

Title:

**THICK-TARGET (α,n) NEUTRON YIELDS AND
SPECTRA CALCULATED WITH ORIGEN-S AND
SOURCES**

Author(s):

T. H. Brown, R. T. Perry, W. B. Wilson and
W. S. Charlton

Submitted to:

<http://lib-www.lanl.gov/la-pubs/00796343.pdf>

THICK-TARGET (α ,n) NEUTRON YIELDS AND SPECTRA CALCULATED WITH ORIGEN-S AND SOURCES

T. H. Brown
Los Alamos National
Laboratory
Group ESH-12, MS K483
Los Alamos, NM 87545
505-665-4846
tombrown@lanl.gov

R. T. Perry
Los Alamos National
Laboratory
Group ESH-12, MS K483
Los Alamos, NM 87545
505-665-3521
rtperry@lanl.gov

W. B. Wilson
Los Alamos National
Laboratory
Group T-16, MS B243
Los Alamos, NM 87545
505-667-7749
wbw@lanl.gov

W. S. Charlton
Los Alamos National
Laboratory
Group NIS-7, MS E541
Los Alamos, NM 87545
505-665-8576
charlton@lanl.gov

ABSTRACT

Thick-target neutron yields and energy spectra from (α ,n) reactions in homogeneous media calculated with ORIGEN-S and SOURCES were compared. SOURCES calculates yields and spectra from fundamental physics and data and is applicable to any combination of α -particle slowing-down, emitting, and target elements. ORIGEN-S employs approximations for both its UO_2 and borosilicate-glass options that limit its applicability. Yields from both ORIGEN-S options compare favorably with yields from SOURCES. For UO_2 -like slowing down media, spectra using the UO_2 option compare moderately well with SOURCE spectra. ORIGEN-S with the borosilicate-glass option is often employed for any non- UO_2 -like slowing down media to generate spectra, but any agreement with SOURCES spectra is coincidental.

I. INTRODUCTION

Both ORIGEN-S¹ and SOURCES^{2,3} generate thick-target neutron yields and energy spectra from (α ,n) reactions in homogeneous materials. SOURCES calculates yield and spectra for any material containing α -emitting and (α ,n) target elements by simulating reaction physics, using α -emission energy spectra, elemental stopping cross sections, (α ,n) cross sections for target nuclei, and branching fractions to product-nuclide energy levels. This methodology results in accurate yield and spectra. ORIGEN-S has two options for calculating yields and spectra. The UO_2 option (default) estimates yields and spectra assuming the input α -emitters to be infinitely dilute in a UO_2 slowing-down medium. The borosilicate-glass option estimates yields from the total input material composition and generates spectra purportedly representative of spectra generated by ^{238}Pu , ^{241}Am , ^{242}Cm , and ^{244}Cm infinitely dilute in borosilicate glass, even if none of these four α -emitters are present in the input material composition.

Because yields from the borosilicate-glass option in ORIGEN-S are based on entire input material composition and

are reasonably accurate, the same is often assumed to be true for spectra. The input/output functionality of the borosilicate-glass option, along with ambiguity in ORIGEN-S documentation, gives the incorrect impression that spectra representative of input compositions are generated. This impression is reinforced by wide usage of the SCALE code system and its ORIGEN-S module and their sponsorship by the U. S. Nuclear Regulatory Commission.¹

II. COMPARISON OF NEUTRON YIELDS

In a given material, the probability of an α -particle of energy E undergoing an (α ,n) reaction with target element i while slowing down from E to $E - dE$ is given by³

$$f_i(E) = - \left(\frac{N_i}{N_m} \right) \frac{\sigma_i(E)}{\varepsilon_m(E)} dE$$

where $\varepsilon_m(E) = - \frac{1}{N_m} \left[\frac{dE}{dx}(E) \right]_m$ is the stopping cross

section for the material, $\sigma_i(E)$ is the (α ,n) reaction cross section for target element i , N_i is the atom density of target element i in the material, and N_m is the total atom density of the material. The thick-target yield from all α particles emitted by a given nuclide k is:

$$\begin{aligned} Y_k &= \sum_{j=1}^{J_k} g_{jk} \sum_{i=1}^I \int_0^{E_{jk}^o} f_i(E) dE \\ &= \sum_{j=1}^{J_k} g_{jk} \sum_{i=1}^I \left(\frac{N_i}{N_m} \right) \int_0^{E_{jk}^o} \frac{\sigma_i(E)}{\varepsilon_m(E)} dE \end{aligned} \quad (1)$$

where I is the number of (α ,n) target elements in the material, J_k is the number of α -emission energies of nuclide k , E_{jk}^o is the j -th α -emission energy of nuclide k , and g_{jk} is the fraction of α -emissions from nuclide k occurring at energy E_{jk}^o .

In SOURCES, neutron yield is calculated according to Eq. (1). The origins and quality of the data used in the calculation of Eq. (1) are discussed elsewhere.³ SOURCES uses the Bragg-Kleeman relationship⁴ of additivity of stopping cross sections of elements as separate constituents to calculate material stopping cross section, ε_m , i.e.

$$\varepsilon_m(E) = \sum_{i=1}^M \left(\frac{N_i}{N_m} \right) \varepsilon_i(E) = \sum_{i=1}^M \left(\frac{\rho_i}{\rho_m} \right) P_i(E) \quad \text{where}$$

$$\varepsilon_i(E) = - \frac{1}{N'_i} \left[\frac{dE}{dx} \right]_i \quad \text{and} \quad P_i(E) = - \frac{1}{\rho'_i} \left[\frac{dE}{dx} \right]_i$$

are the stopping cross section and mass stopping power, respectively, of element i as a separate constituent, N'_i and ρ'_i are the atom density and mass density of element i as a separate constituent, N_i and ρ_i are the atom density and mass density of element i in the composite material, and M is the total number of elements in the composite material. The $\varepsilon_i(E)$ are incorporated in SOURCES as parametric fits.^{5,6}

