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Alpha-n and Spontaneous Fission Sources and Spectra from Individual Plutonium Isotopes in PuF₄ and PuO₂

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Introduction

Plutonium-containing compounds vary widely in isotopic content, and as a result, the dose as function of isotopic content also varies considerably. Determination of the dose from neutrons, decay and capture gammas from plutonium in the form of compounds, thus requires that the spontaneous fission (SF) and alpha-n (α,n) source and spectra from each individual isotope be known.

To facilitate dose calculations from plutonium-containing compounds, we have calculated the spontaneous fission and (α,n) sources and spectra for 1 g of each of the plutonium isotopes in the form of either PuF₄ or PuO₂. As ²⁴¹Am is often a component in a mixture of plutonium isotopic compounds, the source and spectra from 1 g of ²⁴¹Am mixed with PuF₄ or PuO₂ has also been determined. Using these results, the neutron source and spectra may be determined for any sample composition.

Methods

The neutron sources and spectra were calculated using the SOURCES¹ code and library. The code is under development. The code determines (β,n) delayed, spontaneous fission and (α,n) neutron sources and spectra due to the decay of a radionuclides in homogeneous media. Spontaneous fission spectra are calculated with evaluated half life, SF branching, and ν data using Watt spectrum parameters for 43 actinides. The (α,n) spectra are calculated with a library of nuclide decay alpha spectra, evaluated (α,n) cross sections and product nuclide level branching fractions, and functional α stopping cross sections using an assumed isotropic neutron angular distribution in the center-of-mass system.

Results

The neutron source strength from spontaneous fission and (α,n) reactions for 1 g of each isotope are given in Table 1. The alphas from ^{241}Am are assumed to slow down in either PuF_4 or PuO_2 . The source for ^{243}Pu is from spontaneous fission alone as this isotope decays only by beta emission.

The combined SF and (α,n) spectra for each isotope, in the form PuO_2 or PuF_4 , is given in Tables 2 and 3. The first 22 energy groups correspond to those of the MATXS769 group neutron transport cross section library.² The broad final group contains so few neutrons as to be insignificant. Each spectra is normalized to the total source strength for 1 g of a particular isotope. The spectra for ^{241}Am are those produced from alpha particles slowing down in either PuO_2 or PuF_4 . The spectra for ^{243}Pu are the spontaneous fission spectra alone.

Conclusion

The absolute source and spectra for any combination of isotopes may be obtained using the results in Tables 1, 2, and 3. This source, spectra, and a coupled neutron-gamma library input into a transport code, then, would provide a method of determining dose from samples of PuO_2 or PuF_4 .

References

1. W.B. Wilson, R.T. Perry, J.F. Stewart, T.R. England, D. G. Madland, and F.D. Arthur, "Development of the SOURCES Code and Data Library for the Calculation of Neutron Sources and Spectra from (α,n) Reactions, Spontaneous Fission, (β,n) Delayed Neutrons," Los Alamos National Laboratory progress report LA-9841-PR, pp. 65-66, Los Alamos National Laboratory (August 1993).
2. RSIC Data Library Collection, "MATXS7A - 69 Neutron Group Cross Section Library in MATXS." Oak Ridge National Laboratory report OLC-117 (December 1985).

Table 1
Alpha-n and Spontaneous Fission Neutron Source Strength
for One Gram of Metal for Plutonium Isotopes in the Form of
PuO₂ or PuF₄ and ²⁴¹Am in PuO₂ or PuF₄

Isotope	Spontaneous Fission neutrons/gm	PuO ₂ alpha-n neutrons/gm	PuO ₂ total source neutrons/gm	PuF ₄ alpha-n neutrons/gm	PuF ₄ total source neutrons/gm
²³⁸ Pu	2.587E+03	1.412E+04	1.671E+04	2.175E+06	2.178E+06
²³⁹ Pu	2.181E-02	4.013E+01	4.015E+01	5.653E+03	5.653E+03
²⁴⁰ Pu	1.026E+03	1.481E+02	1.174E+03	2.091E+04	2.194E+04
²⁴¹ Pu	4.939E-02	1.357E+00	1.406E+00	1.704E+02	1.705E+02
²⁴² Pu	1.717E+03	2.150E+00	1.719E+03	2.696E+02	1.987E+03
²⁴³ Pu	6.618E-02	0.000E+00	6.618E-02	0.000E+00	6.618E-02
²⁴⁴ Pu	1.900E+03	7.531E-03	1.900E+03	7.628E-01	1.901E+03
²⁴¹ Am	1.182E+00	2.817E+03	2.818E+03	4.331E+05	4.331E+05

