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Solving point burnup equations by Magnus method

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ABSTRACT

The burnup equation of nuclides is one of the most equations in nuclear reactor physics, which is generally coupled with transport calculations. The burnup equation describes the variation of the nuclides with time. Because of its very stiffness and the need for large time step, this equation is solved by special methods, for example transmutation trajectory analysis (TTA) or the matrix exponential methods where the matrix exponential is approximated by CRAM. However, TTA or CRAM functions well when the flux is constant. In this work, a new method is proposed when the flux changes. It's an improved method compared to TTA or CRAM. Furtherly, this new method is based on TTA or CRAM, and it is more accurate than them. The accuracy and efficiency of this method are investigated. Several cases are used and the results show the accuracy and efficiency of this method are great.

1. Introduction

Burnup calculation plays a very important role in the nuclear reactor analysis. Burnup equations describe how the material components of the fuel or absorbers in the nuclear reactor will change with the time. They are Bateman equations [1], and usually a set of first order linear ordinary differential equations, which is very stiff. Burnup calculation often appears in the lattice calculation and is coupled with the neutron transport calculation. Whatever the neutron transport equations are calculated by deterministic or Monte Carlo, burnup calculation must be considered.

Recently, there is a trend in the burnup calculation that the detailed burnup chain is used instead of pseudo fission product nuclide(s). There are about 1500 nuclides in the burnup calculation coupled with Monte Carlo, and many nuclides own a very short half-life, such as ^{212}Po owns the half life of about 1e-7 s. Therefore, the burnup equations are large and very stiff, which makes them difficult to be solved.

There are mainly three kinds of methods to deal with the burnup equations. These methods are transmutation trajectory analysis (TTA), Runge-kutta or linear multi step methods, and matrix exponential methods. Bateman [2] proposed TTA method, which gives the explicitly analytical expression of every nuclide when the reaction of the burnup chain is only decay reaction. When there exists cycles in the burnup chain, TTA method has to cut a cycle into a relative long chain, which will has a bad effect on the efficiency and accuracy of TTA. Because burnup equations belong to ordinary differential equations, any common methods like Runge-kutta methods and linear multi step methods can also be used. Runge-Kutta-Gear methods were applied to the burnup calculations. Matrix exponential methods are very popular in the burnup calculation, since matrix exponential methods are stable and suitable for stiffness problem. The matrix exponential methods express the solution in the matrix exponential form. There are many methods about how to compute the matrix exponential. Taylor series method with scaling and squaring technique was implemented in the ORIGEN code [3]. However, some special care should be taken for the short-lived nuclides in this method. Other methods for calculating matrix exponential are the Quadrature-based rational approximation method (QRAM) (Trefethen et al., 2006), the Krylov Subspace Method [4], the Chebyshev rational approximation method (CRAM) [5], the Laguerre polynomial approximation method (LPAM) [6].

In the matrix exponential methods, the burnup equations are supposed to be a set of linear ordinary differential equations with constant coefficient in one time step. Usually, the reaction rates are taken by the average value in the time step. However, the matrix exponential methods do not give the analytical solution even if the reaction rates are linear in one time step. The Matrix exponential methods give the analytical solution when the reaction rates are unchanged, and they usually own accuracy of second order.

The Magnus expansion is a kind of exponential methods, which contains the matrix exponential method introduced above. The

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Magnus expansion has been successfully applied in many areas such as classical Hamilton mechanics, atomic and molecular physics, nuclear magnetic resonance, etc [7]. From the view of mathematics, the Magnus expansion is a kind of efficient numerical integrators.

Magnus expansion is a widespread tool to construct approximate exponential representations of the solution of the non-autonomous linear differential equations [8]. Though Magnus method is a powerful tool, it's not suitable for burnup calculation because a lot of the commutators in Magnus method destroy the stability of the exponential solver like CRAM. A special Magnus variant without commutators or CF (commutators free) method [12] can improve the accuracy while keeping the stability.

In this present work, the Magnus without the commutator expansions is presented for solving the point burnup equations. The numerical results of this method are compared with the referenced methods.

2. Method description

2.1. The Bateman equations

The general Bateman equation is written as following:

$$\frac{dn_i}{dt} = \sum_{i\neq j}(b_{i,j}\lambda_i + \sigma_{i,j}\phi)n_j - \left(\lambda_i + \phi\sum_j \sigma_{i,j}\right)n_i \tag{1}$$

Where, n_i is the nuclide i density, λ_i is the decay constant of the nuclide i, $\sigma_{i,j}$ is the microscopic cross section of a reaction where nuclide i generates j, ϕ is the neutron flux, $b_{i,j}$ is the generation fraction of the nuclide j when the nuclide i decays.

