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▶ To cite this version:

Sébastien Lahaye, Alexis Anne, Remi Baron, Tan-Dat Huynh, Aime Tsilanizara. New Bateman equation solvers in MENDEL version 3.1. ICNC 2023 - The 12th International Conference on Nuclear Criticality Safety, Oct 2023, Sendai, Japan. cea-04468927

HAL Id: cea-04468927 https://cea.hal.science/cea-04468927v1

Submitted on 10 Feb 2025 $\,$

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NEW BATEMAN EQUATION SOLVERS IN MENDEL VERSION 3.1 S. Lahaye^{(1)*}, A. Anne⁽¹⁾, R. Baron⁽²⁾, T.D. Huynh⁽¹⁾, A. Tsilanizara⁽¹⁾

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ABSTRACT

Accurate determination of isotopic densities in nuclear materials is of first importance for the Criticality-Safety, as all other quantities of interest (Decay Heat, Spectra, Masses, Reactivity factor...) depend on the composition of the considered material. Isotopic densities are computed by solving the Generalized Bateman equations.

MENDEL is the new generation of CEA's fuel cycle code systems, successor to DARWIN/PEPIN2. Its solvers of the Generalized Bateman Equations are used by the stand-alone code MENDEL itself (for applications such as Decay Heat or particle spectra calculations) but also inside deterministic APOLLO3[®] and stochastic TRIPOLI-4[®] transport code systems. They can use any pre-processed nuclear data evaluation.

MENDEL aims to compute as precisely and as rapidly as possible the isotopic densities during irradiation or cooling periods. For this purpose, several algorithms are available in MENDEL, and two new approaches have been added in MENDEL version 3.1.

The analytical approach, was extended from cooling period computations only to in-flux calculations as long as the depletion matrix can be triangularized. Being naturally faster than any numerical approach, this enables, in adequate configurations, an exact and quick way to compute isotopic densities. Nevertheless, fuel cycle depletion chains are often too complete to be triangularized.

When only part of the matrix can be triangularized, only Runge-Kutta 4th order and CRAM methodologies were possible until MENDEL v3.0. From version 3.1, we added a mixed approach between CRAM and analytical method, which can offer for some depletion matrices a high speedup.

This paper describes the different solvers used in MENDEL version 3.1.

KEYWORDS

Fuel Cycle, Criticality safety, Algorithm, Bateman

1. INTRODUCTION

Accurate determination of isotopic densities in nuclear materials is of first importance for Fuel Cycle Studies and for Criticality-Safety. Indeed, all other physical quantities of interest (Decay Heat, Spectra, Masses, Activities, Reactivity factor...) depend on the composition of the considered material at the considered time.

As it is the fundamental physical quantity, one needs to compute isotopic densities as accurately as possible, and, for a practical point of view, as fast as possible. Isotopic densities are computed by solving the Generalized Bateman equations [1]. This accuracy is of utmost importance for nuclear power plant and nuclear installation safety, in normal or accidental operation, as well as for all nuclear industry process, from mine extraction to retreatment and storage.

Nuclear fuel cycle code systems exist all over the world to compute accurate concentrations, such as ORIGEN [2], FISPACT [3] or DARWIN/PEPIN2 [4, 5]. MENDEL [6] is the new generation depletion code system developed at CEA, dedicated to the calculation of all physical quantities of interest for fuel cycle studies. MENDEL already offers iso-capacity with the French current reference fuel code system used by French industry, DARWIN/PEPIN2.

MENDEL can be used as a standalone code system for fuel cycle studies, enabling the computation of isotopic densities with precise depletion chains, but also the computation of isotopic masses, activities, global decay heat, α , β or γ components, particle spectra, radiotoxicity parameters... MENDEL can also provide its depletion solvers as an informatics library to both Monte Carlo TRIPOLI-4[®] [7] and deterministic APOLLO3[®] [8] transport code systems. Here in this paper, we limit our results to the use of the standalone fuel cycle code system.

Bateman equation can be solved by several well-known methodologies [9]. MENDEL depletion solvers until version 3.0 allowed to compute with an analytical approach in the case of cooling periods, and with a choice between Runge-Kutta 4th order and CRAM solvers for in neutron flux periods [10]. This last point covers irradiation of materials inside of the reactor core, being fuel depletion or material activation. The comparison between Runge-Kutta and CRAM [11] has already been described in [10] and will not be discussed in this paper.

