Estimates of Dose Rates from the Plutonium-bearing Fuel Pellets Fabricated at Carborundum

White Paper

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1.0 INTRODUCTION

The Carborundum Company in Niagara Falls, NY, performed reactor fuel development work for the Atomic Energy Commission (AEC). From 1961 through 1967, Carborundum operated a facility for the study and fabrication of plutonium refractory materials for an AEC fuel development program (NIOSH 2015).

This report provides estimates of dose rates from the plutonium-bearing fuel pellets fabricated at the facility. The support files used in this analysis are available in ORAUT (2018).

2.0 GEOMETRY AND MATERIALS

2.1 Pellet Geometry

The radiation source for these dose calculations is a small pellet of Pu-U carbide, having a density of 12.8 g/cm^3 (Strasser and Stahl 1965). The carbide fuel pellets produced by Carborundum had a range of densities. This density (12.8 g/cm^3) was mentioned in the text of the final report as starting material for experiments related to out-of-pile properties of mixed uranium-plutonium carbides (Stahl and Strasser 1963, PDF p. 14). The density chosen was identified as the density of fuel that had been sintered without sintering aids. This density was chosen to maximize the amount of fuel in a single fuel pellet. Fuel that had been sintered with a sintering aid (0.1 w/o Ni) had a higher density of 13.1 g/cm^3 (Stahl and Strasser 1963, PDF p. 14). Doses estimates were made for a single pellet that is a cylinder 0.2 inches in diameter and 0.2 inches tall (Strasser and Stahl 1965). The pellets were assumed to be unclad.

2.2 Glovebox Geometry

The pellets were assumed to be housed in a glovebox three ft high, five ft long, and 3.5 ft deep (Saulino et al. 1962). The side walls, roof, and floor of the glovebox were ¼-in. welded aluminum plate. The front and back walls were ¼-in. safety glass. The safety glass consisted of two ⅛-in. layers of plate glass and a 15-mil inner layer of polyvinyl butyral that inhibits shattering of the glass.

In addition to the glovebox, the model also contained a concrete back wall, a ceiling, and a floor to generate albedo. A 1/16-in. thick steel panel was implemented on the ceiling and walls of the room.

3.0 RADIATION SOURCE TERMS

3.1 Isotopic Compositions of U and Pu

The plutonium was assumed to be weapons grade plutonium having the weight fractions shown in Table 1 (Strasser and Taylor 1962). The uranium was enriched uranium as shown in Table 2. Three isotopic compositions for plutonium were used during the course of the experiments at Carborundum. The isotopic composition shown in Table 1 is that of plutonium that was provided by Hanford (Strasser and Taylor 1962, PDF p. 15). The isotopic composition was chosen because this material had the highest weight fraction of ^{241}Pu. Plutonium-241 decays to ^{241}Am, which drives the photon dose because it emits a 60 keV photon that penetrates shielding more readily than the lower energy photons emitted by plutonium isotopes.

The isotopic composition of the uranium used for the carbide fuel pellets was not explicitly stated in the reports. An enrichment of 24% was chosen to be consistent with peer review calculations performed by SC&A (2016) and consistent with material known to have been used at Carborundum (Strasser and Taylor 1962). The calculated dose rate from the fuel pellets is not strongly influenced by the isotopic composition of the uranium used for the fuel pellets.

Table 1. Isotopic Analysis of Pu.

Nuclide	Mass Fraction
^{238}Pu	0.00
^{239}Pu	0.907
^{240}Pu	0.079
^{241}Pu	0.012
^{242}Pu	0.001

Table 2. Isotopic Analysis of U.

Nuclide	Mass Fraction
^{234}U	0.00132
^{235}U	0.24
^{238}U	0.75868

3.2 Photon Source Calculations

The photon component of the dose rates from $(U_{0.8}\ Pu_{0.2})C$ is due mainly to the radioactive progeny of the nuclides listed in Table 1 and Table 2. The activities of the progeny radionuclides at five years after separation were calculated using the Radiological Toolbox 3.0.0 (Eckerman and Sjoreen 2013). The photon intensities and energies emitted by the radionuclides were obtained from Be et al. (2004) and the International Commission on Radiological Protection (ICRP 2008).

3.3 Neutron Source Calculations

The neutron source term was calculated using the SOURCES 4C computer code (Wilson et al. 2002). The volumetric source considered for these calculations considered included neutrons emitted from within the volume of the source region by spontaneous fission and by alpha-neutron (alpha,n) interactions.

Input to SOURCES 4C included the desired energy structure of the neutron spectrum, the number density of the actinide isotopes, and the fraction of the (alpha,n) target nuclides to the total number density. Neutron spectra were calculated in 760 groups with a minimum energy of 0 MeV and a maximum energy of 4.49 MeV. SOURCES 4C reports that ^{240}Pu produces about 73% of all spontaneous fission and (alpha,n) neutrons that form the source term.

