Fission Product Yield Data Adjustment in a Prototype Version of TSURFER

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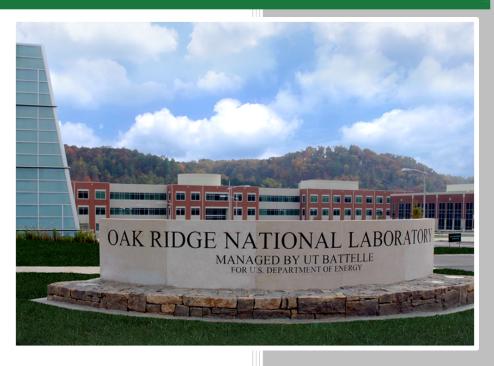MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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ABSTRACT

The TSURFER (Tool for Sensitivity/Uncertainty analysis of Response Functionals using Experimental Results) module of Oak Ridge National Laboratory's (ORNL's) SCALE code system has been updated to perform nuclear data adjustments for fixed-source irradiation/depletion problems. TSURFER uses a generalized linear least squares (GLLS) approach to consolidate a prior set of measured responses and corresponding calculated values to create the most self-consistent set of nuclear data. Traditionally, TSURFER adjustments have been performed for multigroup nuclear data such as reaction cross sections. In this work, TSURFER is expanded to perform adjustments to independent fission product yields and branching ratios that need equality constraints. To preserve equality constraints after the data adjustment procedure, an updated GLLS formulation includes a new Lagrange multiplier that forces data adjustment to sum to 0 for a given fission yield/branching ratio parent. A test problem illustrates that the newly updated TSURFER module satisfies the required constraint that adjustments for fission yield data sum to 0.

1. INTRODUCTION

The TSURFER) module of the ORNL's SCALE code system [1] uses the generalized linear least squares (GLLS) approach to reduce discrepancies between the measured and calculated responses by adjusting nuclear data and experimental values [2]. Traditionally, TSURFER has only been applied to criticality experiments. As a part of the ORNL Laboratory Directed Research and Development project "Establishing the High Flux Isotope Reactor as a Unique Nuclear Data Measurement Facility," TSURFER was recently extended to perform data adjustment for fixed-source irradiation/depletion experiments. For these problems, maximum consistency between simulation and experiment requires the adjustment of fission product yield and branching ratio data. However, yields and branching ratios cannot be adjusted independently because they must satisfy equality constraints—the branching ratios for a given parent isotope must sum to 1.0, and the fission yields for a given parent isotope must sum to 2.0 (for a description of the fission yield sum see Ref. [3]). In this work we derive a new expression for the GLLS adjustment in which new Lagrange multiplier terms are introduced into the GLLS formulation to ensure that for a given parent, the sum of the adjustments is 0. By ensuring the adjustment sum to 0, we ensure that the equality constraints remain satisfied.

Section 2 reviews the standard TSURFER GLLS methodology, and Section 3 presents the new methodology that ensures that equality constraints are satisfied after the data adjustment. A numerical case to test the new GLLS methodology is reported in Section 4.

2. STANDARD GLLS METHODOLOGY

In this section we present a brief review of standard TSURFER GLLS methodology. This follow the derivation given in Ref. [2]. A more thorough review is given in Ref. [3]. The goal of the GLLS method in TSURFER is to adjust both the nuclear data from α to α' and the measured integral responses from m to m' such that they are most consistent with their respective uncertainty matrices, $C_{\alpha\alpha}$ and C_{mm}. According to the chi-squared metric (χ^2), this is done by minimizing the relation

$$\chi^2 = \Delta\alpha^T C_{\alpha\alpha}^{-1}\Delta\alpha + \Delta m^T C_{mm}^{-1}\Delta m, \tag{1}$$

where $\Delta\alpha$ is an $n \times 1$ vector containing the adjustments to the n nuclear data, $C_{\alpha\alpha}$ is an $n \times n$ covariance matrix of nuclear data, Δm is an $I \times 1$ vector of adjustments to the I measured responses, and C_{mm} is an $I \times I$ covariance matrix.

