Application of Depletion Perturbation Theory for Sensitivity Analysis in the High Flux Isotope Reactor

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https://dx.doi.org/10.13182/T31230

INTRODUCTION

Oak Ridge National Laboratory's (ORNL) Standardized Computer Analyses for Licensing Evaluation (SCALE) code system [1] has long had the capability to perform nuclear data adjustment/ uncertainty reduction for criticality experiments. In this work, we expand this capability to perform nuclear data adjustments for activation experiments. This is accomplished using the prototype SCALE module ORSEN [2], which applies depletion perturbation theory (DPT) [3] to nuclear data to calculate the relative sensitivities of post-irradiation nuclide concentrations. The DPT approach uses an adjoint differentiation method to calculate the sensitivities that are required by the nuclear data adjustment codes. ORSEN applies DPT by performing forward and adjoint transmutation calculations in SCALE's Oak Ridge Isotope Generation (ORIGEN) code [4]. Here we present an application of ORSEN to determine sensitivities for ^{226}Ra targets irradiated in ORNL's High Flux Isotope Reactor (HFIR).

MULTIGROUP SENSITIVITY COEFFICIENTS

Depletion Perturbation Theory to Generate Multigroup Sensitivity Coefficients

The ORIGEN code in SCALE solves the *transmutation equation*, the set of coupled differential equations defining the time-dependent concentrations of nuclides in a sample undergoing irradiation and decay. An individual member of this set, describing the time-dependent concentration of nuclide i, is

$$\frac{dN_i}{dt} = \sum_{j \neq i} \left(l_{ij}\lambda_j + f_{ij}\sigma_j\phi\right)N_j(t) - (\lambda_i + \sigma_i\phi)N_i(t) + S_i(t) \quad (1)$$

where

$N_i =$ amount of nuclide i (atoms),
$\lambda_i =$ decay constant of nuclide i (1/s),
$l_{ij} =$ fractional yield of nuclide i from decay of nuclide j,
$f_{ij} =$ fractional yield of nuclide i from neutron-induced removal of nuclide j,
$\phi =$ space- and energy-averaged time-dependent neutron flux (neutrons/cm^2-s),
$S_i =$ time-dependent source/feed term (atoms/s),
$\sigma_i =$ spectrum-averaged removal cross section for nuclide i (cm^2). These are obtained by collapsing problem-independent multigroup cross sections, $\sigma_{i,g}$, with a neutron multigroup flux, $\phi_{i,g}$:

$$\sigma_i = \frac{\sum_g \sigma_{i,g}\phi_{i,g}}{\sum_g \phi_{i,g}}, \quad (2)$$

The transmutation equation (1) can be expressed in matrix notation as

$$\frac{d\mathbf{N}}{dt} = \mathbf{M}(\alpha)\mathbf{N}(t) + \mathbf{S}(t)\,, \quad (3)$$

where $\mathbf{N}$ and $\mathbf{S}$ are vectors containing nuclide quantities and source terms, respectively, and $\mathbf{M}(\alpha)$, called the *transition matrix*, is a matrix containing the nuclear data relevant to the problem.

The relative sensitivity coefficient $S_\alpha^{(R)}$ of a particular response R to a nuclear data parameter α is

$$S_\alpha^{(R)} = \frac{\alpha}{R}\frac{\partial R}{\partial \alpha}\,; \quad (4)$$

$$\frac{\partial R}{R} = S_\alpha^{(R)}\frac{\partial \alpha}{\alpha}. \quad (5)$$

Eq. (5) is used to determine the percent change in a response caused by a 1% change in the data parameter α. In this work, the response is a measured post-irradiation and decay measurement of nuclide concentration, and α is a reaction cross section (σ_i), decay constant (λ_i), branching ratio (l_{ij}), or yield from fission or other removal processes (f_{ij}).

