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memorandum

Computational Physics
XCP–8

Activation Product Inverse Calculations with NDI

Summary

`NDI` based forward calculations of activation product concentrations can be systematically used to infer structural element concentrations from measured activation product concentrations with an iterative algorithm. The algorithm converges exactly for the `basic` production-depletion chain with explicit activation product production and approximately, in the least-squares sense, for the `full` production-depletion chain with explicit activation product production and `nosub` production-depletion chain. The algorithm is suitable for automation.

Background

In the previous memo[1] I described how `NDI`'s `ENDF/B` based and benchmarked neutron reaction data could be used with its `nosub` or `full` production-depletion chains to calculate activation product concentrations given initial nuclide concentrations and time dependent neutron scalar fluxes. The suggested solution method provides estimates of the end-of-chain biases of importance to activation products, and solves the forward calculation problem to the accuracy of the underlying data.

For activation products the inverse problem is of greater interest: given the measured amounts of activation products produced by a specified time dependent neutron scalar flux irradiation, infer the initial concentrations of the elements that produced those activation products.

In this memo I briefly review the formal forward and inverse production-depletion solutions and use them as the basis for an iterative algorithm, using the capabilities in `NDI` in a series of forward calculations, for the approximate solution of the inverse problem.

Investigation

Formal Solution: The time rate of change of a vector of nuclide amounts is given by the total production-depletion rate equation:

$$\frac{d\overrightarrow{|N|}}{dt} = \mathbf{R}\overrightarrow{|N|}, \tag{1}$$

where $|N|$ is the amount of nuclide N and $\mathbf{R}$ is the production-depletion matrix. The entries in the production-depletion matrix $\mathbf{R}$ are given by:

$$R_{P,T} \doteq \sum_g \phi_g \left\{ \sum_{r \notin \{n,n'\}} \sigma_{T(n,r)P,g} - \delta_{P,T} \sum_{X \neq P} \sum_{r \notin \{n,n'\}} \sigma_{P(n,r)X,g} \right\}, \tag{2}$$

where ϕ_g is the neutron scalar flux in group g and $\sigma_{T(n,r)P,g}$ is the microscopic cross section for reaction $n + T \longrightarrow r + P$ in group g subject to the constraints of charge and nucleon balance. An `NDI` call to `ndi2_get_float64_vec_x` with the keyword `NDI_TOT_PROD_XS` and `ndi2_get_float64_vec` with the keyword `NDI_TOT_DEPL_XS` provides the production and depletion sum terms in the production-depletion matrix, respectively. The `NDI full` production-depletion library for `mtmg01ex` contains 181 targets.

If the production-depletion matrix is approximately constant over a small time step, $\Delta t_i \doteq t_{i+1} - t_i$, then Equation 2 can be solved to first order for that time step:

$$\overrightarrow{|N|}(t_{i+1}) \simeq \overrightarrow{|N|}(t_i) + \Delta t_i \mathbf{R}(t_i)\overrightarrow{|N|}(t_i). \tag{3}$$

This forward Euler approximation is efficient for a large production-depletion chain and is the forward solution method typically used in codes.

If the production-depletion matrix is actually constant in time then a finite time interval t can be divided into n equal time steps and Equation 3 can be iterated; in the limit:

$$\begin{aligned} \overrightarrow{|N|}(t_f) &= \lim_{n\to\infty} \left\{ \mathbf{I} + \frac{t\mathbf{R}}{n} \right\}^n \overrightarrow{|N|}(t_0) \\ &= e^{t\mathbf{R}}\overrightarrow{|N|}(t_0), \end{aligned} \tag{4}$$

where $e^{\mathbf{X}} \doteq \sum_{i=0}^{\infty} \frac{\mathbf{X}^i}{i!}$ is the matrix exponential. Physically, the $i = 1$ term in the exponential matrix expansion is the single reaction production of the N_i's from their parents and destruction to their children, the $i = 2$ term is the two step reaction production of N_i's including its destruction followed by subsequent production, etc.

