SECOND DERIVATIVE OF AN (α,n) NEUTRON SOURCE WITH RESPECT TO CONSTITUENT ISOTOPE DENSITIES

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ABSTRACT

This paper derives the second derivative of the (α,n) neutron source rate density and spectrum with respect to the atom densities of the constituents of a homogeneously mixed material. Like the first derivatives derived previously, the second derivatives are analytic. The SOURCES4C neutron source generation code has been modified to output quantities needed to compute the first and second derivatives in post-processing. A numerical test problem verifies the derivation and implementation and demonstrates the difficulty of computing these second derivatives with a finite difference.

KEYWORDS: (alpha,n) neutron source; sensitivity; second derivative; SOURCES4C

1. INTRODUCTION

We are deriving adjoint-based second-order sensitivities of a response in transport theory [1, 2]. Neutron problems [2] need the second-order sensitivity of the inhomogeneous source with respect to the constituent isotope densities. We have previously derived and verified the first-order sensitivity of an (α,n) neutron source with respect to isotope densities and the subsequent first-order sensitivity of a leakage response [3]. In this paper, we derive and verify the second-order sensitivity of an (α,n) neutron source with respect to constituent isotope densities. We show that the second-order sensitivity of a spontaneous-fission source with respect to constituent isotope densities is zero. The propagation of these second-order sensitivities to the second-order sensitivity of a response [2] awaits the completion of the coding.

As in [3], 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 [4]; specifically, a material's α-particle stopping power is given by the Bragg-Kleeman relationship [5]. Also, 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 [6, 7] is also not addressed.

The equations derived herein were implemented in the SENSMG multigroup sensitivity code [8].

The next section of this paper derives the equations for the second derivatives, including the derivative of the spectrum. Sections 3 and 4 briefly discuss modifications to SOURCES4C and SENSMG, respectively. Section 5 presents a six-isotope test problem. Section 6 is a summary and conclusions.

2. DERIVATION

2.1. Derivative of the (α,n) Source Rate Density with Respect to Isotopic Density

The neutron source rate density $Q_{(\alpha,n),k,i}$ due to α-particle source isotope *k* and target isotope *i* is [3, 4]

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

where

N_k = atom density of α-particle source isotope *k*
λ_k = decay constant for isotope *k*
f_{kl}^{α} = fraction of all decays of nuclide *k* resulting in an α particle of energy E_l
$P_i(E_l)$ = probability of an α particle of energy E_l undergoing an (α,n) reaction with nuclide *i* before stopping
L = number of discrete α particle energies emitted by nuclide *k*.

The derivative of $Q_{(\alpha,n),k,i}$ with respect to the density N_j of isotope *j* in the material is [3]

$$\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} = \delta_{ji} \frac{Q_{(\alpha,n),k,i}}{N_i} + \delta_{jk} \frac{Q_{(\alpha,n),k,i}}{N_k} - \lambda_k \frac{N_k}{N} \sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l), \tag{2}$$

where

N_i = atom density of target isotope *i*
N = atom density of the material
$P_{i,j}^{(1)}(E_l)$ = part of the derivative of $P_i(E_l)$ with respect to N_j; defined below.

The derivative of $P_i(E_l)$ with respect to the density N_j is [Eq. (23) of [3]]

$$\frac{\partial P_i\left(E_l\right)}{\partial N_j} = \frac{P_i(E_l)}{N_i} \delta_{ji} - \frac{P_{i,j}^{(1)}(E_l)}{N}, \tag{3}$$

where $P_{i,j'}^{(1)}(E_l)$ is defined as [Eq. (22) of [3]]

$$P_{i,j}^{(1)}(E_l) \equiv \frac{N_i}{N} \sum_{g=1}^{G_\alpha} \frac{1}{2} \left[\frac{\sigma_{i,g+1} \varepsilon_{j,g+1}}{\varepsilon_{g+1}^2} + \frac{\sigma_{i,g} \varepsilon_{j,g}}{\varepsilon_g^2} \right] \left(E_{g+1} - E_g \right), \tag{4}$$

where the sum is a trapezoid-rule evaluation of an integral over the α-particle energy using G_α partitions of the energy domain. In addition,

E_{g+1}, E_g = upper and lower energies corresponding to α-particle energy group *g*
$\sigma_{i,g}$ = microscopic (α,n) cross section for nuclide *i* in α particle energy group *g*
$\varepsilon_{j,g}$ = stopping power in α-particle group *g* for isotope *j*

ε_g = material stopping power in α-particle group g.