Let k be an $I \times 1$ vector of calculated responses (in the GLLS process these will also be adjusted from k to k'). A unique solution to the minimization of χ^2 can be obtained if we enforce the constraint that the adjusted calculated and measured responses agree ($k' = m'$) and we satisfy the linearity condition $\Delta k = S_{k\alpha}\Delta\alpha$, where Δk is an $I \times 1$ vector of calculated response adjustments. The linearity condition is enforced using a Lagrange multiplier:

$$\chi^2 = \Delta\alpha^T C_{\alpha\alpha}^{-1}\Delta\alpha + \Delta m^T C_{mm}^{-1}\Delta m + 2\lambda_1(S_{k\alpha}\Delta\alpha - \Delta k). \tag{2}$$

The constraint that adjusted responses agree ($k' = m'$) means that

$$\Delta k = F_{m/k}\Delta m - d, \tag{3}$$

where $F_{m/k}$ is a diagonal $I \times I$ matrix with ratios of m_i/k_i; $i = 1,...,I$; and d is the discrepancy vector

$$d = \left\{\frac{k_i - m_i}{k_i}, i = 1,...,I. \tag{4}$$

The linearity condition can therefore be expressed as $d = F_{m/k}\Delta m - S_{k\alpha}\Delta\alpha$, and χ^2 can be written as

$$\chi^2 = \Delta\alpha^T C_{\alpha\alpha}^{-1}\Delta\alpha + \Delta m^T C_{mm}^{-1}\Delta m + 2\lambda_1\left(S_{k\alpha}\Delta\alpha - F_{m\backslash k}\Delta m + d\right). \tag{5}$$

To minimize χ^2, we take the derivative of χ^2 with respect to $\Delta\alpha$ and Δm and set them to 0:

$$\frac{\partial\chi^2}{\partial\Delta\alpha} = 0 = 2C_{\alpha\alpha}^{-1}\Delta\alpha + 2S_{k\alpha}^T\lambda_1^T, \tag{6}$$

$$\frac{\partial\chi^2}{\partial\Delta m} = 0 = 2C_{mm}^{-1}\Delta m - 2F_{\frac{m}{k}}\lambda_1^T. \tag{7}$$

We will also use the relative uncertainty matrix of the discrepancy vector d, which can be obtained by standard propagation of error assuming no correlation between k and m:

$$C_{dd} = S_{k\alpha}C_{\alpha\alpha}S_{k\alpha}^T + F_{m/k}C_{mm}F_{m/k}. \tag{8}$$

Here $S_{k\alpha}$ is a vector of sensitivities of response k to nuclear data α. This comes from a sensitivity analysis tool such as ORSEN, meaning all terms on the right side of Eq. (8) are known.

From Eq. (6) we have

$$\Delta\alpha = -C_{\alpha\alpha}S_{k\alpha}^T\lambda_1^T, \tag{9}$$

and from Eq. (7) we have

$$\Delta m = C_{mm}F_{m/k}\lambda_1^T. \tag{10}$$

Inserting Eqs. (9) and (10) into Eq. (8) yields

$$C_{dd} = -S_{k\alpha}\Delta\alpha\left(\lambda_1^T\right)^{-1} + F_{m/k}\Delta m\left(\lambda_1^T\right)^{-1}, \quad \Rightarrow \lambda_1^T = C_{dd}^{-1}\left(F_{\frac{m}{k}}\Delta m - S_{k\alpha}\Delta\alpha\right). \tag{11}$$

Using the linearity condition, this becomes

$$\lambda_1^T = C_{dd}^{-1}d. \tag{12}$$

We can now express $\Delta\alpha$ and Δm in terms of known quantities:

$$\Delta\alpha = -C_{\alpha\alpha}S_{k\alpha}^T C_{dd}^{-1}d \tag{13}$$

$$\Delta m = C_{mm}F_{m/k}C_{dd}^{-1}d \tag{14}$$

3. EXPANDED GLLS METHODOLOGY

Before GLLS adjustment, fission product yields for a given parent isotope sum to 2.0:

$$\sum_i Y_i = 2.0 \tag{15}$$

After GLLS adjustment, the fission product yields become $Y_i' = Y_i + \Delta Y_i$. The sum of the updated fission product yields is

$$\sum_i Y_i' = \sum_i Y_i + \Delta Y_i = \sum_i Y_i + \sum_i \Delta Y_i = 2.0 + \sum_i \Delta Y_i. \tag{16}$$

Equation (16) shows that if the sum of adjustments equals 0 (i.e., $\sum_i \Delta Y_i = 0$), then the adjusted fission product yields still sum to 2.0. Similarly, post-adjustment branching ratios will sum to 1.0 if the individual adjustments sum to 0.

