LA-UR-17-26202
Approved for public release; distribution is unlimited.

Title: Tabulated Neutron Emission Rates for Plutonium Oxide

Author(s): Shores, Erik Frederick

Intended for: Interim report to sponsor
Report

Issued: 2017-07-24

**Tabulated Neutron Emission Rates for Plutonium Oxide**

Erik F. Shores, XTD-Safety and Surety Group
Los Alamos National Laboratory

**Summary**

This work tabulates neutron emission rates for 80 plutonium oxide samples as reported in the literature. Plutonium-238 and plutonium-239 oxides are included and such emission rates are useful for scaling tallies from Monte Carlo simulations and estimating dose rates for health physics applications.

**Introduction**

The Los Alamos Monte Carlo code MCNP [Goor12] provides tallies on a "per source particle" basis. Radiation transport application modelers may account for the number of source particles within a given system by exploiting source and/or tally multipliers within MCNP or, alternatively, scaling results in a post-processing step.

Multiplicative scalars are application dependent and may be calculated or measured quantities. Neutron emission rates for several plutonium oxide samples (Figure 1) are tabulated below and represent candidate multipliers for MCNP models of this material, whether modeling the source explicitly or as a point. The plutonium-238 oxide samples are readily apparent in Figure 1 (the first 24 indices) since their neutron output is orders of magnitude higher than that from plutonium-239 oxide.