If the production-depletion matrix varies in time, the formal solution can still be expressed through the limit of iterated Equations 3 as a matrix exponential:

$$\begin{aligned} \overrightarrow{|N|}(t_f) &= \lim_{\substack{n\to\infty \\ \Delta t_i \to 0}} \left[\prod_{i=1}^{n} \{ \mathbf{I} + \Delta t_{n-i} \mathbf{R}(t_{n-i}) \} \right] \overrightarrow{|N|}(t_0) \\ &= e^{\mathbf{\Omega}(t_f,t_0)}\overrightarrow{|N|}(t_0), \end{aligned} \tag{5}$$

where $\mathbf{\Omega}(t_f, t_0) \doteq \sum_{i=1}^{\infty} \mathbf{\Omega}_k(t_f, t_0)$ is the Magnus expansion[3] of $\mathbf{R}(t)$ on the interval $[t_f, t_0]$. The first few terms of the Magnus expansion are:

$$\begin{aligned}
\mathbf{\Omega}_1(t_f, t_0) &\doteq \int_{t_0}^{t_f} dt_1\, \mathbf{R}(t_1), \\
\mathbf{\Omega}_2(t_f, t_0) &\doteq \frac{1}{2}\int_{t_0}^{t_f} dt_1 \int_{t_0}^{t_1} dt_2\, \left[\mathbf{R}(t_1), \mathbf{R}(t_2)\right], \\
\mathbf{\Omega}_3(t_f, t_0) &\doteq \frac{1}{6}\int_{t_0}^{t_f} dt_1 \int_{t_0}^{t_1} dt_2 \int_{t_0}^{t_2} dt_3\, \left[\mathbf{R}(t_1), [\mathbf{R}(t_2), \mathbf{R}(t_3)]\right] + \left[\mathbf{R}(t_3), [\mathbf{R}(t_2), \mathbf{R}(t_1)]\right],
\end{aligned}$$

where $[\mathbf{A}, \mathbf{B}] \doteq \mathbf{AB} - \mathbf{BA}$ is the matrix commutator of $\mathbf{A}$ and $\mathbf{B}$. The second and higher order terms in the Magnus expansion correct for the non-commutative nature of the product of production-depletion matrices evaluated at different times.

Given the general forward solution in Equation 5, which gives final nuclide concentrations in terms of initial nuclide concentrations, the inverse problem specifying initial nuclide concentrations given final nuclide concentrations is easily solved:

$$\overrightarrow{|N|}(t_0) = e^{-\mathbf{\Omega}(t_f, t_0)}\overrightarrow{|N|}(t_f). \tag{6}$$

Approximate Solution: Equation 6, typical of formal solutions, is nearly useless for actually computing the inverse solution, because the former requires that:

1. the forward solution is known
2. the forward solution provides either $\mathbf{\Omega}$ or $e^{\mathbf{\Omega}}$
3. the large (181×181 in the case of `mtmg01ex`'s `full` chain) matrix $\mathbf{\Omega}$ or $e^{\mathbf{\Omega}}$ is exponentiable or invertable, respectively

And, of course, if the forward solution **is** known, then the initial concentrations **are** already known, too, making the inverse problem moot.

However, the formal solution does suggest a possible iterative approach for activation products: use a trial forward solution like Equation 5 and approximate inverse like Equation 6 of a reduced production-depletion set like Figure 1 to iterate to the inverse solution.

Consider the four activation products of Iron and Cobalt shown in Figure 1 as a subset of the "Chart of the Nuclides"[2] with the reactions available in the `NDI` `mtmg01ex` neutron transport library. This is the nearest neighbor sub-chain, corresponding to `mtmg01ex` `full` chain with explicit daughter production, for these activation products.

If we restrict both the matrix exponential and Magnus expansions in Equation 5 to first order we get:

$$\overrightarrow{|N|}(t_f) \simeq \left\{\mathbf{I} + \int_{t_0}^{t_f} dt_1\, \mathbf{R}(t_1)\right\}\overrightarrow{|N|}(t_0). \tag{7}$$