The DPT equation for $S_\alpha^{(R)}$is

$$S_{\alpha}^{(R)} = \frac{\alpha}{R}\int_0^{T_F} N_R^*(t)\frac{\partial \mathbf{M}(\alpha)}{\partial \alpha}N(t)dt, \quad (6)$$

where T_F is the total time of the irradiation/decay and $N_R^*(t)$ is the solution of the adjoint depletion equation at time t. The adjoint equation is solved in ORIGEN using the Chebyshev rational approximation method and stepping backward through each time step [5][6].

The transition matrix contains one-group cross section values collapsed from multigroup values according to Eq. (2). This multigroup cross-section data can be used to generate multigroup sensitivity coefficients. The multigroup sensitivity coefficient is given by

$$S_{\sigma_{i,g}}^{(R)} = \frac{\sigma_{i,g}}{R}\frac{\partial R}{\partial \sigma_{i,g}}. \quad (7)$$

The derivative on the right side of Eq. (7) is evaluated by the chain rule:

$$\frac{\partial R}{\partial \sigma_{i,g}} = \frac{\partial R}{\partial \sigma_i}\frac{\partial \sigma_i}{\partial \sigma_{i,g}} \quad (8)$$

Using Eq. (2) we have

$$\frac{\partial \sigma_i}{\partial \sigma_{i,g}} = \frac{\phi_{i,g}}{\sum_g \phi_{i,g}}, \quad (9)$$

so that

$$S_{\sigma_{i,g}}^{(R)} = \frac{\sigma_{i,g}}{R}\frac{\partial R}{\partial \sigma_i}\frac{\phi_{i,g}}{\sum_g \phi_{i,g}}. \quad (10)$$

Inserting Eq. (4) into Eq. (10), we obtain

$$S_{\sigma_{i,g}}^{(R)} = S_{\sigma_i}^{(R)}\frac{\sigma_{i,g}}{\sigma_i}\frac{\phi_{i,g}}{\sum_g \phi_{i,g}}. \quad (11)$$

Finally, we use Eq. (2) again to arrive at

$$S_{\sigma_{i,g}}^{(R)} = S_{\sigma_i}^{(R)}\frac{\sigma_{i,g}\phi_{i,g}}{\sum_g \sigma_{i,g}\phi_{i,g}}. \quad (12)$$

Eq. (12) is slightly more convenient than Eq. (11) because it requires tallying only energy-dependent reaction rates ($\sigma_{i,g}\phi_{i,g}$) in a Monte Carlo simulation.

The derivative terms $\partial\mathbf{M}(\alpha)/\partial\alpha$ in Eq. (6) can be determined by considering the individual terms of **M**:

$$M_{ij} = \begin{cases} l_{ij}\lambda_j + f_{ij}\sigma_j\phi, & i \neq j \\ -\lambda_i - \sigma_i\phi & \text{otherwise.} \end{cases} \quad (13)$$

If, for example, $\alpha = \sigma_{n,\gamma}^{Ra-226}$ (radiative capture cross section of ^{226}Ra), then all terms M_{ij} not containing $\sigma_{n,\gamma}^{Ra-226}$ have derivatives equal to zero. The remaining terms are

$$\frac{\partial M_{ij}}{\partial \sigma_{n,\gamma}^{Ra-226}} = \begin{cases} f_{ij}\phi, & i \neq j \\ -\phi & \text{otherwise.} \end{cases} \quad (14)$$

RESULTS FOR IRRADIATION OF ^{226}Ra

Radium-226 is used as a target material to produce medical isotopes actinium-227, thorium-228, and thorium-229 in HFIR. This demonstration examines a 9-day irradiation performed in 2014 of microgram amounts of ^{226}Ra in an effectively infinitely dilute configuration.

Multigroup neutron flux and cross sections at the target location that were used for the ORSEN analyses were calculated in a 252-group structure using a 3-D high-fidelity representative model of the HFIR [7]. The 252-group energy structure is the same as that used in SCALE.

