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# Sources–3A: A Code for Calculating (α,n), Spontaneous Fission, and Delayed Neutron Sources and Spectra

R. T. Perry, W. B. Wilson, W. S. Charlton

**Abstract**

SOURCES–3A is a computer code that determines neutron production rates and spectra from (α,n) reactions, spontaneous fission, and delayed neutron emission due to the decay of radionuclides in homogeneous media (i.e., a mixture of α-emitting source material and low-Z target material) and in interface problems (i.e., a slab of α-emitting source material in contact with a slab of low-Z target material). The code is also capable of calculating the neutron production rates due to (α,n) reactions induced by a monoenergetic beam of α-particles incident on a slab of target material. Spontaneous fission spectra are calculated with evaluated half-life, spontaneous fission branching, and Watt spectrum parameters for 43 actinides. The (α,n) spectra are calculated using an assumed isotropic angular distribution in the center-of-mass system with a library of 89 nuclide decay α-particle spectra, 24 sets of measured and/or evaluated (α,n) cross sections and product nuclide level branching fractions, and functional α-particle stopping cross sections for Z<106. The delayed neutron spectra are taken from an evaluated library of 105 precursors. The code outputs the magnitude and spectra of the resultant neutron source. It also provides an analysis of the contributions to that source by each nuclide in the problem.

## 1. INTRODUCTION

In many systems, it is imperative to have accurate knowledge of all significant sources of neutrons due to the decay of radionuclides. These sources can include neutrons resulting from the spontaneous fission of actinides, the interaction of actinide decay α-particles in (α,n) reactions with low- or medium-Z nuclides, and/or delayed neutrons from the fission products of actinides. Numerous systems exist in which these neutron sources could be important. These include, but are not limited to, clean and spent nuclear fuel ($UO_2$, $ThO_2$, MOX, etc.), enrichment plant operations ($UF_6$, $PuF_4$, etc.), waste tank studies, waste products in borosilicate glass or glass-ceramic mixtures, and weapons-grade plutonium in storage containers. The SOURCES–3A code was designed to calculate neutron sources (magnitude and spectra) resulting from any of the aforementioned interactions and decays.

The SOURCES–3A code is capable of calculating neutron sources in homogeneous problems (i.e., homogeneous mixtures of a-emitting and low-Z materials), interface problems (i.e., composite material consisting of two separate slab regions), or α-beam problems (i.e., a monoenergetic α-beam incident on a low-Z slab. However, systems that include combinations of these problems must be run separately and then compiled by the user.

SOURCES–3A consists of a FORTRAN 77 source code and four library files. It requires a user created input file and produces up to five output files. The SOURCES-3A manual[1] contains detailed input instructions, sample problems, benchmarking information and complete theoretical derivations. The SOURCES–3A code has been under development for several years with continuing improvements made in methods and data. The original version of SOURCES–3A (SOURCES 1x) was actually named POFEAL, which was primarily used for calculating P$_i$ Of E-ALpha [i.e., the probability of an (α,n) interaction with nuclide i by an α-particle prior to stopping in the material].[2] SOURCES 2x was an improvement of the original POFEAL code, which included spectra calculations.[3] Also, improvements in the calculational algorithm were implemented in this version. The most recent addition to the code (SOURCES–3A) was the ability to handle interface problems. SOURCES–3A will continue to be updated and improved as more experimental data and computational methods become available.

## 2. Theory

The SOURCES–3A code is capable of calculating neutron production rates for three different problem configurations (interface, homogeneous, and beam problems) with three different neutron sources: (α,n), spontaneous fission, and delayed neutrons. In the following section, the theory leading to each of these sources and problems is described. As noted, the SOURCES-3A manual contains more detailed derivations.

## 2.1 HOMOGENOUS MIXTURE PROBLEMS

A homogeneous mixture problem is one in which the α-emitting material and spontaneous fission sources are intimately mixed with the low-Z target material (i.e., atoms of α-emitting material are directly adjacent to the target atoms). Three sources of neutrons may exist in these problems, namely spontaneous fission neutrons, delayed neutrons, and neutrons emitted as a result of (α,n) reactions during the slowing down of α-particles. The theory pertaining to calculations for each of these neutron sources is described below. For homogeneous mixture problems, the neutron source (spectra and magnitude) are calculated as neutrons produced per second per unit volume. It is assumed in all homogeneous mixture calculations that the target is thick (i.e., that the dimensions of the target are much smaller than the range of the α-particles); thus, all α-particles are stopped within the mixture.