When we add a constraint that some of the nuclear data must sum to a certain constant (1.0 for branching ratio, 2.0 for fission yields), we add a second Lagrange multiplier to χ^2:

$$\chi^2 = \Delta\alpha^T C_{\alpha\alpha}^{-1}\Delta\alpha + \Delta m^T C_{mm}^{-1}\Delta m + 2\lambda_1\left(S_{k\alpha}\Delta\alpha - F_{m\backslash k}\Delta m + d\right) + 2\lambda_2(h\Delta\alpha), \tag{17}$$

where h is an $M \times n$ matrix and M is the number of fission yield plus branching ratio parents. Each row of h contains either 1s or 0s. The constraint that $h\Delta\alpha = 0$ forces the adjustments for fission yields and branching ratios to sum to 0, thus maintaining their initial constraints (i.e., if the prior values summed to 2.0, the adjusted values will still sum to 2.0).

The derivatives of χ^2 become

$$\frac{\partial\chi^2}{\partial\Delta\alpha} = 0 = 2C_{\alpha\alpha}^{-1}\Delta\alpha + 2S_{k\alpha}^T\lambda_1^T + 2h^T\lambda_2^T, \tag{18}$$

and

$$\frac{\partial\chi^2}{\partial\Delta m} = 0 = 2C_{mm}^{-1}\Delta m + 2F_{m/k}\lambda_1^T. \tag{19}$$

Solving Eq. (18) for $\Delta\alpha$ yields

$$\Delta\alpha = -C_{\alpha\alpha}\left(S_{k\alpha}^T\lambda_1^T + h^T\lambda_2^T\right), \tag{20}$$

and solving Eq. (19) for Δm yields

$$\Delta m = C_{mm} F_{m/k} \lambda_1^T. \tag{21}$$

From Eq. (20), we get

$$C_{\alpha\alpha} S_{k\alpha}^T = \left(\Delta\alpha + h^T \lambda_2^T\right)\left(\lambda_1^T\right)^{-1}, \tag{22}$$

and from Eq. (21) we get

$$C_{mm} F_{m/k} = \Delta m \left(\lambda_1^T\right)^{-1}. \tag{23}$$

Inserting these results into Eq. (8) yields

$$C_{dd} = -S_{k\alpha}\left(\Delta\alpha + h^T \lambda_2^T\right)\left(\lambda_1^T\right)^{-1} + F_{m/k}\Delta m \left(\lambda_1^T\right)^{-1}. \tag{24}$$

From the linearity condition $F_{m/k}\Delta m - \Delta k = F_{m/k}\Delta m - S_{k\alpha}^T \Delta\alpha = d$, we get

$$C_{dd} = \left(d - S_{k\alpha} h^T \lambda_2^T\right)\left(\lambda_1^T\right)^{-1}, \tag{25}$$

$$\Rightarrow \lambda_1^T = C_{dd}^{-1}\left(d - S_{k\alpha} h^T \lambda_2^T\right). \tag{26}$$

Now using the condition that $h\Delta\alpha = 0$,

$$h\Delta\alpha = 0 = -hC_{aa}\left(S_{k\alpha}^T \lambda_1^T + h^T \lambda_2^T\right) = -hC_{aa}\left(S_{k\alpha}^T C_{dd}^{-1}(d - S_{k\alpha} h^T \lambda_2^T) + h^T \lambda_2^T\right)$$

$$= -hC_{aa} S_{k\alpha}^T C_{dd}^{-1} d + hC_{aa} S_{k\alpha}^T C_{dd}^{-1} S_{k\alpha} h^T \lambda_2^T - hC_{\alpha\alpha} h^T \lambda_2^T. \tag{27}$$

(Note, $C_{aa} S_{k\alpha}^T C_{dd}^{-1} d$ is the adjustment $\Delta\alpha$ for the unconstrained problem.) Solving for λ_2^T yields

$$\lambda_2^T = \left(hC_{aa} S_{k\alpha}^T C_{dd}^{-1} S_{k\alpha} h^T - hC_{\alpha\alpha} h^T\right)^{-1} hC_{\alpha\alpha} S_{k\alpha}^T C_{dd}^{-1} d. \tag{28}$$

λ_2^T is now expressed in terms of known quantities. With this, we can determine λ_1, $\Delta\alpha$, and Δm.

