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PHOTON DOSE RATES FROM SPENT FUEL ASSEMBLIES
WITH RELATION TO SELF-PROTECTION*

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ABSTRACT

Photon dose rates as a function of fission product decay times have been calculated for spent fuel assemblies typical of MTR-type research and test reactors. Based upon these dose rates, the length of time that a spent fuel assembly will be self-protecting (dose rate greater than 100 rem/h at 1 m in air) can be estimated knowing the mass of fuel burned, the fraction of fuel burned, and the fuel assembly specific power density.

The calculated dose rates cover 20 years of fission product decay, spent fuel with up to 80% ^{235}U burnup and assembly power densities ranging from 0.089 to 2.857 MW/kg ^{235}U . Most of the results are unshielded dose rates at 1 m in air with some shielded dose rates at 40 cm in water. Dose rate sensitivity estimates have been evaluated for a variety of MTR fuel assembly designs and for uncertainties in both the physical and analytical models of the fuel assemblies.

INTRODUCTION

The photon dose rate from spent nuclear fuel is a factor, which in combination with the material form and quantity, determines the physical protection requirements^[1] for the spent fuel material. In this paper, the photon dose rate from spent fuel assemblies is calculated for the purpose of estimating the radiation level. The dose rate data are evaluated as functions of specific power density and burnup in the fuel assembly, and as a function of fission product decay time in the spent fuel.

A fuel assembly is considered to be self-protecting when the dose rate is greater than 100 rem/h at a distance of 1 m in air. It is important to know if spent nuclear fuel is self-protecting or when self-protection is lost since significant and costly additional physical protection requirements could be necessary.

Because of the high-radiation fields, it is not always convenient to measure fuel assembly dose rates in an unshielded configuration. As an aid to assess spent fuel radiation levels, dose rates in air have been calculated to cover a broad range of MTR-type fuel assembly designs and burnup histories. Some dose rate calculations also have been made in a shielded configuration. These data could be useful in correlating dose rates in water to corresponding dose rates in air.

FUEL ASSEMBLY MODEL

A functional model of an MTR-type fuel assembly and the locations where dose rates were calculated is shown in Fig. 1. The fuel assembly has aluminum-clad fuel plates and aluminum side plates. The locations are typical of where dose rate measurements could be made relative to the fuel assembly. In the criterion for radiation protection, the location is unspecified other than at 1 m. The dose rate in general will be a function of the fuel assembly orientation.

