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SENSITIVITY OF A RESPONSE TO THE COMPOSITION OF AN (α,n) NEUTRON SOURCE

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We derive the sensitivity of a response to the composition of a homogeneously mixed (α,n) neutron source. We use the mathematical descriptions of the (α,n) neutron source rate density and spectrum that are used by SOURCES4C; specifically, a material's α-particle stopping power is given by the Bragg-Kleeman relationship. The (α,n) neutron source is computed for each combination of α particle emitter (source isotope) and (α,n) target isotope. In addition, each isotope in the material has an effect on the (α,n) neutron source because of its contribution to the α particle stopping power. The derivative of the total (α,n) source rate density with respect to each isotope density is exact, but the derivative of the energy spectrum is approximated using an appropriate spectrum. Numerical results using the total neutron leakage from the BeRP ball demonstrate the correctness and accuracy of the sensitivities.

I. INTRODUCTION

The theory for adjoint-based sensitivities of responses to nuclear data in shielding problems is well established. We are also interested in the sensitivities of responses with respect to material composition[1] (i.e., isotopic weight fractions or atom densities). In this paper, we derive the sensitivity of a response to the composition of an (α,n) neutron source. We use the mathematical descriptions of the (α,n) neutron source rate density and energy spectrum that are used by SOURCES4C and described in the SOURCES4C manual;[2] specifically, a material's α-particle stopping power is given by the Bragg-Kleeman relationship.[3]

This paper addresses the (α,n) neutron source generated in a homogeneously mixed material. This paper does not account for the sensitivity of the (α,n) neutron source strength or spectrum to the crystal form of the material. The sensitivity to (α,n) cross sections or stopping powers[4] is also not addressed.

This paper is organized as follows. Section II summarizes the equations for an (α,n) neutron source that are implemented in SOURCES4C. In Sec. III, we derive the sensitivity of the (α,n) neutron source strength and spectrum to the isotopic density of one of the material constituents. In Sec. IV, we derive the sensitivity of the (α,n) neutron source strength and spectrum to the material atom density. In Sec. V, we propagate the sensitivities to the sensitivity of the response. Section VI discusses the modifications made to SOURCES4C that facilitate calculating the sensitivity in post-processing. Section VII contains a test problem. Section VIII is a summary and conclusions.

II. COMPUTATION OF AN (α,n) NEUTRON SOURCE

This section describes how SOURCES4C computes the (α,n) source for a homogeneously mixed material. (There are other methods available.[5]) This section generally follows the SOURCES4C manual.[2]

The stopping cross section ε of a material for α particles of energy E is defined as

$$\varepsilon(E) = -\frac{1}{N}\frac{dE}{dx}, \tag{1}$$

where N is the total atom density of the material and x is the path length of the α particle through the material. The stopping cross section can be calculated using the Bragg-Kleeman relationship[3]

$$\varepsilon(E) \cong \frac{1}{N}\sum_{j=1}^{J} N_j \varepsilon_j(E), \tag{2}$$

where $\varepsilon_j(E)$ and N_j are the stopping power and atom density of isotope j, respectively, and

$$N = \sum_{j=1}^{J} N_j. \tag{3}$$

Stopping powers depend on the element, not the isotope, so all isotopes of an element have the same stopping power.

The probability of an α particle of energy E_α undergoing an (α,n) reaction with nuclide i before stopping can be represented by the function

$$P_i(E_\alpha) = \frac{N_i}{N}\int_0^{E_\alpha} \frac{\sigma_i(E)}{\varepsilon(E)} dE. \tag{4}$$

In SOURCES4C, Eq. (4) is approximated as

$$P_i(E_\alpha) = \frac{N_i}{N}\sum_{g=1}^{G_\alpha} \frac{1}{2}\left[\frac{\sigma_{i,g+1}}{\varepsilon_{g+1}} + \frac{\sigma_{i,g}}{\varepsilon_g}\right](E_{g+1} - E_g), \tag{5}$$

where G_α, the number of α-particle energy bins, is a user input. The number of quadrature points in the numerical integral is, of course, $G_\alpha + 1$. For each α-emitting isotope in the material, the α-particle energy range is the largest discrete α-particle energy emitted minus the minimum allowable α-particle energy. This range is divided by G_α to determine the α-particle energy group structure. Therefore, $E_{g+1} - E_g$ is the same for each energy bin and Eq. (5) is a simple trapezoid rule. For smaller energies (for each α-emitting isotope), the code computes P_i by interpolating on the group structure resulting from the maximum α-particle energy and the number of bins. The multigroup form of Eq. (2) is used:

$$\varepsilon_g = \frac{1}{N}\sum_{j=1}^{J} N_j \varepsilon_{j,g}. \tag{6}$$

In Eqs. (5) and (6), g indexes the α-particle energy discretization used to evaluate Eq. (4).

The fraction of decays of nuclide k resulting in an (α,n) reaction in isotope i is

$$R_{k,i}(\alpha,n) = \sum_{l=1}^{L} f_{kl}^{\alpha} P_i(E_l), \tag{7}$$

where f_{kl}^{α} is the fraction of all decays of nuclide k resulting in an α particle of energy E_l and L is the number of discrete α particle energies emitted by nuclide k. The quantities L, f_{kl}^{α}, and E_l (l = 1,…,L) are given in the nuclear data for each α particle emitter (source isotope) k (specifically, these are given in SOURCES4C's "tape5" data file[2]).

The total neutron source rate density due to target i and α source k is

$$\begin{aligned} Q_{(\alpha,n),k,i} &= \lambda_k N_k R_{k,i}(\alpha,n) \\ &= \lambda_k N_k \sum_{l=1}^{L} f_{kl}^{\alpha} P_i(E_l), \end{aligned} \tag{8}$$

where λ_k is the decay constant for nuclide k. Note that P_i contains the density of every nuclide [see Eqs. (5), (3), and (2)]. The total (α,n) neutron source rate density is the sum of $Q_{(\alpha,n),k,i}$ over all targets and sources:

$$Q_{(\alpha,n)} = \sum_k \sum_i Q_{(\alpha,n),k,i}. \tag{9}$$

We now turn to the calculation of the (α,n) neutron energy spectrum. This section differs slightly from the discussion in Ref. 2. The fraction of α particles from source isotope k reacting with target nuclide i resulting in product level m reactions occurring in α-particle energy group g is

$$F_{k,i,g}(m) = S_{i,g}(m) H_{i,g,l}, \tag{10}$$

where $H_{i,g,l}$ is the fraction of target i reactions of α particles of emission energy l in α-particle energy group g, equal to*

$$H_{i,g,l} = \frac{P_i(E_{g+1}) - P_i(E_g)}{P_i(E_l)}, \tag{11}$$

and $S_{i,g}(m)$ is the branching fraction to product level m of α particles of energy E_g, calculated using the interpolation

$$\begin{aligned} S_{i,g}(m) = f_i(m,m'-1) + \left(f_i(m,m') - f_i(m,m'-1)\right) \\ \times \frac{E_g - E(m'-1)}{E(m') - E(m'-1)}, \end{aligned} \tag{12}$$

where $f_i(m,m')$ is the fraction of (α,n) reactions with target i at energy $E(m')$ resulting in the production of product level m. The energy levels and fractions used in Eq. (12) are nuclear data for each target nuclide and do not depend on densities. [When the code computes Eq. (12) for each of the α-particle energies in the G_α-group structure, E_g is actually the midpoint each energy group rather than the endpoint.]

