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Comparison of depletion algorithms for large systems of nuclides

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ABSTRACT

In this work five algorithms for solving the system of decay and transmutation equations with constant reaction rates encountered in burnup calculations were compared. These are Chebyshev rational approximation method (CRAM), which is a new matrix exponential method, the matrix exponential power series with instant decay and a secular equilibrium approximations for short-lived nuclides, which is used in ORIGEN, and three different variants of transmutation trajectory analysis (TTA), which is also known as the linear chains method. The common feature of these methods is their ability to deal with thousands of nuclides and reactions. Consequently, there is no need to simplify the system of equations and all nuclides can be accounted for explicitly.

The methods were compared in single depletion steps using decay and cross-section data taken from the default ORIGEN libraries. Very accurate reference solutions were obtained from a high precision TTA algorithm. The results from CRAM and TTA were found to be very accurate. While ORIGEN was not as accurate, it should still be sufficient for most purposes. All TTA variants are much slower than the other two, which are so fast that their running time should be negligible in most, if not all, applications. The combination of speed and accuracy makes CRAM the clear winner of the comparison.

1. Introduction

Being able to accurately solve changes in the compositions of materials under irradiation is important in many applications. One particularly challenging application is in burnup calculations, which aim at following the time development of material compositions and core parameters of a nuclear reactor.

There are two factors responsible for the difficulty. First, material compositions and neutron flux affect each other leading to a complex combined problem. This is typically handled by sequentially solving neutronics and changes in material compositions while assuming the other to remain constant. Second, irradiated nuclear fuel contains thousands of different nuclides with widely varying half-lives, resulting in an extremely large and stiff problem.

Most of the nuclides are produced only in small amounts and can be handled by lumping, i.e., representing a large number of low importance or short-lived nuclides with one pseudo nuclide that has suitably averaged properties. However, there are also a few methods that can be used to solve the full system even when all nuclides are explicitly represented. Such methods provide greater flexibility and problem independence as the need to average lumped nuclides is removed. They also directly solve the proportions of those nuclides, which would otherwise be combined to pseudo nuclides, simplifying result handling.

The equations governing decay and transmutation of an arbitrary mixture of N different nuclides in a homogenized material region can be written as

$$\frac{dx_i}{dt} = -\lambda_i^{\text{eff}} x_i + \sum_j^N b_{j,i}^{\text{eff}} \lambda_j^{\text{eff}} x_j \quad \text{for} \quad i = 1, \ldots, N, \tag{1}$$

where x_i is the atomic density of nuclide i, λ_i^{eff} the effective decay constant of nuclide i, and $b_{i,j}^{\text{eff}}$ the effective branching ratio from nuclide i to nuclide j. These are defined as

$$\lambda_i^{\text{eff}} = \lambda_i + \phi \sum_j \sigma_{i,j} \tag{2}$$

and

$$b_{i,j}^{\text{eff}} = \frac{b_{i,j}\lambda_i + \sigma_{i,j}\phi}{\lambda_i^{\text{eff}}}, \tag{3}$$

where λ_i is the decay constant of nuclide i, ϕ the one-group neutron flux, $\sigma_{i,j}$ the microscopic one-group cross-section for transmutation of nuclide i to nuclide j and $b_{i,j}$ the branching ratio from nuclide i to nuclide j, i.e., the fraction of natural decays of nuclide i that produce nuclide j.

With the assumption of constant reaction rates during a time step, the decay and transmutation equations are a system of linear first order differential equations with constant coefficients.

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Numerous ways of solving such systems exist, but in burnup calculations the size and stiffness of the system render most of them practically useless, especially when the number of nuclides is not reduced by approximations.

2. Matrix exponential methods

The decay and transmutation equations, Eq. (1), can be written in a matrix form $d\vec{x}/dt = A\vec{x}$, where $A_{i,j} = -\lambda_i^{\text{eff}}\delta_{i,j} + b_{j,i}^{\text{eff}}\lambda_j^{\text{eff}}$. The solution to this equation is

$$\vec{x}(t) = e^{At}\vec{x}(0), \tag{4}$$

which employs the matrix exponential notation

$$e^{At} = \sum_{m=0}^{\infty} \frac{1}{m!}(At)^m. \tag{5}$$

So-called matrix exponential methods are based on different numerical approximations to the matrix exponential, which in a general case cannot be evaluated exactly. Numerous methods have been developed for evaluating the matrix exponential (Mole and Van Loan, 2003), but only a few are applicable in burnup calculations, and even fewer when solving the full system.