Eq. (1) is a form of the linear equations, which can be simplified into a compact form in Eq. (2) by the help of the matrix.

$$\frac{d\vec{n}}{dt} = \mathbf{A}\vec{n} \tag{2}$$

Where $\vec{n}$ is the vector of the nuclides, and A is the matrix consisting of the coefficient in Eq. (1). If the coefficients in Eq. (1) are constant, namely A is constant, the solutions can be written in the form of matrix exponential vector multiplication, seen in Eq. (3).

$$\vec{n} = e^{Ah}\vec{n}_0 \tag{3}$$

Where h is the time step, and $\vec{n}_0$ is the initial value. If ϕ depends on the time t, the matrix exponential in the literature [9] gives the solution in the following form:

$$\vec{n} = e^{\bar{A}h}\vec{n}_0 \tag{4}$$

Where $\bar{A}$ is the average value of A(t) in one time step. Notice that the exact solution of Eq. (2) cannot be written as the simple form of Eq. (5). And this method owns the accuracy of second order.

$$\vec{n} = e^{\int_0^h A(\tau)d\tau}\vec{n}_0 \tag{5}$$

There are many ways to compute the matrix exponential in Eq. (4), such as Taylor expansion, matrix decomposition, and rational approximation, etc. Chebyshev rational approximation method (CRAM) [5] belongs to the rational approximation. The formulas of CRAM are computed as following:

$$e^{Ah} = \alpha_0 I + \mathrm{Re}\sum_{i=1}^{k}\frac{\alpha_i}{Ah - \theta_i I} \tag{6}$$

Where α_i and θ_i are the parameters of CRAM, k is the CRAM order, I is the identity matrix, Re represents computing the real part of the complex number. The larger the CRAM order, the more accurate the calculation precision. Usually, the CRAM order k is chosen 14 in burnup calculations. The disadvantage of Eq. (5) is that there exists the complex number in this formulas.

Mini-Max polynomial approximation method (MMPA) [10] uses the similar formulas of Eq. (5), but there is no complex number in this method.

TTA method [11] is an analytical method to solve the burnup equation. TTA method has been successfully applied in the field of burnup chain, especially when there is only the decay process.

2.2. The Magnus expansion

Here, we consider the more general model than Eq. (1), where the flux can be changed now:

$$\frac{dn_i}{dt} = \sum_{i\neq j}(b_{i,j}\lambda_i + \sigma_{i,j}\phi(t))n_j - \left(\lambda_i + \phi(t)\sum_j \sigma_{i,j}\right)n_i \tag{7}$$

Or Eq. (6) is written as the following form:

$$\frac{d\vec{n}}{dt} = \mathbf{A}(t)\vec{n} \tag{8}$$

According to the literature [8], if the solution above is represented in the form of matrix exponential

$$Y(t) = e^{\Omega(t)}Y_0 \tag{9}$$

then Ωsatisfies the differential equation by the means of the Lie algebra:

$$\frac{d}{dt}\Omega(t) = d\exp_\Omega^{-1}(A(t)), \Omega(0) = 0. \tag{10}$$

Here

$$d\exp_\Omega^{-1}(C) = \sum_{k=0}^{\infty}\frac{B_k}{k!}ad_\Omega^k C, \tag{11}$$

Where B_k is the kth Bernoulli number and the definition of ad^m can be recursively written:

$$ad_\Omega^0 C = C, \qquad ad_\Omega^{m+1}C = [\Omega, ad_\Omega^m C] \tag{12}$$

where [X,Y] = XY-YX is the commutator of X and Y. Eq. (11) is solved by Picard's iteration and Ωis approximated

$$\Omega^{[m+1]}(t) = \int_{t_0}^{t}\sum_{k=0}^{\infty}\frac{B_k}{k!}ad_{\Omega^{[m]}(s)}^k A\left(s, e^{\Omega^{[m]}(s)}Y_0\right)ds, \Omega^{[0]}(t) = 0 \tag{13}$$