The (α,n) neutron energy spectrum $\chi_{k,i}^{(\alpha,n)}(E^g)$ is†

$$\begin{aligned} \chi_{k,i}^{(\alpha,n)}(E^g) = R_{k,i}(\alpha,n) \\ \times \sum_{l=1}^{L}\sum_{m=1}^{M}\sum_{g_\alpha=1}^{G_\alpha} F_{k,i,g_\alpha}(m) \frac{E^{g+1} - E^g}{E_{n,m}^{+}(E_{g_\alpha}) - E_{n,m}^{-}(E_{g_\alpha})}, \end{aligned} \tag{13}$$

where $E_{n,m}^{-}(E_{g_\alpha})$ and $E_{n,m}^{+}(E_{g_\alpha})$ are the minimum and maximum permissible neutron kinetic energies from an incident α particle of energy E_{g_α} that generates a product nuclide with level m, M is the number of product levels for target k, and the neutron group boundaries E^{g+1} and E^g are between $E_{n,m}^{-}(E_{g_\alpha})$ and $E_{n,m}^{+}(E_{g_\alpha})$. It is assumed that the neutrons are isotropically emitted and contribute uniformly to all groups between $E_{n,m}^{-}(E_{g_\alpha})$ and $E_{n,m}^{+}(E_{g_\alpha})$. Define

$$\Delta E_{m,g_\alpha}^{g} \equiv \frac{E^{g+1} - E^g}{E_{n,m}^{+}(E_{g_\alpha}) - E_{n,m}^{-}(E_{g_\alpha})}. \tag{14}$$

The total neutron source rate density in energy group g due to α emitter k and target i is

* Equation (32) in Ref. 2 indicates that H is a function of the product level m, but in the code, H is calculated for each of the G_α + 1 energy bin boundaries in the group structure and does not depend on the product level.

† Equation (35) in Ref. 2 lacks the sums over discrete α energies l, product levels m, and α energy groups g_α.

$$Q^g_{(\alpha,n),k,i} = \lambda_k N_k \chi^{(\alpha,n)}_{k,i}(E^g) = Q_{(\alpha,n),k,i} \sum_{l=1}^{L}\sum_{m=1}^{M}\sum_{g_\alpha=1}^{G_\alpha} F_{k,i,g_\alpha}(m)\Delta E^g_{m,g_\alpha} \quad (15)$$

[using Eq. (8)]. In Eqs. (13), (14), and (15), g indexes neutron energy groups and g_α indexes α energy groups.

The total neutron source rate density in energy group g due to α emitter k is the sum of $Q^g_{(\alpha,n),k,i}$ over targets; the total neutron source rate density in energy group g due to target i is the sum of $Q^g_{(\alpha,n),k,i}$ over α emitters. The total (α,n) neutron source rate density in energy group g, $Q^g_{(\alpha,n)}$, is the sum of $Q^g_{(\alpha,n),k,i}$ over α emitters and targets. The total (α,n) neutron source rate density $Q_{(\alpha,n)}$ is the sum of $Q^g_{(\alpha,n)}$ over neutron energy groups.

For comparison, the total neutron source rate density due to the spontaneous fission of isotope k is

$$Q_{s.f.,k} = \lambda_k N_k R_k(s.f.), \quad (16)$$

where $R_k(s.f.)$ is the average number of spontaneous-fission neutrons produced per decay of nuclide k. The total neutron source rate density in energy group g due to the spontaneous fission of isotope k is

$$Q^g_{s.f.,k} = \lambda_k N_k \chi^{s.f.}_k(E^g), \quad (17)$$

where the spontaneous-fission spectrum $\chi^{s.f.}_k(E^g)$ is a Watt spectrum normalized to $R_k(s.f.)$.

III. DERIVATIVE OF THE (α,n) SOURCE RATE DENSITY WITH RESPECT TO ISOTOPIC DENSITIES

From Eqs. (6) and (3), the derivative of $1/\varepsilon_g$ with respect to N_j is

$$\frac{\partial(1/\varepsilon_g)}{\partial N_j} = \frac{\partial}{\partial N_j}\left(\frac{\sum_{j=1}^{J} N_j}{\sum_{j=1}^{J} N_j \varepsilon_{j,g}}\right) = \frac{\varepsilon_g - \varepsilon_{j,g}}{N\varepsilon_g^2}. \quad (18)$$

From Eq. (5), and using Eq. (18), the derivative of $P_i(E_l)$ with respect to N_j is

$$\begin{aligned}\frac{\partial P_i(E_l)}{\partial N_j} &= \frac{N_i}{N}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\left(\frac{\partial}{\partial N_j}\frac{\sigma_{i,g+1}}{\varepsilon_{g+1}}\right)+\left(\frac{\partial}{\partial N_j}\frac{\sigma_{i,g}}{\varepsilon_g}\right)\right]\left(E_{g+1}-E_g\right) \\ &\quad+\left(\frac{\partial}{\partial N_j}\frac{N_i}{N}\right)\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}}{\varepsilon_{g+1}}+\frac{\sigma_{i,g}}{\varepsilon_g}\right]\left(E_{g+1}-E_g\right) \\ &= \frac{N_i}{N^2}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}(\varepsilon_{g+1}-\varepsilon_{j,g+1})}{\varepsilon_{g+1}^2}+\frac{\sigma_{i,g}(\varepsilon_g-\varepsilon_{j,g})}{\varepsilon_g^2}\right] \\ &\quad\times\left(E_{g+1}-E_g\right)+P_i(E_l)\left[\frac{\delta_{ji}}{N_i}-\frac{1}{N}\right].\end{aligned} \quad (19)$$

Equation (19) can be reduced to

$$\begin{aligned}\frac{\partial P_i(E_l)}{\partial N_j} &= \frac{N_i}{N^2}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}}{\varepsilon_{g+1}}+\frac{\sigma_{i,g}}{\varepsilon_g}\right]\left(E_{g+1}-E_g\right) \\ &\quad-\frac{N_i}{N^2}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}}{\varepsilon_{g+1}^2}+\frac{\sigma_{i,g}\varepsilon_{j,g}}{\varepsilon_g^2}\right]\left(E_{g+1}-E_g\right) \\ &\quad+P_i(E_l)\left[\frac{\delta_{ji}}{N_i}-\frac{1}{N}\right]\end{aligned} \quad (20)$$

or, finally, identifying $P_i(E_l)$ of Eq. (5) in the first term on the right side of Eq. (20),

$$\begin{aligned}\frac{\partial P_i(E_l)}{\partial N_j} &= \frac{P_i(E_l)}{N_i}\delta_{ji} \\ &\quad-\frac{N_i}{N^2}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}}{\varepsilon_{g+1}^2}+\frac{\sigma_{i,g}\varepsilon_{j,g}}{\varepsilon_g^2}\right]\left(E_{g+1}-E_g\right).\end{aligned} \quad (21)$$