DPT was used to calculate one-group and multigroup sensitivity coefficients of the amount of ^{228}Th produced to the fission and radiative capture (n,γ) cross sections of the nuclides shown in Fig. 1, which shows the dominant paths to production of ^{228}Th during irradiation of ^{226}Ra. To verify the accuracy of the DPT approach, these results were compared to direct perturbation calculations. Table I shows that the DPT results were highly consistent with direct perturbation, matching to better than 99% for all reactions considered (the table shows five reactions chosen for importance; DPT calculated the sensitivities for over 90 reactions). As expected, there is a strong positive sensitivity to radiative capture in ^{226}Ra, as this reaction must occur in all the dominant paths to ^{228}Th. Because this is only a 9-day irradiation and the half-life for decay from ^{228}Ac to ^{228}Th is very short compared to the half-life for decay from ^{227}Ac to ^{227}Th, the production of ^{228}Th is much more sensitive to radiative capture in ^{227}Ac than radiative capture in ^{227}Th. Also, as expected, there are negative sensitivities to radiative capture and fission of ^{228}Th, both of which remove ^{228}Th from the final sample.

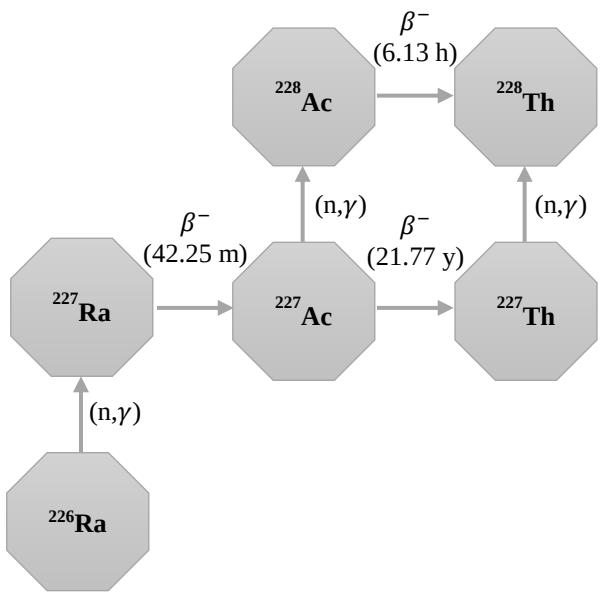

Fig. 1. Dominant paths for production of ^{228}Th during irradiation of ^{226}Ra.

TABLE I. Comparison of DPT and Direct Perturbation Results of Sensitivity of ^{228}Th Concentration to Select Nuclides/Reactions

Nuclide/ Reaction	DPT Sensitivity	Direct Perturbation	Difference (%)
^{226}Ra (n,γ)	0.985005	0.984999	0.001
^{227}Ac (n,γ)	0.705336	0.705743	−0.577
^{227}Th (n,γ)	0.000196	0.000195	0.513
^{228}Th (n,γ)	−0.072635	−0.072633	0.003
^{228}Th Fission	−0.000161	−0.000161	0.000

A comparison of computational burdens shows the advantage of the DPT approach. ORSEN, which requires only one forward and one adjoint calculation, took 54 seconds to calculate sensitivities of ^{228}Th concentration for 93 reactions. Direct perturbation requires one (forward differencing) or two (central differencing) forward transport calculations *per reaction*. The direct perturbation method using forward differencing required 87 minutes to calculate the 93 sensitivities.

The one-group sensitivities were converted to a 252-group structure using Eq. (12). The sensitivity of the concentration of ^{228}Th to the radiative capture cross section is given as a function of energy in Fig. 2 and the normalized neutron flux at the target location is illustrated in Fig. 3. Comparison of Fig. 2 to Fig. 3 shows that the region of largest sensitivity corresponds to the region of highest flux. A large drop in sensitivity around 30 eV can be understood by considering Fig. 4, which shows the ^{227}Ac radiative capture cross section as a function of energy. The large decrease in sensitivity around 30 eV corresponds to a large drop in the ^{227}Ac cross section.