### 2.1.1 (α,n) Sources

The calculation of the (α,n) neutron production in a material requires knowledge of the stopping powers for the α-particles and the probability of neutron production, $\sigma(E)$ as a function of the α-particle at energy, $E$. Stopping powers are used here in the form of stopping cross sections, $\varepsilon(E)$. Given these two items and the slowing down material's number density, $N$, and the neutron producing material number density, $N_i$, the probability of producing a neutron, $P_i(E_\alpha)$, from an alpha particle with energy $E_\alpha$, from material $i$, can be determined by the following expression:

$$P_i(E_\alpha) = \frac{N_i}{N}\int_0^{E_\alpha}\frac{\sigma_i(E)}{\varepsilon(E)}\,dE. \tag{1}$$

In general, any material involved in a homogeneous problem will be composed of any number of different elements (e.g., H, C, and O). In these cases, the stopping cross section $\varepsilon(E)$ of a material composed of more than one elemental constituents may be calculated using the Bragg-Kleeman[4] relationship.

The value for $P_i(E_\alpha)$ is determined in the code using a discrete form of Eq. (1) for each energy alpha that results from the decay of the alpha producing material. The total neutron source is found by multiplying the total alpha source strength for each energy alpha times the respective value of $P_i(E_\alpha)$.

### 2.1.2 (α,n) Spectra

The (α,n) spectra are determined assuming an isotropic neutron angular distribution in the center-of-mass system.[5] The maximum and minimum kinetic energy of a neutron emitted from an (α,n) reaction with an alpha of specific energy may be determined by a mass, momentum and energy balance. These maximum and minimum energies are related to the Q values for the reaction and the final excitation level of the recoil nucleus.

For the neutron spectral calculation, the alpha spectra for each alpha particle is user discretized into $L$ energy groups. The number, $N_l$, of neutrons occurring from an alpha in energy group $l$, the result of an alpha of original energy $E_\alpha$, slowing down is:

$$N_l = P_i(E_{l+1}) - P_i(E_l) \tag{2}$$

where $P_i(E_l)$ was defined in Equation 1. This number, $N_l$ neutrons for each alpha group $l$, is further divided using the branching fractions associated with each energy level, $m$, into $N_{l,m}$ neutrons. There is a maximum and minimum neutron energy associated with each set composed of energy group, $l$, and energy level, $m$, and neutron producing material, $i$. These $N_{l,m}$ neutrons are evenly divided, using a user defined energy structure between the maximum and minimum neutron energy for this alpha energy group and level. The maximum and minimum neutron energy is calculated at the midpoint of the alpha energy group. The product nuclide levels, the number of product level branching data points, the ($\alpha$,n) reaction Q-values, the excitation energy of each product nuclide level, and the branching fraction of ($\alpha$,n) reactions resulting in the production of product levels are available from the library.

The process is repeated for each source alpha in the problem. The number of neutrons in each neutron energy group for each alpha particle is then summed resulting in the neutron spectra.

## 2.2 SPONTANEOUS FISSION SOURCES AND SPECTRA

The spontaneous fission (SF) of an actinide nuclide $k$ is accompanied by the emission of an average $\nu_k$(SF) neutrons. The fraction of nuclide $k$ decays that are spontaneous fission events are given by the SF branching fraction:

$$F_k^{SF} = \frac{\lambda_k^{SF}}{\lambda_k}. \tag{3}$$

Here $\lambda_k^{SF}$ and $\lambda_k$ are the SF and total decay constant respectively.

Thus, the average number of SF neutrons emitted per decay of nuclide k (by any mode) is:

$$R_k(SF) = F_k^{SF} \nu_k(SF). \tag{4}$$

Therefore, to compute the neutron production due to spontaneous fission per decay of nuclide $k$, the SF branching fraction and average number of neutrons per spontaneous fission must be known. These quantities are available to SOURCES–3A from a library file.

The spontaneous fission neutron spectra are approximated by a Watt's fission spectra using two evaluated parameters (a and b):

$$\chi_k^{SF}(E) = R_k(SF) e^{-E/a} \sinh\sqrt{bE}\,. \tag{5}$$

Evaluated parameters are provided for 43 fissioning nuclides in a library file.