Define the component of $\partial P_i(E_l)/\partial N_j$ accounting for the contribution of isotope j to the stopping power as

$$P^{(1)}_{i,j}(E_l) \equiv \frac{N_i}{N}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}}{\varepsilon_{g+1}^2}+\frac{\sigma_{i,g}\varepsilon_{j,g}}{\varepsilon_g^2}\right]\left(E_{g+1}-E_g\right) \quad (22)$$

so that Eq. (21) becomes

$$\frac{\partial P_i(E_l)}{\partial N_j} = \frac{P_i(E_l)}{N_i}\delta_{ji} - \frac{P^{(1)}_{i,j}(E_l)}{N}. \quad (23)$$

Using Eq. (6), it can be shown that the relationship between $P_i(E_l)$ of Eq. (5) and $P^{(1)}_{i,j}(E_l)$ of Eq. (22) is

$$\sum_j \frac{N_j}{N} P^{(1)}_{i,j}(E_l) = P_i(E_l). \quad (24)$$

From Eq. (7), the derivative of $R_{k,i}(\alpha,n)$ with respect to N_j is

$$\frac{\partial R_{k,i}}{\partial N_j} = \sum_{l=1}^{L} f^\alpha_{kl}\frac{\partial P_i(E_l)}{\partial N_j}. \quad (25)$$

From Eq. (8), the derivative of $Q_{(\alpha,n),k,i}$ with respect to N_j is

$$\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} = \lambda_k N_k \frac{\partial R_{k,i}}{\partial N_j} + \delta_{jk}\lambda_k R_{k,i}(\alpha,n). \tag{26}$$

Applying Eqs. (25) and (7) to Eq. (26) yields

$$\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} = \lambda_k N_k \sum_{l=1}^{L} f_{kl}^{\alpha} \frac{\partial P_i(E_l)}{\partial N_j} + \delta_{jk}\lambda_k \sum_{l=1}^{L} f_{kl}^{\alpha} P_i(E_l). \tag{27}$$

Using Eq. (23), Eq. (27) becomes

$$\begin{aligned}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} &= \delta_{ji}\lambda_k \frac{N_k}{N_i} \sum_{l=1}^{L} f_{kl}^{\alpha} P_i(E_l) + \delta_{jk}\lambda_k \sum_{l=1}^{L} f_{kl}^{\alpha} P_i(E_l) \\ &\quad -\lambda_k \frac{N_k}{N} \sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l).\end{aligned} \tag{28}$$

Finally, using Eq. (8), Eq. (28) becomes

$$\begin{aligned}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} &= \delta_{ji} \frac{Q_{(\alpha,n),k,i}}{N_i} + \delta_{jk} \frac{Q_{(\alpha,n),k,i}}{N_k} \\ &\quad -\lambda_k \frac{N_k}{N} \sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l).\end{aligned} \tag{29}$$

Equation (29) is the derivative of the neutron source rate density due to α-particle source isotope *k* and target isotope *i* with respect to the density of isotope *j* in the material. The derivative $\partial Q_{(\alpha,n),k,i} / \partial N_j$ consists of three terms: The first term on the right side of Eq. (29) applies when isotope *j* is a target; the second term applies when isotope *j* is an α emitter; and the third term accounts for isotope *j*'s contribution to the material's α stopping power. (All isotopes of an element have the same value for the third term, for a given source isotope *k* and target isotope *i*.)

The derivative of the total (α,n) neutron source rate density with respect to N_j is the sum of Eq. (29) over all sources and targets:

$$\frac{\partial Q_{(\alpha,n)}}{\partial N_j} = \sum_k \sum_i \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}. \tag{30}$$

We now turn to the derivative of the (α,n) neutron energy spectrum. From Eq. (15), the derivative of the total neutron source rate density in energy group *g* due to α emitter *k* and target *i* with respect to the density of isotope *j* is

$$\begin{aligned}\frac{\partial Q_{(\alpha,n),k,i}^{g}}{\partial N_j} &= \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} \sum_{l=1}^{L} \sum_{m=1}^{M} \sum_{g_\alpha=1}^{G_\alpha} F_{k,i,g_\alpha}(m) \Delta E_{m,g_\alpha}^{g} \\ &\quad + Q_{(\alpha,n),k,i} \sum_{l=1}^{L} \sum_{m=1}^{M} \sum_{g_\alpha=1}^{G_\alpha} \frac{\partial F_{k,i,g_\alpha}(m)}{\partial N_j} \Delta E_{m,g_\alpha}^{g}.\end{aligned} \tag{31}$$

From Eqs. (10), (11), and (12), the derivative of the fraction $F_{k,i,g_\alpha}(m)$ is

$$\begin{aligned}\frac{\partial F_{k,i,g_\alpha}}{\partial N_j} &= S_{i,g_\alpha}(m) \frac{\partial H_{i,g_\alpha,l}}{\partial N_j} \\ &= \frac{S_{i,g_\alpha}(m)}{P_i(E_l)} \\ &\quad \times \left[\frac{\partial P_i\left(E_{g_\alpha+1}\right)}{\partial N_j} - \frac{\partial P_i\left(E_{g_\alpha}\right)}{\partial N_j} - \frac{\partial P_i(E_l)}{\partial N_j} H_{i,g_\alpha,l} \right].\end{aligned} \tag{32}$$

The branching fraction $S_{i,g_\alpha}(m)$ does not depend on N_j. Using Eq. (23) in Eq. (32) yields

$$\begin{aligned}\frac{\partial F_{k,i,g_\alpha}}{\partial N_j} &= \frac{S_{i,g_\alpha}(m)}{P_i(E_l)} \\ &\quad \times \left[\frac{P_i(E_{g_\alpha+1}) - P_i(E_{g_\alpha})}{N_i} \delta_{ji} - \frac{P_i(E_l) H_{i,g_\alpha,l}}{N_i} \delta_{ji} \right. \\ &\quad \left. - \frac{P_{i,j}^{(1)}(E_{g_\alpha+1}) - P_{i,j}^{(1)}(E_{g_\alpha}) - P_{i,j}^{(1)}(E_l) H_{i,g_\alpha,l}}{N} \right] \\ &= S_{i,g_\alpha}(m) \left[\frac{P_i(E_{g_\alpha+1}) - P_i(E_{g_\alpha})}{P_i(E_l) N_i} \delta_{ji} - \frac{H_{i,g_\alpha,l}}{N_i} \delta_{ji} \right. \\ &\quad \left. - \frac{P_{i,j}^{(1)}(E_{g_\alpha+1}) - P_{i,j}^{(1)}(E_{g_\alpha}) - P_{i,j}^{(1)}(E_l) H_{i,g_\alpha,l}}{P_i(E_l) N} \right].\end{aligned} \tag{33}$$

Using Eq. (11), the first two terms in the brackets evaluate to zero, leaving

$$\begin{aligned}&\frac{\partial F_{k,i,g_\alpha}}{\partial N_j} = \\ &-S_{i,g_\alpha}(m) \left[\frac{P_{i,j}^{(1)}(E_{g_\alpha+1}) - P_{i,j}^{(1)}(E_{g_\alpha}) - P_{i,j}^{(1)}(E_l) H_{i,g_\alpha,l}}{P_i(E_l) N} \right].\end{aligned} \tag{34}$$