In MENDEL v3.1, we introduced two new approaches in order to compute isotopic densities. The first approach consists of extending the analytic computation to irradiation configurations, when possible. The second approach is a mixed algorithm segregating the isotopes between those calculated by an analytical way, and those calculated by a numerical way. In this paper, we focus on the description of those new methodologies, and show the speedup obtained for several configurations. The main goal of this work is a decrease of time consumption.

2. BATEMAN EQUATION SOLVERS IN MENDEL

Let us consider a material submitted to a neutron flux $\phi(E, t)$, the flux can potentially be null in case of the simulation of a cooling period. The depletion (evolution) in time of the nuclei concentrations $N_i(t)$ is described by the generalized Bateman [1] equations:

$$\frac{dN_i(t)}{dt} = -\left(\lambda_i + \tau_{i,i}\right)N_i(t) + \sum_{j\neq i} \left(b_{j,i}\lambda_j + \tau_{j,i}^r\right)N_j(t) + \sum_k \gamma_{k,i}\tau_k^f N_k(t)$$
(1)

with:

- λ_i the radioactive decay constant of nuclide *i*,
- $\tau_{i,i} = \int_E \sigma_i^d(E)\phi(E)dE$ the global disappearance reaction rate by neutronic reactions, with *E* the energy of incident neutron,
- $b_{i,i}$ the radioactive decay branching ratio from father nuclide *j* to daughter *i*,
- $\tau_{j,i}^r = \int_E \sigma_j^r(E)\phi(E)dE$ the neutronic reaction rate from nuclide *j* to nuclide *i* with reaction *r*, except fission,
- $\tau_k^f = \int_E \sigma_k^f(E)\phi(E)dE$ the fission reaction rate of fissile nuclide k,
- $\gamma_{k,i}$ the independent fission product yields from fissile nuclide k to fission product nuclide i.

Bateman equation (1) can be written down in matrix form:

$$\begin{cases} \frac{dN(t)}{dt} = A(t)N(t) \\ N(t_0) = N_0 \end{cases}$$
(2)

where A(t) is called the *depletion matrix* and N_0 is the isotopic density vector at initial time t_0 .

In MENDEL stand-alone fuel cycle code system, nuclear data are given by libraries based on international nuclear data evaluations such as JEFF-3.1.1 [12] (by default), ENDF/B-VII.1 [13] or JENDL-4 [14]. Those data are completed, when necessary and available by reaction rates issued from self-shielded cross sections and computed using:

- Microscopic multigroup cross sections from transport codes APOLLO2 [15], APOLLO3[®], ERANOS [16].
- Multigroup neutron flux computed by APOLLO2 (SAPHYB file), APOLLO3[®] (MPO file) ERANOS (XML file) or TRIPOLI-4[®] (INTERPEP file).

Another possibility is to use ORIGEN2 library format for one-group cross sections and neutron flux given by any code in one-group description and given to MENDEL by the INTERPEP format [17].

2.1. Bateman Equation solvers in MENDEL v3.0

We had in MENDEL 3.0 three ways to compute isotopic densities.

2.1.1 Analytic computation

For cooling periods only, we can transform the depletion matrix in Eq. 2 to be triangular. In this particular case, the matrix resolvant, which is the exact solution of the equation, is the exponential of matrix and Bateman equation reads:

$$N(t) = \exp(A * (t - t_0)) N(t_0)$$
(3)

When we consider cooling period only, the matrix A is constant in time as it does not depend on neutron flux (which varies in time) nor on self-shielded cross-sections (which can change from one self-shielding calculation to another). The exponential of the matrix [9] is easily computed, and the calculation time is quite fast, compared to any numerical computation.

For this reason, MENDEL uses - by default - the analytical method for all cooling period.

2.1.2 Runge-Kutta method

4th order Runge-Kutta method is the original way to compute isotopic densities in-flux, for both MENDEL and DARWIN/PEPIN2 code systems. This method offers the advantage the possibility of dependence in time for the depletion matrix. This possibility is used in MENDEL (not in DARWIN/PEPIN2) where the depletion matrix can be constant, linear or parabolic in time, depending the choice of the user.

The disadvantage of the Runge-Kutta method is the necessity of very short depletion steps for short radioactive decay period isotopes, leading to very high time consuming computations. MENDEL and DARWIN/PEPIN2 both fixed this issue with the introduction of the idea of *saturated isotopes*, treated by a specific algorithm. Using an adequate criteria, one can achieve a very good approximation of all isotopes, while performing a fast calculation.

