron Source (n/s.bundle)	
			473 kW	650 kW
1	20.0	13.215	7.74E+03	6.40E+03
2	6.43	4.715	5.66E+04	4.68E+04
3	3.00	2.425	6.40E+04	5.29E+04
4	1.85	1.625	4.86E+04	4.02E+04
5	1.40	1.15	5.35E+04	4.42E+04
6	0.90	0.65	4.62E+04	3.82E+04
7	0.40	0.25	2.01E+04	1.66E+04
8	0.10	5.85E-02	4.05E+03	3.35E+03
9	1.70E-02	1.00E-02	1.93E+02	1.60E+02
10	3.0E-03	1.78E-03	2.17E+01	1.79E+01
11	5.50E-04	3.25E-04	1.02E+00	8.40E-01
12*	1.00E-04	6.50E-05	9.30E-02	7.69E-02
		TOTAL	**3.01E+05**	**2.49E+05**

* Lower limit is 3.0E-5 MeV.

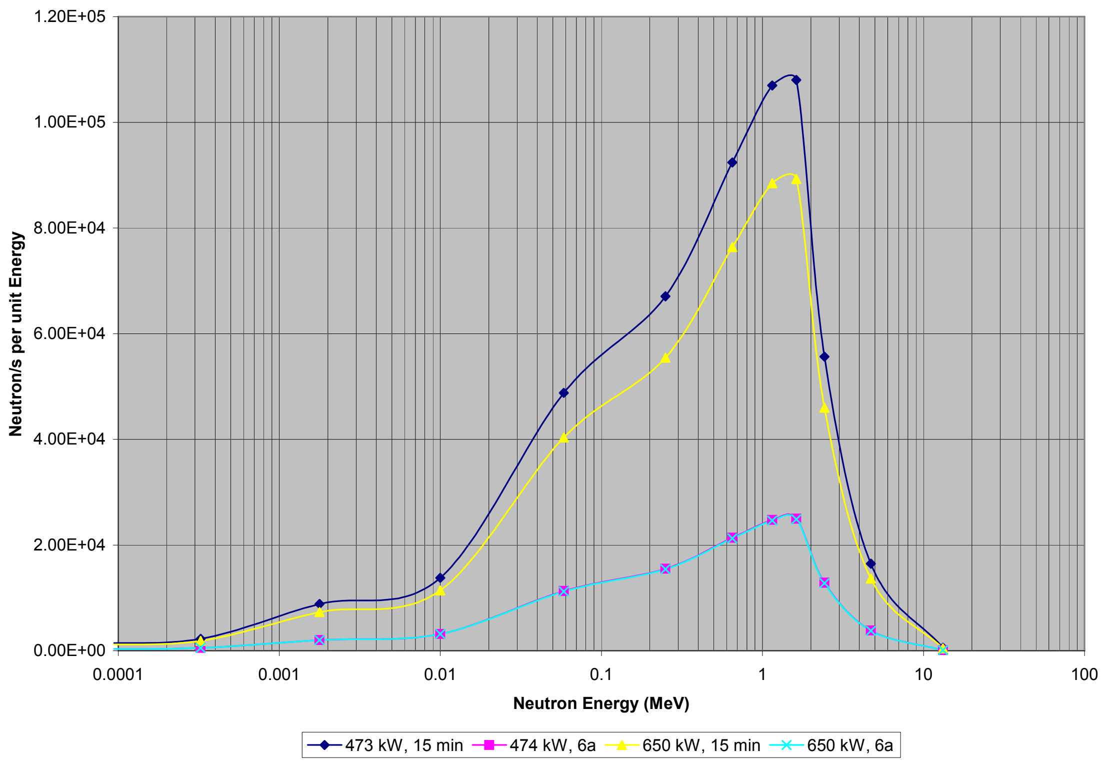


Figure 2 Neutron source spectra in spent fuel, 15 minutes and six years after discharge (P=473 kW & 650 kW, burnup = 7800 MW.d/MgU))

3. MCNP-4C CALCULATIONS

To calculate the gamma and neutron dose rate and their spectra from spent fuel, a series of MCNP-4C calculations were performed. The 37-element fuel bundle is modelled as shown in Figures 3 and 4, in the MCNP-4C calculations.

Spent fuel bundles are transferred in air from the FM magazine (after the FM is attached to the spent fuel port and the D_2O level is reduced below the port level in the FM) onto the elevator ladle in the spent fuel discharge room (see Figure 5). There are water spray nozzles at the elevation of the port; they are used for emergency cooling if the elevator fails. Consequently, the dose rates were calculated for spent fuel being transferred in air.