For the borosilicate-glass option in ORIGIN-S, Eq. (1) has been approximated as^{1,7}

$$Y_k^{ORG} = \frac{\sum_{i=1}^I N_i \varepsilon_{ik}(E) y_{ik}(\bar{E}_k^o)}{\sum_{i=1}^M N_i \varepsilon_{ik}(E)} = \frac{\sum_{i=1}^I \rho_i P_{ik}(E) y_{ik}(\bar{E}_k^o)}{\sum_{m=1}^M \rho_i P_{ik}(E)} \quad (2)$$

where $\bar{E}_k^o = \sum_{j=1}^{J_k} g_{jk} E_{jk}^o$ is the average energy of α particles

emitted by nuclide k , M is the total number of elements in the composite material, $\varepsilon_{ik}(E)$ and $P_{ik}(E)$ are the stopping cross section and mass stopping power, respectively, of element i as a separate constituent selected at $E \leq \bar{E}_k^o$ for which (α, n) reactions occur for α particles from nuclide k , $y_{ik}(\bar{E}_k^o)$ is the yield in separate-constituent element i from α emission of nuclide k , and the other parameters have the same meanings as before. Three approximations are incorporated in Eq. (2): (1) Bragg-Kleeman relationship, (2) elemental stopping cross sections have same energy variation, apart from constant factors, and (3) all α emission from one nuclide occurs at one energy equal to the average emission energy. The first two approximations taken together constitute the West approximation.⁸ Because these approximations are physically reasonable and data for stopping cross section and yields are available for many separate-constituent elements, yields estimated with Eq. (2) compare favorably with those generated by SOURCES.

For the UO_2 option in ORIGIN-S, thick-target yields were precalculated for many actinide nuclides emitting α -particles in UO_2 .¹ Thin target cross sections for $^{17}\text{O}(\alpha, n)$ and $^{18}\text{O}(\alpha, n)$ reactions in an oxygen slowing-down medium were used to

calculate the individual actinide yields in UO_2 .⁹ The yield for multiple actinide nuclides in UO_2 is presumably calculated using Eq. (2) for oxygen being the only target element. Again, because of the physical reasonableness of the assumptions, yields in a UO_2 or similar slowing-down medium, e. g. PuO_2 , are expected to compare favorably with those generated by SOURCES.

Differences in yields calculated by SOURCES and ORIGIN-S for various materials and α -emitters are shown in Table I. As surmised, the deviation of the ORIGIN-S yields from the SOURCES yields are reasonable.

Table I
Differences in (α, n) Neutron Yields calculated by
ORIGIN-S and SOURCES

α -emitter(s)	Material	ORIGIN-S option	Yield Difference (%)*
^{238}Pu	^{238}Pu infinitely dilute in borosilicate glass†	borosilicate glass	-3.5
^{242}Cm	^{242}Cm infinitely dilute in borosilicate glass†	borosilicate glass	-13.0
^{244}Cm	^{244}Cm infinitely dilute in borosilicate glass†	borosilicate glass	-5.0
^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am	Weapons Pu metal with (α, n) target impurities	borosilicate glass	11.6
^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am	Weapons Pu infinitely dilute in Be	borosilicate glass	-10.7
^{238}Cm	^{238}Cm infinitely dilute in UO_2	UO_2	8.9
^{244}Cm	^{244}Cm infinitely dilute in UO_2	UO_2	9.3
^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am	Weapons Pu in form of PuO_2	UO_2	6.6

*Deviation of ORIGIN-S yield relative to SOURCES yield.

†Fixed composition given in Table F7.2.1 of Ref. 1.

III. COMPARISON OF NEUTRON ENERGY SPECTRA

SOURCES calculates spectra from fundamental physics formulations and data. The α -particle slowing-down energy range is divided into energy bins. The neutron production for each α -particle energy bin is determined for each product nuclide energy level and then uniformly distributed between the maximum and minimum neutron energies for that level. The validity of uniform distribution in energy follows from the assumption of isotopic neutron emission in the center-of-mass system. The total neutron energy spectrum is formed by summing contributions from all targets and product nuclide levels, each integrated over the α -particle energy range. The details and basis of this procedure are described elsewhere.^{2,3,10,11}

ORIGEN-S with the borosilicate-glass option calculates the spectrum for a given input material using individual precalculated spectra for ^{238}Pu , ^{241}Am , ^{242}Cm , and ^{244}Cm α -emission in borosilicate glass of fixed composition.¹ The spectrum for the input material is determined by averaging these four precalculated spectra according to neutron production from the above four α -emitters in the input material. The emission probability in each energy group of the input-material spectrum, χ_g , is calculated by:^{1,7}

$$\chi_g = \frac{\sum_{k=1}^4 S'_k \frac{\varphi_{kg}}{\sum_{g=1}^G \varphi_{kg}}}{\sum_{k=1}^4 S'_k} \quad ; \quad S'_k = N_k \lambda_k Y_k^{\text{ORG}} \quad (3)$$

where k indicates each of the four α -emitters ^{238}Pu , ^{241}Am , ^{242}Cm , or ^{244}Cm ; S'_k is neutron production rate in the input material from the k -th α -emitter; and φ_{kg} is the borosilicate-glass yield into the g -th energy group from the k -th α -emitter. A spectrum will always be generated if at least one α -emitter and one target element are included in the input material, even if none of the α -emitters ^{238}Pu , ^{241}Am , ^{242}Cm , or ^{244}Cm are present in the input material. For the latter situation, ORIGEN-S documentation does not indicate the makeup of the input-material spectrum from the four precalculated spectra. A user employing the borosilicate-glass option to generate a spectrum for any arbitrary material incorrectly assumes, probably unknowingly, that spectra for all materials are similar to those produced by the above four α -emitters in borosilicate glass.

ORIGEN-S documentation does not indicate how the borosilicate-glass spectra for the four α -emitters, $\{\varphi_{kg}\}$, were precalculated. It only states that the four spectra are based on measured spectra from six target elements as separate constituents, B, O, F, Mg, Al, and Si, impinged with discrete-energy α particles.^{1,7} It is unknown how the code developers

made the transition from these measured spectra to the $\{\varphi_{kg}\}$. It is suspected that the $\{\varphi_{kg}\}$ were calculated by weighting the spectra from the six target elements according to their atom densities in borosilicate glass.