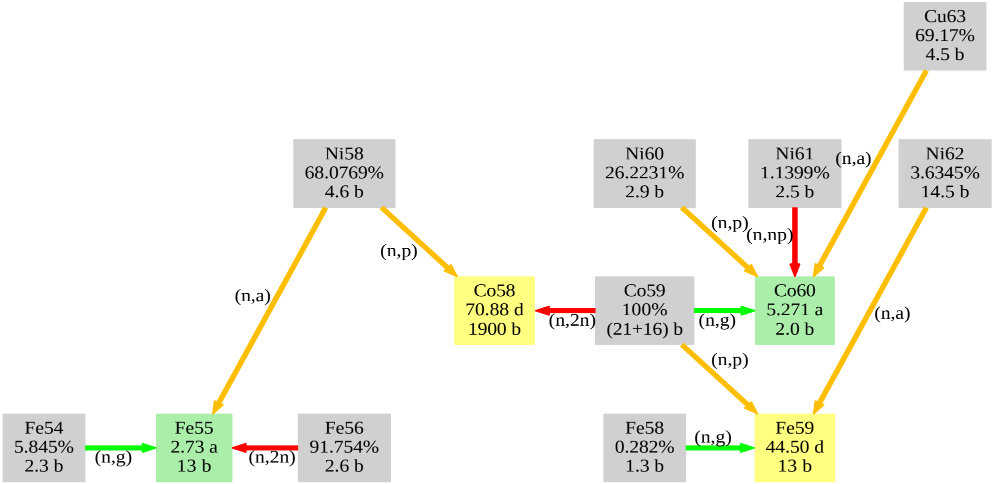

Figure 1: The Activation Products of Iron and Cobalt with Their Parents. Stable nuclides are shown in gray with their natural isotopic abundance and thermal capture cross section below their symbol and mass number. Activation products are shown in green ($100\,\mathrm{d} < t_{1/2} \leq 10\,\mathrm{a}$) or yellow ($10, \mathrm{d} < t_{1/2} \leq 100\,\mathrm{d}$) with their half-life and thermal capture cross section below their symbol and mass number. The capture ((n,γ)), low threshold ((n,p), (n,α)), and high threshold ((n,np), $(n,2n)$) arrows indicate the production reactions for the activation products.

Projecting this matrix equation to the outgoing subspace containing only the activation products in Figure 1 and reducing the system to only the non-zero, i.e. parent, matrix entries on the right, we obtain a reduced approximation to the forward equation that is suitable for an iterative solve of the inverse problem:

$$\Delta\overrightarrow{|P|} \simeq \int_{t_0}^{t_f} dt_1\, \mathbf{R}(t_1)\overrightarrow{|I|}(t_0), \tag{8}$$

where $\Delta\overrightarrow{|P|} \doteq \overrightarrow{|P|}(t_f) - \overrightarrow{|P|}(t_0)$ are the change in concentrations of the four activation products, and $\overrightarrow{|I|}(t_0)$ are the initial concentrations of the nine stable nuclides in Figure 1.

This reduced chain is exact provided the sub-chain in Figure 1 includes all the first order production-depletion reactions available in the chain and the parent populations are unchanged, which is exactly the case for `NDI`'s `basic` production-depletion chain for the `mtmg01ex` library with explicit daughter production[1].

We can construct initial nuclide concentrations from initial element concentrations, assuming only natural abundances, by using the natural abundance matrix:

$$\overrightarrow{|I|}(t_0) = \mathbf{A}\overrightarrow{|E|}(t_0), \tag{9}$$

where $A_{j,k}$ is the natural isotopic abundance of isotope I_j in element E_k. Thus:

$$\Delta\overrightarrow{|P|} \simeq \mathbf{M}\overrightarrow{|E|}(t_0), \tag{10}$$

where $\mathbf{M} \doteq \int_{t_0}^{t_f} dt_1\, \mathbf{R}(t_1)\mathbf{A}$.

```
1. Given $\Delta\overrightarrow{|P|}_E$ guess $\overrightarrow{|E|}(t_0)$, where $|E_i|(t_0) \neq 0$
2. $\overrightarrow{|I|}(t_0) \leftarrow \mathbf{A}\overrightarrow{|E|}(t_0)$; forward calculate $|\mathbf{I}(\mathbf{n},\mathbf{r})\mathbf{P}|(t_f)$ and $\Delta\overrightarrow{|P|}$
3. If $\Delta\left|\Delta\overrightarrow{|P|} - \Delta\overrightarrow{|P|}_E\right| < \epsilon\left|\Delta\overrightarrow{|P|}_E\right|$ then stop
4. $M_{i,k} \leftarrow \sum_{I_j \in E_k} \frac{|I_j(n,r)P_i|(t_f)}{|I_j|(t_0)} A_{j,k}$
5. $\overrightarrow{|E|}(t_0) \leftarrow \mathbf{M}^+\Delta\overrightarrow{|P|}_E$
6. Goto 2
```