A precise description of the Runge-Kutta method has been given in reference [10].

2.1.3 CRAM method

The second algorithm introduced for in-flux calculation is the Chebyshev Rational Approximation Method (CRAM), based on the method described by Maria Pusa [11]. This algorithm has been

introduced in MENDEL and has proved to be rather consistent in term of accuracy, with Runge-Kutta method, and slightly faster for some applications.

A precise description of the CRAM approach has been given in reference [10], as well as comparison with Runge-Kutta.

2.2. New Bateman Equation solvers in MENDEL v3.1

In the new 3.1 version of MENDEL released in 2022, two new approaches for solving the Generalized Bateman Equations have been introduced.

2.2.1 Extension of the analytical solver

The first idea is that, for some very particular depletion chains, the depletion matrix can be triangularized. It is not possible for the complete and accurate depletion chain generally used for fuel cycle studies, but it applies to some industrial depletion chains where only several isotopes of interest have been selected. In particular, for core level calculation in some CRONOS2 [18] applications, users decide, for time-consuming reason, to get rid of several isotopes or reactions. In this way, they obtain some triangular matrices on the isotopes of interest, for example on the Xenon/Iodine chain.

If the matrix is triangular, we solve the Bateman equations by computing directly the exponential of matrix on the depletion time of interest, using the same algorithm we historically only used for cooling periods.

As shown is section 3.1, one can achieve a huge speedup, for accurate (analytical) result.

Nevertheless, such triangularized matrices are quite rare, and this algorithm can be used only for very specific applications.

2.2.2 Hybrid algorithm

As the depletion matrix cannot always be considered triangularized, a suggestion would be to divide it in several blocks, and use a "per block" resolution of the Bateman equation.

For a general depletion matrix *A*, we can always distribute lines and columns in a new order such as *A* reads:

$$A = \begin{pmatrix} L & 0\\ S & B \end{pmatrix} \tag{4}$$

where:

- *L* is a triangular-inferior matrix;
- *B* is a non-triangular matrix;
- *S* is the source term from isotopes in the *L* block to isotopes in the *B* block.

S can be, depending on the methodology used for the subdivision of the *A* matrix, either null, either non-null.

While using Eq. 4, we can retrieve the Bateman equations as a system of two new sets of Bateman equations on two segregated isotope vectors. If we note N the isotopic vector related to full matrix A, N^L the isotopic vector related to matrix A and N^B the isotopic vector related to matrix B, Eq. 2 reads:

$$\begin{cases} \frac{dN^{L}(t)}{dt} = LN^{L}(t) \\ \frac{dN^{B}(t)}{dt} = SN^{L}(t) + BN^{B}(t) \end{cases}$$
(5)

Eq. 5 first line is directly a Bateman equations in the sense of Eq. 2, and, as matrix L is a low triangular matrix, it can be solved analytically using the new extension of the MENDEL's analytical solver to influx calculations. By using this approach, one will obtain a speedup proportional to the relative size of vector $N^L(t)$ relatively to the total number of isotopes in N(t).

By considering this point of view, one will tend to maximize the size of L. One can observe that the second line of Eq. 5 is made of two terms on the right-hand side. The second term can be considered as the classical term for Bateman equation, while the first one can be considered as a source term from the isotopes of $N^{L}(t)$. Using this approach, we can also use the already existing solvers in MENDEL, namely the Runge-Kutta and the CRAM solvers. Nevertheless, in current MENDEL approach, source terms are considered constant in time on one depletion step.

This new source is time-dependent, as these concentrations depend on time (as well as the *S* matrix if we consider the dependence of the flux in the reaction rate calculation). By taking a "constant approach" of this source term, we introduce a bias in our numerical calculation.

To better estimate the potential discrepancy, we describe in Section 3 the effect of this approximation. We decided to implement two options in MENDEL v3.1.

The first option is to accept to run fast but biased calculation. Users know they are doing an approximation, and the corresponding option is not to be run by default. In this case, the *S* matrix is non-zero, and the N_L vector size is maximized so that the *L* matrix is as important as possible. The first line of Eq. 5 is resolved by the analytical solver, the second line by the CRAM solver. CRAM solver order can be chose by the user (16 by default).