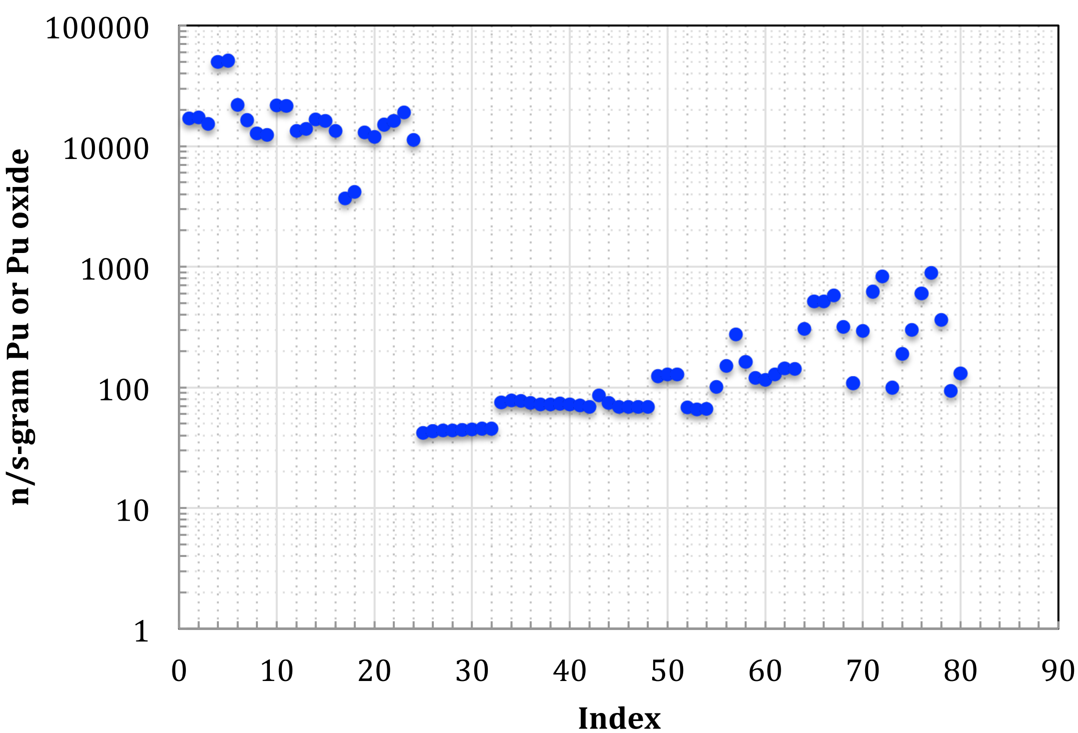


Figure 1. Plutonium oxide neutron emission rates

Table 1. Plutonium Oxide Neutron Emission Rates

| Index | Oxide | Reference | Emission Rate | Units | Comments |
|---|---|---|---|---|---|
| 1 | Pu238 | [Bair73] | 1.705E+04 | n/s-g Pu-238 | 5.09 g sample; isotopics, impurities provided; 90.37 wt% 238; measured to 1% accuracy |
| 2 | | [Bair73] | 1.72e+04 | n/s-g Pu-238 | [Bair73] cites communication with Anderson |
| 3 | | [Bube68] | 1.52E+04 | n/s-g Pu-238 | 10-20 g samples |
| 4 | | [Shor02] | 4.97E+04 | n/s-g oxide | Calculation; molten salt oxidation study |
| 5 | | [Enss91] | 5.1E+04 | n/s-g oxide | PANDA manual; (0.003e6n/s-Ci)x(17Ci/g) |
| 6 | | [Ande69] | 2.20E+04 | n/s-g 238 | Estimate (n,f) in such a large source would be 12-18% of yield; (a,n) as much as 10%; correcting for this → ~1.65e4 n/s per g 238 |
| 7 | | [Hero68] | 1.65E+04 | n/s-g 238 | 73.5 g powdered oxide sample; “81% 238Pu”; 51.8 g 238 via calorimetric assay; impurities and energy spectrum also included |
| 8 | | [Pans01] | 1.2756E+04 | n/s-g oxide | First six samples were prepared with “ion exchange" during aqueous purification; six more values "without ion exchange” are available and range from 1.8034e4 to 2.5524e4 n/s-g oxide |
| 9 | | | 1.2337E+04 | | |
| 10 | | | 2.1655E+04 | | |
| 11 | | | 2.1436E+04 | | |
| 12 | | | 1.3319E+04 | | |
| 13 | | | 1.3889E+04 | | |
| 14 | | [Crof92] | 1.6616E+04 | n/s-g 238 | Calculated |
| 15 | | [Tahe72] | 1.62E+04 | n/s-g oxide | Calculated |
| 16 | | | 1.34E+04 | | |
| 17 | | [Ruth67] | 3.7E+03 | n/s-g 238 | Oxygen depletion study (varied 18O content); neutron counting accuracy +/- 13% |
| 18 | | | 4.2E+03 | | |
| 19 | | | 1.3E+04 | | |
| 20 | | | 1.2E+04 | | |
| 21 | | | 1.5E+04 | | |
| 22 | | [Tahe72a] | 1.62E+04 | n/s-g oxide | Calculated |
| 23 | | [Stod65] | 1.9E+04 | n/s-g oxide | |
| 24 | | [Matl67] | 1.13E+04 | n/s-g 238 | Calculated; 81 (wt?)% 238 |
| 25 | Pu239 | [Menl86] | 4.2021E+01 | n/s-g Pu | Los Alamos National Laboratory (June 1984) oxide samples; mass varied from 59.97 to 876.58 g plutonium and isotopics included—generally ~16 wt% 240; all samples were in cylindrical geometry and the varied diameter-to-height ratio affected measured and calculated multiplication. These sample cans were 8.3 cm diameter and 10.5 cm high and densities were not precisely known. |
| 26 | | | 4.3445E+01 | | |
| 27 | | | 4.3689E+01 | | |
| 28 | | | 4.3831E+01 | | |
| 29 | | | 4.4565E+01 | | |
| 30 | | | 4.5146E+01 | | |
| 31 | | | 4.5330E+01 | | |
| 32 | | | 4.5441E+01 | | |
| 33 | | | 7.5214E+01 | | BNFL Sellafield, UK (Feb 1985) samples; mass varied from 2.476 to 7.156 kg plutonium and isotopics included—generally ~22wt% 240; all |
| 34 | | | 7.7998E+01 | | |
| 35 | | | 7.7493E+01 | | |