Using Eqs. (15) and (34) and rearranging slightly, Eq. (31) becomes

$$\begin{aligned}\frac{\partial Q_{(\alpha,n),k,i}^{g}}{\partial N_j} &= \frac{Q_{(\alpha,n),k,i}^{g}}{Q_{(\alpha,n),k,i}} \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} \\ &\quad - \frac{Q_{(\alpha,n),k,i}}{N} \sum_{l=1}^{L} \sum_{m=1}^{M} \sum_{g_\alpha=1}^{G_\alpha} S_{i,g_\alpha}(m) \\ &\quad \times \left[\frac{P_{i,j}^{(1)}(E_{g_\alpha+1}) - P_{i,j}^{(1)}(E_{g_\alpha}) - P_{i,j}^{(1)}(E_l) H_{i,g_\alpha,l}}{P_i(E_l)} \right] \Delta E_{m,g_\alpha}^{g}.\end{aligned} \tag{35}$$

The first term in Eq. (35) is the derivative of the neutron source rate density due to α-particle source isotope *k* and target isotope *i* with respect to the density of isotope *j* [Eq. (29)] multiplied by the normalized energy spectrum

of the source rate density due to source k and target i. In the second term, the factor in brackets has terms that are $P_{i,j}^{(1)}/P_i(E_l)$ (where the numerator is computed at various α-particle energies). We expect the second term to be small relative to the first. We hypothesize that a good approximation for the derivative of $Q_{(\alpha,n),k,i}^g$ with respect to N_j is

$$\frac{\partial Q_{(\alpha,n),k,i}^g}{\partial N_j} \approx \frac{Q_{(\alpha,n),k,i}^g}{Q_{(\alpha,n),k,i}} \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}. \tag{36}$$

This hypothesis is tested in Sec. VII.

For comparison, from Eq. (16), the derivative of the neutron source rate density due to spontaneous-fissioning isotope k with respect to the density of isotope j in the material is

$$\frac{\partial Q_{s.f.,k}}{\partial N_j} = \delta_{jk} \lambda_k R_k(s.f.), \tag{37}$$

and from Eq. (17), the derivative of the neutron source rate density in group g due to spontaneous-fissioning isotope k with respect to the density of isotope j in the material is

$$\frac{\partial Q_{s.f.,k}^g}{\partial N_j} = \delta_{jk} \lambda_k \chi_k^{s.f.}(E^g). \tag{38}$$

Because neither $\chi_k^{s.f.}(E^g)$ nor $R_k(s.f.)$ is a function of any isotopic densities, these derivatives are straightforward.

IV. DERIVATIVE OF THE (α,n) SOURCE RATE DENSITY WITH RESPECT TO MATERIAL ATOM DENSITY

Assume that the material atom density is perturbed by perturbing each isotopic density the same relative amount p_N:

$$N_j = N_{j,0}(1 + p_N), \tag{39}$$

where subscript zero indicates the initial, reference material. The total material atom density is

$$N = N_0(1 + p_N) \tag{40}$$

and p_N can be written

$$p_N = N_j / N_{j,0} - 1 = N / N_0 - 1. \tag{41}$$

The derivative of arbitrary quantity W with respect to the atom density of the material is

$$\frac{\partial W}{\partial N} = \frac{\partial W}{\partial p_N} \frac{\partial p_N}{\partial N} = \frac{1}{N_0} \frac{\partial W}{\partial p_N}. \tag{42}$$

From Eqs. (6) and (3), the derivative of $1/\varepsilon^g$ with respect to N is

$$\frac{\partial (1/\varepsilon_g)}{\partial N} = \frac{1}{N_0} \frac{\partial (1/\varepsilon_g)}{\partial p_N} = \frac{1}{N_0} \frac{\partial}{\partial p_N} \left(\frac{(1+p_N)\sum_{j=1}^{J} N_{j,0}}{(1+p_N)\sum_{j=1}^{J} N_{j,0}\varepsilon_{j,g}} \right) = 0. \tag{43}$$

From Eq. (5), and using Eq. (43), the derivative of $P_i(E_l)$ with respect to N is

$$\frac{\partial P_i(E_l)}{\partial N} = \frac{1}{N_0} \frac{N_i}{N} \sum_{g=1}^{G_\alpha} \frac{1}{2} \left[\left(\frac{\partial}{\partial p_N} \frac{\sigma_{i,g+1}}{\varepsilon_{g+1}} \right) + \left(\frac{\partial}{\partial p_N} \frac{\sigma_{i,g}}{\varepsilon_g} \right) \right] \left(E_{g+1} - E_g \right) + \frac{1}{N_0} \left(\frac{\partial}{\partial p_N} \frac{N_i}{N} \right) \sum_{g=1}^{G_\alpha} \frac{1}{2} \left[\frac{\sigma_{i,g+1}}{\varepsilon_{g+1}} + \frac{\sigma_{i,g}}{\varepsilon_g} \right] \left(E_{g+1} - E_g \right) = 0. \tag{44}$$

From Eq. (7), the derivative of $R_{k,i}(\alpha,n)$ with respect to N is

$$\frac{\partial R_{k,i}}{\partial N} = 0. \tag{45}$$

From Eq. (8), the derivative of $Q_{(\alpha,n),k,i}$ with respect to N is

$$\frac{\partial Q_{(\alpha,n),k,i}}{\partial N} = \frac{N_{k,0}}{N_0} \lambda_k R_{k,i}(\alpha,n) = \frac{1}{N_0} Q_{(\alpha,n),k,i}. \tag{46}$$

The derivative of the total (α,n) neutron source rate density with respect to N is the sum of Eq. (46) over all sources and targets. Using Eq. (9) yields

$$\frac{\partial Q_{(\alpha,n)}}{\partial N} = \frac{1}{N_0} \sum_k \sum_i \frac{\partial Q_{(\alpha,n),k,i}}{\partial N} = \frac{1}{N_0} Q_{(\alpha,n)}. \tag{47}$$

The derivative of the total (α,n) neutron source rate density with respect to N is also

$$\frac{\partial Q_{(\alpha,n)}}{\partial N} = \sum_j \frac{N_{j,0}}{N_0} \sum_k \sum_i \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}. \tag{48}$$

Using Eq. (28) and rearranging (and dropping the subscript zero) yields

$$\frac{\partial Q_{(\alpha,n)}}{\partial N} = \frac{1}{N} \left\{ \sum_j \sum_k \sum_i \left(\delta_{ji} + \delta_{jk} \right) \lambda_k N_k \sum_{l=1}^{L} f_{kl}^{\alpha} P_i(E_l) - \sum_j \sum_k \sum_i \frac{N_j}{N} \lambda_k N_k \sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l) \right\}. \tag{49}$$

The first term in the braces in Eq. (49) is the sum of the total (α,n) neutron source rate density due to targets and due to α emitters. Those two terms are both equal to the total (α,n) neutron source rate density. Also using Eq. (24) and then Eq. (8), Eq. (49) becomes

$$\frac{\partial Q_{(\alpha,n)}}{\partial N} = \frac{1}{N}\left\{2Q_{(\alpha,n)} - \sum_k \sum_i \lambda_k N_k \sum_{l=1}^{L} f_{kl}^{\alpha} P_i(E_l)\right\}. \quad (50)$$

Using Eq. (9), Eq. (50) becomes Eq. (47).