The material stopping power in α-particle group g is calculated using the Bragg-Kleeman relationship [5],

$$\varepsilon_g = \frac{1}{N}\sum_{j=1}^{J} N_j \varepsilon_{j,g}. \tag{5}$$

The first term on the right side of Eq. (2) 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. For later convenience, define

$$\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{target}} = \delta_{ji}\frac{Q_{(\alpha,n),k,i}}{N_i}, \tag{6}$$

$$\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\alpha\text{-emitter}} = \delta_{jk}\frac{Q_{(\alpha,n),k,i}}{N_k}, \tag{7}$$

$$\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{stopping}} = -\lambda_k \frac{N_k}{N}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l). \tag{8}$$

Equation (2) becomes

$$\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} = \left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{target}} + \left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\alpha\text{-emitter}} + \left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{stopping}}. \tag{9}$$

The derivative of $\partial Q_{(\alpha,n),k,i}/\partial N_j$ with respect to the atom density of another isotope j' is

$$\frac{\partial^2 Q_{(\alpha,n),k,i}}{\partial N_{j'}\partial N_j} = \frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{target}} + \frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\alpha\text{-emitter}} + \frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{stopping}}. \tag{10}$$

The first term on the right side of Eq. (10) is, from Eq. (6),

$$\begin{aligned}\frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{target}} &= \frac{\partial}{\partial N_{j'}}\left\{\delta_{ji}\frac{Q_{(\alpha,n),k,i}}{N_i}\right\} = \delta_{ji}\left\{\frac{\partial}{\partial N_{j'}}\left(\frac{1}{N_i}\right)Q_{(\alpha,n),k,i} + \frac{1}{N_i}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_{j'}}\right\} \\ &= \delta_{ji}\left\{-\delta_{j'i}\frac{1}{N_i^2}Q_{(\alpha,n),k,i} + \frac{1}{N_i}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_{j'}}\right\}.\end{aligned} \tag{11}$$

Using Eq. (2) with j' rather than j, Eq. (11) becomes

$$\frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{target}} = \delta_{ji}\delta_{j'k}\frac{Q_{(\alpha,n),k,i}}{N_i N_k} - \delta_{ji}\lambda_k\frac{N_k}{N_i N}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j'}^{(1)}(E_l). \tag{12}$$

The second term on the right side of Eq. (10) is, from Eq. (7),

$$\frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{α-emitter}} = \frac{\partial}{\partial N_{j'}}\left\{\delta_{jk}\frac{Q_{(\alpha,n),k,i}}{N_k}\right\} = \delta_{jk}\left\{\frac{\partial}{\partial N_{j'}}\left(\frac{1}{N_k}\right)Q_{(\alpha,n),k,i} + \frac{1}{N_k}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_{j'}}\right\}$$
$$= \delta_{jk}\left\{-\delta_{j'k}\frac{1}{N_k^2}Q_{(\alpha,n),k,i} + \frac{1}{N_k}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_{j'}}\right\}. \quad (13)$$

Again using Eq. (2) with j' rather than j, Eq. (13) becomes

$$\frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{α-emitter}} = \delta_{jk}\delta_{j'i}\frac{Q_{(\alpha,n),k,i}}{N_k N_i} - \delta_{jk}\frac{\lambda_k}{N}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j'}^{(1)}(E_l). \quad (14)$$

The third term on the right side of Eq. (10) is, from Eq. (8),

$$\frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{stopping}} = \frac{\partial}{\partial N_{j'}}\left\{-\lambda_k\frac{N_k}{N}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l)\right\}$$
$$= -\lambda_k\left\{\frac{\partial}{\partial N_{j'}}\left(\frac{N_k}{N}\right)\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l) + \frac{N_k}{N}\sum_{l=1}^{L} f_{kl}^{\alpha}\frac{\partial P_{i,j}^{(1)}(E_l)}{\partial N_{j'}}\right\}$$
$$= -\lambda_k\left\{\left(\delta_{j'k}\frac{1}{N} - \frac{N_k}{N^2}\right)\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l) + \frac{N_k}{N}\sum_{l=1}^{L} f_{kl}^{\alpha}\frac{\partial P_{i,j}^{(1)}(E_l)}{\partial N_{j'}}\right\}. \quad (15)$$

From Eq. (4), the derivative of $P_{i,j}^{(1)}(E_l)$ with respect to $N_{j'}$ is

$$\frac{\partial P_{i,j}^{(1)}(E_l)}{\partial N_{j'}} = \frac{\partial}{\partial N_{j'}}\left(\frac{N_i}{N}\right)\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}}{\varepsilon_{g+1}^2} + \frac{\sigma_{i,g}\varepsilon_{j,g}}{\varepsilon_g^2}\right]\left(E_{g+1} - E_g\right)$$
$$+ \frac{N_i}{N}\sum_{g=1}^{G_\alpha}\frac{1}{2}\frac{\partial}{\partial N_{j'}}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}}{\varepsilon_{g+1}^2} + \frac{\sigma_{i,g}\varepsilon_{j,g}}{\varepsilon_g^2}\right]\left(E_{g+1} - E_g\right). \quad (16)$$