| Index | Oxide | Reference | Emission Rate | Units | Comments |
|---|---|---|---|---|---|
| 36 | | | 7.4429E+01 | | samples were in cylindrical geometry and the varied diameter-to-height ratio affected measured and calculated multiplication. These sample cans were 12.5 cm diameter and 26 cm high and densities were not precisely known.<br><br>For all samples in [Menl86], plutonium mass and total count rate were provided (n/s). The table entries here summarize (cnt rate, n/s)/(mass). |
| 37 | | | 7.2197E+01 | | |
| 38 | | | 7.2395E+01 | | |
| 39 | | | 7.3733E+01 | | |
| 40 | | | 7.2237E+01 | | |
| 41 | | | 7.0879E+01 | | |
| 42 | | | 6.9024E+01 | | |
| 43 | | | 8.5583E+01 | | |
| 44 | | | 7.4294E+01 | | |
| 45 | | | 6.8734E+01 | | |
| 46 | | | 6.8889E+01 | | |
| 47 | | | 6.8772E+01 | | |
| 48 | | | 6.8923E+01 | | |
| 49 | | | 1.2454E+02 | | |
| 50 | | | 1.2852E+02 | | |
| 51 | | | 1.2829E+02 | | |
| 52 | | | 6.8419E+01 | | Belgonucleaire, Belgium (Sept 1984) oxide samples; mass range from 1.589 to 2782 kg plutonium and isotopics included—generally ~25 wt% 240; all samples were in cylindrical geometry and the varied diameter-to-height ratio affected measured and calculated multiplication. These sample cans were 8.5 cm diameter and 33 cm high and densities were not precisely known. |
| 53 | | | 6.5991E+01 | | |
| 54 | | | 6.6223E+01 | | |
| 55 | | | 1.0139E+02 | | |
| 56 | | | 1.5127E+02 | | |
| 57 | | | 2.7565E+02 | | |
| 58 | | | 1.6141E+02 | | |
| 59 | | | 1.1902E+02 | | |
| 60 | | | 1.1568E+02 | | |
| 61 | | | 1.2861E+02 | | |
| 62 | | | 1.4304E+02 | | |
| 63 | | | 1.4254E+02 | | |
| 64 | | [Faus74] | 3.06E+02 | n/s-g Pu | 1975 BWR, cycle 1 (6 months old) |
| 65 | | | 5.21E+02 | | 1985 PWR, cycle 2 |
| 66 | | | 5.16E+02 | | 1985 PWR, 60 d after separation |
| 67 | | | 5.76E+02 | | 1985 PWR, 5 yr after separation |
| 68 | | [Brac70] | 3.1724E+02 | n/s-g Pu | 725 g source, 22.7 wt% 240 |
| 69 | | [Menl98] | 1.0877E+02 | n/s-g Pu | Low burnup, ~14 wt% 240 (10yr after sep) |
| 70 | | | 2.9477E+02 | | Intermediate burnup, ~14 wt% 240 |
| 71 | | | 6.2556E+02 | | Medium burnup, ~22 wt% 240 |
| 72 | | | 8.3673E+02 | | High burnup, ~25 wt% 240 |
| 73 | | [Ande82] | 1.0E+02 | n/s-g Pu | Low burnup <8000 MWD/t (~6 wt % 240) |
| 74 | | | 1.9E+02 | | 8-10 1000 MWD/t (~10 wt % 240) |

| Index | Oxide | Reference | Emission Rate | Units | Comments |
|---|---|---|---|---|---|
| 75 | | | 3.0E+02 | | 16-18 1000 MWD/t (~18 wt % 240) |
| 76 | | | 6.0E+02 | | 25-27 1000 MWD/t (~25 wt % 240) |
| 77 | | | 8.90E+02 | | 38-40 1000 MWD/t (~27 wt % 240) |
| 78 | | [Kimu86] | 3.61668e+02 | n/s-g Pu | 0.947 g sample, ~20 wt% 240 |
| 79 | | | 9.42482E+01 | | 0.991 g sample, ~4.5 wt% 240 |
| 80 | | [Shor15] | 1.31E+02 | n/s-g oxide | 3437 g sample, ~6 wt% 240 |