However, the summation needs to be truncated, and the practical implement is the formulas:

$$\Omega^{[1]}(t) = \int_{t_0}^{t} A(s)ds$$
$$\Omega^{[m]}(t) = \sum_{k=0}^{m-2} \frac{B_k}{k!} \int_{t_0}^{t} ad^k_{\Omega^{[m-1]}(s)} A(s)ds, \quad m \geq 2 \tag{14}$$

According to the literature [8], the formulas above can achieve the order up to m

$$\Omega^{[m]}(t) - \Omega(t) = O\left(t^{m+1}\right) \tag{15}$$

The integrals are computed by quadrature rules with equispaced points along the interval. Eq. (14) with m up to 3 is reduced to the following form:

$$\Omega^{[2]}(t) = \Omega^{[1]}(t) = \int_{t_0}^{t} A(s)ds$$
$$\Omega^{[3]}(t) = \int_{t_0}^{t} A(s)ds + \frac{1}{2}\int_{t_0}^{t}\int_{t_0}^{s} dsd\tau[A(s), A(\tau)] \tag{16}$$

In this situation, $\Omega^{[3]}(t)$ can indeed achieve the order 4. From Eq. (15), the Magnus expansion of order 2 is indeed the formulas of Eq. (4). So Eq. (4) or the method now usually used is the special 2-order Magnus method. When the commutator of A(s) and A(τ) of the any different moments is zero, then Eq. (4) gives the exact solutions.

$\Omega^{[k]}(t)$ needs much more calculation when k becomes larger, so only no more than $\Omega^{[3]}(t)$ is considered. In fact, $\Omega^{[4]}(t)$ can be written as follows:

$$\Omega^{[4]}(t) = \Omega^{[3]}(t) + \frac{1}{6}\int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \int_{t_0}^{t_2} dt_3([A(t_1), [A(t_2), A(t_3)]] + [A(t_3), [A(t_2), A(t_1)]]) \tag{17}$$

The integrals of Eq. (15) can be computed by Gaussian quadrature

$$\Omega^{[3]} = \frac{h}{2}(A_1 + A_2) + \frac{\sqrt{3}}{12}h^2[A_2, A_1] \tag{18}$$

$$c_1 = \frac{1}{2} + \frac{\sqrt{3}}{6}, c_2 = \frac{1}{2} - \frac{\sqrt{3}}{6}$$
$$A_i = A(t_0 + c_i h), \quad i = 1, 2 \tag{19}$$

There is a commutator in Eq. (17), which may be a disadvantage for burnup calculation. In burnup calculation, the time step h is usually quite large, the term associated with h^2 may not a small quantity. In that case, the eigenvalues of $\Omega^{[3]}$ may not distribute near the negative real axis, and CRAM will fail if CRAM is used to calculate $e^{\Omega^{[3]}}$.

The Magnus without commutator is investigated in this work. A product of exponentials of linear combinations of the A(t) are used which avoids the presence of commutators. It's different from the standard Magnus method, but it owns the same order with the standard one. For fourth order Magnus method without commutator, the formulas is the product of two matrix exponetials, seen in Eq. (19).

$$\exp(a_1 hA_1 + a_2 hA_2)\exp(a_2 hA_1 + a_1 hA_2) = \exp\left(\Omega^{[3]}\right) + \left(O\left(h^5\right)\right) \tag{20}$$

with $a_1 = \frac{3-2\sqrt{3}}{12}$, $a_2 = \frac{3+2\sqrt{3}}{12}$.

In constant flux mode, the flux is unchanged, then Eq. (19) is the exp(Ah). In non-constant flux case, the calculation of 4th-order Magnus without commutator is twice of the calculation of 2th-order Magnus. And this method is called ME3/CF3, while Eq. (4) is called ME2.

2.3. The stability of Magnus without commutator

From Eq. (1), the matrix A(t) can be written as two parts:

$$A(t) = D + \phi(t)N \tag{21}$$

Where D is the matrix associated with decay processes, and it is unchanged with time; N is the matrix associated with neutron reactions, and it's also unchanged. For any flux, all of the eigenvalues of the matrix A(t) are near the negative real axis, where CRAM can be used. Because the first exponent of Eq. (19) can also be written in the similar way of Eq. (20), seen in Eq. (21), the eigenvalues of the exponent are also near the negative real axis. The second exponent of Eq. (19) is the similar situation. That is to say, CRAM can be used to approximate Eq. (19).

$$a_1 hA_1 + a_2 hA_2 = (a_1 h + a_2 h)\left(D + \frac{a_1\phi_1 + a_2\phi_2}{a_1 + a_2}N\right) \tag{22}$$

3. Calculations and results

Consider that the matrix A in Eq. (2) owns so many zeros, the matrix A is stored in the sparse matrix format.