Note, the possibility for simplification is based on the form of the covariance matrix of the data. If $C_{\alpha\alpha}$ has rows and columns that sum to 0 (in the locations where h contains 1s) then $h^T C_{\alpha\alpha} = 0$ and thus $\lambda_2 = 0$, $\lambda_1 = C_{dd}^{-1} d$. The adjustments for $\Delta\alpha$ and Δm are the same as in standard TSURFER. This row/column sum zero approach has been used for equality-constrained GLLS in the past [5], but as noted in Ref. [4], obtaining a covariance matrix of this form for fission product yields is generally not possible.

4. NUMERICAL TEST CASE

The procedure for obtaining covariance matrices for independent fission product yields and related constraints is detailed in Refs. [4,6]. Here those covariance matrices are used with the updated GLLS approach to perform fission product yield data adjustment using "measured" data from two simulated irradiation experiments. Fission yield covariance data are currently available at three incident neutron energies, 0.0253, 500, and 1.4×10^7 eV; however, in these calculations we used only the 500 eV covariance data. In future work we will include extrapolation between files to generate covariance data to match the average fission energy of the simulation.

For this test case, two experiments were simulated using the ORIGEN module of SCALE. In the first simulation (Experiment 1) a mixture of 1,000 g of ^{235}U and 500 g of ^{238}Np was irradiated for 12 hours with a flux of 3.77 × 10^{12} n/cm^{2}s and allowed to decay for 12 hours. In the second simulation (Experiment 2), a 1,000 g sample of ^{235}U was irradiated for 12 hours with the same flux and allowed to decay for 12 hours. The amount of ^{135}Xe was the quantity measured at the end of the decay time. At the end of the Experiment 1, 0.00175 g of ^{135}Xe was present, and at the end the Experiment 2, 0.00068 g of ^{135}Xe was present. Measured values were then created by adding a random noise term to these simulated values. This resulted in "measured" values of 0.00161 g of ^{135}Xe for Experiment 1 and 0.00071 g of ^{135}Xe for the Experiment 2.

The sensitivities of the final ^{135}Xe masses with respect to fission yield data were calculated using the depletion perturbation theory method in a prototype ORIGEN sensitivity module [7]. The largest sensitivities are given in Table 1.

Table 1. Sensitivities of final mass ^{135}Xe to fission product yields.

Reaction	Experiment 1 Sensitivities	Experiment 2 Sensitivities
^{235}U → ^{135}Sb	7.04 × 10^{-3}	1.81 × 10^{-2}
^{235}U → ^{135}Te	1.87 × 10^{-1}	4.81 × 10^{-1}
^{235}U → ^{135}I	1.74 × 10^{-1}	4.48 × 10^{-1}
^{235}U → ^{135}Xe	1.26 × 10^{-2}	3.23 × 10^{-2}
^{235}U → ^{136}Sn	7.64 × 10^{-7}	1.96 × 10^{-6}
^{235}U → ^{136}Sb	3.41 × 10^{-4}	8.76 × 10^{-4}
^{235}U → ^{136}Te	1.04 × 10^{-4}	2.66 × 10^{-3}
^{235}U → ^{137}Sb	5.45 × 10^{-7}	1.40 × 10^{-6}
^{235}U → ^{137}Sn	5.85 × 10^{-5}	1.50 × 10^{-4}
^{238}Np → ^{135}Sb	2.27 × 10^{-2}	< 10^{-10}
^{238}Np → ^{135}Te	3.36 × 10^{-1}	< 10^{-10}
^{238}Np → ^{135}I	2.40 × 10^{-1}	< 10^{-10}
^{238}Np → ^{135}Xe	1.68 × 10^{-3}	< 10^{-10}
^{238}Np → ^{136}Sn	3.33 × 10^{-6}	< 10^{-10}
^{238}Np → ^{136}Sb	2.27 × 10^{-3}	< 10^{-10}
^{238}Np → ^{136}Te	2.49 × 10^{-3}	< 10^{-10}
^{238}Np → ^{137}Sb	4.17 × 10^{-8}	< 10^{-10}
^{238}Np → ^{137}Sn	5.12 × 10^{-6}	< 10^{-10}
^{238}Pu → ^{135}Sb	7.27 × 10^{-6}	< 10^{-10}
^{238}Pu → ^{135}Te	3.63 × 10^{-4}	< 10^{-10}
^{238}Pu → ^{135}I	6.55 × 10^{-4}	< 10^{-10}
^{238}Pu → ^{135}Xe	3.39 × 10^{-5}	< 10^{-10}
^{238}Pu → ^{136}Sb	3.90 × 10^{-7}	< 10^{-10}
^{238}Pu → ^{136}Te	1.67 × 10^{-6}	< 10^{-10}