## Discussion

Inspection of Table 1 reveals a relatively large mass range (1 to 7200 g) although sample mass was not always reported. Neutron emission rates for over 40 plutonium-239 oxide samples in this 7 kg range are shown in Figure 2. The higher neutron rates for smaller mass samples are indicative of higher burnup isotopics.

Multiplication and other secondary reactions, e.g. (n,2n), may be significant in larger plutonium oxide sources and should be considered in any application of reported neutron measurements.

An example shielding application is shown in Figure 3. In 1974, Faust and Brackenbush demonstrated six inches of polyethylene reduce the plutonium oxide neutron dose by an order of magnitude [Faus74]. “A [similar] rule of thumb for neutron dose reduction is that 10 cm of polyethylene will reduce the neutron dose by roughly a factor of 10” [Rina91]. Note 15 cm (six inches) is mentioned elsewhere [DOE98] and the 10 cm rule of thumb was applied in [Shor01].

The results from two MCNP6 models, effectively reproducing the [Faus74] measurements, are also shown in Figure 3. The “polyethylene” curve is from an explicit model of a 3.4 kg plutonium oxide source [Shor15] while the “point source” simulations represent a self-explanatory version of the same material. Both MCNP6 models applied a source weight multiplier of 4.5e5 n/s (e.g. see Table 1, index 80; 3437 g oxide x 131 n/s-g oxide = 4.5e5 n/s).

While general trends in Figure 3 agree, model deviation from the measurements increases as the shield thickness increases and this could be an artifact of our model geometry assumptions. For simplicity and symmetry (e.g. ring-tally application), the polyethylene shield was modeled as a relatively close fitting cylindrical or spherical shell while the [Faus74] measurements were made in unspecified slab geometry.

Treading the polyethylene shield as an MCNP “attenuator set” (i.e. not explicitly modeling material) overestimated the dose reduction and only 3” were necessary to achieve an order of magnitude drop. One arbitrary slab geometry (15.24 cm thick) simulation was considered and the shielded dose was 13% of the unshielded case.

All simulations exercised MCNP6.2.0 ld=06/01/17 on LANL’s MOONLIGHT machine.