Equation (16) requires the derivative of the inverse squared of the material stopping power. It is

$$\frac{\partial(1/\varepsilon_g^2)}{\partial N_{j'}} = \frac{\partial}{\partial N_{j'}}\left[\frac{\left(\sum_{j=1}^{J} N_j\right)^2}{\left(\sum_{j=1}^{J} N_j\varepsilon_{j,g}\right)^2}\right] = \frac{1}{\left(\sum_{j=1}^{J} N_j\varepsilon_{j,g}\right)^2}\frac{\partial}{\partial N_{j'}}\left[\left(\sum_{j=1}^{J} N_j\right)^2\right] + \left(\sum_{j=1}^{J} N_j\right)^2\frac{\partial}{\partial N_{j'}}\left[\left(\sum_{j=1}^{J} N_j\varepsilon_{j,g}\right)^{-2}\right]$$
$$= \frac{2(\varepsilon_g - \varepsilon_{j',g})}{N\varepsilon_g^3}. \quad (17)$$

Working out the derivative in the first term of Eq. (16) and using Eq. (17) in the second term, Eq. (16) becomes

$$\frac{\partial P_{i,j}^{(1)}(E_l)}{\partial N_{j'}} = \left(\delta_{j'i}\frac{1}{N} - \frac{N_i}{N^2}\right)\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}}{\varepsilon_{g+1}^2} + \frac{\sigma_{i,g}\varepsilon_{j,g}}{\varepsilon_g^2}\right]\left(E_{g+1} - E_g\right)$$
$$+\frac{N_i}{N}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\sigma_{i,g+1}\varepsilon_{j,g+1}\frac{2(\varepsilon_{g+1} - \varepsilon_{j',g+1})}{N\varepsilon_{g+1}^3} + \sigma_{i,g}\varepsilon_{j,g}\frac{2(\varepsilon_g - \varepsilon_{j',g})}{N\varepsilon_g^3}\right]\left(E_{g+1} - E_g\right). \tag{18}$$

Using Eq. (4) in the first term of Eq. (18) and expanding the second term yields

$$\frac{\partial P_{i,j}^{(1)}(E_l)}{\partial N_{j'}} = \left(\delta_{j'i}\frac{1}{N} - \frac{N_i}{N^2}\right)\frac{N}{N_i}P_{i,j}^{(1)}(E_l) + 2\frac{N_i}{N^2}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}}{\varepsilon_{g+1}^2} + \frac{\sigma_{i,g}\varepsilon_{j,g}}{\varepsilon_g^2}\right]\left(E_{g+1} - E_g\right)$$
$$-2\frac{N_i}{N^2}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}\varepsilon_{j',g+1}}{\varepsilon_{g+1}^3} + \frac{\sigma_{i,g}\varepsilon_{j,g}\varepsilon_{j',g}}{\varepsilon_g^3}\right]\left(E_{g+1} - E_g\right). \tag{19}$$

Define

$$P_{i,j,j'}^{(2)}(E_l) \equiv \frac{N_i}{N}\sum_{g=1}^{G_\alpha}\frac{1}{2}\left[\frac{\sigma_{i,g+1}\varepsilon_{j,g+1}\varepsilon_{j',g+1}}{\varepsilon_{g+1}^3} + \frac{\sigma_{i,g}\varepsilon_{j,g}\varepsilon_{j',g}}{\varepsilon_g^3}\right]\left(E_{g+1} - E_g\right). \tag{20}$$

Using Eqs. (4) and (20), Eq. (19) becomes

$$\frac{\partial P_{i,j}^{(1)}(E_l)}{\partial N_{j'}} = \left(\frac{\delta_{j'i}}{N_i} - \frac{1}{N}\right)P_{i,j}^{(1)}(E_l) + \frac{2}{N}P_{i,j}^{(1)}(E_l) - \frac{2}{N}P_{i,j,j'}^{(2)}(E_l)$$
$$= \frac{\delta_{j'i}}{N_i}P_{i,j}^{(1)}(E_l) + \frac{1}{N}P_{i,j}^{(1)}(E_l) - \frac{2}{N}P_{i,j,j'}^{(2)}(E_l). \tag{21}$$

Using Eq. (21), Eq. (15) becomes

$$\frac{\partial}{\partial N_{j'}}\left[\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}\right]_{\text{stopping}} = -\lambda_k\left\{\left(\delta_{j'k}\frac{1}{N} - \frac{N_k}{N^2}\right)\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j}^{(1)}(E_l)\right.$$
$$\left.+\frac{N_k}{N}\sum_{l=1}^{L}f_{kl}^{\alpha}\left[\frac{\delta_{j'i}}{N_i}P_{i,j}^{(1)}(E_l) + \frac{1}{N}P_{i,j}^{(1)}(E_l) - \frac{2}{N}P_{i,j,j'}^{(2)}(E_l)\right]\right\}$$
$$= -\lambda_k\left\{\delta_{j'k}\frac{1}{N}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j}^{(1)}(E_l) + \delta_{j'i}\frac{N_k}{N_iN}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j}^{(1)}(E_l)\right.$$
$$\left.-\frac{2N_k}{N^2}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j,j'}^{(2)}(E_l)\right\}. \tag{22}$$