## 3.3 DELAYED NEUTRON SOURCES

During the fissioning process, a number of products are formed including neutrons, gamma rays, beta rays, neutrinos, fission products, and an appreciable amount of energy. Some of the fission products formed as a result of fission can decay by $\beta^-$ emission to a highly excited state, which can then decay by emitting a neutron. These neutrons are called "delayed neutrons" because they appear within the system with some appreciable time delay. The nuclide emitting the neutron is referred to as the "delayed neutron emitter," and the nuclide, which $\beta^-$ decays to the emitter is referred to as a "delayed neutron precursor." It is customary to assume that one neutron is emitted per decay and that the emitter decays almost instantaneously. The fraction of decays by fission product nuclide $k$ (by any mode) leading to the emission of a delayed neutron is given by the DN branching fraction, $F_k^{DN}$, and the number of neutrons per decay is:

$$R_k(DN) = F_k^{DN}. \tag{6}$$

The values for $F_k^{DN}$ is provided to SOURCES–3A in a library file.

A series of evaluated delayed neutron spectra are provided in a library file for 105 precursor nuclides. These evaluated spectra are provided in a discrete form. They are read directly into SOURCES–3A and then adjusted so that the default spectra energy mesh correlates with the user desired energy mesh. They are then normalized to the source strength.

## 2.3 BEAM PROBLEMS

A beam problem is one in which a monoenergetic α-beam is incident upon a slab containing low-Z target material. The slab could also contain higher mass isotopes; however, actinides (i.e., α-emitting or spontaneous fissioning material) will not be used to calculate a source. It is a necessary condition that the thickness of the slab of target material be significantly larger than the range of the α-particles in the beam (i.e., that all a-particles come to rest within the target slab).

The neutron production rate within the slab per incident α-particle is a function of the α-particle beam energy ($E_\alpha$) and the probability of an (α,n) interaction with any nuclide $i$ within the slab by an α-particle from the beam prior to stopping in the material. The thick-target neutron production function, $P_i(E_\alpha)$, is calculated using Equation 1. The beam energy, $E_\alpha$, must be supplied by the user. The neutron spectra are calculated using the same procedure as described in 2.1.2 above.

## 2.4 INTERFACE PROBLEMS

Interface problems exist when a slab (Region I) of α-emitting material is in close contact with a low-Z target material (Region II.) In these problems, α-particles are emitted from the Region I materials and travel across the interface junction into the Region II materials. In Region II, the α-particles can interact through (α,n) reactions and generate a neutron source. It is necessary to assume that in all interface problems the thickness of each region is significantly larger than the range of the α-particles within it.

Given a uniform volumetric source of α-particles in Region I, where $\lambda_k$ is the decay constant for source nuclide $k$, $N_k$ is the atom density of source nuclide $k$, and $f^{\alpha}_{kl}$ is the fraction of all decays of nuclide $k$ resulting in an α-particle of energy $E_\alpha$, the following equation may be derived:

$$\Phi^g = \frac{\lambda_k f^{\alpha}_{kl}}{4} \frac{N_k}{N} \int_{E_g}^{E_{g+1}} \frac{dE}{\varepsilon(E)} . \tag{7}$$

The quantity, $\Phi^g$, is the number of α-particles between energies $E_g$ and $E_{g+1}$ which pass into the low-Z target material (Region II) per unit area and per unit time from decay α-particles with energy $E_\alpha$. The quantity $\Phi^g$ is then used by SOURCES–3A as the source strength of a monoenergetic beam with energy at the midpoint of $E_g$ *and* $E_{g+1}$.

SOURCES–3A can then use the same procedure developed in Section 2.3 to solve for the neutron production rate due to the α-particles crossing the junction with energies between $E_g$ and $E_{g+1}$. SOURCES–3A then repeats this procedure for all α-particle energies and all source nuclides.

# 3. DATA

The libraries contain 89 nuclide decay α-particle spectra, 24 sets of evaluated (α,n) cross sections and product nuclide level branching fractions. The recommended cross sections sets are listed in Table 1. The library contains additional data. A maximum α-particle energy of 6.5 MeV is allowed by SOURCES–3A. This restriction is required because of the limitations of the cross section libraries.

**Table 1. The (α,n) target isotopes available in SOURCES-3A**

| Isotope | Level Branching Fraction Data Source | Cross Section Data Source |
|---|---|---|
| $^{7}Li$ | GNASH | Gibbons and Macklin[6] |
| $^{9}Be$ | Geiger and Van der Zwan[7] | Geiger and Van der Zwan[7] |
| $^{10}B$ | GNASH | Bair *et al.*[8] |
| $^{11}B$ | GNASH | Bair *et al.*[8] |
| $^{13}C$ | GNASH[a] | Bair and Haas[9] |
| $^{14}N$ | N/A[b] | GNASH |
| $^{17}O$ | Lesser and Schenter[10] | Perry and Wilson[2] |
| $^{18}O$ | Lesser and Schenter[10] | Perry and Wilson[2] |
| $^{19}F$ | Lesser and Schenter[10] | Norman *et al.*[11] |
| $^{21}Ne$ | N/A[b] | GNASH |
| $^{22}Ne$ | N/A[b] | GNASH |
| $^{23}Na$ | GNASH | GNASH[a] |
| $^{25}Mg$ | GNASH | GNASH |
| $^{26}Mg$ | GNASH | GNASH |
| $^{27}Al$ | GNASH | GNASH[a] |
| $^{29}Si$ | GNASH | GNASH[a] |
| $^{30}Si$ | GNASH | GNASH[a] |
| $^{31}P$ | GNASH | GNASH |
| $^{37}Cl$ | GNASH | Woosley *et al.*[12] |