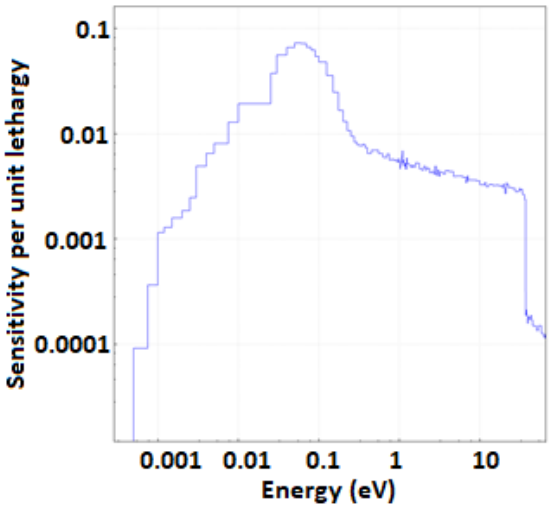

Fig. 2. Sensitivity of post-irradiation ^{228}Th concentration to the radiative capture cross section of ^{227}Ac as a function of energy.

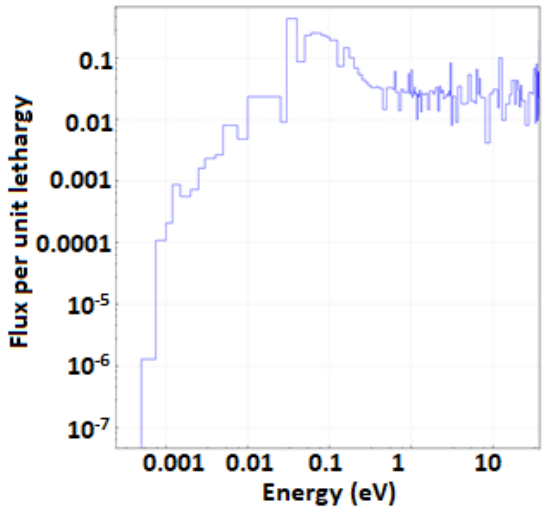

Fig. 3. Normalized flux at target locations as a function of energy.

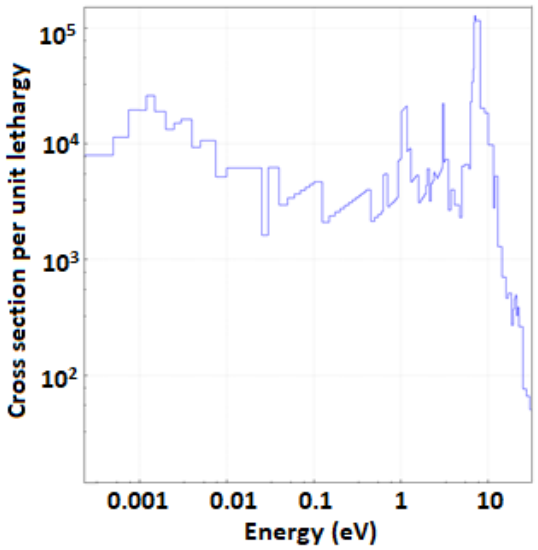

Fig. 4. ^{227}Ac radiative capture cross section as a function of energy.

CONCLUSIONS

DPT capabilities in the ORSEN prototype module in SCALE have been applied to determine relative sensitivity coefficients of nuclide concentrations to nuclear data in a model of an activation experiment performed in HFIR. DPT results match direct perturbation to better than 99% for all considered reactions of the analyzed experiment. Sensitivity results have been extended from one-group to multiple groups using multigroup fluxes at the target locations as calculated with a 3-D high-fidelity HFIR model. As expected, sensitivities are largest in energy groups where HFIR flux and target cross sections are both higher. In future work, the sensitivities calculated by DPT in ORSEN will serve to inform nuclear data adjustment/uncertainty reduction using the generalized linear least squares methodology in SCALE's TSURFER module.

ACKNOWLEDGEMENT

Research sponsored by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U. S. Department of Energy.

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