Using Eqs. (12), (14), and (22) in Eq. (10) yields

$$\frac{\partial^2 Q_{(\alpha,n),k,i}}{\partial N_{j'}\partial N_j} = \delta_{ji}\delta_{j'k}\frac{Q_{(\alpha,n),k,i}}{N_iN_k} - \delta_{ji}\lambda_k\frac{N_k}{N_iN}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j'}^{(1)}(E_l) + \delta_{jk}\delta_{j'i}\frac{Q_{(\alpha,n),k,i}}{N_kN_i} - \delta_{jk}\frac{\lambda_k}{N}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j'}^{(1)}(E_l)$$
$$-\delta_{j'k}\frac{\lambda_k}{N}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j}^{(1)}(E_l) - \delta_{j'i}\frac{\lambda_kN_k}{N_iN}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j}^{(1)}(E_l) + \frac{2\lambda_kN_k}{N^2}\sum_{l=1}^{L}f_{kl}^{\alpha}P_{i,j,j'}^{(2)}(E_l). \tag{23}$$

Finally, rearranging Eq. (23) yields

$$\begin{aligned}\frac{\partial^2 Q_{(\alpha,n),k,i}}{\partial N_{j'}\partial N_j} &= \left(\delta_{ji}\delta_{j'k}+\delta_{j'i}\delta_{jk}\right)\frac{Q_{(\alpha,n),k,i}}{N_i N_k} - \frac{\lambda_k N_k}{N_i N}\left(\delta_{ji}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j'}^{(1)}(E_l) + \delta_{j'i}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l)\right) \\ &\quad - \frac{\lambda_k}{N}\left(\delta_{jk}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j'}^{(1)}(E_l) + \delta_{j'k}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l)\right) + 2\frac{\lambda_k N_k}{N^2}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j,j'}^{(2)}(E_l).\end{aligned} \tag{24}$$

Note the symmetry of Eq. (24): j and j' can be reversed, and this symmetry applies to each term individually.

The second derivative of $Q_{(\alpha,n),k,i}$ with respect to the density N_j of isotope j in the material is obtained by setting $j' = j$ in Eq. (24):

$$\frac{\partial^2 Q_{(\alpha,n),k,i}}{\partial N_j^2} = -\delta_{ji}\, 2\frac{\lambda_k N_k}{N_i N}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l) - \delta_{jk}\, 2\frac{\lambda_k}{N}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l) + 2\frac{\lambda_k N_k}{N^2}\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j,j}^{(2)}(E_l). \tag{25}$$

In this case, the first term in Eq. (24) becomes zero because j cannot be both i and k, for physical reasons: α-particle emitters are not (α,n) targets.

The second derivative of the total (α,n) source rate density with respect to the densities of isotopes j and j' is the sum of Eq. (24) over all sources and targets:

$$\frac{\partial^2 Q_{(\alpha,n)}}{\partial N_{j'}\partial N_j} = \sum_k \sum_i \frac{\partial^2 Q_{(\alpha,n),k,i}}{\partial N_{j'}\partial N_j}. \tag{26}$$

2.2. Derivative of the (α,n) Source Rate Spectrum with Respect to Isotopic Density

In [3], a good approximation for the derivative of the (α,n) source rate density in neutron energy group g, $Q_{(\alpha,n),k,i}^{g}$, with respect to the density N_j of isotope j in the material was found to be

$$\frac{\partial Q_{(\alpha,n),k,i}^{g}}{\partial N_j} \approx \frac{Q_{(\alpha,n),k,i}^{g}}{Q_{(\alpha,n),k,i}}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j}. \tag{27}$$

The derivative of $\partial Q_{(\alpha,n),k,i}^{g} / \partial N_j$ with respect to the atom density of another isotope j' is

$$\frac{\partial^2 Q_{(\alpha,n),k,i}^{g}}{\partial N_{j'}\partial N_j} \approx \frac{1}{Q_{(\alpha,n),k,i}}\frac{\partial Q_{(\alpha,n),k,i}^{g}}{\partial N_{j'}}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} - \frac{Q_{(\alpha,n),k,i}^{g}}{Q_{(\alpha,n),k,i}^{2}}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_{j'}}\frac{\partial Q_{(\alpha,n),k,i}}{\partial N_j} + \frac{Q_{(\alpha,n),k,i}^{g}}{Q_{(\alpha,n),k,i}}\frac{\partial^2 Q_{(\alpha,n),k,i}}{\partial N_{j'}\partial N_j}. \tag{28}$$

Using Eq. (27) (with j' instead of j) for $\partial Q_{(\alpha,n),k,i}^{g} / \partial N_{j'}$ in the first term on the right side of Eq. (28) yields

$$\frac{\partial^2 Q_{(\alpha,n),k,i}^{g}}{\partial N_{j'}\partial N_j} \approx \frac{Q_{(\alpha,n),k,i}^{g}}{Q_{(\alpha,n),k,i}}\frac{\partial^2 Q_{(\alpha,n),k,i}}{\partial N_{j'}\partial N_j}, \tag{29}$$

a convenient result. The second derivative of the total (α,n) source rate density in group g with respect to the densities of isotopes j and j' is the sum of Eq. (29) over all sources and targets:

$$\frac{\partial^2 Q^g_{(\alpha,n)}}{\partial N_{j'} \partial N_j} = \sum_k \sum_i \frac{\partial^2 Q^g_{(\alpha,n),k,i}}{\partial N_{j'} \partial N_j}. \tag{30}$$