The second option consists of having no other approximation than the use of a CRAM numerical solver for the second line of Eq. 5. First line is still resolved by the analytical methodology. To avoid any effects of the source term, we construct the division between L and B so that S = 0. By using this option, we obtain two independent Bateman equation, with no links between them, and no discrepancy in the resolution. This second option ensures to obtain accurate results for all isotopes. We choose the N_L as large as possible, but the constraint of S = 0 can reduce drastically its size.

3. Approximation induced by the source term

3.1. Bias with non-zero source term on a short depletion chain

In order to show the discrepancies obtained by the Hybrid Method with a non-zero *S* matrix, we implemented a high burnup depletion calculation with a short chain of only 7 isotopes. Among those seven isotopes, the analytical solver applies on 3 of them, and isotopic densities at end of depletion are similar to any numerical solver. The CRAM solver treats the 4 others: Thorium 231, Thorium 232, Uranium 236 and Uranium 237 (231 Th, 236 U and 237 U).

As explained in Section 2.2.2, the source term containing the *S* matrix implies a discrepancy. Indeed, one needs to compute its integration in time taking into account its accurate time dependency, which is currently not possible in MENDEL. We should have a time depending source term, when MENDEL only offers constant source terms per depletion step. Two approaches have been used in MENDEL development version to correctly measure the bias introduced by the Hybrid Method with non-zero source term.

On the first hand, we use the trapezoidal rule to compute the integration of the $SN^{L}(t)$ term. We subdivise all depletion steps by *n* substeps, *n* between 1 and 10, and on each substep we compute a

trapezoidal form for the integration. This approach corresponds to an integration of the source term on the depletion step which will converge to the exact value when n tends to infinity.

On the second hand, we introduced in MENDEL an analytical solution of the computation of the integration of the $SN^{L}(t)$ term.

We show in Figure 1. the discrepancy between the Hybrid Method and the CRAM 16^{th} order solver (CRAM16). The full lines represent the convergence when *n* goes from 1 to 10. The cross on the right corresponds to the analytical integration. If there were no discrepancies due to the implementation of a constant source $SN^{L}(t)$ term on all depletion steps, all isotopic densities would tend to zero.



Figure 1. Relative discrepancy between Hybrid Method and CRAM on a toy box example.

We observe a good convergence of the trapezoidal rule to the analytical integration. For this reason, only analytical integration, which is less time consuming, has been introduced in MENDEL v3.1.

Nevertheless, the discrepancy with CRAM solver can be rather important, like Thorium 231 whose discrepancy is more than 1 %. Thorium 232 gives on the contrary very accurate results, with a less than 10^{-4} % bias.

3.2. Bias with non-zero source term on a complete depletion chain

To analyze the discrepancies with depletion chain generally used for fuel cycle studies, we run an existing OECD benchmark [18] with the complete structure chain of MENDEL, containing 3851 isotopes. Nuclear data are based on JEFF-3.1.1 evaluation. The irradiation history is constituted of 52 depletion steps. We compare the Hybrid Method and the Chebyshev Rational Approximation Method isotopic densities at the end of the last cycle.

At the end of the last cycle, 43 isotopes present discrepancies (less than 7 identical digits), and around 20 present more than 10% discrepancy, as shown in Figure 2.

At present time in MENDEL v3.1, we have no estimator of the discrepancy introduced by this bias, isotope per isotope. Due to this point, users need to compare with an accurate solution, and decide by themselves if they can use or not this faster but less accurate methodology.



Figure 2. Relative bias between Hybrid Method and CRAM using a depletion chain with 3851 isotopes.

On the current example, isotopes with discrepancy greater than 10% all have very low densities (roughly 10^{-12} gram per ton). For this reason, they have a negligible impact on quantities of interest for safety criticality applications. The only exception to low concentrations is the Lead 206 (²⁰⁶Pb), which is a stable isotope, and for this reason also have a negligible importance.

4. Time consumption of new methodologies

Those new methodologies have for main goal a time reduction of depletion calculation. We provide hereafter some details about time consumption.

4.1. Analytical solver

Triangular depletion chains with physical sense are always very short depletion chains. For this reason, the effect is difficult to assess. Nevertheless, we observe that a 2000 isotopes triangular depletion chain leads to a time reduction of a factor 50 when compared to CRAM 16th order.

If the matrix is triangular, users are encouraged to apply this new approach, more accurate and faster.