The $\{\varphi_{kg}\}$ spectra calculated from ORIGEN-S are plotted in Fig. 1 for ^{238}Pu , ^{242}Cm , and ^{244}Cm for the fixed borosilicate-glass composition. (The curve for ^{241}Am would be almost identical to that for ^{238}Pu because the two nuclides have nearly identical α -emission energies.) Due to the similarity of the three curves, Eq. (3) will generate similar spectra for all mixtures of α -emitting and target nuclides and slowing down elements. There is no experimental or theoretical basis for this similarity. An example of dissimilarity is given in Fig. 2 that depicts the spectra from weapons Pu infinitely dilute in Be as generated by ORIGEN-S and SOURCES. The ORIGEN-S curve is similar to the curves in Fig. 1 and highly dissimilar to the more accurate SOURCES curve.

The $\{\varphi_{kg}\}$ spectra in Fig. 1 are not even representative of the borosilicate glass composition¹ assumed for their determination. This is evident from comparing these spectra to the SOURCES-generated spectra in Fig. 3. The dissimilarity is due largely to omission of the spectral contributions of Li and Na to the $\{\varphi_{kg}\}$. Their spectral contributions are evident from comparing the ORIGEN-S and SOURCES spectra shown in Fig. 4. From Fig. 5, it is evident that the SOURCES spectrum with these contributions excluded resembles the ORIGEN-S spectrum. This resemblance suggests that the unknown method of determining the $\{\varphi_{kg}\}$ might give reasonable results if applied directly to the input material composition, thus eliminating the need for the precalculated $\{\varphi_{kg}\}$, provided that separate-constituent spectra for all significant targets are incorporated in the ORIGEN-S module.

For the UO_2 option in ORIGEN-S, spectra for ^{238}Pu , ^{242}Cm , and ^{244}Cm in UO_2 have been precalculated by extrapolating, in an unspecified manner, the spectrum measured from a Po- α -O solution source¹² to the α -emission energies of the three nuclides.¹ (The extrapolation apparently did not account for the difference in spectra between the Po- α -O solution, with a presumably low Po:O atom ratio, and UO_2 , with its higher U:O atom ratio.) The spectrum for multiple input α -emitters, not necessarily any of the above three nuclides, is calculated from some unspecified weighting of the three precalculated spectra. ORIGEN-S documentation does not provide the recipe for this weighting.

From the small number of comparisons performed, for α -emission in UO_2 or similar slowing-down media, spectra generated by ORIGEN-S with the UO_2 option are similar to spectra generated by SOURCES. The comparison examples shown in Figs. 6-8 indicate adequate similarity of the spectra.

IV. CONCLUSIONS

Neutron yields generated with the ORIGEN-S borosilicate glass and UO_2 options are expected to be sufficiently accurate for most material compositions. Neutron spectra generated with the UO_2 option for UO_2 and UO_2 -like slowing-down media are expected to be adequate. Neutron spectra generated with the borosilicate glass option are unacceptable for three reasons: (1) no basis for the method of calculating material spectra from precalculated borosilicate glass spectra, (2) no prescription provided on how the precalculated borosilicate glass spectra were generated from separate-element target spectra, and (3) the precalculated borosilicate-glass spectra exclude the contributions of Li and Na.

The (α, n) source module in ORIGEN-S and its documentation are inadequate. The module should be greatly expanded or replaced with SOURCES methodology, and documentation should clearly indicate limitations of the module. The module should be generalized to provide accurate neutron spectra for all materials. The spectra should be calculated only from the composition of the input materials without the user having to specify a UO_2 or borosilicate glass option.

REFERENCES

1. O. W. Hermann 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," Oak Ridge National Laboratory report NUREG/CR-0200, Rev. 6, Vol. 2, Sec. F7, (September 1998).
2. W. B. Wilson, R. T. Perry, W. S. Charlton, E. D. Arthur, M. Bozoian, T. H. Brown, T. R. England, D. G. Madland, T. A. Parish, and J. E. Stewart, "SOURCES-3A: A Code for Calculating (α, n) , Spontaneous Fission, and Delayed Neutron Sources and Spectra, Los Alamos National Laboratory Report LA-UR-97-4365 (October 1997).
3. W. B. Wilson, M. Bozoian, and R. T. Perry, "Calculated α -Induced Thick Target Neutron Yields and Spectra, with Comparison to Measured Data," *Proc. Int. Conf. on Nuclear Data for Science and Technology*, May 30 - June 3, 1988, Mito, Japan (1988) pp. 1193-1197.
4. W. H. Bragg and R. Kleeman, "On the Alpha Particles of Radium and their Loss of Range in Passing through Various Atoms and Molecules," *Phil. Mag.* **10**, 318 (1905).
5. J. F. Ziegler, *Helium Stopping Powers and Ranges in All Elemental Matter*, Vol. 4 of *The Stopping Power and Ranges of Ions in Matter Series*, Pergamon Press, New York (1977).
6. W. B. Wilson, R. T. Perry, J. E. Stewart, T. R. England, D. G. Madland, and E. D. Arthur, "Development of the SOURCES Code and Data Library for the Calculation of Neutron Sources and Spectra from (α, n) Reactions, Spontaneous Fission, and (β, n) Delayed Neutrons," Los Alamos National Laboratory Progress Report LA-9841-PR (August 1983), pp. 65-66.
7. O. W. Hermann, C. V. Parks, and S. B. Ludwig, "ORIGEN-S (α, n) Neutron Source Spectra in Borosilicate Glass Containing HLW," *Trans. Am. Nucl. Soc.* **54**, 63 (1987).
8. D. West, "The Calculation of Neutron Yields in Mixtures and Compounds from the Thick Target (α, n) Yields in the Separate Constituents," *Ann. Nucl. Energy* **6**, 549 (1979).
9. J. K. Bair and J. Gomez del Campo, "Neutron Yields from Alpha-Particle Bombardment," *Nucl. Sci. Eng.* **71**, 18 (1979).
10. B.G. Whitmore and W.B. Baker, "The Energy Spectrum of Neutrons from a PoBe Source," *Phys. Rev.* **78**, 799 (1950).
11. R. T. Perry, W. B. Wilson, W. S. Charlton, "Sources-3A: A Code for Calculating (α, n) , Spontaneous Fission, and Delayed Neutron Sources and Spectra," *Proc. of the 1998 American Nuclear Society Topical Meeting on Radiation Protection and Shielding*, Nashville, Tennessee, April 19-23, 1998, Vol. 2, pp 348-355.
12. A. G. Khabakhpashev, "Neutron Spectrum of a Po- α -O Source," *Atomnaya Energiya* **7**, 71 (1959).