[a] GNASH[13] calculated and experimental data (in that order) are available for these nuclides in the library file. By default, the GNASH calculation is used (actually SOURCES-3A uses the first dataset that it encounters during the reading of the file). To use an alternate dataset, the library file must be altered by reversing the order in which these datasets occur in the file.

[b] Nuclide level branching data for these isotopes are absent from the library files. Thus problems containing these isotopes can be executed only for neutron source magnitudes and not for neutron source spectra .

Stopping-power coefficients (which are a function of atomic number only) are included for all elemental constituents with $Z \leq 105$. The data by Ziegler *et al.*[14] was used for all $Z \leq 92$. The stopping power coefficients calculated by Perry and Wilson[2] were used for $92 < Z \leq 105$.

The spontaneous fission spectra are calculated with evaluated half-life, spontaneous fission branching, and ν data using Watt spectrum parameters for 43 actinides. The delayed neutron sources are calculated from a library of evaluated delayed neutron branching fractions and half-lives for 105 precursors.

## 4. SAMPLE RESULTS / BENCHMARKS

The SOURCES code has been extensively benchmarked against experimental data. Details of the results may be found in the manual. In this section, two examples of benchmark calculations will be given. The first example is a yield and spectral calculation. The second is an example of thick target yields as a function of initial alpha energy.

## 4.1 SPECTRAL CALCULATION

This problem illustrates the neutron source magnitudes and spectra from a $PuBe_{13}$ source (elemental constituents are 13/14 Be and 1/14 Pu) with six isotopes of Pu ($^{237}$Pu, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, and $^{242}$Pu) and one isotope of Be ($^{9}$Be) present, which is an appropriate model of the experimental measurement performed by L. Stewart.[15]

A comparison of the data measured by the experimenters and the SOURCES-3A calculation is presented in Fig. 1. To construct this plot, the histogram output from SOURCES-3A is plotted using only the midpoint energy for each energy group. The line is simply a guide to the eye. From Fig. 1, reasonable agreement between the SOURCES-3A spectrum calculation and the measured values is found. The total neutron source magnitude calculated by SOURCES-3A was 2.69 x $10^5$ neutrons/s-cm$^3$, whereas the experimenters reported a total neutron source rate of 2.28 x $10^5$ neutrons/s-cm$^3$. This magnitude of agreement (±17%) is standard for a SOURCES-3A calculation. The calculation neglected any source contaminants (especially $^{241}$Am) because they were not specified in the published experiment.