We now turn to the derivative of the (α,n) neutron energy spectrum. From Eqs. (10), (11), and (12), the derivative of the fraction $F_{k,i,g_\alpha}(m)$ is

$$\begin{aligned}\frac{\partial F_{k,i,g_\alpha}}{\partial N} &= S_{i,g_\alpha}(m)\frac{\partial H_{i,g_\alpha,l}}{\partial N} \\ &= \frac{S_{i,g_\alpha}(m)}{P_i(E_l)} \\ &\times\left[\frac{\partial P_i\left(E_{g_\alpha+1}\right)}{\partial N} - \frac{\partial P_i\left(E_{g_\alpha}\right)}{\partial N} - \frac{\partial P_i(E_l)}{\partial N} H_{i,g_\alpha,l}\right].\end{aligned} \quad (51)$$

Applying Eq. (44) yields

$$\frac{\partial F_{k,i,g_\alpha}}{\partial N} = 0. \quad (52)$$

From Eq. (15), and applying Eq. (52), the derivative of the total neutron source rate density in energy group g due to α emitter k and target i with respect to the material atom density is

$$\begin{aligned}\frac{\partial Q^g_{(\alpha,n),k,i}}{\partial N} &= \frac{\partial Q_{(\alpha,n),k,i}}{\partial N}\sum_{l=1}^{L}\sum_{m=1}^{M}\sum_{g_\alpha=1}^{G_\alpha} F_{k,i,g_\alpha}(m)\Delta E^g_{m,g_\alpha} \\ &= \frac{Q^g_{(\alpha,n),k,i}}{Q_{(\alpha,n),k,i}}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N}.\end{aligned} \quad (53)$$

Equation (53) is the same as Eq. (35) except that it lacks the second term involving $P^{(1)}_{i,j}(E_l)$ terms.

V. DERIVATIVE OF THE RESPONSE WITH RESPECT TO THE (α,n) SOURCE RATE DENSITY

The relative sensitivity of a response R to the atom density of isotope j, N_j, is defined as

$$S^g_{R,N_j} \equiv \frac{N_j}{R}\frac{\partial R}{\partial N_j}. \quad (54)$$

The adjoint-based formula for the relative sensitivity is[6,7]

$$\begin{aligned}S^g_{R,N_j} &= N_j\int_V dV\int_{4\pi} d\hat{\mathbf{\Omega}}\,\psi^{*g}(r,\hat{\mathbf{\Omega}})\frac{\partial Q^g(r)}{\partial N_j} \\ &-N_j\int_V dV\int_{4\pi} d\hat{\mathbf{\Omega}}\,\psi^{*g}(r,\hat{\mathbf{\Omega}})\sigma^g_{t,j}(r)\psi^g(r,\hat{\mathbf{\Omega}}) \\ &+N_j\int_V dV\sum_{g'=1}^{G}\varphi_0^{*g}(r)\chi_j^{g'\to g}(r)\nu_j^{g'}(r)\sigma^{g'}_{f,j}(r)\varphi_0^{g'}(r) \\ &+N_j\sum_{l=0}^{L}\int_V dV\sum_{g'=1}^{G}\varphi_l^{*g}(r)(2l+1)\sigma^{g'\to g}_{s,l,j}(r)\varphi_l^{g'}(r),\end{aligned} \quad (55)$$

where

$\psi^g(r,\hat{\mathbf{\Omega}})$ and $\psi^{*g}(r,\hat{\mathbf{\Omega}})$ = forward and adjoint neutron fluxes, respectively, at energy group g, position r, and angle $\hat{\mathbf{\Omega}}$

$\varphi_l^g(r)$ and $\varphi_l^{*g}(r)$ = forward and adjoint neutron flux moments, respectively, of order l at energy group g and position r

$\sigma^g_{t,j}$ = total cross section of isotope j at energy group g

$\sigma^{g'\to g}_{s,l,j}$ = scattering cross section moment of order l of isotope j from energy group g' to g

$\chi_j^{g'\to g}$ = fission spectrum of isotope j from energy group g' to g

$\nu_j^{g'}$ = number of neutrons from isotope j in energy group g'

$\sigma^{g'}_{f,j}$ = fission cross section of isotope j at energy group g'.

This paper is concerned with the first term on the right side of Eq. (55).

The source Q^g is the sum of the spontaneous fission source in group g and the (α,n) source in group g: $Q^g = Q^g_{s.f.} + Q^g_{(\alpha,n)}$. The derivative of the spontaneous fission neutron source rate density in group g with respect to the density of isotope j is straightforward and is unaffected by the presence of other isotopes [Eq. (38)]. The derivative of the (α,n) source rate density in group g with respect to the density of isotope j is

$$\frac{\partial Q^g_{(\alpha,n)}}{\partial N_j} = \sum_k\sum_i \frac{\partial Q^g_{(\alpha,n),k,i}}{\partial N_j}. \quad (56)$$

VI. MODIFICATION OF SOURCES4C

SOURCES4C was modified to print quantities needed to compute the derivatives. Where the material stopping powers are computed from Eq. (6), the elemental stopping powers ε_j^g are now retained. Where $P_i(E_l)$ of Eq. (5) is computed, $P^{(1)}_{i,j}(E_l)$ of Eq. (22) is now also computed for all α energies. The quantities λ_k, N_k, N_i, and $Q_{(\alpha,n),k,i}$ are now printed for each combination of source isotope k and target isotope i in a new output file. The quantity $\sum_{l=1}^{L} f_{kl}^{\alpha} P^{(1)}_{i,j}(E_l)$ is now printed for each stopping element for each combination of source isotope k and target isotope i. Then $\partial Q_{(\alpha,n),k,i}/\partial N_j$ can be computed

(in post-processing) using Eq. (29) for each combination of source isotope k and target isotope i.

Equations (29) and (30) were implemented in the SENSMG multigroup neutron sensitivity code.[8] Equation (36) was also implemented, but with a further approximation. SOURCES4C was modified to compute and print the spectrum due to each source isotope k as the sum over all target isotopes:

$$X_k^g \equiv \frac{\sum_i Q_{(\alpha,n),k,i}^g}{\sum_i Q_{(\alpha,n),k,i}}. \tag{57}$$

Likewise, SOURCES4C computes and prints the spectrum due to each target isotope i as the sum over all source isotopes:

$$X_i^g \equiv \frac{\sum_k Q_{(\alpha,n),k,i}^g}{\sum_k Q_{(\alpha,n),k,i}}. \tag{58}$$

The overall spectrum is also output:

$$X^g \equiv \frac{\sum_k \sum_i Q_{(\alpha,n),k,i}^g}{\sum_k \sum_i Q_{(\alpha,n),k,i}}. \tag{59}$$

Equation (59) is a standard SOURCES4C output, of course.