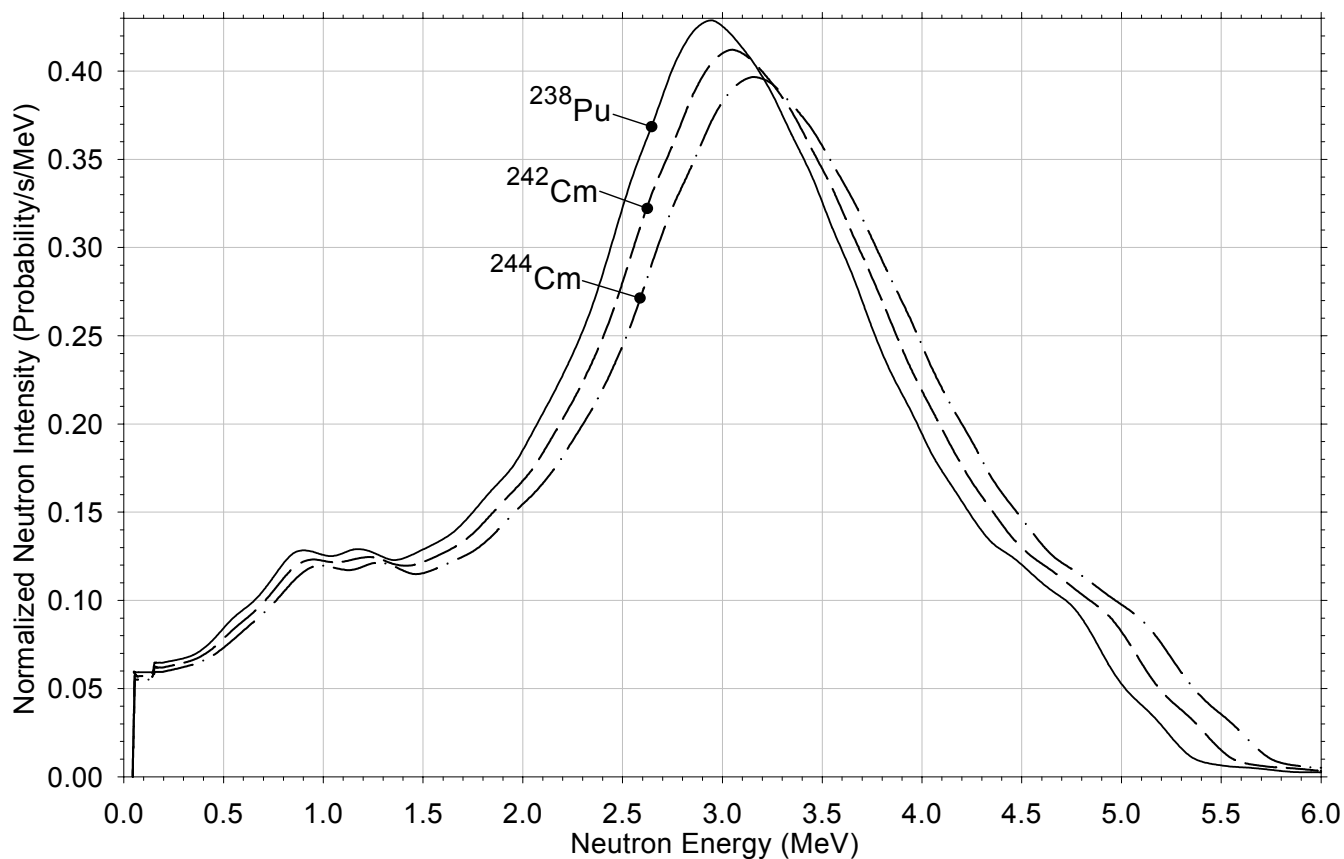


Fig. 1. Normalized (α, n) ORIGEN-S Spectra from ^{238}Pu , ^{242}Cm , and ^{244}Cm Infinitely Dilute in Borosilicate Glass

