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Title: COMPARISON OF (ALPHA, N) THICK-TARGET NEUTRON YIELDS AND SPECTRA FROM ORIGEN-S AND SOURCES

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Comparison of (α,n) Thick-Target Neutron Yields and Spectra from ORIGEN-S and SOURCES

Introduction

Both ORIGEN-S[1] 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 UO_2. 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]

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 by[3]

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

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:

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

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 relationship[4] of additivity of stopping cross sections of elements as separate constituents to calculate material stopping cross section, ε_m, i.e.

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

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

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 ORIGEN-S, Eq. (1) has been approximated as[1,7]

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

where $\overline{E_k^o} = \sum_{j=1}^{J_k} g_{jk} E_{jk}^o$ 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 $E \leq \overline{E_k^o}$ for which (α,n) reactions occur for α particles from nuclide k, $y_{ik}(\overline{E_k^o})$ 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 assumptions 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 assumptions taken together constitute the West approximation.[8] Because these assumptions 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.

Comparison of Neutron Energy Spectra

SOURCES calculates spectra from fundamental physics formulations and data. The α slowing-down energy range is divided into energy bins. The neutron production for each α energy bin is determined for each product nuclide energy level and then divided evenly into

neutron energy bins between the maximum and minimum neutron energies for that level. The total neutron spectrum is formed by summing all contributions from all targets and product nuclide levels, each integrated over the α energy range. The details and basis of this procedure are described elsewhere.[2,3,9,10]

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]

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

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 are ^{238}Pu, ^{241}Am, ^{242}Cm, or ^{244}Cm. 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 by their atom densities in borosilicate glass.

Even though Li and Na are significant target elements in the borosilicate-glass composition[1] used to determine the $\{\varphi_{kg}\}$, they were omitted in the determination of the $\{\varphi_{kg}\}$. The significance of their spectral contributions to the $\{\varphi_{kg}\}$ is evident from comparing the ORIGEN-S and SOURCES spectra shown in Fig. 1 for the borosilicate glass. 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.

Conclusions

Yields generated with the ORIGEN-S borosilicate glass option are expected to be sufficiently accurate for most material compositions, but the spectra generated are unacceptable for following three reasons: (1) no basis for the method of calculating material spectra from precalculated borosilicate glass spectra, (2) no prescription provided of 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.

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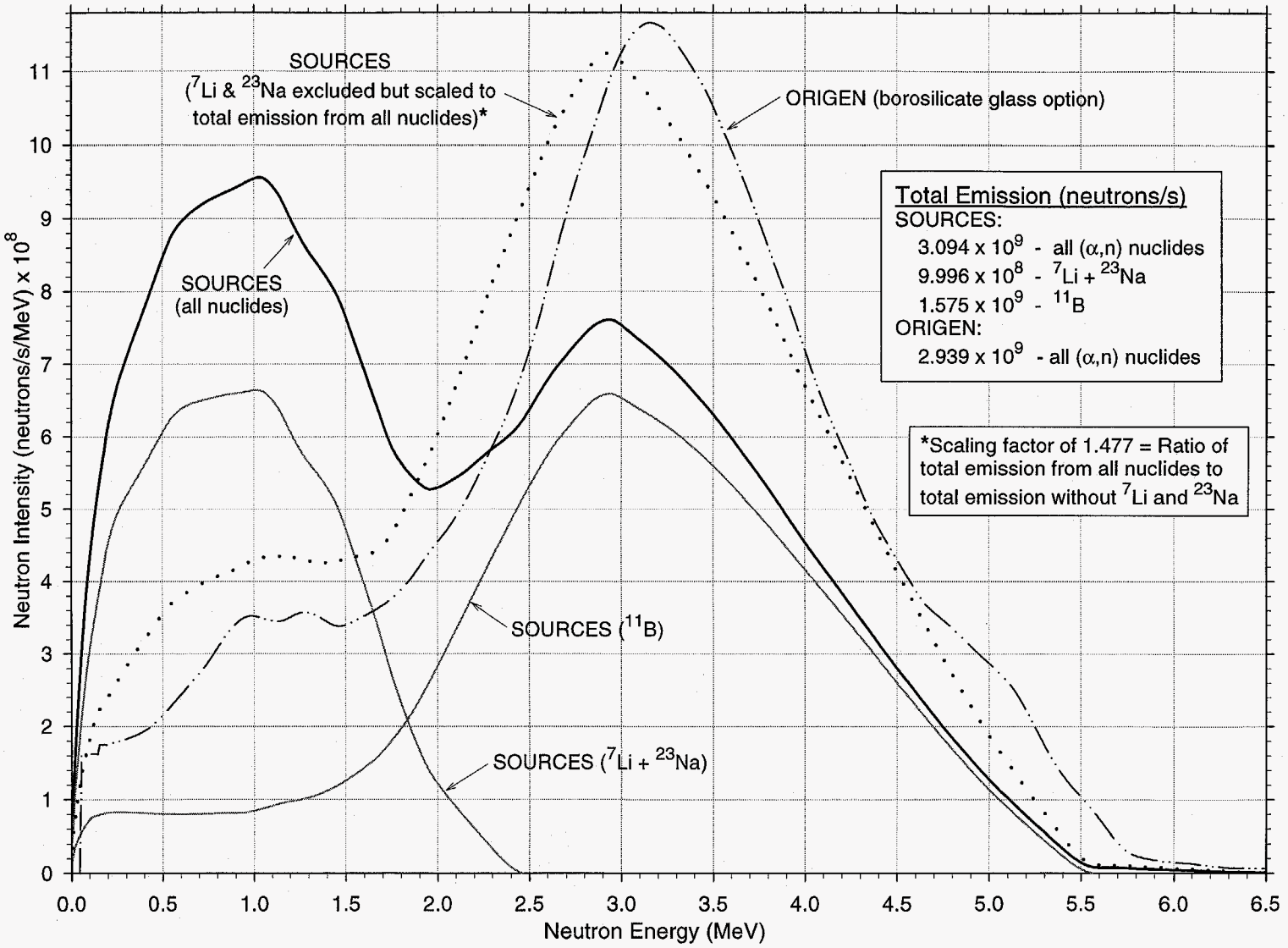

Fig. 1. (α,n) Neutron Spectrum from 1 kg of ^{244}Cm Infinitely Dilute in Borosilicate Glass