2.3. Derivative of the (α,n) Source Rate Density with Respect to Material Density

The derivative of the total (α,n) neutron source rate density with respect to the material atom density N is [3]

$$\frac{\partial Q_{(\alpha,n)}}{\partial N} = \frac{1}{N} Q_{(\alpha,n)}. \tag{31}$$

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

$$\frac{\partial^2 Q_{(\alpha,n)}}{\partial N^2} = \frac{\partial}{\partial N}\left(\frac{1}{N}\right) Q_{(\alpha,n)} + \frac{1}{N}\frac{\partial Q_{(\alpha,n)}}{\partial N} = -\frac{1}{N^2} Q_{(\alpha,n)} + \frac{1}{N}\frac{\partial Q_{(\alpha,n)}}{\partial N}. \tag{32}$$

Using Eq. (31) yields

$$\frac{\partial^2 Q_{(\alpha,n)}}{\partial N^2} = 0. \tag{33}$$

The material mass density ρ is related to the material atom density N through

$$N = \rho N_A \sum_{j=1}^{J} \frac{w_j}{A_j}, \tag{34}$$

where w_j and A_j are the weight fraction and atomic weight of isotope j, respectively, and N_A is Avogadro's number. Using Eq. (34) and the chain rule, it can be shown that

$$\frac{\partial^2 Q_{(\alpha,n)}}{\partial \rho^2} = 0. \tag{35}$$

Equations (33) and (35) provide an internal consistency check. The second derivative of the total (α,n) neutron source rate density with respect to N is also

$$\frac{\partial^2 Q_{(\alpha,n)}}{\partial N^2} = \sum_{j=1}^{J} \sum_{j'=1}^{J} \frac{\partial^2 Q_{(\alpha,n)}}{\partial N_j \partial N_{j'}} \frac{N_j N_{j'}}{N^2}. \tag{36}$$

2.4. Comparison with Derivatives of the Spontaneous Fission Source Rate Density

For comparison, the total neutron source rate density due to the spontaneous fission of isotope k is [4]

$$Q_{s.f.,k} = \lambda_k N_k R_k(s.f.), \tag{37}$$

where $R_k(s.f.)$ is the average number of spontaneous-fission neutrons produced per decay of nuclide *k*. The first derivative of $Q_{s.f.,k}$ with respect to the density N_j of isotope *j* is

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

The average number of spontaneous-fission neutrons produced per decay of nuclide *k* does not depend on any atom densities. The derivative of $\partial Q_{s.f.,k} / \partial N_j$ with respect to the atom density of another isotope *j′* is

$$\frac{\partial^2 Q_{s.f.,k}}{\partial N_{j'} \partial N_j} = 0. \tag{39}$$

3. MODIFICATION OF SOURCES4C

Previously [3], SOURCES4C was modified to output $\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j}^{(1)}(E_l)$ for each stopping element for each combination of source isotope *k* and target isotope *i* into a new output file. For second derivatives, SOURCES4C has now been modified to output $\sum_{l=1}^{L} f_{kl}^{\alpha} P_{i,j,j'}^{(2)}(E_l)$ for each combination of source isotope *k* and target isotope *i* and all combinations of isotopes contributing to the stopping power (i.e., all elements).

Also previously [3], SOURCES4C was modified to output the average (α,n) neutron spectrum for each target isotope and (separately) each source isotope as well as the overall average (α,n) neutron spectrum. These averages are no longer used (see Sec. 4), so they are no longer printed. The isotope-dependent spontaneous-fission and (α,n) source rate spectra are now written with more digits in SOURCES4C output files `tape7` and `tape8`.

A bug in which some totals were written to `tape7` instead of `tape8` was identified and corrected.

4. MODIFICATION OF SENSMG

The SENSMG sensitivity code was modified to compute the second derivatives of the (α,n) source rate density with respect to isotopic atom densities in the same place that it computes the first derivatives. The formulas of Sec. 2 were implemented in SENSMG. We are presently implementing in SENSMG second derivatives of the response with respect to cross sections and densities, which will use the results of this paper.

In addition, SENSMG was modified to use the isotope-dependent spontaneous-fission and (α,n) source rate spectra rather than make the approximations described in previous work [3, 8]. The isotope-dependent spectra are read from SOURCES4C output files `tape7` and `tape8`.

5. TEST PROBLEM

The test problem material had the composition shown in Table I. It had a mass density of 19.6 g/cm^3 and an atom density of 9.017136107E-02 at/b·cm. Using 618 neutron energy groups, the SOURCES4C value for the (α,n) neutron source rate density for this material is 1.5773819E+05 neutrons/cm^3/s.

Table I. Test problem material.