2.1. CRAM

Chebyshev rational approximation method (CRAM) (Pusa and Leppänen, 2010) is a new matrix exponential method that is based on the observation that the eigenvalues of the depletion coefficient matrix A appear to be clustered around the negative real axis. This can be exploited by making a Chebyshev rational approximation of the exponential function for the interval $(-\infty, 0]$. The resulting rational function is then decomposed into a pole-residue form to avoid numerical instability. When the denominator and numerator orders are selected equal and even in the Chebyshev approximation, the poles form conjugate pairs and the imaginary parts cancel out for a real valued variable. Thus, an order (k,k) approximation becomes

$$e^z \approx \frac{P_k(z)}{Q_k(z)} = a_0 + \sum_{i=1}^{k} \frac{a_i}{z+\theta_i} = a_0 + 2\text{Re}\left[\sum_{i=1}^{k/2} \frac{a_i}{z+\theta_i}\right]. \tag{6}$$

where P_k and Q_k are polynomials of order k, whose coefficients have been selected to minimize absolute deviation from exponential function on the negative real axis, a_0 is the limiting value of the approximation at infinity, and a_i and θ_i are the residues and poles. When this approximation is applied to the matrix exponential in Eq. (4) it becomes

$$\vec{x}(t) \approx a_0\vec{x}(0) + 2\text{Re}\left[\sum_{i=1}^{k/2} a_i(At + \theta_i I)^{-1}\right]\vec{x}(0), \tag{7}$$

where the matrix inversions can be calculated efficiently thanks to the sparse structure of the matrix A.

There is no reason to select any particular value of k as long as it is even. Thus, the order of the approximation can be used to scale the accuracy versus running time. Since the number of operations required for an order (k,k) approximation scales linearly with k, a wide range of values might be viable. However, only an order (14,14) approximation is considered in this work following the choice of Pusa and Leppänen (2010).

2.2. ORIGEN

ORIGEN (Croff, 1983, 1980a) is a well known and widely used program for depletion and transmutation calculations. It is used for solving depletion steps in numerous linked burnup codes such as Monteburns (Posfon and Trellue, 1999) and MOCUP (Moore et al., 1995). The more modern ORIGEN-S of the SCALE package also uses the same method of solution (Hermann and Westfall, 2009). While solving depletion steps is the most central function of ORIGEN, it also provides a wide range of other features including stand-alone burnup calculations using pre-calculated cross-section libraries.

The method of solution for depletion steps in ORIGEN is a power series approximation of the matrix exponential with instant decay and secular equilibrium approximations for handling short-lived nuclides (Croff, 1980b).

First, contributions to the final concentration from short-lived ($\lambda^{\text{eff}} \leqslant \ln(0.001)/t$, i.e., $T_{1/2}^{\text{eff}} \lesssim 0.1t$, where t is the step length) nuclides present initially are calculated. This is done by constructing for each nuclide all the populating chains consisting of short-lived nuclides, and solving these chains in a similar way as in the TTA methods described later. The contributions to final concentrations of short-lived nuclides are saved and the contributions to long-lived nuclides are added to the initial concentrations of those nuclides.

Second, a reduced version of the coefficient matrix A in Eq. (4) is constructed by assuming short-lived nuclides to decay instantly,[1] thus removing them from the system. The reduced system is then evaluated by truncating the power series for the exponential:

$$e^{At} = \sum_{m=0}^{\infty} \frac{(At)^m}{m!} \approx \sum_{m=0}^{M} \frac{(At)^m}{m!}. \tag{8}$$

The convergence and numerical stability problems usually encountered when using the power series (Mole and Van Loan, 2003) are avoided because the reduced system does not contain the large matrix elements associated with the short-lived nuclides.

Finally, the contributions from long-lived nuclides to short-lived nuclides are solved by assuming these decay and transmutation chains to be in a secular equilibrium at the end of the step:

$$\frac{dx_i}{dt} = \sum_{i=1}^{N} a_{ij}x_j = 0. \tag{9}$$

This results in a greatly reduced form of the system that is solved by iterating

$$x_i^{k+1} = \frac{1}{-a_{ii}} \sum_{\substack{j=1 \\ j\neq i}}^{N} a_{ij}x_j^k, \tag{10}$$

where x_i are the final concentrations obtained from the matrix solution for the long-lived nuclides and unknowns to be solved for the rest. The final concentrations of short-lived nuclides are superposition of the contributions calculated from other short-lived nuclides and from the long-lived nuclides.

3. Transmutation trajectory analysis

Transmutation trajectory analysis (TTA) (Cetnar, 2006), also known as the linear chains method, is an alternative method for solving the decay and transmutation equations. The core of the method is that a complex web of decay and transmutation reactions can be decomposed into a set of linear chains consisting of all possible routes, or trajectories, through the web.

[1] The approximation is actually more complex than just instant decay. There is an additional correction that attempts to account for the effects of non-zero half-lives of the removed nuclides by modifying the effective decay constants of their long-lived parents. This feature is not mentioned in the documentations of ORIGEN.