4.2. Hybrid algorithm

The hybrid method is tested on the 3851 isotopes chain of MENDEL, containing all isotopes and reactions needed for a reference activation calculation.

In this section, we will compare time consumption when running the Hybrid Algorithm comparatively to the standard CRAM solver. We will present five consumption times :

- The *Matrix creation*, meaning the subdivision of the depletion matrix, only done in the Hybrid Algorithm and which could be a source of increase of the algorithm cost;
- The time spent in the Analytical solver (only for Hybrid Algorithm);
- The time spent in the *CRAM* solver (in both approaches);

- The time spent in the *Depletion* calculation (from the matrix initialization to the output concentration calculation);
- The time spent in the *Total* fuel cycle calculation (meaning depletion and calculation of decay heat or other quantities of interest, printing results to output files...).

Therefore, the three first lines are included in the *Depletion* line and the *Total* is not a summation of the other lines.

4.2.1 Hybrid algorithm without source matrix

Taking into account an Hybrid Method with no approximation, out of the 3851 isotopes, only 21 isotopes are computed analytically. This approach is a worse case, where we create two new matrices even if the matrix associated to the fast solver is of a very small size. The observed speedup is roughly between 1 % and 3 % depending on the runs. We conclude that, even for very small numbers of isotopes treated analytically, the use of this Hybrid Method does not cost more than a standard CRAM approach. Therefore, it can be used on all matrices.

To observe a better speedup, we removed all the fission reactions from the depletion chain. By doing this, which is valid for non-fissile materials, 340 isotopes are computed by the analytical method. We observe the speedup shown in Table I.

Function	Time with Hybrid Method	Time with CRAM16		
Matrix creation	0.3 s			
Analytical	0.08 s			
CRAM	4.32 s	4.94 s		
Depletion	5.66 s	6.00 s		
Total	10.40 s	10.77 s		

Table I. Time comparison between Hybrid Method without bias and CRAM16

We observe a 6% acceleration on the depletion part of the computation, and a 3% acceleration of the complete calculation, which also includes decay heat computation.

Even if quite few isotopes are used in the analytical part of the Hybrid Method (roughly 10%), this new methodology gives a small speedup for the calculation, without any bias on the numerical results.

4.2.2 Hybrid algorithm with source matrix

On the 3851 isotopes standard depletion chain, the division of the matrix allowing bias leads to 2441 isotopes computed analytically (roughly 63%). Speedup is given in Table II.

Table II.	Time com	parison	between	Hybrid	Method	with bia	s and	CRAM16
				•/				

Function	Time with Hybrid Method	Time with CRAM16		
Matrix creation	0.40 s			
Analytical	0.26 s			
CRAM	4.26 s	12.83 s		
Depletion	5.95 s	13.68 s		
Total	15.53 s	23.30 s		

We observe an important time reduction of 57% of the time consumption for the depletion computation itself, which confirms the fact that the algorithm enables to gain a time factor nearly proportional to the fraction of isotopes computed analytically.

We made the same kind of calculations for several orders of CRAM solver, used both as full CRAM computation or CRAM for the numerical part of the Hybrid Method. By using orders 16, 32 or 48, we obtained, for all cases, between 58% and 55% of acceleration. We remind the readers that the numerical results are biased.

5. CONCLUSION

We introduced in MENDEL the possibility to use, for triangular depletion matrices, the analytical method for nuclear fuel irradiations and neutron activation problems. This methodology leads to more accurate results, as there are no more numerical approximation due to the numerical solver: approximation order in CRAM, or time subdivision in Runge-Kutta algorithm. Furthermore, calculation time is lower than in the pure numerical approaches. When possible, this methodology is recommended.

We also introduced a Hybrid Method between analytical and CRAM.

The approach with bias can lead to a huge time reduction, but users need to evaluate the approximation induced by this choice. It is to be used with care.

The approach without bias leads to nearly no acceleration, but there is also no real disadvantage to use it, as computing time is similar or slightly faster. The approach can be used.

In conclusion, all three approaches are considered valid, and can be used for time reduction. To continue the investigation on the Hybrid Method, it would be interesting to focus on the estimation and then reduction of the bias associated with the Hybrid fastest approach. One could think about introducing time dependent source terms in MENDEL.

ACKNOWLEDGMENTS

The authors thank EDF for its continuous financial support to the development of MENDEL code system and Framatome for its partial financial support to the specific topics of this paper.

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