Isotope	Weight Fraction	Atom Density (at/b·cm)
^{9}Be	0.02	2.619426605E-02
^{23}Na	0.02	1.026851546E-02
^{239}Pu	0.90	4.443823802E-02
^{242}Pu	0.02	9.752520195E-04
^{58}Ni	0.02	4.074654487E-03
^{56}Fe	0.02	4.220435034E-03

5.1. ^{9}Be

The (α,n) neutron source rate density as a function of the ^{9}Be atom density perturbation is shown in Figure 1. Seven points are plotted and fit to a quadratic polynomial that is used to estimate the first and second derivatives. In the fit equation shown in Figure 1, *x* is the perturbation parameter and the ^{9}Be atom density $N_{^9Be}$ is

$$N_{^9Be} = N_{^9Be,0}(1+x), \tag{40}$$

where subscript 0 represents the initial, unperturbed configuration. Solving for *x* and using the result in a generic quadratic equation for $y = Q_{(\alpha,n)}$ yields

$$Q_{(\alpha,n)} = a\left(\frac{N_{^9Be}}{N_{^9Be,0}}-1\right)^2 + b\left(\frac{N_{^9Be}}{N_{^9Be,0}}-1\right) + c = a\left(\frac{N_{^9Be}}{N_{^9Be,0}}\right)^2 - (2a-b)\left(\frac{N_{^9Be}}{N_{^9Be,0}}\right) + a - b + c. \tag{41}$$

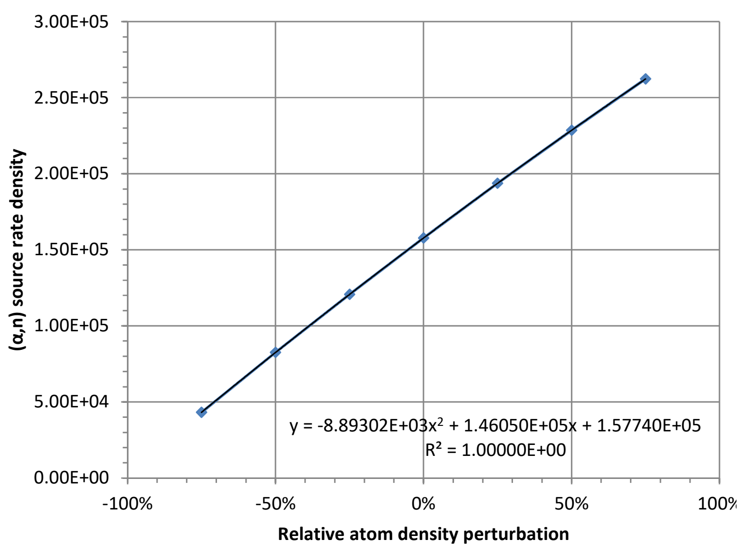


Figure 1. (α,n) neutron source rate density as a function of ^{9}Be atom density perturbation for the six-isotope material.

The derivative of $Q_{(\alpha,n)}$ of Eq. (41) with respect to $N_{^9Be}$ is

$$\frac{\partial Q_{(\alpha,n)}}{\partial N_{^9Be}} = \frac{2a}{N^2_{^9Be,0}} N_{^9Be} - \frac{2a-b}{N_{^9Be,0}}. \tag{42}$$

Evaluating Eq. (42) at $N_{^9Be} = N_{^9Be,0}$ yields

$$\frac{\partial Q_{(\alpha,n)}}{\partial N_{^9Be}} = \frac{b}{N_{^9Be,0}}. \tag{43}$$

From Eq. (42), the second derivative is

$$\frac{\partial^2 Q_{(\alpha,n)}}{\partial N^2_{^9Be}} = \frac{2a}{N^2_{^9Be,0}}. \tag{44}$$

The first and second derivatives computed using the formulas of [3] and Sec. 2 as implemented in SENSMG are compared with those estimated from the fit shown on Figure 1 [using Eqs. (42) and (44)] on Table II. The differences are within 0.23%. The SENSMG derivatives are compared with central difference formulas using perturbations of ±25% in Table III. The differences are within 0.023%. These results highlight the difficulty of estimating numerical derivatives.

Two mixed partial second derivatives of $Q_{(\alpha,n)}$ that include $N_{^9Be}$ are shown in Table IV. The central differences were computed using perturbations of ±50% for both isotopes in each case. The difference of ~0.3% was not analyzed for its dependence on the perturbation.

The spectra $\partial^2 Q^g_{(\alpha,n)} / \partial N^2_{^9Be}$ and $\partial^2 Q^g_{(\alpha,n)} / \partial N_{^9Be} \partial N_{^{58}Ni}$ obtained from Eq. (30) are compared with a central difference in Figure 2. The agreement is excellent.

Table II. Derivatives with Respect to ^{9}Be Atom Density (Quadratic Fit).