3.1. The decay problem of ^{237}Np

The decay library of ORIGEN-2 is used in this case. The aim is to get the main products of ^{237}Np when ^{237}Np decays about 1million years. At the initial moment, the quantity of ^{237}Np is 1 mol.

The results obtained by ME3 are shown in Table 1. The references are the results obtained by ME2. From Table 1, it shows that the results obtained by ME3 and ME2 are the same. In fact, ME3 and ME2 are the same in the theory when flux is constant, and the numerical results verify this point.

Table 1
The results of the^{237}Np decay problem.

Nuclides	Half-lives	ME3 /mole	ME2/mol
Np237	2.14E6 a	7.23911364E-01	7.23911364E-01
Pa233	27.0 d	2.49410552E-08	2.49410552E-08
U233	1.592E5 a	5.70317235E-02	5.70317235E-02
Th229	7340 a	2.63600152E-03	2.63600152E-03
Ra225	14.9 d	1.46483142E-08	1.46483142E-08
Ac225	10.0 d	9.83395345E-09	9.83395345E-09
Fr221	4.8 min	3.34632217E-12	3.34632217E-12
At217	3.2E-3 s	3.67627640E-16	3.67627640E-16
Bi213	46.5 min	3.11257111E-11	3.11257111E-11
Po213	3.72E-6 s	4.67985371E-20	4.67985371E-20
Tl209	2.2 min	3.13971653E-14	3.13971653E-14
Pb209	3.25 h	1.33276457E-10	1.33276457E-10
Bi209	1.9E19 a	2.16399220E-01	2.16399220E-01

Table 2
The important nuclides of the UO2 problem.

Nuclides	ME3 /mole	ME2/mole
Se79	5.19524710E-06	5.19524710e-06
Kr83	5.36479101E-05	5.36479101e-05
Sr90	5.34309655E-04	5.34309655e-04
Zr94	7.10937646E-04	7.10937646e-04
Mo95	7.20574486E-04	7.20574486e-04
Tc99	7.10230322E-04	7.10230322e-04
Ru101	6.45169750E-04	6.45169750e-04
Ag109	3.90544780E-05	3.90544780e-05
Sn126	1.00741521E-05	1.00741521e-05
I129	8.78890248E-05	8.78890248e-05
Xe136	1.35436484E-03	1.35436484e-03
Cs133	7.72658576E-04	7.72658576e-04
Ba138	7.88870889E-04	7.88870889e-04
La139	7.43163639E-04	7.43163639e-04
Ce142	6.70799499E-04	6.70799499e-04
Eu153	4.08641882E-05	4.08641882e-05
U234	8.24830641E-07	8.24830641e-07
U235	1.92874770E-02	1.92874770e-02
U236	1.96025582E-03	1.96025582e-03
U238	9.61257565E-01	9.61257565e-01
Np237	1.15241567E-04	1.15241567e-04
Pu239	4.11853275E-03	4.11853275e-03
Am241	6.61403064E-05	6.61403064e-05
Cm244	2.13149043E-07	2.13149043e-07

3.2. The UO_2 problem

Consider that UO2 is exposed in the environment for one year where the neutron flux is 3.0E14, then let them decay for three years. The initial component of UO2 is that: U235 0.03 mol, U238 0.97 mol, O16 2 mol.

The results obtained by ME3 and ME2 are shown in Table 2. From Table 2, it shows again that ME3 and ME2 are the same when flux is constant.

3.3. The linear flux problem

Consider that UO2 is exposed in the environment for one year, however the neutron flux decreases from 3.0E14 to 3.0E13, that is $\phi(t) = 3e14 \times (1 - 0.9t/T), T = 1\ year$. The reference is obtained by ME2 with 40 time step. Table 3 gives the results obtained by ME3 with 8 time steps and ME2 with 16 time steps. The cost time of ME2 and ME3 are about the same, but the relative errors of Am241 obtained by ME2 are much larger than that of ME3.

Table 4 gives the maximum relative error of results obtained by ME2 and ME3 under the different time steps. The relative error obtained by ME3 drops more quickly than ME2, and the results obtained by ME3 with 4 time steps are even more accurate than those obtained by ME2 with 16 time steps.

