



Qualitative Analysis of Physical and Mathematical Approximations Necessary for Induced Radioactivity Calculations in Fusion Devices

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November 1995

UWFDM-997

To be published in *Fusion Engineering and Design*.

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**Qualitative Analysis of Physical and
Mathematical Approximations Necessary for
Induced Radioactivity Calculations in Fusion
Devices**

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Abstract

When engineers and designers of fusion research and power reactors require information related to the induced radioactivity in the first wall, blanket and shield, they turn to one of many codes to generate a solution. Unfortunately, these codes all make physical and mathematical approximations in order to turn a functionally infinite and highly complicated problem into a more manageable, simple problem. This paper examines these approximations, namely, the handling of loops in the decay scheme, the truncation of the decay scheme, and the modeling of pulsed/intermittent operation histories, as physical approximations and the choice of a solution method as a mathematical approximation. The final recommendation is a code that uses an exact representation of the pulsing history, handles loops by unraveling, truncates the decay schemes with a relative sub-tree production rule modified by a stable truncation restriction and uses an adaptively chosen analytical or Laplace inverse mathematical solution method.

1.2 The Mathematical Representation

This problem is governed by very simple, linear, first order, ordinary differential equations. In its most simple mathematical representation, the time rate of change of density of an isotope is the destruction rate of the isotope subtracted from a sum of production rates from all parents. If we combine the equations for all the isotopes of interest, we get a set of coupled equations which, when written in vector and matrix notation, are of the form:

$$\dot{\vec{N}}(t) = \mathbf{A}\vec{N}(t) + \vec{S}(t)$$

where the matrix elements $a_{ij}, i \neq j$, are the production rates of isotope i from isotope j , the elements a_{ii} are the destruction rates of element i , and the vector \vec{S} is an arbitrary source term.

1.3 The Solution

Analogous to the scalar equation, the formal solution has the matrix exponential form,

$$\vec{N}(t) = e^{\mathbf{A}t}\vec{N}_o + e^{\mathbf{A}t} \int_0^t e^{-\mathbf{A}t'} \vec{S}(t') dt',$$

the calculation of which is a more daunting task than might be expected. Based on the physical representation of the system, this matrix \mathbf{A} can range in complexity from a simple lower-triangular, diagonalizable matrix to an arbitrarily filled defective matrix. In general, increased complexity of matrix \mathbf{A} represents improved physical modeling but requires more mathematical and/or computational approximation. The challenge is to use a physical representation which does not jeopardize the accuracy and allows a relatively simple mathematical solution method. Solutions to variations of this problem have been in existence for over 80 years, beginning with the analytical Bateman[1] equations for simple linear chains through to the modern computational methods for more complicated transmutation/decay schemes.

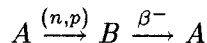
Due to the simplicity of the equations, a variety of independently developed codes exist[2][3][4][5][6][7] (to name a few), all using different physical and mathematical approximations which will be addressed and assessed here with the hope of providing suggestions of an optimum set of approximations.

2 Physical Approximations

The reason for most physical approximations is to preserve the tractability of the problem. As data libraries have become more comprehensive, including more isotopes and neutron reaction types, the possible size of the physical problem becomes unmanageable. Without these approximations, the problem would become simply too large or too mathematically complex, due to complicated matrices, both resulting in slow codes. There are three main sources of physical approximation: transmutation/decay loops, decay chain or scheme truncation, and pulsed/intermittent operation. Other sources of approximation exist, such as inaccuracies in nuclear data libraries, but are not within the scope of this analysis.

2.1 Loop Handling

During periods of irradiation, there is the potential for loops to occur. These loops are not possible in pure decay periods for the simple reason that since radioactive decay is a transition to a more stable state, it cannot be reversed without an energy source. With a neutron bombardment as an energy source, however, it is possible to create loops in the decay scheme. One such example is:



by which an isotope, A, is transmuted by a neutron with the release of a proton to an isotope, B, which decays by β^- emission back to A. There are other loops with different reactions and different lengths, that is, involving more than just two isotopes.