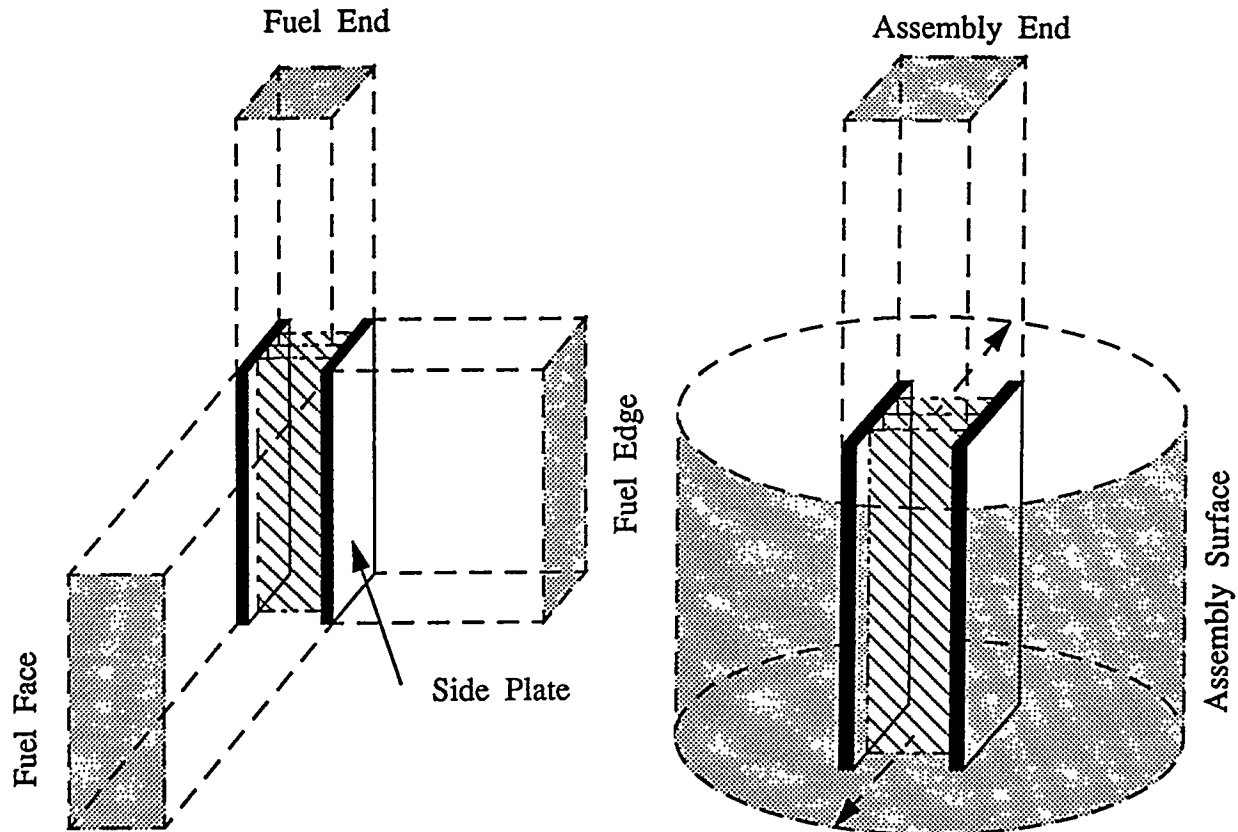


Figure 1. Model of Dose Rate Locations Relative to a Fuel Assembly

The orientation of the fuel assembly in Fig. 1 represent locations towards the fuel-plate face, the fuel-plate edge, and the assembly end - each 1 m from the side plates of the fuel assembly. The cylindrical surface is located 1 m from the axial center line and represents an average dose rate around the fuel assembly. In all cases the dose rates were averaged over the indicated surface areas as defined by the fuel assembly dimensions.

Three MTR fuel assemblies with 16-, 19- and 23-fuel plates were modeled (Fig. 2). The assembly with 16 fuel plates has two additional outside aluminum plates. These three models represent most standard and control, plate-type fuel assemblies. In all cases the fuel assembly was modeled as 60 cm long without end pieces.

CALCULATIONAL MODEL

The fission product photon source was calculated using the isotope generation and depletion code, ORIGEN^[2]. The source was calculated for a ^{235}U mass with up to 80% burnup and for six power densities from 0.089 to 2.857 MW/kg ^{235}U . Burnup in these calculations is equal to the product of the fuel assembly specific power density and the exposure time in days,

times the constant $1.25\text{E-}3 \text{ kg}^{235}\text{U}$ burned per megawatt-day. At each burnup level, the fission product photon source was calculated in yearly increments through 20 years of fission product decay.

These photon source data as functions of ^{235}U burnup, assembly specific power density, and fission product decay time were then introduced into an MCNP^[3] Monte Carlo model to calculate the photon flux in select regions around the fuel assembly. The photon source in all cases was uniformly distributed in the fuel meat of all fuel plates in a fuel assembly. The fuel, clad, side plates, gaps, etc. were all modeled so as to fully account for photon transport in the fuel assembly.

Based upon the calculated photon flux per unit photon source, the dose rates in rem/h were then calculated using the American National Standard fluence-to-dose factors given in Ref. 4.