Table 2
Neutron Spectra Normalized to Total Source Strength for
Plutonium Isotopes in the Form of PuO₂ and ²⁴¹Am in PuO₂

Group Upper Energy Boundary, MeV	²³⁸ PuO ₂ neutrons per group	²³⁹ PuO ₂ neutrons per group	²⁴⁰ PuO ₂ neutrons per group	²⁴¹ PuO ₂ neutrons per group	²⁴² PuO ₂ neutrons per group	²⁴³ PuO ₂ neutrons per group	²⁴⁴ PuO ₂ neutrons per group	²⁴¹ Am in in PuO ₂ neutrons per group
1.000E+01	5.453E+01	5.260E-04	1.702E+01	9.961E-04	3.119E+01	7.996E-04	1.809E+01	3.406E-02
6.065E+00	9.529E+02	7.400E-01	1.067E+02	1.733E-02	1.802E+02	5.819E-03	1.503E+02	1.302E+02
3.679E+00	6.533E+03	1.661E+01	2.827E+02	5.551E-01	3.737E+02	1.373E-02	3.828E+02	1.189E+03
2.231E+00	5.233E+03	1.297E+01	2.952E+02	4.519E-01	4.116E+02	1.622E-02	4.706E+02	9.219E+02
1.353E+00	2.302E+03	6.332E+00	2.125E+02	2.383E-01	3.133E+02	1.275E-02	3.767E+02	3.693E+02
8.210E-01	9.679E+02	2.324E+00	1.250E+02	9.520E-02	1.927E+02	7.936E-03	2.361E+02	1.377E+02
5.000E-01	4.084E+02	8.270E-01	6.805E+01	3.320E-02	1.076E+02	4.446E-03	1.325E+02	5.057E+01
3.025E-01	1.413E+02	2.129E-01	3.417E+01	8.491E-03	5.530E+01	2.285E-03	6.808E+01	1.728E+01
1.830E-01	5.994E+01	7.331E-02	1.673E+01	3.349E-03	2.729E+01	1.127E-03	3.353E+01	4.101E+00
1.110E-01	2.756E+01	2.951E-02	8.114E+00	1.537E-03	1.329E+01	5.479E-04	1.629E+01	1.673E+00
6.734E-02	1.285E+01	1.257E-02	3.896E+00	6.775E-04	6.392E+00	2.634E-04	7.827E+00	7.215E-01
4.085E-02	5.983E+00	5.306E-03	1.858E+00	2.990E-04	3.053E+00	1.257E-04	3.734E+00	3.137E-01
2.478E-02	2.833E+00	2.481E-03	8.832E-01	1.390E-04	1.452E+00	5.978E-05	1.775E+00	1.464E-01
1.503E-02	1.354E+00	1.196E-03	4.188E-01	6.608E-05	6.885E-01	2.834E-05	8.412E-01	7.100E-02
9.118E-03	6.462E-01	5.739E-04	1.985E-01	3.197E-05	3.262E-01	1.342E-05	3.985E-01	3.466E-02
5.530E-03	2.869E-01	2.561E-04	8.754E-02	1.418E-05	1.439E-01	5.923E-06	1.758E-01	1.555E-02
3.519E-03	1.463E-01	1.301E-04	4.451E-02	7.208E-06	7.309E-02	3.012E-06	8.935E-02	7.981E-03
2.239E-03	7.470E-02	6.650E-05	2.259E-02	3.678E-06	3.714E-02	1.526E-06	4.537E-02	4.119E-03
1.425E-03	3.812E-02	3.403E-05	1.145E-02	1.878E-06	1.890E-02	7.768E-07	2.302E-02	2.138E-03
9.069E-04	2.921E-02	2.624E-05	8.780E-03	1.444E-06	1.436E-02	5.926E-07	1.766E-02	1.670E-03
5.673E-04	7.626E-03	6.793E-06	2.290E-03	3.732E-07	3.674E-03	1.520E-07	4.504E-03	4.457E-04
3.487E-04	1.573E-03	1.510E-06	4.901E-04	8.392E-08	8.521E-04	3.653E-08	1.033E-03	1.002E-04
2.250E-04	8.904E-04	8.182E-07	2.717E-04	4.481E-08	4.343E-04	1.713E-08	5.291E-04	5.382E-05