History

Currently, some codes model the transmutation/decay scheme (sample in Figure 1) with linear chains (see Figure 2), handling loops by simply truncating the chains at the onset of a loop, guaranteeing lower triangular matrices \mathbf{A} in the rate equations[3][4]. Another way is to model all the isotopes produced by a single input isotope in a tree structure, with each “branch” in the tree representing a transmutation or decay path. There are two methods by which to handle loops when using the decay scheme model. One is to include

the loops explicitly by modifying the rate equation of the first isotope in the loop to include production from its own child, guaranteeing that \mathbf{A} will be non-lower triangular[6][7] (see Figure 1). Alternatively, the loops can be “straightened”, or unravelled, where the production of the parent from the child is treated as the production of a pseudo-unique isotope and the different occurrences of the same isotope (see Figure 3) are combined after computation, resulting in a defective lower-triangular matrix \mathbf{A} , for which there exist efficient and accurate solution methods[2]. Finally, these trees with straightened loops can be separated into linear chains and solved one chain at a time, using a series of matrices, \mathbf{A} , all of which are bi-diagonal[8].

Related to loop handling is the issue of an isotope which may have two independent production paths. The different schemes will treat them all in different ways, but none should affect the level of accuracy.

There is much debate as to whether or not the contributions from loops are significant. In general the production of a grandchild (which is the closest relationship that an isotope may have to itself) is very small and due to the less likely neutron reactions which must take place for loops to occur, the contributions to the final result are thought, by some, to be negligible. In most cases this is a valid engineering approximation which can greatly lower the complexity of the problem and allow for

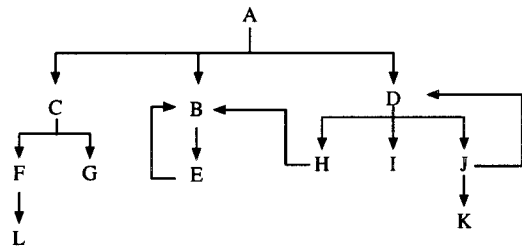


Figure 1. Sample reaction tree (explicit loop representation).

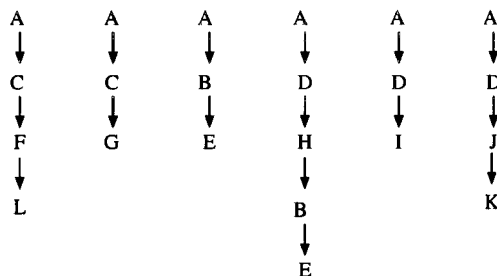


Figure 2. Linear chain representation.

very rapid solution methods, and thus, should not be dismissed. Mathematical purists feel that the contributions could be important in some low tolerance safety calculations and that they should always be handled with some accuracy, rather than discarded entirely. In addition, new methods are being developed to allow for the fast and efficient solution of systems that do handle loops.

Important Considerations and Discussion

The decision whether or not to handle loops is dependent on the nature of the problem. For rapid and numerous engineering calculations, the lack of loops is likely to play a small, if not negligible, role. On the other hand, for some studies that require precise calculation of certain isotopes to very small concentrations, the choice of how to handle loops may have important consequences on the final results. By not including the feedback from the loop, there is the risk that a significant contribution might be missed. On the other hand, this choice is often dictated by (or, in turn, dictates) the mathematical methods that can be used for the solution. This is significant if a method that accommodates loops introduces more mathematical error than is gained by increased accuracy.

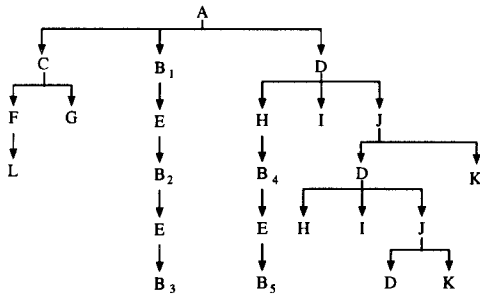


Figure 3: Straightened loops representation.