It should be noted that only a single bundle is modelled. The dose point is assumed to be located at a radius that circumscribes the fuel elements in the outer ring, i.e., at a radial distance of about 5.6 cm from fuel axis. The f2 tally (surface fluence) and f4 tally (average fluence in the cell) with fm cards (tally multiplier cards), and *f8 tally (pulse height tally in MeV) were used to calculate the dose rates. The f2 and *f8 tally results showed the same dose rate while the f4 tally dose rate was slightly higher, but nevertheless indicating consistent results. The f4 tally dose rates were used in this study.

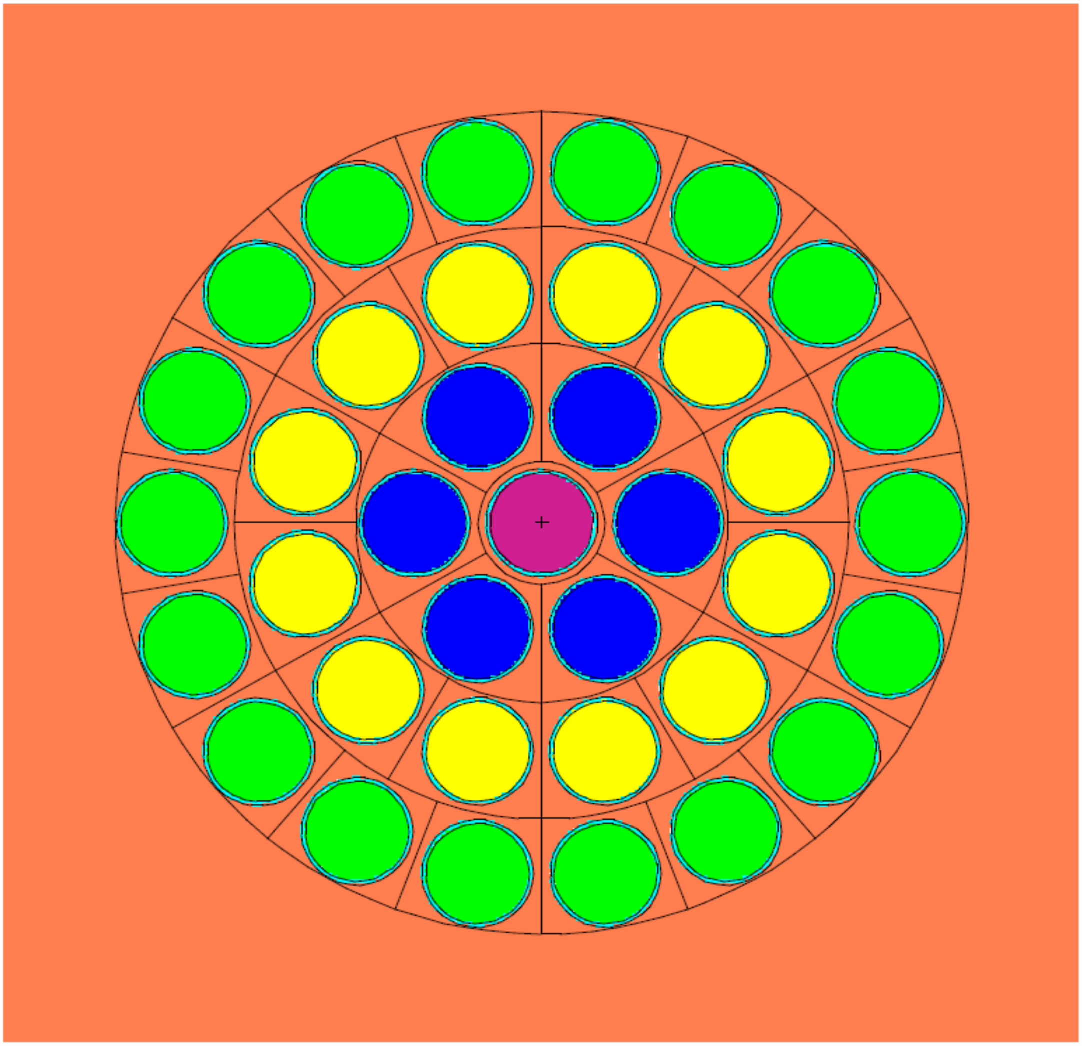


Figure 3 MCNP-4C model of single 37-element fuel bundle in air (x-y plane)