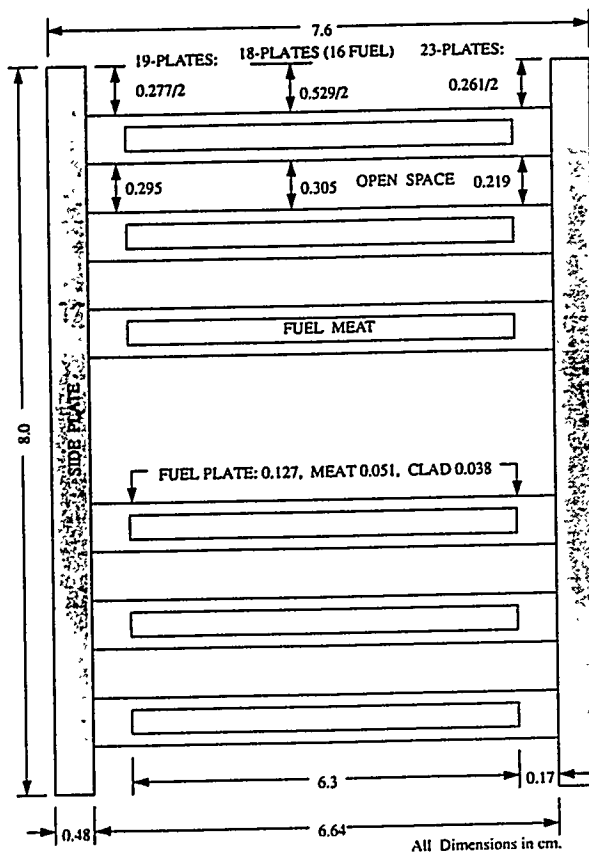


Figure 2. Cross Section of 18-, 19- and 23-Plate Fuel Assemblies

GAMMA DOSE RATE vs DECAY TIME
as functions of U-235 burnup, 60%
and assembly power density, 1.43 MW/kgU-235

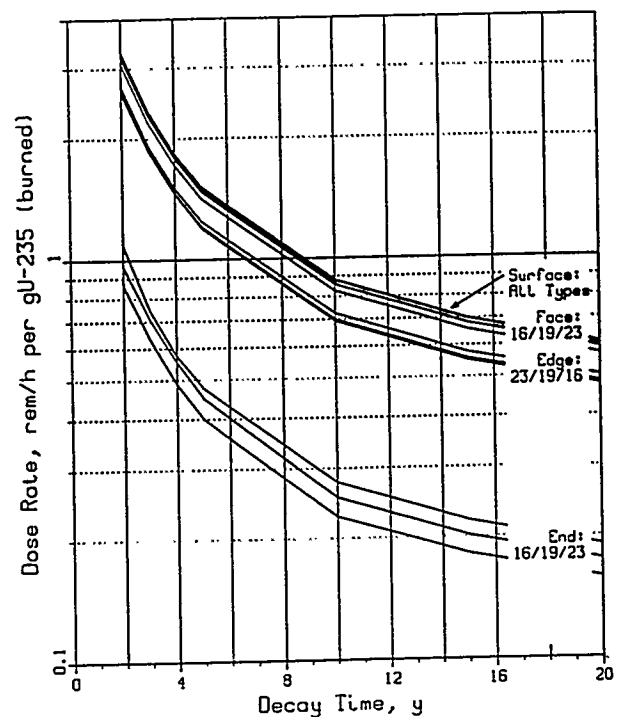


Figure 3. Surface-, Face-, Edge- and End-Location Dose Rates for Three Fuel Assembly Types

DOSE RATE SENSITIVITIES

MTR Fuel Assembly Design Variation

Shown in Fig. 3 are dose rates per g^{235}U burned for the three MTR fuel assembly designs shown in Fig. 2 and for the four "1-meter" regions represented in Fig. 1. Figure 3 shows the variation between the dose rates at 1 m from the face, edge and end of an assembly. It also shows the dose rate variation as a function of the number of fuel plates in an assembly.

Figure 3 also shows that as a function of fission product decay time, the radial surface area dose rate is, within statistics, the same for all fuel assembly designs. Since the same total photon source is assumed in each fuel assembly design, the average dose rate around an assembly is approximately the same. The insensitivity of the dose rate to the fuel assembly design is the basis for presenting all calculated dose rate data as assembly surface area dose rates.

Constant Factors Relating Dose Rate And Photon Source

To a good approximation the calculated surface area dose rate correlates by a constant to the calculated fission product photon source. This simple correlation exists because the major contribution to both the dose rate and the photon source comes from the fission product ^{137m}Ba with minor contributions from its parent ^{137}Cs , and from ^{90}Y and its parent ^{90}Sr . The photon source from these fission product chains includes bremsstrahlung radiation for beta decay in uranium.

Table 1. Ratio of Photon Dose Rate (rem/h) to Photon Source Rate (ph/s)

Assembly Power Density, MW/kg ^{235}U	Average Ratio at 1 m in Air	
	1 to 20% Burnup 2 to 4 years Decay	20 to 80% Burnup 2 to 20 years Decay
2.857	7.95-12 \pm 4.9%	8.75-12 \pm 4.4%
1.429	7.97-12 \pm 4.9%	8.83-12 \pm 3.5% ^a
0.714	8.01-12 \pm 5.1%	8.83-12 \pm 3.7%
0.357	8.04-12 \pm 5.4%	8.86-12 \pm 3.2%
0.179	8.15-12 \pm 5.2%	8.92-12 \pm 2.5%
0.089	8.28-12 \pm 5.2%	8.96-12 \pm 1.9%
Average	8.07-12 \pm 5.1%	8.86-12 \pm 3.3%

^a Average ratio in water at 1 m, 40 cm and 42.5 cm: 4.80-14 \pm 8.0%, 9.80-12 \pm 3.2% and 7.76-12 \pm 3.2%.