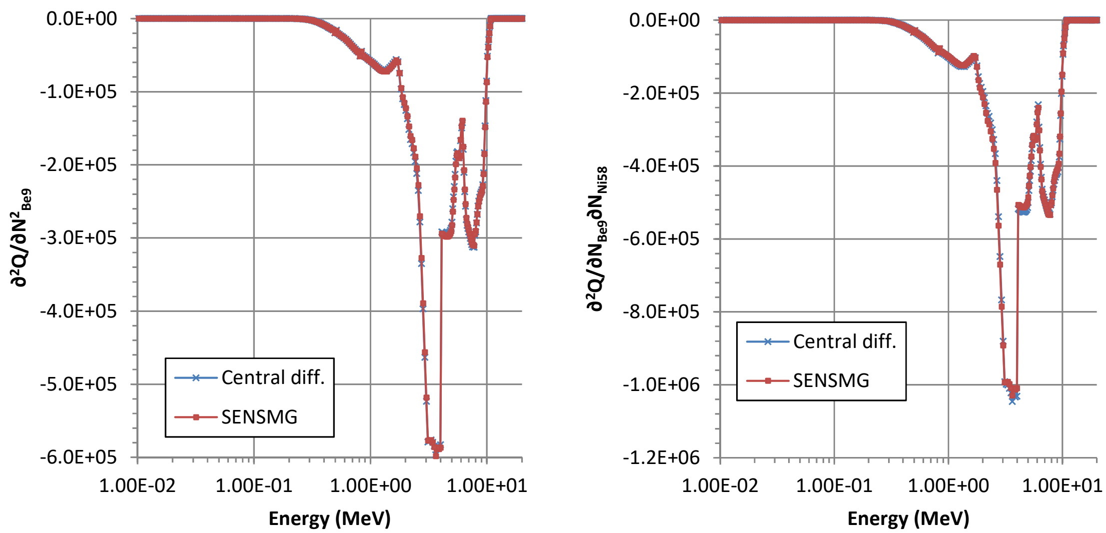
Derivative	Fit	This Paper	Difference
1st	5.57565E+06	5.56662E+06	–0.162%
2nd	–2.59219E+07	–2.58644E+07	–0.222%

Table III. Derivatives with Respect to ^{9}Be Atom Density (Central Difference).

Derivative	Central Diff.	This Paper	Difference
1st	5.56791E+06	5.56662E+06	–0.023%
2nd	–2.58700E+07	–2.58644E+07	–0.021%

Table IV. Mixed Partial Derivatives that Include the ^{9}Be Atom Density.

Derivative	Central Diff.	This Paper	Difference
$\partial^2 Q_{(\alpha,n)} / \partial N_{^9Be} \partial N_{^{242}Pu}$	–8.68608E+07	–8.65902E+07	–0.311%
$\partial^2 Q_{(\alpha,n)} / \partial N_{^9Be} \partial N_{^{58}Ni}$	–4.45812E+07	–4.44401E+07	–0.317%



(a) (b)

Figure 2. (a) $\partial^2 Q^g_{(\alpha,n)} \big/ \partial N^2_{^9Be}$ (b) $\partial^2 Q^g_{(\alpha,n)} \big/ \partial N_{^9Be} \partial N_{^{58}Ni}$.

5.2. ^{58}Ni

The (α,n) neutron source rate density as a function of the ^{58}Ni atom density perturbation is shown in Figure 3. The first and second derivatives computed using the formulas of [3] and Sec. 2 are compared with those estimated from the fit shown on Figure 3 on Table V. The differences are within 0.08%.

Mixed partial second derivatives of $Q_{(\alpha,n)}$ that include $N_{^{58}Ni}$ are shown in Table IV (Sec. 5.1) and Table VI. The central differences were computed using perturbations of ±50% for both isotopes in each case. The difference of ~0.07% for $\partial Q_{(\alpha,n)} \big/ \partial N_{^{58}Ni} \partial N_{^{242}Pu}$ on Table VI was not analyzed for its dependence on the perturbation.

5.3. Material

The (α,n) neutron source rate density as a function of the material atom density perturbation is shown in Figure 4. The first and second derivatives computed using the formulas of [3] and Sec. 2 are compared with those estimated from the fit shown on Figure 4 on Table VII. The first derivative matches almost perfectly. The second derivative has a large difference, over 50%.

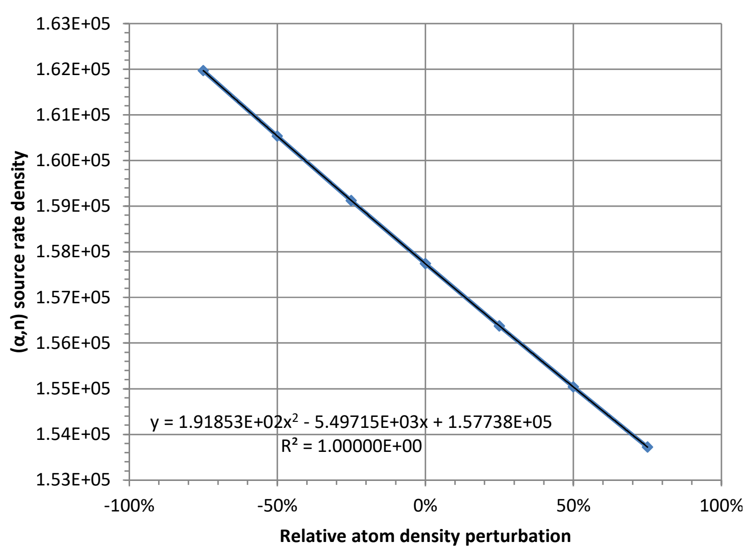


Figure 3. (α,n) neutron source rate density as a function of ^{58}Ni atom density perturbation for the six-isotope material.