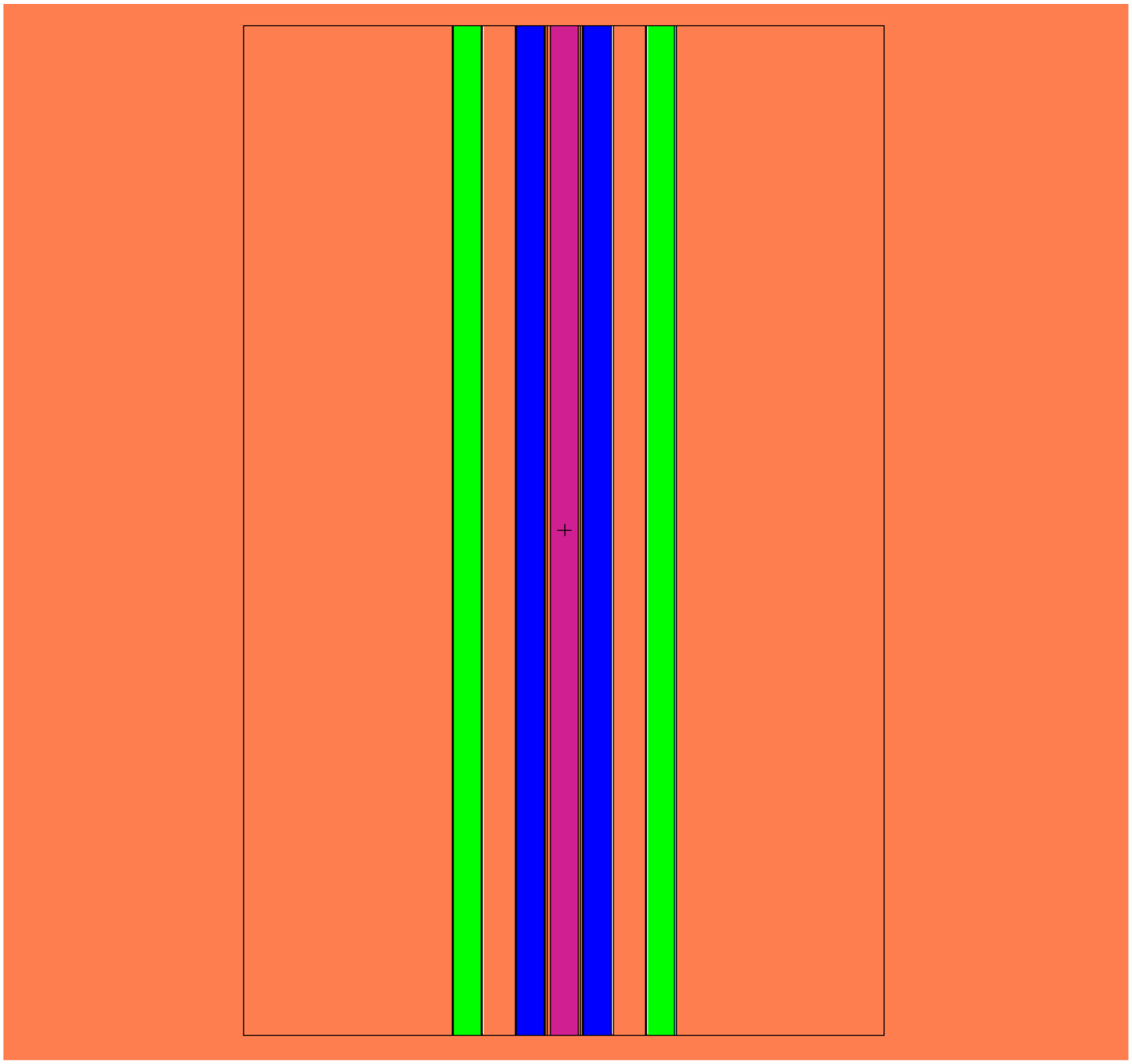


Figure 4 MCNP-4C model of single 37-element fuel bundle in air (x-z plane)