Table 1 shows the ratio of the calculated dose rate in rem/h per g ^{235}U burned to the calculated fission product photon source in ph/s per g ^{235}U burned. The ratio is tabulated as a function of the fuel assembly specific power density for two groupings of burnup and decay time. Ratios for water at a power density of 1.429 MW/kg ^{235}U are given in the footnote.

replace the time-consuming task of Monte Carlo dose rate calculations by simple, photon source calculations. The ratios are fairly insensitive to the fuel assembly specific power densities in Table 1. The approximate 4% uncertainty in the average ratio constant will translate into a similar uncertainty in the dose rate.

The use of these dose-to-source ratios can

DOSE RATE RESULTS

Air-Water Dose Rate Correlation

As an alternative to making photon dose rate measurements of spent fuel assemblies in air, a more convenient and natural medium may be a water environment. Water has the advantage that spent fuel assemblies are usually stored in water which provides reasonable shielding for safe dosimetry. Water also provides a natural attenuation for particle decay that can otherwise affect dose rate measurements made in air.

From the data in Table 1, which list the ratio of the surface dose rate to the photon source rate in both air and water, it is possible to establish the location in water where the shielded dose rate will be the same as the unshielded dose rate at 1 m. A linear interpolation of the water ratio data at 40 and 42.5 cm shows that at 41.2 cm, the shielded dose rate in water should be the same as the unshielded dose rate in air.

Self-Protecting Dose Rates

Based upon the calculated dose rates per g^{235}U burned, the total mass of ^{235}U burned per fuel assembly that is required to achieve a dose rate of 100 rem/h can be easily determined. Figure 4 shows the mass of ^{235}U burned that is necessary for a spent fuel assembly to be self-protecting as a function of fission product decay time. These data are for four fuel assembly burnups from 20 to 80%, and for six fuel assembly power densities from 0.089 to 2.857 MW/kg ^{235}U . Interpolation of these data to other fuel assembly burnups and power densities can be easily made.

The data in Fig. 4 show either the minimum mass of ^{235}U burned per fuel assembly for a given number of self-protected years or the maximum number of years spent fuel will be self-protecting for a given mass of ^{235}U burned per fuel assembly. To use these figures, three characteristics of the fuel assembly should be known or estimated: (1) - the mass of ^{235}U burned, (2) - the percentage of ^{235}U burned, and (3) - the time-average specific power density.

For example, in a fuel assembly with 40% burnup that initially contained 280 g ^{235}U , the mass of ^{235}U burned is 112 g. If irradiated at a time-averaged power density of 0.089 MW/kg ^{235}U (0.025 MW), this fuel assembly would be self-protecting for a maximum of 6 years after discharge from the reactor. At 2.857 MW/kg ^{235}U (0.8 MW), the self-protection would increase to approximately 9 years after reactor discharge. To increase the number of self-protecting years at a given power density, the fuel assembly burnup would need to be increased. An increase in burnup to 60% (168 g ^{235}U burned) would increase the minimum number of self-protected years from about 15 years to more than 20 years for the range of power densities from 0.089 to 2.857 MW/kg ^{235}U .

Throughout this paper the fuel assembly power density has been assumed to be the time-average power density that would be equivalent to the actual fuel assembly irradiation history. This assumption may not be accurate, however, for decay times less than about two years. For decay times less than two years, calculations using actual irradiation histories need to be performed since assemblies retain a "memory" of how they were irradiated. For decay times greater than two years, a fuel assembly loses virtually all memory of its irradiation history and the curves shown in Fig. 4 are independent of the fuel assembly irradiation history.