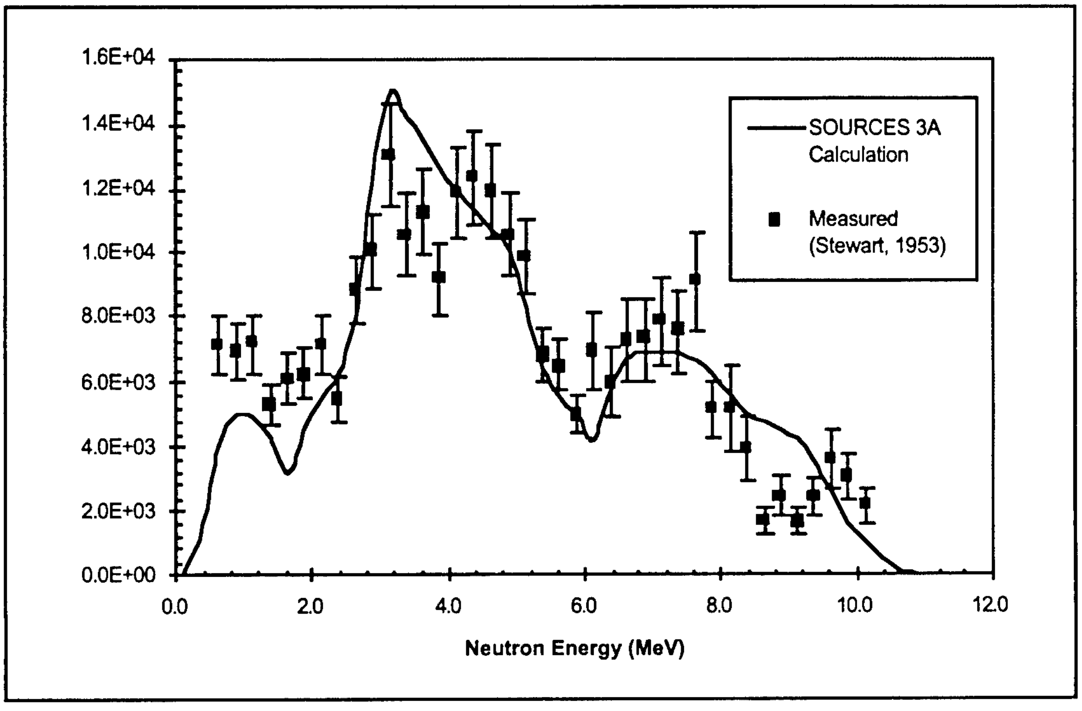


Fig. 1. Energy-dependent neutron source strength in $PuBe_{13}$ homogeneous problem as calculated by SOURCES-3A and compared with measured data.

## 4.2 THICK TARGET YIELD CALCULATION

In this section, calculational thigh target yield results for carbon are compared with experiment data. The experimental data was taken from References 8, 16, 17, 18, and 19. The results are presented in Figure 2. Thick target calculations for the following materials appear in the manual: Natural Lithium Target, $^{9}$Be Target, Natural Boron Target, Natural Carbon Target, Natural Oxygen Target, Fluorine Target, Natural Magnesium Target, Natural Aluminum Target, Natural Silicon Target, Uranium Dioxide, and Uranium Carbide. In general, the comparison of calculational results with experiment of thick target yields are within 10-20% of each other.

# 5. CONCLUSION

SOURCES-3A is an extremely versatile code for calculating ($\alpha$,n), spontaneous fission, and delayed neutron sources and spectra. It has been extensively benchmarked. The libraries contain data for most problems of interest.

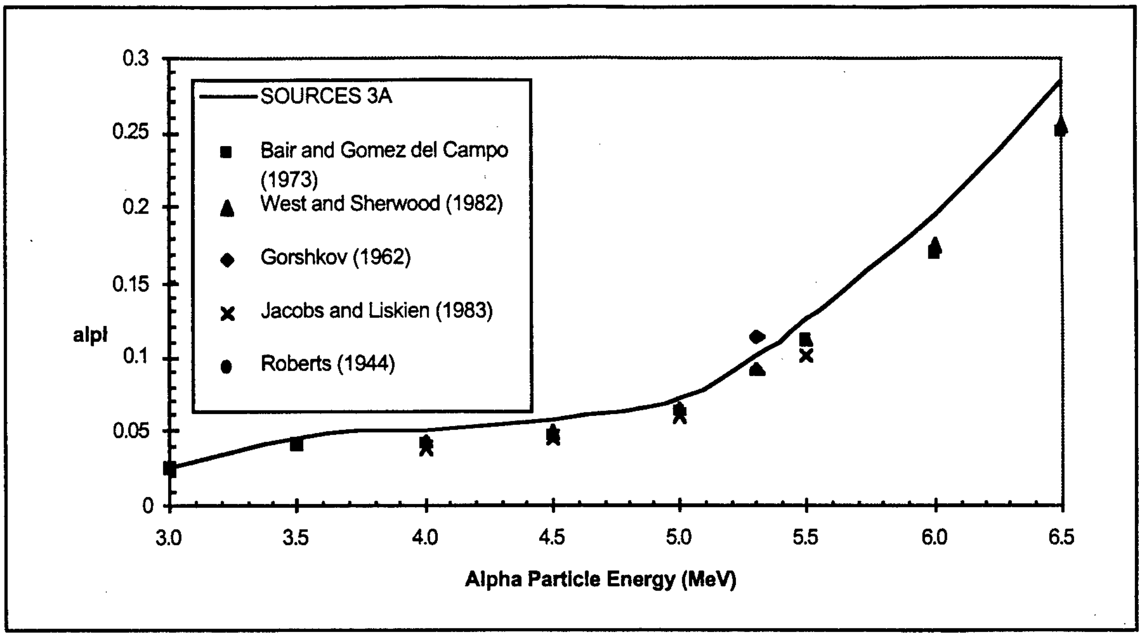


Fig. 2. Energy-dependent thick-target yields as calculated by SOURCES–3A and compared to measured data for carbon.

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