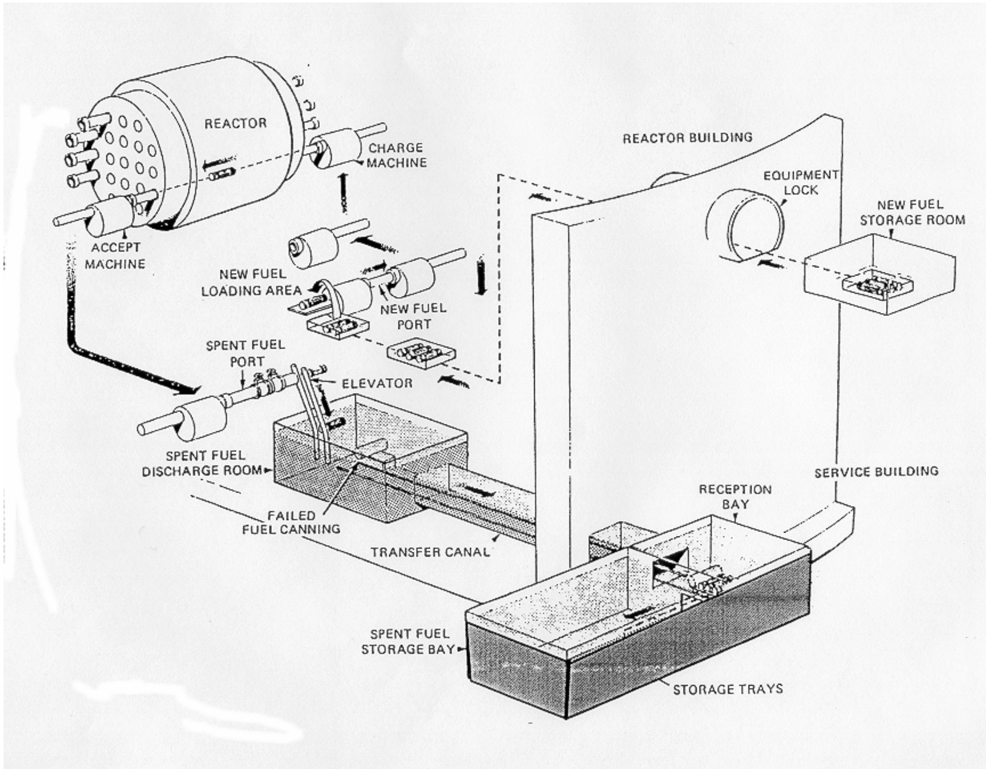


Figure 5 Re-fuelling sequence in CANDU 6 reactors

Twenty MCNP runs were performed for the gamma dose-rate calculations. The source was specified in each energy group, and the resulting dose rates as a function of 20 gamma groups and the total dose rate at that energy E_i were calculated. The total dose rates (based on a fixed source) for each energy group were normalized to the actual gamma spectra from the ORIGEN-S calculations. This resulted in a dose rate that was almost the contact dose rate for the fuel bundle. The contact gamma dose rate was calculated to be about 9.6E4 Sv/h (9.6E6 rem/h), 15 minutes after discharge, for the average-power bundle. (The dose rate was higher, 1.3E5 Sv/h, for the higher-power bundle (650 kW).) The gamma dose rate for each energy group is divided by the width of the group, to calculate the gamma dose rate in mSv/h per unit energy in MeV. This is shown in Figure 6 at 15 minutes, three hours, 24 hours, seven days, three months and six years after discharge for the average-power bundle. The dose rates are associated with a fuel bundle in air.

The corresponding dose-rate spectra for the higher-power bundle irradiated to the same burnup were similar, i.e., dose rates were higher at shorter decay times, but gradually became the same as those of the average-power bundle at longer decay times.