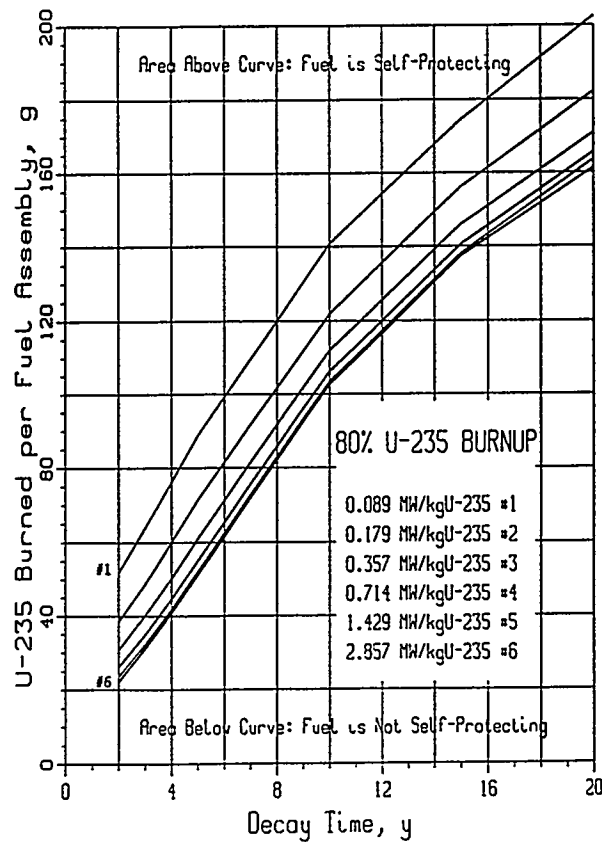
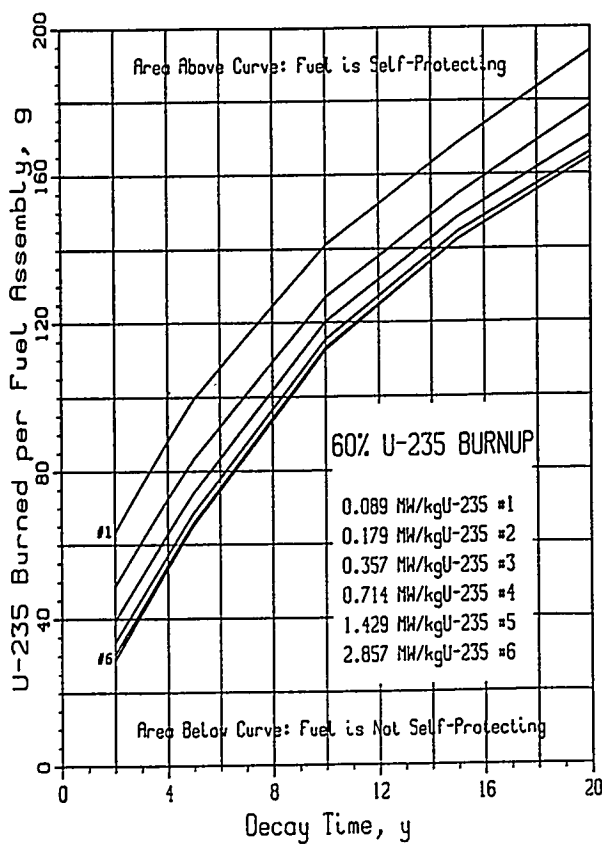
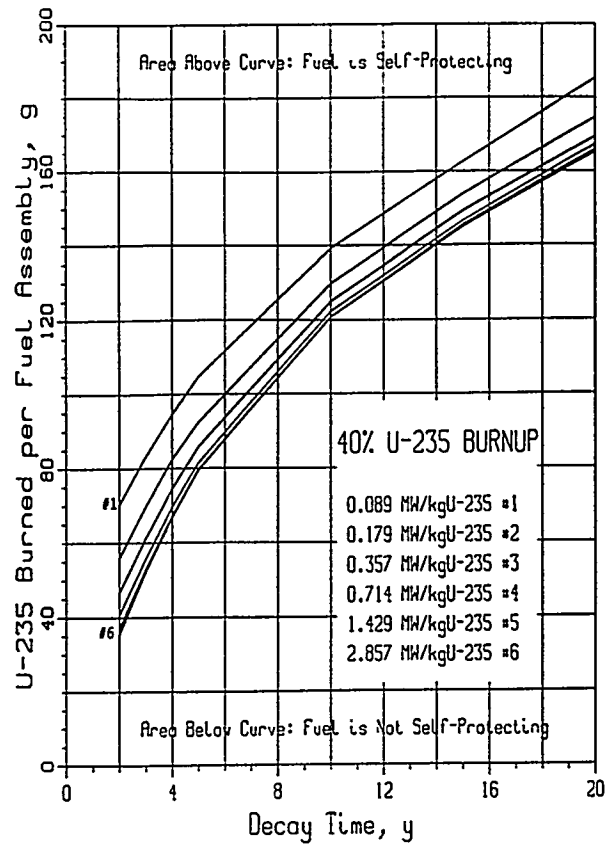
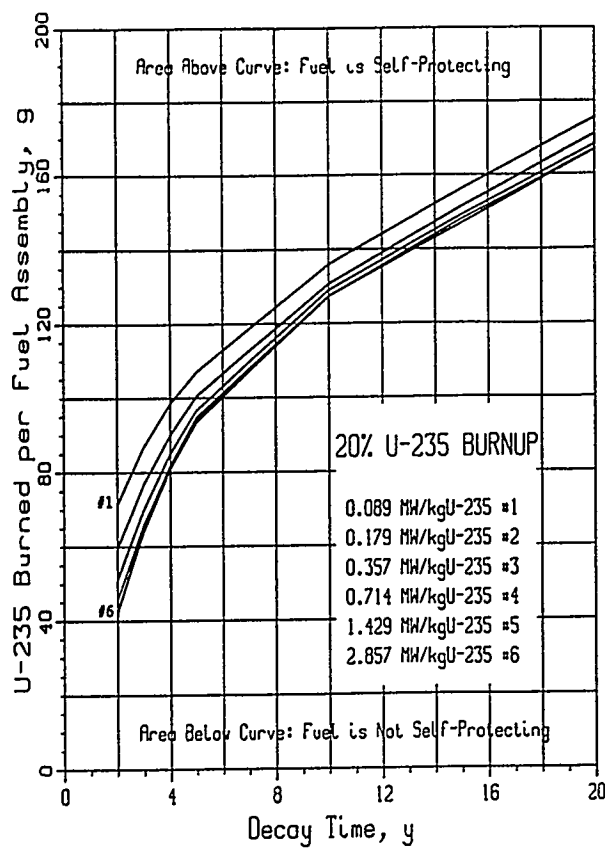