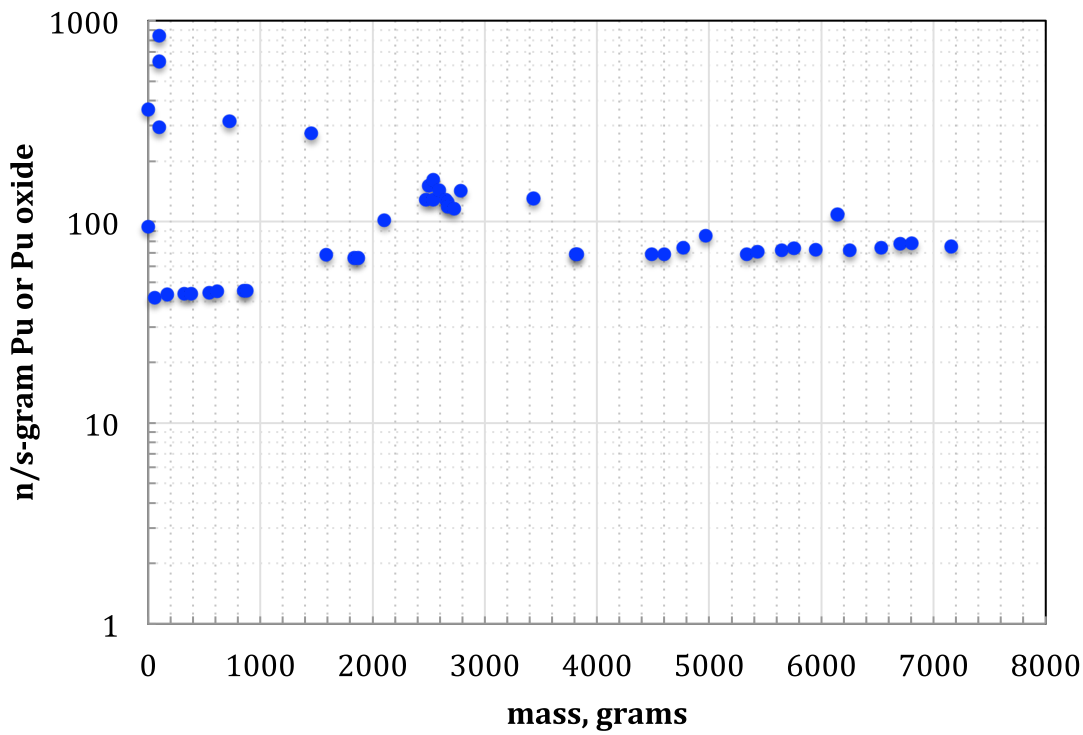


Figure 2. Plutonium-239 oxide neutron emission rates as a function of sample mass

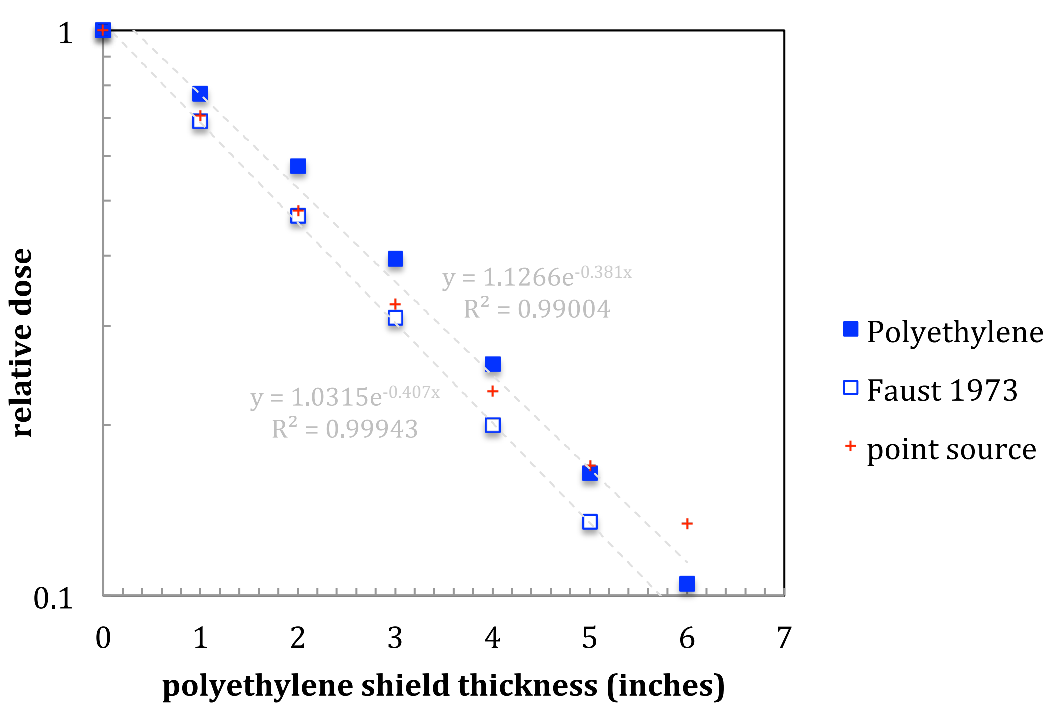


Figure 3. Plutonium-239 oxide neutron dose reduction from polyethylene

## Conclusions

This work tabulated neutron emission rates from 80 plutonium oxide sources spanning a seven-kilogram mass range. Such emission rates may be applied to Monte Carlo simulations and one such shielding application was demonstrated. Other applications are found in [Shor01], [Shor04], and [Shor15].