4. Conclusion

The standard Magnus expansions gives the exact exponential form of the solution, although the exponent is expressed as an infinite series. The Magnus expansion of order 2 (ME2) is equivalent with the matrix exponential method now used widespread. However, the higher order Magnus methods are not suitable for the burnup calculation because the commutators in standard Magnus expansions will destroy the stability of the exponential methods. Notice that the eigenvalues distribution of the burnup matrix is along the negative axis, while the eigenvalues of the commutators may appear in the positive half plane. Here, the special Magnus method without commutators is analyzed in this work, which is denotes as ME3 in this work. This method keeps the characteristic of eigenvalues distribution of the matrix appearing in ME3, which CRAM or other methods can be used to approximate the matrix exponential. Compared with the 2th-order Magnus, the cost time increases twice for ME3. ME3 is tested by several numerical cases.

Table 4
The maxmium relative error.

STEP	Relative error of ME2	Relative error of ME3
2	1.13e-01	3.77e-03
4	2.92e-02	1.01e-03
8	7.17e-03	2.82e-04
16	1.58e-03	2.98e-04
32	1.69e-04	–

Table 3
The important nuclides of the linear flux problem.

Nuclides	ME3	Relative error of ME3	ME2	Relative error of ME2	Reference
Se79	2.86835258E-06	−2.58E-06	2.86836097E-06	3.42E-07	2.86835999E-06
Kr83	3.16836657E-05	−1.14E-05	3.16833215E-05	−2.23E-05	3.16840270E-05
Sr90	3.31760412E-04	−3.01E-06	3.31765042E-04	1.09E-05	3.31761410E-04
Zr94	3.99832729E-04	−2.25E-06	3.99833396E-04	−5.78E-07	3.99833627E-04
Mo95	3.07918949E-04	1.11E-04	3.07710087E-04	−5.67E-04	3.07884643E-04
Tc99	3.94043103E-04	−1.18E-04	3.93962264E-04	−3.24E-04	3.94089765E-04
Ru101	3.51357600E-04	−3.53E-06	3.51358523E-04	−8.99E-07	3.51358839E-04
Ag109	1.49987755E-05	−6.95E-05	1.49980221E-05	−1.20E-04	1.49998179E-05
Sn126	4.88873990E-06	−5.59E-06	4.88878598E-06	3.84E-06	4.88876722E-06
I129	4.48667744E-05	−1.87E-05	4.48662722E-05	−2.98E-05	4.48676113E-05
Xe136	6.85465066E-04	1.24E-04	6.85316263E-04	−9.27E-05	6.85379823E-04
Cs133	4.29618624E-04	−7.20E-05	4.29450426E-04	−4.63E-04	4.29649539E-04
Ba138	4.35001474E-04	−3.73E-06	4.35001991E-04	−2.54E-06	4.35003096E-04
La139	4.11340161E-04	−5.42E-06	4.11339705E-04	−6.53E-06	4.11342391E-04
Ce142	3.71783138E-04	−5.47E-06	3.71782160E-04	−8.10E-06	3.71785170E-04
Eu153	1.74077600E-05	−1.59E-04	1.74042373E-05	−3.61E-04	1.74105252E-05
U234	3.30389917E-07	7.32E-06	3.30373677E-07	−4.18E-05	3.30387499E-07
U235	2.35288942E-02	0.00E+00	2.35288941E-02	−4.25E-09	2.35288942E-02
U236	1.20781585E-03	0.00E+00	1.20781583E-03	−1.66E-08	1.20781585E-03
U238	9.65181560E-01	1.04E-09	9.65181559E-01	0.00E+00	9.65181559E-01
Np237	4.84230546E-05	−4.95E-05	4.83944157E-05	−6.41E-04	4.84254520E-05
Pu239	2.95661811E-03	−1.41E-04	2.95590199E-03	−3.83E-04	2.95703427E-03
Am241	2.42758546E-06	2.82E-04	2.42307417E-06	−1.58E-03	2.42690109E-06
Cm244	9.06089197E-09	−1.01E-04	9.06251803E-09	7.82E-05	9.06180941E-09

The numerical results show that ME3 are in great agreement with CRAM when the flux is constant. The results also show that ME3 is more accurate than CRAM when the flux changes with time. This paper shows the applicability of the Magnus expansions for solving the linear point burnup mode.

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