If isotope j is source isotope k of α particles, the derivative of the (α,n) neutron source rate density with respect to N_j in group g is approximated in SENSMG as

$$\frac{\partial Q_{(\alpha,n)}^g}{\partial N_j} \approx X_k^g \sum_k \sum_i \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}. \tag{60}$$

If isotope j is (α,n) target isotope i, the derivative of the (α,n) neutron source rate density with respect to N_j in group g is approximated as

$$\frac{\partial Q_{(\alpha,n)}^g}{\partial N_j} \approx X_i^g \sum_k \sum_i \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}. \tag{61}$$

If isotope j is neither a source nor a target but contributes to the (α,n) source only through its contribution to the stopping power, the overall spectrum is applied:

$$\frac{\partial Q_{(\alpha,n)}^g}{\partial N_j} \approx X^g \sum_k \sum_i \frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}. \tag{62}$$

In some circumstances, the isotope may be more important due to its stopping power than to its being an α emitter or a target. The third term of Eq. (29) will be larger (in magnitude) than the other two terms. In this case, Eq. (62) is used rather than Eq. (60) or (61).

The derivative of the (α,n) neutron source rate density with respect to N, the atom density of the material, in group g is calculated using

$$\frac{\partial Q_{(\alpha,n)}^g}{\partial N} \approx X^g \sum_k \sum_i \frac{\partial Q_{(\alpha,n),k,i}}{\partial N} = \frac{1}{N_0} X^g Q_{(\alpha,n)} \tag{63}$$

as an approximation to Eq. (53).

VII. RESULTS

A test problem used the beryllium-reflected plutonium (BeRP) ball reflected by 3.81 cm of polyethylene (rather than beryllium). The BeRP ball is a 4.48-kg sphere of α-phase plutonium. Using the multigroup discrete-ordinates PARTISN code[9] with 618 energy groups, S_{128} quadrature, and a P_3 Legendre scattering expansion, the total neutron leakage for this model [from (α,n), spontaneous fission, and induced fission] is 1.282×10^6 neutrons/s. The neutron source rate density due to (α,n) reactions is 31.8316 neutrons/cm^3/s, compared to 1231.18 neutrons/cm^3/s due to spontaneous fission.

VII.A. Molybdenum

Molybdenum is present as an impurity[10] in the plutonium at 9 ppm or 9×10^{-4} wt%. It is neither an α particle emitter nor an (α,n) target; however, perturbing the molybdenum content perturbs the plutonium's (α,n) source rate density because of molybdenum's contribution to the material's stopping power. The absolute sensitivity of the (α,n) source rate density to the Mo atom density is shown in Table I, where the result from Eq. (30) is compared with the result from a central difference that was computed with a perturbation of ±90%. The agreement is within 0.1% but Eq. (30) is certainly much more accurate than the central difference. The absolute sensitivity of –424.759 corresponds to a relative sensitivity of -1.4777×10^{-5} %/%.

The relative sensitivity of the neutron leakage rate to the Mo atom density is shown in Table II, where the result from Eq. (55) is compared with the result from a central difference that was computed with a perturbation of ±90%. The agreement is within 0.1% but Eq. (55) is certainly much more accurate than the central difference. Although increasing the Mo density decreases the (α,n) source rate density (Table I), overall it increases the neutron leakage rate (Table II).

The four terms on the right side of Eq. (55) are shown in Table III. Although the total cross section and scattering terms are each ~400 times greater than the source term (in magnitude), those terms are of opposite sign, making the source term important. The source term is 4% of the total sensitivity.

TABLE I. Absolute Sensitivity of the (α,n) Source Rate Density to the Mo Atom Density.

Method	Value [(g/cm^3·s)/(atom/b·cm)]
Eq. (30)	–4.24759E+02
Central Difference	–4.24422E+02
Difference	0.0795%

TABLE II. Relative Sensitivity of the Neutron Leakage Rate to the Mo Atom Density.

Method	Value (%/%)
Adjoint [Eq. (55)]	9.44244E-06
Central Difference	9.44519E-06
Difference	–0.0291%

TABLE III. Components of the Relative Sensitivity of the Neutron Leakage Rate to the Mo Atom Density.

Term	Value (%/%)
Source	–3.85754E-07
Total Cross Section	–1.44405E-04
Fission	0.
Scattering	1.54233E-04
Total (Sum)	9.44244E-06

The approximation of Eq. (62) for the derivative of the (α,n) source rate density in each group with respect to the atom density of Mo (N_{Mo}) is compared with a central-difference calculation in Fig. 1. Clearly, Eq. (62) is an excellent approximation for this case.

VII.B. Carbon

Carbon is present as an impurity[10] in the plutonium at 0.023 wt%. Using SOURCES4C with 618 neutron energy groups, the neutron source rate density due to (α,n) reactions on carbon is 3.978 neutrons/cm^3/s, compared to a total (α,n) source rate density of 31.8316 neutrons/cm^3/s and 1231.18 neutrons/cm^3/s due to spontaneous fission. The absolute sensitivity of the (α,n) source rate density to the C atom density is shown in Table IV, where the result from Eq. (30) is compared with the result from a central difference that was computed with a perturbation of ±50%. The agreement is within 0.0003%. The absolute sensitivity of 1.74856×10^4 corresponds to a relative sensitivity of 0.124153 %/%.

The relative sensitivity of the neutron leakage rate to the C atom density is shown in Table V, where the result from Eq. (55) is compared with the result from a central difference that was computed with a perturbation of ±50%. The agreement is within 0.02% but Eq. (55) is certainly more accurate than the central difference. Increasing the C density decreases the (α,n) source rate density (Table IV) and the neutron leakage rate (Table V). If the effect of carbon on the stopping power is ignored, the difference rises to 0.4%.

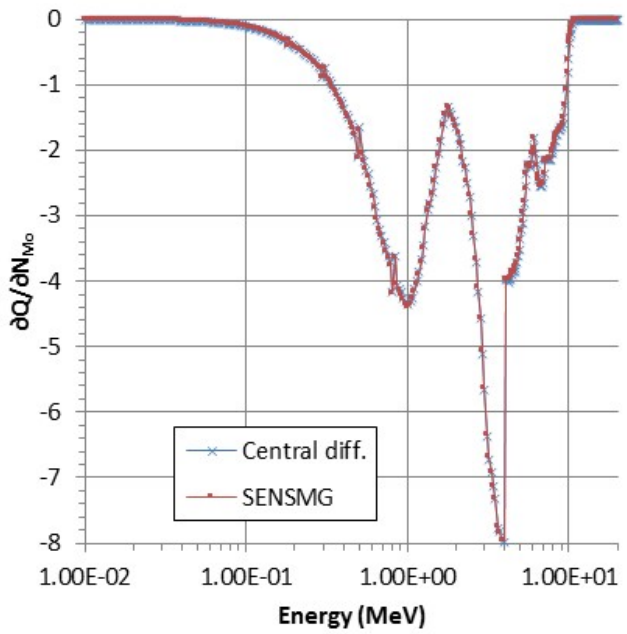

Fig. 1. Energy-dependent derivative of the neutron source rate density with respect to the Mo atom density.