Carbon-13 was the only target isotope considered for calculations of (alpha,n) neutrons because the atom fraction of ^{13}C exceeded the atom fractions of other impurity isotopes by about a factor of 100.

4.0 DOSE EQUIVALENT CALCULATIONS

4.1 Fluence to Dose Conversion Coefficients

MCNP6™ (Pelowitz 2013) was used to calculate the fluence rates of neutrons and photons that entered volume elements that represented dosimeters. Two dosimeters were implemented in the MCNP6 input file; one at 30.48 cm from the center of the fuel pellet and one at 100 cm from the center of the fuel pellet. Conversion coefficients were used to translate the fluence rates to dose equivalent rates. The conversion coefficients were included in the MCNP6 input file and the conversion from fluence to dose equivalent was performed by MCNP6.

The neutron conversion coefficients used to generate the data in Table 3 can be found in Table A.42 of ICRP Publication 74 (ICRP 1996). The photon data in Tables 4 and 5 were derived using conversion coefficients found in Tables A.1, A.21, and A.24 of ICRP (1996).

4.2 Tallies

The neutron and photon doses were accumulated into energy bins as described by the Office of Compensation and Analysis (NIOSH 2007). Photon exposures were divided into three energy bins (< 30 keV, 30-250 keV, and >250 keV). Neutrons were divided into five energy bins (< 10 keV, 10-100 keV, 100-2000 keV, 2-20 MeV, and >20 MeV). For both neutrons and photons, the upper energy bins were divided into two bins to help elucidate the energy spectra of the radiations. In particular, it was desired to determine if the tally energies exceeded the energy of the dose conversion factors.

The tallies were accumulated into two air volumes that represented dosimeters. The air volumes were 5.08 cm × 5.08 cm × 0.2 cm, and the centers were located 30.48 cm (1 ft) and 100 cm from the center of the fuel pellet. The 30.48 cm location approximates the abdomen of the Nuclear Technology Services, Inc. (NTS) Bottle Mannequin Absorber (BOMAB) phantom described in the glovebox Technical Information Bulletin (NIOSH, 2011a). The dosimeter air volumes are approximately the size of a dosimeter.

5.0 MCNP6 RESULTS

Dose equivalent rate information for a single fuel pellet are provided in Tables 3–5. The photon dose rates exceed the neutron dose rates. A plot of the photon spectrum at the 30.48 cm dosimeter location, not shown here, indicates that the major contributor to the photon dose rate is the 59.5 keV photon from ^{241}Am. The values in Tables 3–5 below are based on a single pellet. Section 6.0 provides a dose model based on the amount of material in process and typical working conditions.

Table 3. Neutron Dose Component of Neutron Source Term Calculation.

Neutron energy (MeV)	Hp(10) (mRem.h^{-1})	Error	H*(10) (mRem.h^{-1})	Error
0–0.01[a]	8.7147E-08	0.0099	7.8582E-08	0.0099
0.01–0.1[a]	1.3035E-06	0.0042	1.3058E-06	0.0042
0.1–2.0[a]	3.3814E-04	0.0006	3.2707E-04	0.0006
2.0–20.0[a]	4.8368E-04	0.0005	4.6333E-04	0.0005
Total[a]	8.2322E-04	0.0004	7.9179E-04	0.0004
0–0.01[b]	6.8832E-08	0.0116	6.2225E-08	0.0116
0.01–0.1[b]	2.7467E-07	0.0107	2.7460E-07	0.0107
0.1–2.0[b]	4.3184E-05	0.0017	4.1759E-05	0.0017
2.0–20.0[b]	5.8541E-05	0.0014	5.6074E-05	0.0013
Total[b]	1.0207E-04	0.0011	9.8169E-05	0.0011

a. Dosimeter 30.48 cm (1 ft) from center of fuel pellet.
b. Dosimeter 100 cm from center of fuel pellet.

Table 4. Photon Dose Component of Neutron Source Term Calculation.

Photon Energy (MeV)	Hp(10) (mRem.h^{-1})	Error	H*(10) (mRem.h^{-1})	Error	Air Kerma (mRad.h^{-1})	Error
0.0–0.03[a]	1.7751E-10	0.0469	1.7435E-10	0.1456	2.2925E-10	0.0577
0.03–0.25[a]	1.2270E-08	0.0088	1.1433E-08	0.0088	7.8356E-09	0.009
0.25–10.0[a]	5.9759E-07	0.0024	5.9558E-07	0.0095	5.1669E-07	0.0024
Total[a]	6.1004E-07	0.0023	6.0718E-07	0.0091	5.2476E-07	0.0024
0.0–0.03[b]	1.3352E-11	0.1445	1.3033E-11	0.1456	1.7789E-11	0.1917
0.03–0.25[b]	2.9254E-09	0.0180	2.7187E-09	0.0180	1.8412E-09	0.0183
0.25–10.0[b]	4.7926E-08	0.0095	4.7601E-08	0.0095	4.1110E-08	0.0097
Total[b]	5.0865E-08	0.0023	5.0333E-08	0.0091	4.2969E-08	0.0093

a. Dosimeter 30.48 cm (1 ft) from center of fuel pellet.
b. Dosimeter 100 cm from center of fuel pellet.