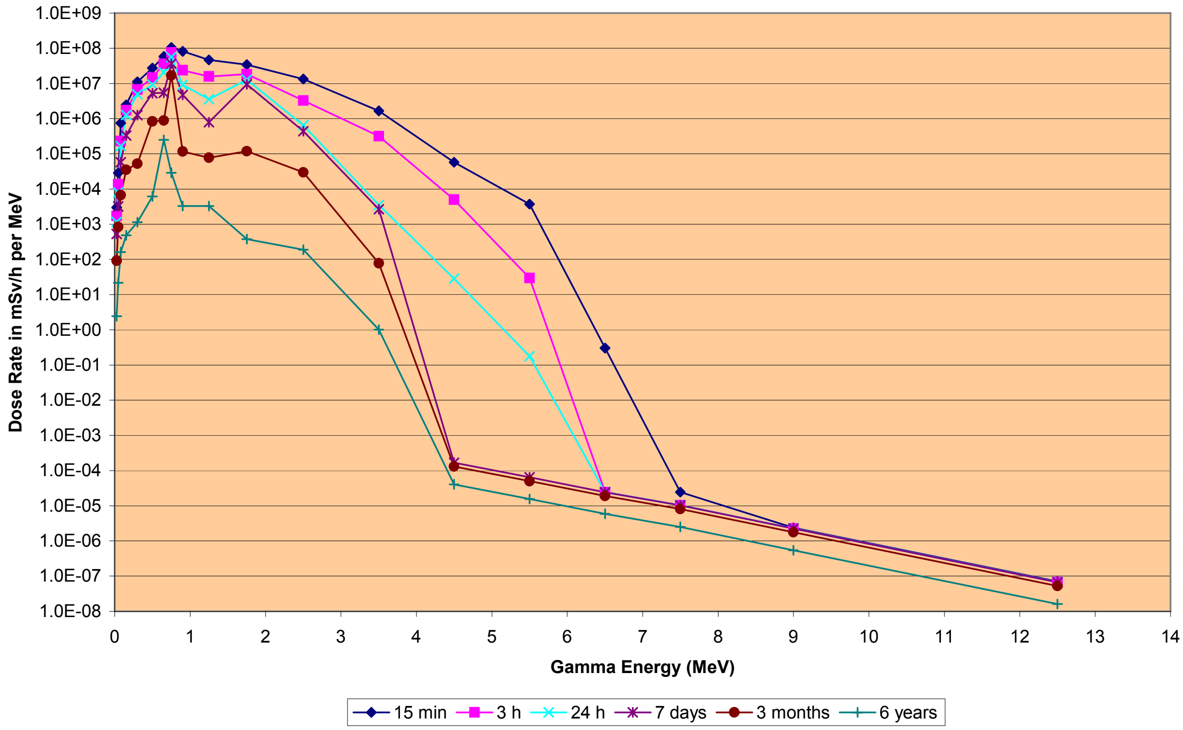


Figure 6 Gamma dose-rate spectra on contact with spent fuel in Air (473 kW bundle, burn-up = 7800 MW.d/MgU)

Nine MCNP-4C runs were performed for source neutrons with the neutron-energy group structure shown in Table 2. The first nine energy groups were considered since there are insignificant amounts of neutrons below 3.0E-3 MeV. Similar to the gamma dose-rate calculations, the neutron source (1 n/s) was put in the first group and the resulting dose rates were summed over all the nine groups to calculate the total dose rate as a result of the neutron source in group 1. These calculations were repeated for the remaining eight groups to calculate the dose rate spectra on contact with the fuel as a function of energy with 1 n/s in each group. These "pseudo" dose rates were normalized to the actual neutron source calculated by the ORIGEN-S code. The dose rate in a given group was divided by the width of that energy group to calculate the neutron spectra as a function of energy in units of mSv/h per unit energy (MeV). The neutron dose rate on contact with the bundle in air was calculated to be 0.35 mSv/h (35 mrem/h). This value was calculated using the f5 tally (point detector tally) and dose-rate conversion factors (dose equivalent per unit fluence) taken from Reference 3. The neutron dose-rate spectra are shown in Figure 7 for various cooling times, i.e., 15 minutes, 14 days, one and six years.