The four terms on the right side of Eq. (55) are shown in Table VI. Although the total cross section and scattering terms are each ~4 times greater than the source term (in magnitude), those terms are of opposite sign, making the source term important. The source term is 70% of the total sensitivity.

TABLE IV. Absolute Sensitivity of the (α,n) Source Rate Density to the C Atom Density.

Method	Value [(g/cm^3·s)/(atom/b·cm)]
Eq. (30)	1.74856E+04
Central Difference	1.74856E+04
Difference	–0.0003%

TABLE V. Relative Sensitivity of the Neutron Leakage Rate to the C Atom Density.

Method	Value (%/%)
Adjoint [Eq. (55)]	4.66672E-03
Central Difference	4.66749E-03
Difference	–0.0165%

TABLE VI. Components of the Relative Sensitivity of the Neutron Leakage Rate to the C Atom Density.

Term	Value (%/%)
Source	3.26724E-03
Total Cross Section	–1.22178E-02
Fission	0.
Scattering	1.36173E-02
Total (Sum)	4.66672E-03

The approximation of Eq. (61) for the derivative of the (α,n) source rate density in each group with respect to the atom density of C (N_C) is compared with a central-difference calculation in Fig. 2. Clearly, Eq. (61) is an excellent approximation for this case.

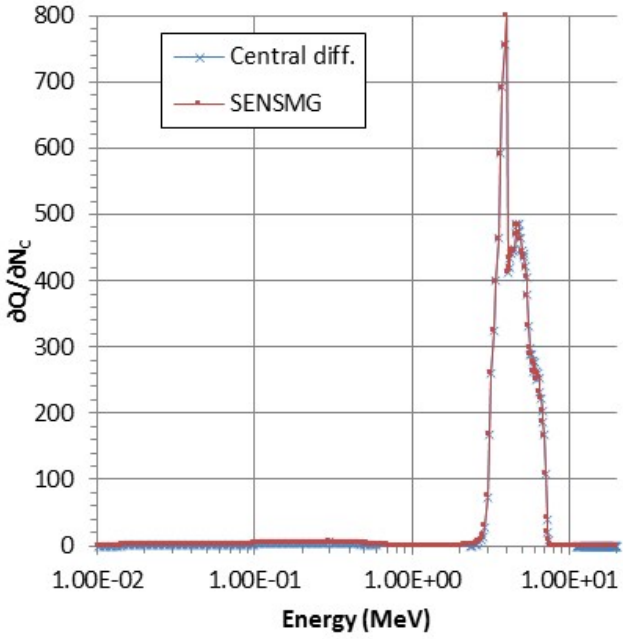

Fig. 2. Energy-dependent derivative of the neutron source rate density with respect to the C atom density.

VII.C. Plutonium-242

Plutonium-242 is present as an impurity[10] in the plutonium at 0.028 wt% (as a percentage of the material). Using SOURCES4C with 618 neutron energy groups, the neutron source rate density due to α-particle emissions from ^{242}Pu is 3.02539×10^{-4} neutrons/cm^3/s, compared to a total (α,n) neutron source rate density of 31.8316 neutrons/cm^3/s. The neutron source rate density due to spontaneous fission of ^{242}Pu is 9.47880 neutrons/cm^3/s, compared to a total spontaneous fission neutron source rate density of 1231.18 neutrons/cm^3/s.

The absolute sensitivity of the (α,n) source rate density to the ^{242}Pu atom density is shown in Table VII, where the result from Eq. (30) is compared with the result from a central difference that was computed with a perturbation of ±50%. The agreement is within 0.15%. The absolute sensitivity of –621.917 corresponds to a relative sensitivity of -2.66371×10^{-4} %/%. It is counterintuitive that increasing the density of an α emitter would decrease the (α,n) source rate density. However, changing the ^{242}Pu density in this material affects the material's stopping power more than it affects the α-particle emission rate. In other words, the third term in Eq. (29) is negative and much larger (absolute value) than the second term: –644 compared to 22.

The relative sensitivity of the neutron leakage rate to the ^{242}Pu atom density is shown in Table VIII, where the result from Eq. (55) is compared with the result from a central difference that was computed with a perturbation of ±50%. The agreement is within 0.05% but Eq. (55) is certainly more accurate than the central difference. As in the Mo case (Sec. VII.A), increasing the ^{242}Pu density decreases the (α,n) source rate density (Table VII) but increases the neutron leakage rate (Table VIII).

The four terms on the right side of Eq. (55) are shown in Table IX. The contributions to the source term from spontaneous fission neutrons [Eq. (37)] and (α,n) neutrons [Eq. (30) with (29)] are both shown. The combination of the total cross section, fission, and scattering terms almost balances to zero; the source term is 97.5% of the total sensitivity. The (α,n) contribution is only 0.1% of the total sensitivity.

TABLE VII. Absolute Sensitivity of the (α,n) Source Rate Density to the ^{242}Pu Atom Density.

Method	Value [(g/cm^3·s)/(atom/b·cm)]
Eq. (30)	–6.21917E+02
Central Difference	–6.21036E+02
Difference	0.1417%

TABLE VIII. Relative Sensitivity of the Neutron Leakage Rate to the ^{242}Pu Atom Density.

Method	Value (%/%)
Adjoint [Eq. (55)]	7.68268E-03
Central Difference	7.68624E-03
Difference	–0.0464%

TABLE IX. Components of the Relative Sensitivity of the Neutron Leakage Rate to the ^{242}Pu Atom Density.

Term	Value (%/%)
Source, Spont. Fiss.	7.49779E-03
Source, (α,n)	–6.95365E-06
Total Cross Section	–3.14530E-03
Fission	1.04025E-03
Scattering	2.29689E-03
Total (Sum)	7.68268E-03

The approximations of Eqs. (62) and (60) for the derivative of the (α,n) source rate density in each group with respect to the atom density of ^{242}Pu (N_{Pu}) are compared with a central-difference calculation in Fig. 3. Equation (60) is a poor approximation for this case, probably because ^{242}Pu is more important because of its contribution to the stopping power than because of its α-particle emissions. Figure 3 demonstrates why we treat ^{242}Pu like a stopping element and apply the overall (α,n) neutron spectrum [Eq. (62), as was done for Mo in Sec. VII.A], yielding the correct spectrum for ^{242}Pu.

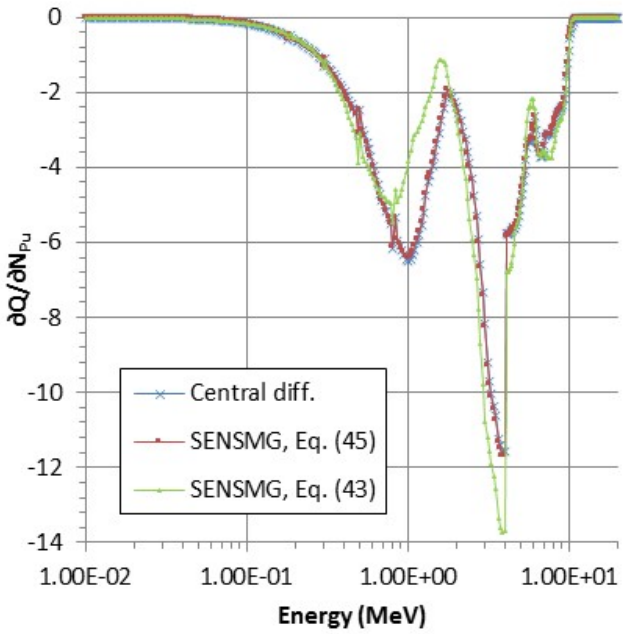

Fig. 3. Energy-dependent derivative of the (α,n) neutron source rate density with respect to the ^{242}Pu atom density.