Figure 4. Mass of Burned ^{235}U per Fuel Assembly Necessary for an Unshielded 100 rem/h Dose Rate at 1 m for Fuel Assemblies with 20, 40, 60 and 80% ^{235}U Burnup and Power Densities from 0.089 to 2.857 MW/kg ^{235}U

CONCLUSIONS

Based upon the dose rates calculated in this paper, the length of time that a spent fuel assembly will be self-protecting (i.e., having a dose rate that is greater than 100 rem/h at 1 m in air) can be estimated knowing the mass of ^{235}U burned, the fraction of ^{235}U burned, and the fuel assembly specific power density. These data for a spent fuel assembly are usually known or can be reasonably estimated. The number of years that a spent fuel assembly will be self-protecting can be read directly from figures that show the $g^{235}\text{U}$ burned as a function of fission product decay time.

These dose rates, however, also can be very sensitive to a number of parameters including such items as the fuel assembly orientation and the location relative to the fuel assembly. Estimates of these dose rate sensitivities have been evaluated for a variety of fuel assembly designs, and shielded (water) and unshielded (air) environments. Of much less importance are dose rate contributions from other sources such as heavy metal formation in the spent fuel, and possible material alloys and impurities in the structure of the fuel assembly. Fission products account for nearly all the dose rate in the spent fuel. These dose rate variations are discussed in the appendix.

A simplification of dose rate calculations can be made using calculated proportionality factors that relate the spent fuel assembly dose rate to the fission product photon source. This approximation can reduce the dose rate calculation to a photon source calculation times a constant, with a small uncertainty in the result.

REFERENCES

1. "The Physical Protection of Nuclear Material," INFCIRC/225/Rev.3, International Atomic Energy Agency, Vienna, Austria (1993).
2. M. J. Bell, "ORIGEN - The ORNL Isotope Generation and Depletion Code," ORNL-4628, Oak Ridge National Laboratory, Oak Ridge, TN (1973).
3. J. F. Briesmeister, Ed., "MCNP - A General Monte Carlo N-Particle Transport Code, Version 4A," LA-12625-M, Los Alamos National Laboratory, Los Alamos, NM (1993).
4. "American National Standard for Neutron and Gamma-Ray Fluence-to-Dose Factors," ANSI/ANS-6.1.1-1991, American Nuclear Society, La Grange Park, IL (1992).