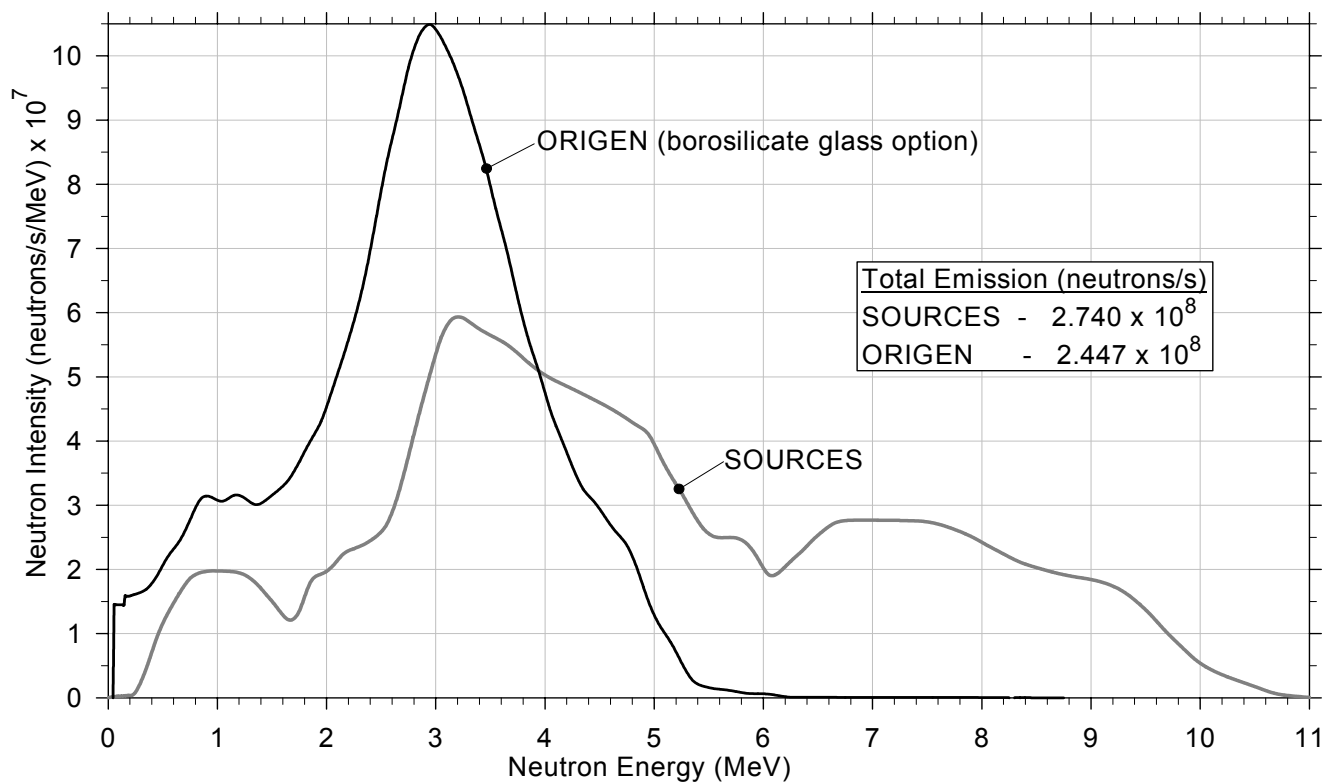


Fig. 2. (α, n) Neutron Energy Spectrum from 1 kg of Pu Infinitely Dilute in Be at 60-y Decay

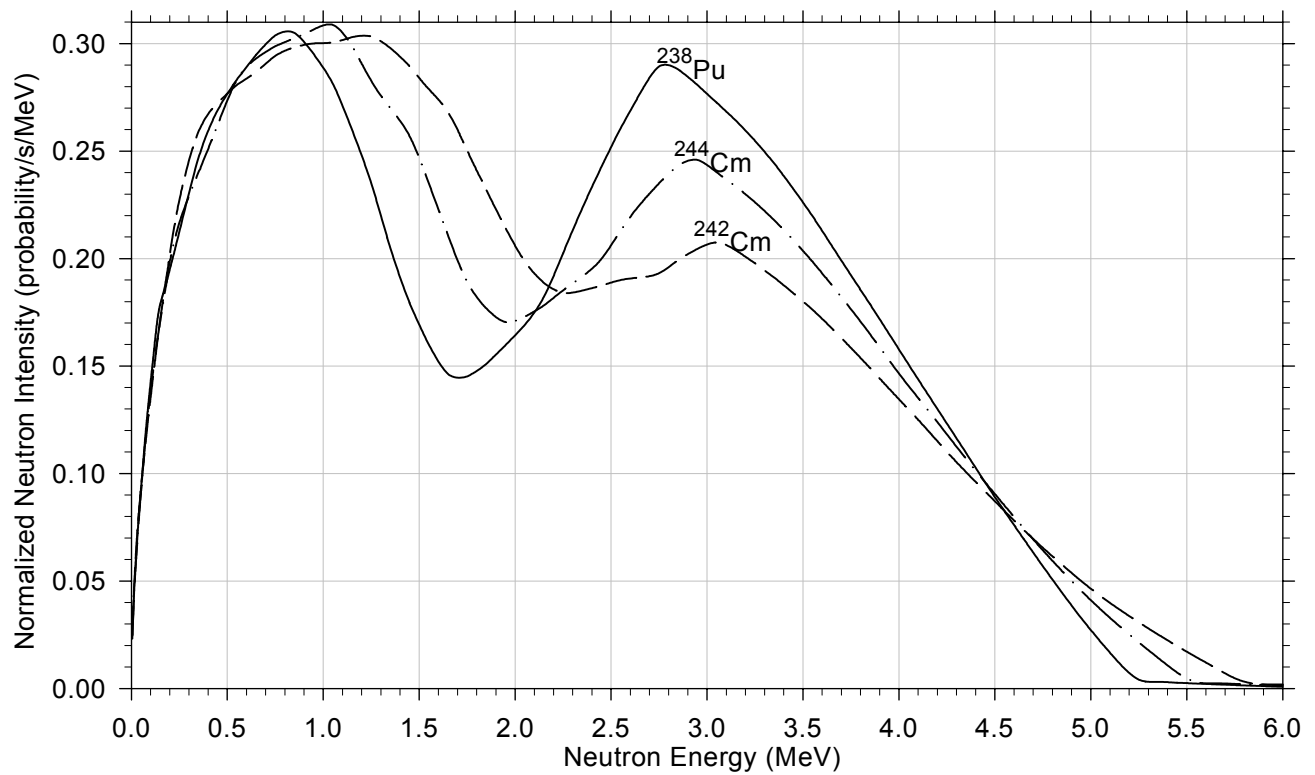


Fig. 3. Normalized SOURCES (α,n) Spectra from ^{238}Pu , ^{242}Cm , and ^{244}Cm Infinitely Dilute in Borosilicate Glass