## References


[Ande82] Anderson, M.E. and J.F. Lemming, Selected Measurement Data for Plutonium and Uranium, MLM-3009 (1982)

[Bair73] Bair, J.K. and H.M. Butler, Neutron Yield from a Small High Purity 238PuO2 Source, Nucl Tech 19 (1973)

[Brac70] Brackenbush, L.W. and L.G. Faust, Neutron Spectra of Plutonium Compounds Part I: 3He and 6Li Spectrometer Measurements, BNWL-1262 (1970)

[Crof92] Croft, S., The (alpha,n) Neutron Yield From Pure 238PuO2, Ann. Nucl. Energy Vol 19 No 8, 451-457 (1992)

[DOE98] DOE STANDARD, Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilitites, DOE-STD-1128-98 (June 1998) Change Notice 2, Dec 2006

[Enss91] Ensslin, N., The Origin of Neutron Radiation, In: Passive Nondestructive Assay of Nuclear Materials, Los Alamos National Laboratory Report LA-UR-90-732, NUREG/CR-5550 (1991)

[Faus74] Faust L.G. and L.W. Brackenbush, High Exposure Plutonium Fuels from Light Water Reactors, BNWL-SA-5044, CONF-741106-1 (1974)

[Goor12] T. Goorley, et al., "Initial MCNP6 Release Overview", Nuclear Technology, 180 (Dec 2012) 298-315

[Hero67] Herold T. R., Neutron Spectrum of 238PuO2, Nucl. Appl. 4 (1968)

[Kimu86] Kimura T. et al., Neutron Yields from Actinide Oxides, Appl. Radiat. Isot. Vol 37 No 2, 121-125 (1986)

[Menl86] Menlove H.O. and P.M. Rinard, Calculation of Fast-Neutron Induced-Fission Multiplication in Large PuO2 Samples, LA-10842-MS (1986)

[Menl98] Menlove, H.O. and T.R. Wenz, Measurements of impure reactor-grade plutonium using the increase in totals neutrons from plutonium-241 decay, Nucl. Instrum. Meth. Phys. Research A 423 (1999) 394-403

[Pans01] Pansoy-Hjelvik et al., Purification and Neutron Emission Reduction of 238Plutonium Oxide by Nitrate Anion Exchange Processing, CP552, AIP (2001)

[Rina91] Rinard, P., Neutron Interactions with Matter, In: Passive Nondestructive Assay of Nuclear Materials, Los Alamos National Laboratory Report LA-UR-90-732, NUREG/CR-5550 (1991)

[Ruth67] Rutherford, W.M. et al., Preparation and Neutron Counting of 238PuO2 Depleted in 18O, Nucl. Appl. 3 (1967)

[Shor01] Shores, Erik F., Shielding Design for a Plutonium Oxide Storage Room, LA-UR-01-2266 (2001)

[Shor02] Shores, Erik F., Calculated Neutron Production Rates from a 238PuO2 Source, LA-UR-02-5872 (2002)

[Shor04] Shores, Erik F., Plutonium oxide benchmark problems for the SOURCES code, App. Radiat. and Iso. 62 (2005) 699–704

[Shor15] Shores et al., Predicted Radiation Output from Plutonium and Uranium Oxide, LA-UR-15-20307 (2015)

[Tahe72] Taherzadeh, M. and P. J. Gingo, Neutron Radiation Characteristics of Plutonium Dioxide Fuel, Nucl. Tech. 15 (1972)

[Tahe72a] Taherzadeh, M., Neutron Emission Characteristics of Plutonium Dioxide Fuel, Trans. Amer. Nucl. Soc. 15, 687 (1972)