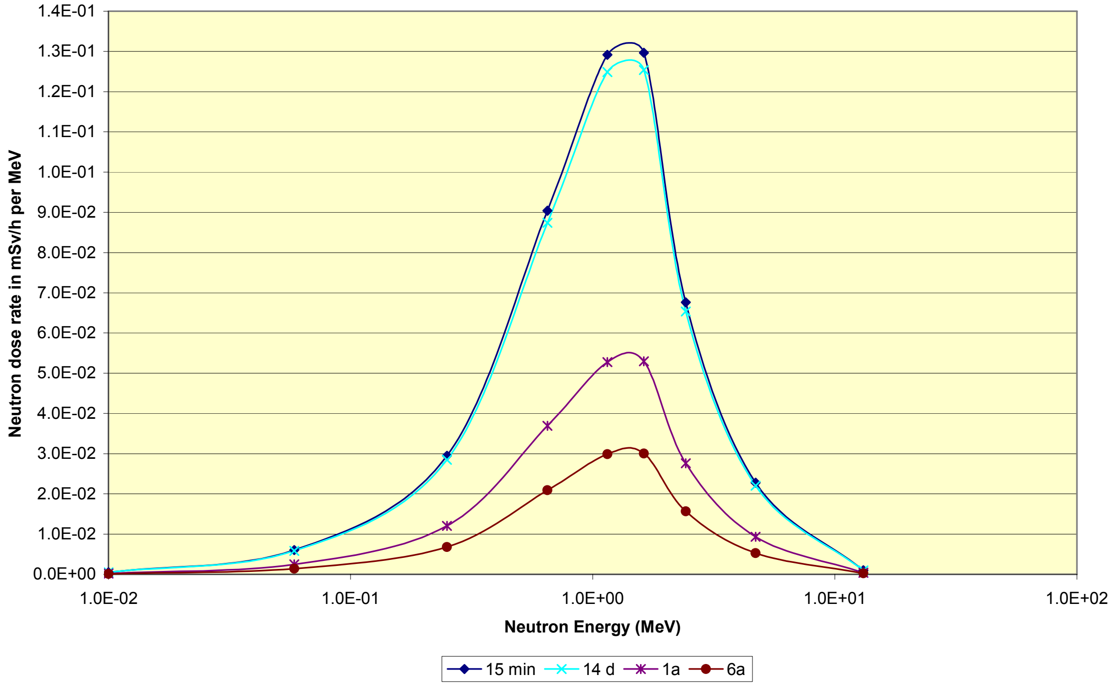


Figure 7 Neutron dose-rate spectra on contact with spent fuel in air (P =473 kW, burn-up = 7,800 MW.d/MgU))

The fission-product decay-gamma dose rate drops by a factor of 3000 within six years, while the neutron dose rate drops by a factor of about 4. The dose rate on contact with a single fuel bundle at discharge is about 9.6E4 Sv/h, while the neutron dose rate on contact is 0.35 mSv/h. Thus, the corresponding dose rates after six years of decay will drop to 32 Sv/h (gamma) and 0.08 mSv/h (neutron). The ratio of gamma to neutron dose rate, which starts at 0.3 billion to one soon after discharge, will be about 0.4 million to one after six years.

The spontaneous fission neutrons are emitted mainly from ^{242}Cm, ^{240}Pu and ^{244}Cm, but also from ^{242}Pu, ^{238}U and ^{238}Pu. The neutrons that are produced by (α,n) reactions are essentially associated with ^{242}Cm, ^{239}Pu and ^{240}Pu, but also with ^{241}Am and ^{244}Cm.

It should be noted that photo-neutrons are generated by high-energy (> 2.23 MeV) fission-product decay gamma interactions with the D_2O inside the FM during re-fuelling operations. However, when the spent fuel is transferred through the spent fuel port in a CANDU 6 reactor, the pair of bundles is transferred in air. Thus the photo-neutron dose rate becomes negligible and the neutron dose rate from the spent fuel is due mainly to spontaneous fission neutrons.

4. FUEL INVENTORIES IN CANDU PLANTS

There are four main inventories of nuclear material in CANDU reactors. These are located in the new-fuel storage room, the reactor core, the irradiated (spent) fuel storage bays and the dry-fuel storage facility.

The new fuel is received and stored in crates in the new-fuel storage area in the service building of a CANDU 6 reactor. When required, the fuel bundles are transferred from the new-fuel storage area through the main airlock to the new-fuel loading area in the reactor building (see Figure 5). The bundles are loaded into the FM magazine through the two new-fuel ports. The two FMs are remotely controlled from the console in the main control room.

5. CONCLUSIONS

This paper presented the gamma and neutron dose-rate spectra for spent fuel from CANDU 6 reactors, calculated using the MCNP-4C and ORIGEN-S codes. Either spectrum can be used to identify and verify the presence of a spent fuel bundle or bundles using appropriate gamma and neutron detection techniques. The neutron spectrum is hard and consists of high-energy neutrons.

6. REFERENCES

1. J.F. Briesmeister, "MCNP – A General Monte Carlo N-Particle Transport Code Version 4C", Los Alamos National Laboratory Report, LA-13709-M, (2000 April).
2. O.W. Herman and R.M. Westfall, "ORIGEN-S: SCALE System Module to Calculate Fuel Depletion, Actinide Transmutation, Fission Product Buildup and Decay, and Associated Radiation Source Terms", NUREG/CR-0200, ORNL/NUREG/CSD-2/V2/R6, Rev. 6, Vol. 2, Section F7, U.S. Nuclear Regulatory Commission, Oak Ridge National Laboratory, (2000 March).
3. National Council on Radiation Protection and Measurements (NCRP), "Protection against Neutron Radiation", NCRP Report 38, 1971.