Table V. Derivatives with Respect to ^{58}Ni Atom Density (Quadratic Fit).

Derivative	Fit	This Paper	Difference
1st	–1.34911E+06	–1.34839E+06	–0.053%
2nd	2.31109E+07	2.30943E+07	–0.072%

Table VI. Mixed Partial Derivatives that Include the ^{58}Ni Atom Density.

Derivative	Central Diff.	This Paper	Difference
$\partial^2 Q_{(\alpha,n)} \big/ \partial N_{^{58}Ni} \partial N_{^{242}Pu}$	4.68014E+07	4.67704E+07	–0.066%

Why does the second derivative of Eq. (36) not match the derivative from the quadratic fit? Recall that the second derivative of $Q_{(\alpha,n)}$ with respect to material density is zero [Eq. (33)]. By this measure, both second derivatives shown on Table VII are much too large! However, it is appropriate to compare the first and second derivatives. A second-order Taylor series for the (α,n) source rate density with respect to the material atom density is

$$Q_{(\alpha,n)}(N) = Q_{(\alpha,n)}(N_0) + \frac{\partial Q_{(\alpha,n)}}{\partial N}(N - N_0) + \frac{1}{2}\frac{\partial^2 Q_{(\alpha,n)}}{\partial N^2}(N - N_0)^2. \tag{45}$$

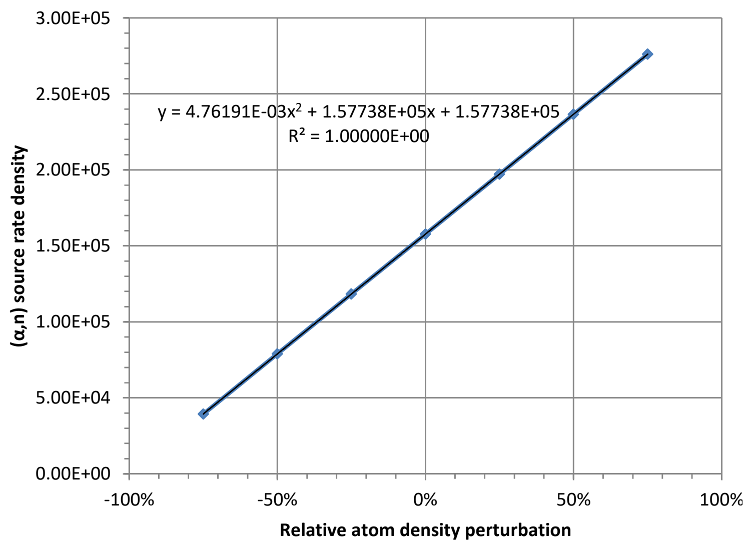


Figure 4. (α,n) neutron source rate density as a function of material atom density perturbation for the six-isotope material.

The ratio of the second-order term to the first-order term for a doubling of the atom density ($N = 2N_0$) is

$$\frac{\frac{1}{2}\frac{\partial^2 Q_{(\alpha,n)}}{\partial N^2}(2N_0 - N_0)^2}{\frac{\partial Q_{(\alpha,n)}}{\partial N}(2N_0 - N_0)} = \frac{\frac{N_0}{2}\frac{\partial^2 Q_{(\alpha,n)}}{\partial N^2}}{\frac{\partial Q_{(\alpha,n)}}{\partial N}}, \tag{46}$$

or 4.56×10^{-8} using the values of Table VII ("This Paper") and the material atom density, 9.017136107E-02 at/b·cm. Thus, the second-order term is effectively zero compared to the first-order term.

Table VII. Derivatives with Respect to Material Atom Density (Quadratic Fit).

Derivative	Fit	This Paper	Difference
1st	1.74931E+06	1.74932E+06	0.000%
2nd	1.17132E+00	1.77032E+00	51.139%

We conjecture that round-off error and other numerical imprecisions cause the second derivative to be non-zero.

6. CONCLUSIONS

This paper has derived the second derivative (including mixed partial derivatives) of an (α,n) neutron source rate density and spectrum with respect to constituent isotope densities and material density. (The first derivative was given previously [3].) The derivation is based on the (α,n) neutron source rate density and spectrum equations implemented in SOURCES4C [4]. The second derivatives have been implemented in the SENSMG multigroup sensitivity code [8]. SOURCES4C was also modified to print data useful for computing second derivatives.

In a simple six-isotope test problem, results from the equations derived in this paper (implemented in SENSMG) compared very well with results computed using numerical derivatives (central differences).

This work has been undertaken so that SENSMG can compute the second derivative of the neutron leakage with respect to isotope densities. We are actively working on other pieces of this project.

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