APPENDIX

This appendix contains dose rate sensitivity data for various parameters and assumptions inherent in the fuel assembly models. These data also may be used to adjust calculated surface dose rates to compare with measured dose rates that are functions of a fuel assembly design and the fuel assembly orientation.

Face-, Edge- And End-Dose Rate Ratios

Table A1 shows the face-, edge- and end-location dose rate ratios relative to the surface area dose rate for three fuel assembly designs. These data may be used to estimate the unshielded, 1-m dose rates at these locations based upon the calculated surface area dose rate.

Table A1. Face-, Edge- and End-Location Dose Rate Ratios

Assembly Type	Dose Rate Ratio ^a with respect to the Surface Dose Rate at 1 m in Air		
	Face	Edge	End
16-Plates	$0.972 \pm 2\%$	$0.790 \pm 2\%$	$0.318 \pm 7\%$
19-Plates	$0.976 \pm 2\%$	$0.793 \pm 2\%$	$0.292 \pm 7\%$
23-Plates	$0.940 \pm 2\%$	$0.824 \pm 2\%$	$0.263 \pm 7\%$

^a For all fuel assembly types at 40 cm in water, the face-, edge- and end-dose rate ratios with respect to the surface dose rate are $0.693 \pm 2\%$, $0.686 \pm 2\%$ and $0.255 \pm 7\%$, respectively.

Radial Dose Rate Variation

The variation of the surface area dose rate as a function of the distance from the center of the fuel assembly is shown in Table A2. These radial dose rate gradients were calculated for both unshielded and shielded fuel assembly configurations.

Table A2. Radial Dose Rate Gradient

Medium	Percent Change per cm Relative to the Surface Dose Rate ^a
Air (1 m)	$-1.9 \pm 1.0\%$
Water (40 cm)	$-9.8 \pm 0.5\%$

Axial Dose Rate Variation

Table A3 shows the axial dose rate relative to the average surface dose rate, over five axial sections of the 60-cm active length of a fuel assembly. These results show that the axial dose rate distribution at 1-m radius is fairly flat with only about $\pm 2\%$ variation. At 40-cm radius in water, the central section (-5 to +5 cm) dose rate is about 16% larger than the average dose rate, with similarly large (+10% and -12%) differences in the two other axial sections.

^a Applicable over a radial distance of ± 5 cm relative to a radius of 1 m in air and 40 cm in water.

Table A3. Axial Dose Rate Distribution

Axial Section Medium/	Percent Difference Relative to the Surface Dose Rate		
	-5 to +5 cm	+5 to +15 cm -5 to -15 cm	+15 to +30 cm -15 to -30 cm
Air (1 m)	2.9 ± 0.3	1.8 ± 0.2	-2.2 ± 0.2
Water (40 cm)	15.8 ± 0.3	10.2 ± 0.3	-12.0 ± 0.2

Aluminum Alloy Materials and Impurities

A number of aluminum alloys containing different alloying materials and impurities are used to manufacture MTR-type fuel assemblies. Table A4 lists a representative sample of the material alloys and impurities commonly found in Al-6061 which is used here as an example. A typical fuel assembly contains about 3 kg of aluminum alloys in the side plates and cladding of the active portion of the fuel plates.

Table A4. Al-6061 Material Alloys and Impurities

Mat'l	ppm	Mat'l	ppm	Mat'l	ppm	Mat'l	ppm
Li	10	Ti	300	Co	10	Ga	100
B	10	Cr	1700	Ni	500	Zr	10
Mg	8800	Mn	300	Cu	2600	Cd	10
Si	7000	Fe	2400	Zn	500	Sn	500

Results of dose rate calculations for Al-6061 shows that the major dose rate contributor is ^{60}Co with minor contributions from ^{65}Zn and ^{54}Mn . The latter materials have half-lives of less than a year which leaves ^{60}Co , with a 5.3 y half-life, the only major component. Relative to the dose rate of fission products per gram of ^{235}U burned, the dose rate of cobalt per kilogram of Al-6061 is an order of magnitude smaller.

Heavy Metals

Dose rate calculations were made for the heavy metals generated in the burning of 93% enriched uranium fuel. These data showed that the contribution of heavy metals to the dose rate per gram of ^{235}U burned was five orders of magnitude smaller than the contribution of fission products. Similar dose rate calculations for LEU fuel showed that the heavy metal contribution was larger, but still three orders of magnitude smaller than the contribution of fission products.