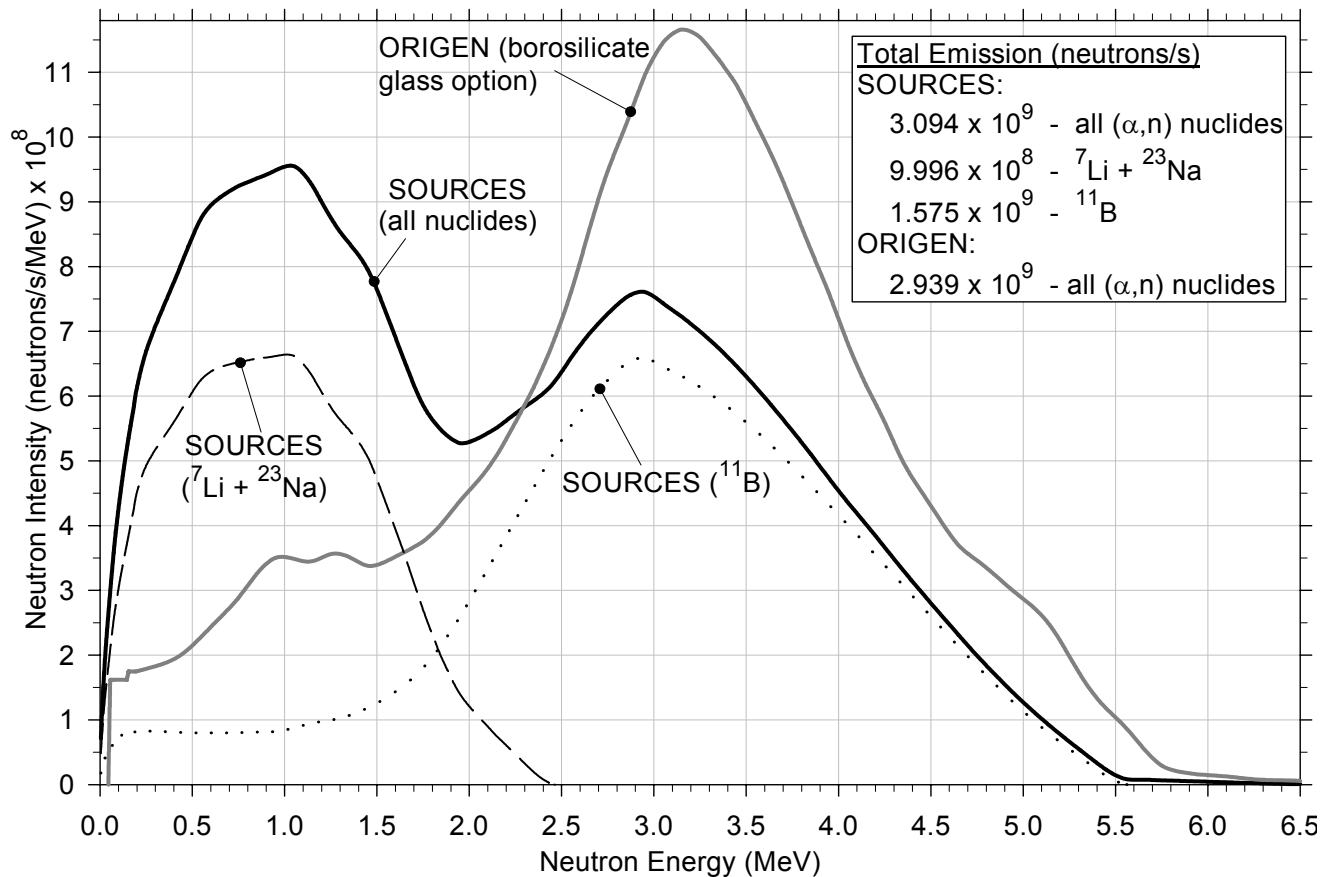


Fig. 4. (α,n) Spectra from 1 kg of ^{244}Cm Infinitely Dilute in Borosilicate Glass.