Table 5. Photon Source Term Dose Calculation.

Photon Energy (MeV)	Hp(10) (mRem.h^{-1})	H*(10) (mRem.h^{-1})	Air Kerma (mRad.h^{-1})
0.0–0.03[a]	5.0170E-05	4.9554E-05	6.9764E-05
0.03–0.25[a]	2.3594E-02	2.1704E-02	1.2798E-02
0.25–10.0[a]	2.4844E-03	2.3750E-03	1.8683E-03
10.0–100[a]	0.0000E+00	0.0000E+00	0.0000E+00
Total[a]	2.6129E-02	2.4129E-02	1.4736E-02
0.0–0.03[b]	5.7146E-06	5.6488E-06	6.5398E-06
0.03–0.25[b]	3.0126E-03	2.7686E-03	1.6304E-03
0.25–10.0[b]	3.0763E-04	2.9422E-04	2.3178E-04
10.0–100[b]	0.0000E+00	0.0000E+00	0.0000E+00
Total[b]	3.3260E-03	3.0685E-03	1.8687E-03

a. Dosimeter 30.48 cm (1 ft) from center of fuel pellet.
b. Dosimeter 100 cm from center of fuel pellet.

Monte Carlo calculations were performed that compared the transmission of 15 keV photons through ¼-in. plate glass and ¼-in. Lucite (an alternative window material). When the dose location is one foot from the source, the fluence through the ¼-in. equivalent plate glass is reduced by a factor of 3.8E-05. When the source-receptor distance was one meter, the transmission of 15 keV (and lower) photons through the ¼-in. plate-glass-equivalent window was zero.

6.0 DOSE SCENARIOS AND ANNUAL DOSE ASSIGNMENT

Dose rates were calculated at one foot (30.48 cm) and one meter from the center of a single pellet (Table 6). The dose rates from a single pellet were scaled upward to account for the maximum amount of plutonium allowed to be processed in any batch. Consistent with the *Technical Basis Document: Site Profiles for Atomic Weapons Employers That Worked Uranium Metals* (NIOSH 2011b) (and following the approach used to calculate uranium exposures above), the following assumptions are made about a worker's exposure conditions:

- Operator: 50% of the workday was spent at one foot (30.48 cm) from the fuel object containing 525 grams of pellets (i.e., 100 grams of Pu).
- Radiation (Rad) Production Support: 50% of the workday was spent at one meter from the fuel object containing 525 grams of pellets (i.e., 100 grams of Pu).
- Non-Rad Production Area: Exposure was equal to 50% of the Rad Production Support exposure.
- Administrative: Exposure was equal to 10% of the Non-Rad Production Area exposure.

Table 6. External Dose from 2nd Operational Period for Plutonium (1959 to 1967).

Job Category[a]	Photons (keV)[b] <30 (rem/y)	Photons (keV)[b] 30–250 (rem/y)	Photons (keV)[b] >250 (rem/y)	Neutrons (MeV)[b] <0.01 (rem/y)	Neutrons (MeV)[b] 0.01–0.1 (rem/y)	Neutrons (MeV)[b] 0.1–2.0 (rem/y)	Neutrons (MeV)[b] 2.0–20 (rem/y)
Operator	0.020	8.551	0.936	N/A[c]	0.001	0.129	0.183
Rad Production Support	0.002	1.091	0.116	N/A[c]	N/A[c]	0.016	0.022
Non-Rad Production Area	0.001	0.545	0.058	N/A[c]	N/A[c]	0.008	0.011
Administrative	<0.001	0.055	0.006	N/A[c]	N/A[c]	N/A[c]	0.001

a. The following assumptions are made about a worker's exposure conditions: Operator: 50% of the workday was spent at one foot (30.48 cm) from a fuel object containing 525 grams of pellets (i.e., 100 grams of Pu), Rad Production Support: 50% of the workday was spent at one meter from a fuel object containing 525 grams of pellets (i.e., 100 grams of Pu), Non-Rad Production Area: Exposure was equal to 50% of the Rad Production Support exposure, Administrative: Exposure was equal to 10% of the Non-Rad Production Area exposure.
b. Assigned as a constant distribution. Specific organ dose values calculated using the applicable H*10 dose conversion factors and ICRP 60 (1991) neutron weighting factors.
c. All dose rates were less than 0.001 rem/year. Therefore, no dose is assigned.

Photon and neutron doses are treated as a constant distribution and then multiplied by the applicable H*10 dose conversion factors. All photon doses are assigned as acute exposures. All neutron doses are assigned as chronic exposures.

7.0 REFERENCES

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