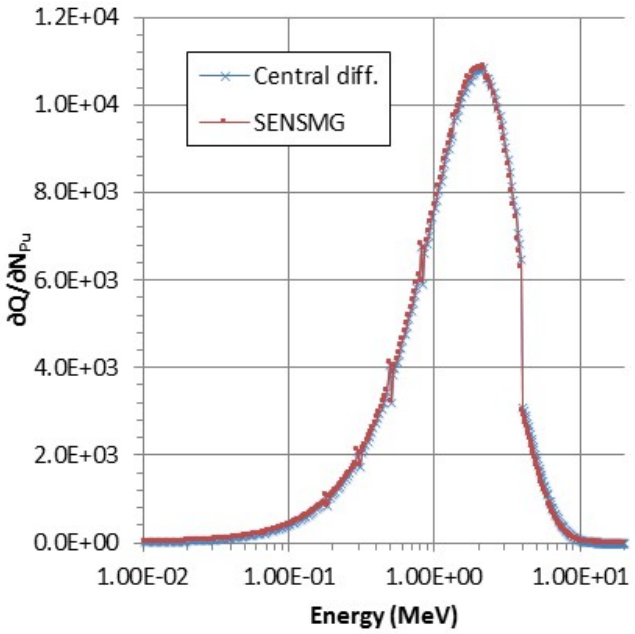

Fig. 4. Energy-dependent derivative of the total neutron source rate density with respect to the ^{242}Pu atom density.

If it is important to study (α,n) source sensitivities in detail, then it will be necessary to consider the spectrum associated with the combination of each α emitter and target separately and to compute the second term of Eq. (35).

However, for isotopes that are both α emitters and spontaneous fissioners, it is likely that neutrons from spontaneous fission are more numerous and more important than neutrons from (α,n) reactions. Figure 4 shows the derivative of the total [(α,n) plus spontaneous fission] neutron source rate density in each group with respect to the atom density of ^{242}Pu. The derivative of the (α,n) neutron source rate density spectrum and the small differences in the spectra of Fig. 3 can only have a very small effect on the spectrum of the total.

VII.D. Material Atom Density

As noted in Secs. VII.B and VII.C, the total (α,n) neutron source rate density is 31.8316 neutrons/cm^3/s. The atom density of the BeRP ball material is 0.04965644533 atom/b·cm. The absolute sensitivity of the (α,n) neutron source rate density to the material atom density is the ratio, as given by Eq. (47). In Table X, this ratio is compared with a central difference estimate that was computed with a perturbation of ±1%. The agreement is within 0.0001%. The absolute sensitivity of 641.036 corresponds to a relative sensitivity of 1 %/%, as expected from using Eq. (47) in Eq. (54).

TABLE X. Absolute Sensitivity of the (α,n) Source Rate Density to the Material Atom Density.

Method	Value [(g/cm^3·s)/(atom/b·cm)]
Eq. (47)	6.41036E+02
Central Difference	6.41036E+02
Difference	0.0001%

The approximation of Eq. (63) for the derivative of the (α,n) source rate density in each group with respect to the atom density of the material (N) is compared with a central-difference calculation in Fig. 5. Clearly, Eq. (63) is an excellent approximation for this case.

VIII. CONCLUSIONS

We have derived the derivative (first-order sensitivity) of a material's (α,n) neutron source rate density with respect to the isotopic density of each of its constituents as well as the total atom density of the material. We have propagated these derivatives to the derivative of a response. Test problem results confirm the accuracy of the equations we derived.

These derivatives with respect to atom densities can easily be converted to derivatives with respect to constrained isotopic weight fractions or atom fractions using the methods of Ref. 1.

We relied on the mathematical description of the (α,n) neutron source generated from a homogeneously mixed material that was implemented in the SOURCES4C code. In that context, the derivatives of the (α,n) neutron source rate density for each combination of

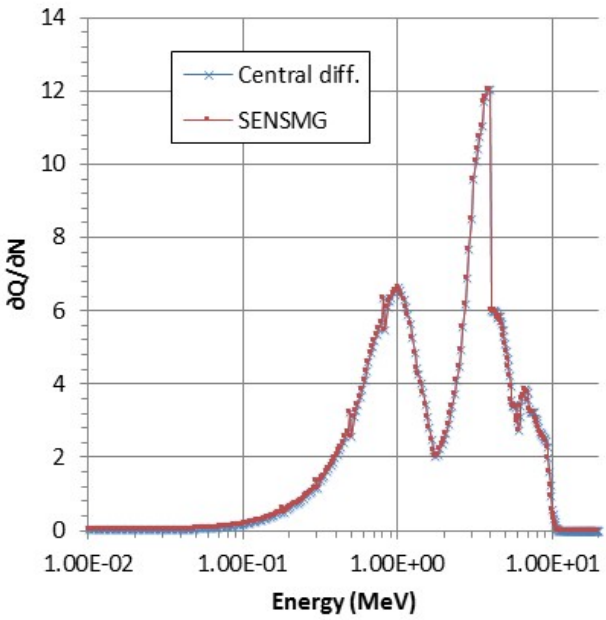

Fig. 5. Energy-dependent derivative of the (α,n) neutron source rate density with respect to the material atom density.

target isotope and α-emitting isotope are exact. We also derived exact expressions for the energy-dependent (α,n) neutron source rate densities (the spectrum).

We modified SOURCES4C to print quantities needed to compute the derivatives, and we implemented the equations for the derivatives in the SENSMG code after SOURCES4C is run.

In the coding, two approximations were made for the spectra. First, a complicated term that is expected to be small was ignored. Second, we used ensemble averages of a spectrum function rather than individual spectra for each combination of target isotope and α-emitting isotope. In comparisons with central differences, any errors associated with these approximations were negligible. However, it would be possible to compute exact spectra if needed.

We are working on deriving and implementing second derivatives of a material's (α,n) neutron source rate density.

The test problem in Sec. VII used the data [α stopping cross sections, (α,n) cross sections, (α,n) product level branching, and decay] that is distributed with SOURCES4C. Minor improvements were made to the decay data in 2001; otherwise, the data seems to be from 1984 or earlier.[2] It would be interesting to compare the results of Sec. VII with results obtained using newer nuclear data. In particular, the ENDF/B-VII.1 evaluation included an updated and improved radioactive decay sublibrary.[12] (The ENDF/B-VIII.0 evaluation seems to have made only minor improvements to the radioactive decay sublibrary.) The changes in the decay data probably change the absolute (α,n) neutron production rates and spectra, but how do they change the sensitivities computed in Sec. VII?

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