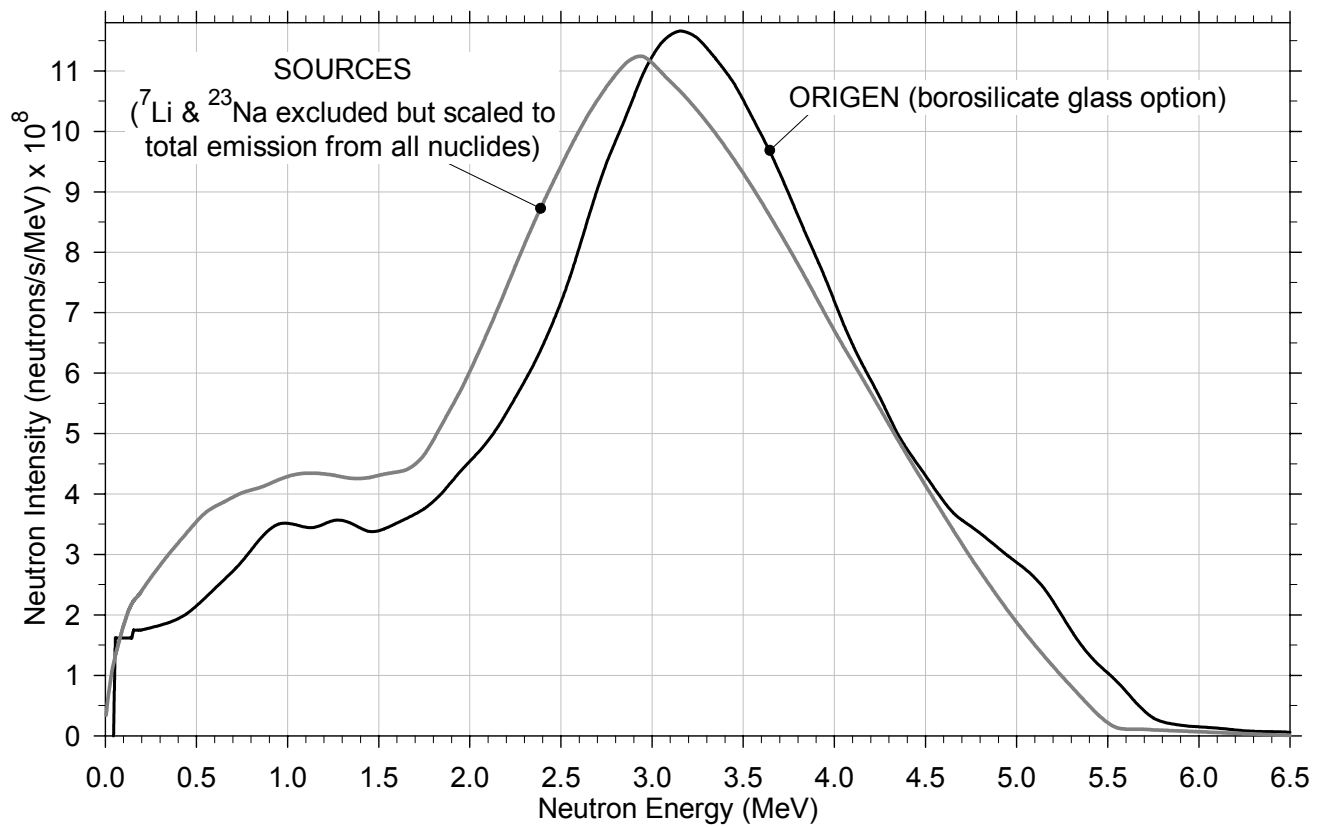


Fig. 5. (α,n) Spectra from 1 kg of ^{244}Cm in Borosilicate Glass with Li and Na Excluded from SOURCES

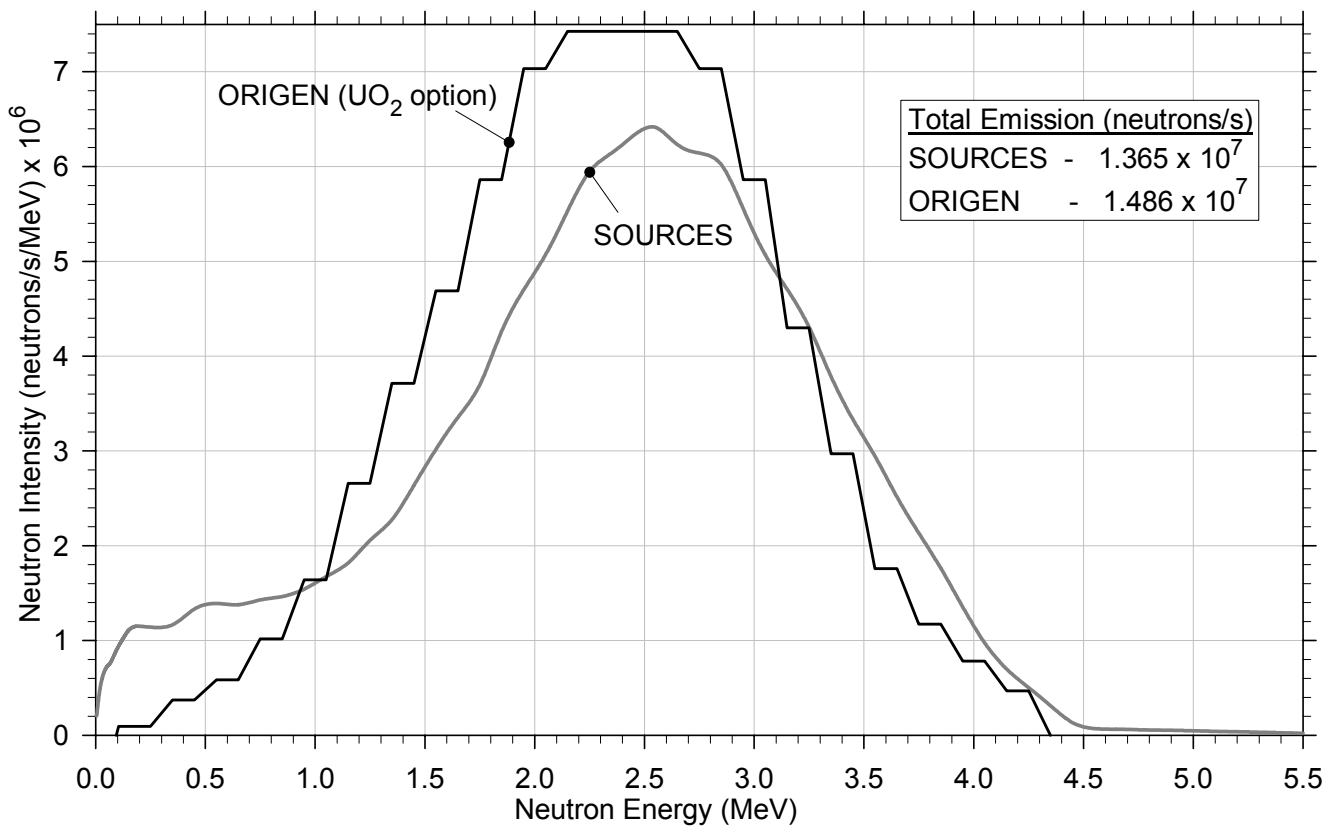


Fig. 6. (α,n) Neutron Energy Spectrum from 1 kg of ^{238}Pu Infinitely Dilute in UO_2 .