Table 3
Neutron Spectra Normalized to Total Source Strength for
Plutonium Isotopes in the Form of PuF₄ and ²⁴¹Am in PuF₄

Group Upper Energy Boundary, MeV	²³⁸ PuF ₄ neutrons per group	²³⁹ PuF ₄ neutrons per group	²⁴⁰ PuF ₄ neutrons per group	²⁴¹ PuF ₄ neutrons per group	²⁴² PuF ₄ neutrons per group	²⁴³ PuF ₄ neutrons per group	²⁴⁴ PuF ₄ neutrons per group	²⁴¹ Am in PuF ₄ neutrons per group
1.000E+01	5.453E+01	5.260E-04	1.702E+01	9.961E-04	3.119E+01	7.996E-04	1.809E+01	3.406E-02
6.065E+00	2.906E+02	2.588E-03	1.039E+02	5.425E-03	1.802E+02	5.819E-03	1.503E+02	1.513E-01
3.679E+00	1.854E+05	2.737E+02	1.251E+03	4.243E+00	3.794E+02	1.373E-02	3.828E+02	3.640E+04
2.231E+00	8.089E+05	2.046E+03	7.831E+03	5.592E+01	4.992E+02	1.622E-02	4.707E+02	1.610E+03
1.353E+00	5.803E+05	1.853E+03	7.008E+03	6.354E+01	4.136E+02	1.275E-02	3.770E+02	1.159E+05
8.210E-01	2.936E+05	7.430E+02	2.855E+03	2.773E+01	2.365E+02	7.936E-03	2.363E+02	5.827E+04
5.000E-01	1.721E+05	3.763E+02	1.464E+03	1.076E+01	1.246E+02	4.446E-03	1.326E+02	3.416E+04
3.025E-01	8.179E+04	1.932E+02	7.542E+02	4.497E+00	6.237E+01	2.285E-03	6.810E+01	1.633E+04
1.830E-01	3.342E+04	9.743E+01	3.782E+02	2.006E+00	3.044E+01	1.127E-03	3.354E+01	6.680E+03
1.110E-01	1.323E+04	4.118E+01	1.598E+02	9.409E-01	1.476E+01	5.479E-04	1.629E+01	2.644E+03
6.734E-02	5.136E+03	1.625E+01	6.389E+01	4.440E-01	7.090E+00	2.634E-04	7.828E+00	1.075E+03
4.085E-02	2.158E+03	7.026E+00	2.789E+01	2.264E-01	3.410E+00	1.257E-04	3.735E+00	4.304E+02
2.478E-02	9.313E+02	3.139E+00	1.256E+01	1.053E-01	1.619E+00	5.978E-05	1.775E+00	1.854E+02
1.503E-02	3.929E+02	1.373E+00	5.475E+00	4.489E-02	7.597E-01	2.834E-05	4.413E-01	7.801E+01
9.118E-03	1.671E+02	5.931E-01	2.368E+00	1.895E-02	3.564E-01	1.347E-05	3.985E-01	3.322E+01
5.530E-03	6.832E+01	2.401E-01	9.860E-01	7.736E-03	1.563E-01	5.923E-06	1.758E-01	1.364E+01
3.519E-03	3.327E+01	1.193E-01	4.844E-01	3.747E-03	7.906E-02	3.012E-06	8.935E-02	6.652E+00
2.239E-03	1.635E+01	5.879E-02	2.389E-01	1.831E-03	4.005E-02	1.576E-06	4.537E-02	3.269E+00
1.425E-03	8.130E+00	2.932E-02	1.189E-01	9.090E-04	2.034E-02	7.768E-07	2.302E-02	1.626E+00
9.069E-04	6.146E+00	2.218E-02	9.000E-02	6.830E-04	1.555E-02	5.936E-07	1.766E-02	1.329E+00
5.673E-04	1.570E+00	5.672E-03	2.306E-02	1.742E-04	3.951E-03	1.520E-07	4.504E-03	3.140E-01
1.487E-04	3.469E-01	1.252E-03	5.070E-03	3.855E-05	9.134E-04	3.653E-08	1.033E-03	6.939E-02
1.550E-05	1.872E-01	6.773E-04	2.753E-03	2.078E-05	4.673E-04	1.713E-08	5.292E-04	3.746E-02

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