Figure 2: Activation Product Inverse Calculation Algorithm. Use an initial guess of element concentrations and forward calculation of activation product concentrations to approximate the projected production-depletion matrix; iterate to solution. Step 2 in this algorithm corresponds to the forward solve of Equation 5 with trial initial concentrations, while Step 5 corresponds to the inverse solve, in the least squares sense with a generalized inverse $\mathbf{M}^+$, of Equation 10 as approximation to Equation 6 with measured activation product concentrations.

Because the forward solution with the `basic` or `full` chains must explicitly calculate the total number of reactions which produce activation products from each source[1], we have the data to approximate the matrix $\mathbf{M}$:

$$M_{i,k} \simeq \sum_{I_j \in E_k} \frac{|I_j(n,r)P_i|(t_f)}{|I_j|(t_0)} A_{j,k}, \tag{11}$$

where $|I_j(n,r)P_i|(t_f)$ is the total number of $n + I_j \rightarrow r + P_i$ reactions[1], provided the initial stable nuclide concentrations are not zero.

This suggests the iterative algorithm shown in Figure 2 for finding initial element concentrations given measured activation product concentrations. For the `basic` chain with explicit activation product production, Equations 10 and 11 are exact and the scheme should converge for a good initial guess; for the `full` chain with explicit activation product production these equations are approximate and ϵ needs to be larger than the error introduced by ignoring the higher order contributions.

We use the generalized inverse of $\mathbf{M}$ in Step 5 both because of the approximations made and to avoid problems with under (more activation products than structural elements) and over (more structural elements than activation products) constraints in the system. In any case, Step 5 returns the best fit, in the least-squared residual sense, to the available data.

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[1] Since $\Delta\overrightarrow{|P|} \doteq |I_j(n,r)P_i|(t_f)$, it includes all terms in the exponential and Magnus expansions, including the production of activation products by second and higher order reactions in the case of `full` and `nosub` chains. Since activation products are primarily produced by their parents in the `full` and `nosub` chains, and only produced by them in the `basic` chain, it suffices as an approximation.

Conclusion

The inverse problem of finding structural element concentrations from measured activation product concentrations can be solved systematically by iterating on an initial guess through a series of forward calculations of activation product concentrations with a least squares fit of structural element concentrations to measured activation product concentrations at each iteration. The algorithm converges exactly for the `basic` production-depletion chain with explicit activation product production, and approximately, in the least-squares sense, for the `full` chain with explicit activation product production and the `nosub` production-depletion chain. The algorithm is suitable for automation.

The algorithm is similar to Jabobi iteration[4, p. 506] scheme in its outer iteration, but in this case the matrix is (weakly) dependent on the solution vector through its effects on the scalar neutron flux and the outer iteration converges the matrix while the inner explicit solve finds a consistent right hand side for the current matrix approximation.

Although the algorithm follows the manual steps one might intuitively use to solve the inverse problem, its rate of convergence and stability have not been formally established. The rate might be accelerated or potential oscillations damped by the introduction of a relaxation parameter which mixes old and new iterates in Step 5, but this modification requires more experience with this algorithm or a more rigorous derivation.

If the time averaged scalar neutron flux for a problem is approximately known, it could be used in Equations 11 and 10 to estimate initial element concentrations for the first step in the algorithm. The TD weight function[5] available from the `NDI` call `ndi2_get_float64_vec` with the keyword `NDI_WGTS` could be suitably scaled to provide a generic initial guess in the absence of more problem specific information.

Because the algorithm converges exactly for the `basic` production-depletion chain and the forward calculation with the `basic` chain is faster than using either the `full` or `nosub` chains, iterating first with the `basic` chain to converge only the first order reactions in the chain, followed by refinement with the `full` or `nosub` chain to include the remaining terms might provide the best results.

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