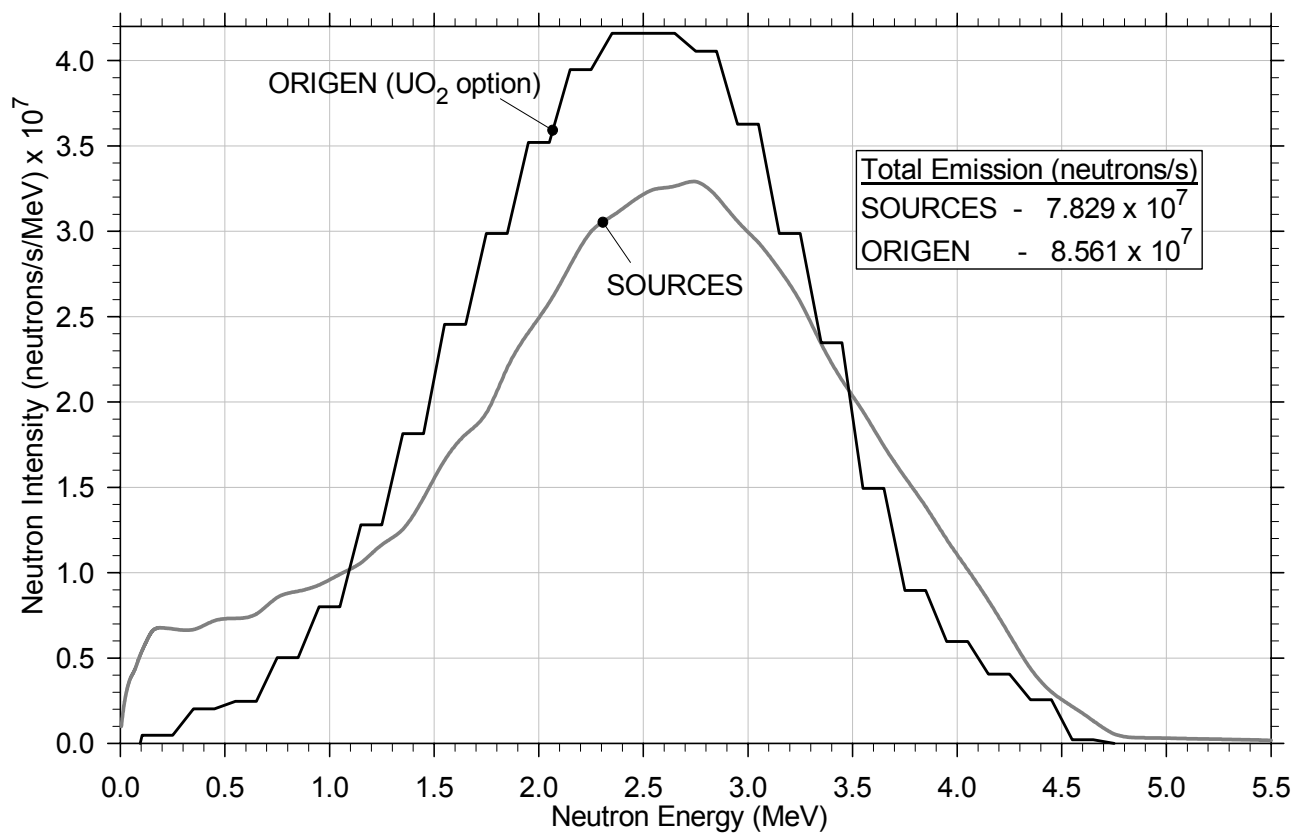


Fig. 7. (α ,n) Neutron Energy Spectrum from 1 kg of ^{244}Cm Infinitely Dilute in UO_2 .

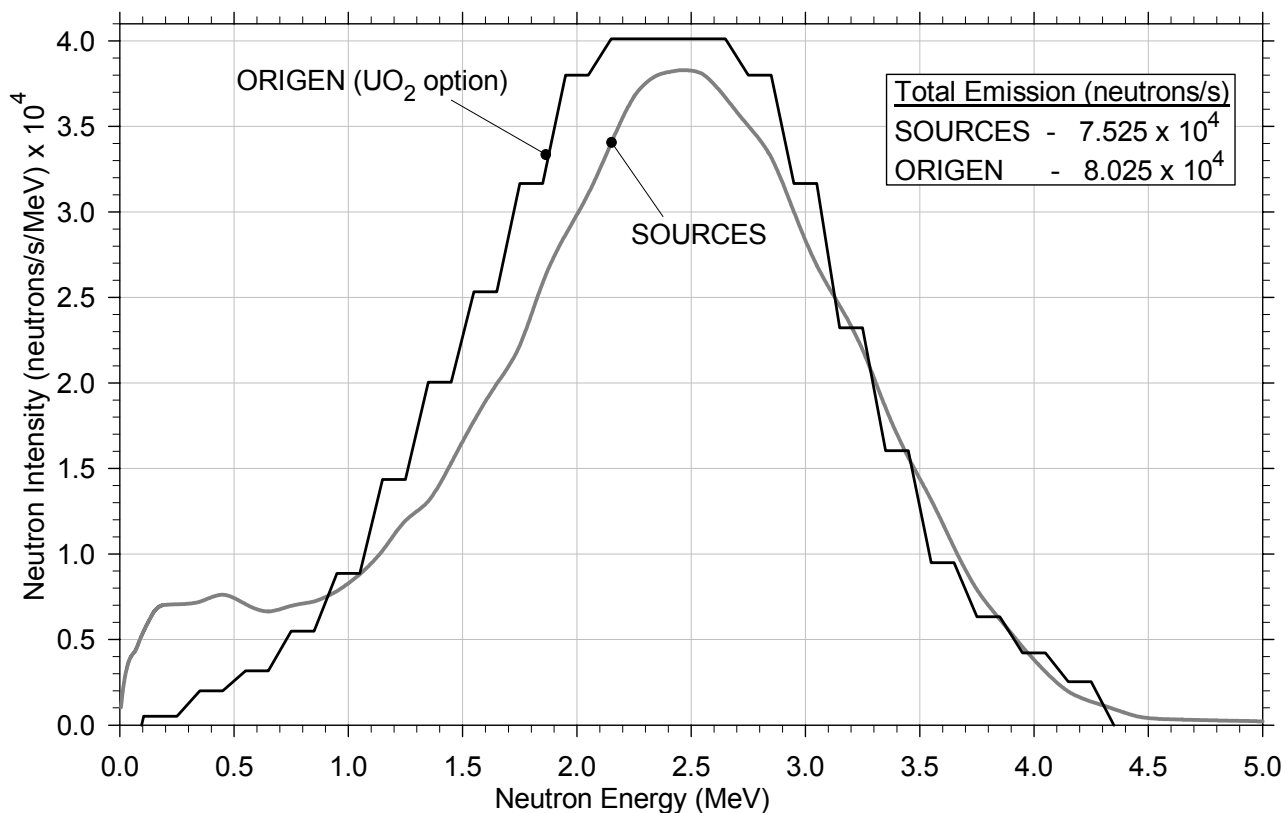


Fig. 8. (α ,n) Neutron Energy Spectrum from 1 kg of 60-y Decayed Weapons-Grade Pu as PuO_2 .