Table 2 shows the absolute adjusted values for fission product yields. Note, for the individual parent isotopes, the adjustments sum to 0 as required.

Table 2. Absolute adjusted value of fission product yields.

Fission Yield	Adjustment
^{235}U Fission Yields	
$^{235}\text{U} \rightarrow {}^{135}\text{Sb}$	0.2414044
$^{235}\text{U} \rightarrow {}^{135}\text{Te}$	−0.1862993
$^{235}\text{U} \rightarrow {}^{135}\text{I}$	−0.0889851
$^{235}\text{U} \rightarrow {}^{135}\text{Xe}$	0.0175893
$^{235}\text{U} \rightarrow {}^{136}\text{Sn}$	0.0025497
$^{235}\text{U} \rightarrow {}^{136}\text{Sb}$	0.0103145
$^{235}\text{U} \rightarrow {}^{136}\text{Te}$	−0.0013855
$^{235}\text{U} \rightarrow {}^{137}\text{Sb}$	0.0025287
$^{235}\text{U} \rightarrow {}^{137}\text{Sn}$	0.0022834
Sum of Adjustments	**0.0000000**
^{238}Np Fission Yields	
$^{238}\text{Np} \rightarrow {}^{135}\text{Sb}$	0.0502742
$^{238}\text{Np} \rightarrow {}^{135}\text{Te}$	−0.1670862
$^{238}\text{Np} \rightarrow {}^{135}\text{I}$	−0.0139868
$^{238}\text{Np} \rightarrow {}^{135}\text{Xe}$	0.0221285
$^{238}\text{Np} \rightarrow {}^{136}\text{Sn}$	0.0206014
$^{238}\text{Np} \rightarrow {}^{136}\text{Sb}$	0.0230141
$^{238}\text{Np} \rightarrow {}^{136}\text{Te}$	0.0239119
$^{238}\text{Np} \rightarrow {}^{137}\text{Sb}$	0.0206002
$^{238}\text{Np} \rightarrow {}^{137}\text{Sn}$	0.0205426
Sum of Adjustments	**0.0000000**
^{238}Pu Fission Yields	
$^{238}\text{Pu} \rightarrow {}^{135}\text{Sb}$	0.0001254
$^{238}\text{Pu} \rightarrow {}^{135}\text{Te}$	−0.0000690
$^{238}\text{Pu} \rightarrow {}^{135}\text{I}$	−0.0004430
$^{238}\text{Pu} \rightarrow {}^{135}\text{Xe}$	0.0001337
$^{238}\text{Pu} \rightarrow {}^{136}\text{Sb}$	0.0001215
$^{238}\text{Pu} \rightarrow {}^{136}\text{Te}$	0.0001313
Sum of Adjustments	**0.0000000**

5. SUMMARY AND FUTURE WORK

A new prototype version of SCALE's TSURFER module has been created to perform nuclear data adjustments for fixed source irradiation/depletion problems. This update was needed because fission product yields and branching ratios must satisfy equality constraints after the adjustment. By introducing a new Lagrange multiplier term into the GLLS method used by TSURFER, we ensured that the equality constraints remain satisfied. This was verified using a numerical example.

Branching ratio covariance data are not currently available and thus could not be used in the numerical example. Generating this data will be the subject of future work.

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