In those codes that use linear chains for their physical model, it is common to truncate at the onset of a loop[3][4]. Without loops, as in the case of pure decay, the analytical Bateman equations can easily be implemented. When these linear chains are formed with straightened loops, accurate methods have recently been developed to solve these systems[8]. Furthermore, when using linear chains, there is the possibility to treat each chain uniquely, determining whether or not it contains a loop, and if not, resorting to the highly accurate and efficient Bateman equations.

In those codes that use decay schemes, the two methods of explicit loop handling and loop straightening are used. The first method will give the most accurate results, since it is guaranteed to include all contributions of feedback from the child to the parent back to the child, and so on[6][7]. However, the need for non-lower triangular matrices results in solution methods that may be more mathematically approximate and/or subject to higher computational error. When straightening loops, the pseudo-unique isotopes are treated with the same truncation rules as the rest of the decay scheme, thus maintaining the same accuracy throughout the decay scheme[2], even though the accuracy of the loop solution is reduced. Since a limit has already been determined for the rest of the calculation, the penalty for choosing straightened loops is negligible while the benefit derived from the use of defective lower-triangular matrices allowing for more simple mathematical methods is significant. The inclusion of 2 or 3 iterations/corrections of the loop by straightening tend to introduce an error *relative* to the exact solution of less than 10^{-9} compared to an error as high as 0.2% when loops are ignored completely[8]. In addition, by straightening loops, as mentioned above, the tree can be separated into linear chains and solved using highly accurate methods.

2.2 Truncation

During periods of irradiation, every isotope, in theory, has some finite probability of transmuting to some other element. Thus, as we follow the decay schemes, they will continue indefinitely to include most isotopes known to exist. Clearly, this problem is far too large to be managed by any code and some way to limit its size must be chosen. Normally, a set of rules are devised to truncate the system while maintaining the accuracy of the physical model.

History

A variety of methods have been used, historically, to effect this truncation of the data schemes. Some codes may simply limit the decay scheme to a certain number of generations relative to the initial input nuclides. While this approach can be very revealing for certain studies, most codes use more complex sets of rules. Since radioactivity is the most important result, this simple rule can be improved by counting only stable isotopes as the generations and only truncating with a stable isotope. Another variant counts only transmutation reactions as generations[6][7]. Yet another alternative is to calculate the production of a child isotope from the initial isotope and compare that to a threshold value. Only an estimate is available[2] unless fast enough mathematical methods are developed to allow solving the entire problem. A modification of this is to calculate the relative production of the entire subtree below a particular child by preventing the transmutation/decay of the child during the reference calculation. A final modification is to perform this calculation and, if below the threshold criterion, truncate any subsequent transmutation branches while keeping subsequent decay branches[3][4]. Currently, these sets of rules, and variations, are used by the leading activation codes. While these rules appear simple and straightforward, their implications can be significant.

Important Considerations and Discussion

There are two main considerations when choosing a set of truncation rules. Primarily, it is important not to reduce the calculated radioactivity by not including enough isotopes in the decay scheme. If a scheme is truncated too early, many radioactive isotopes that may be created in the real system might be ignored, drastically affecting the accuracy of the calculation. Related to this issue is the conservation of atoms in the model. In a real system, there will always be at least as many atoms as there were of the initial isotope, with additional atoms coming from neutron interactions that produce light ions (say tritium (n, t) or helium (n, α)). However, because the model decay scheme is truncated, there is the possibility that atoms can transmute or decay out of the model system, thus “losing” atoms from the model.

The simplest rule is deficient in many areas. Mainly, by truncating with a maximum rank, it is possible to cut off a branch or sub-scheme that would have arbitrarily high production rates. By paying no attention to the production rate as a criterion, important information is bound to be lost while unimportant information will be kept. If we consider the variations based on counting stable isotopes or transmutation reactions[6][7], we will fall into the same problem that information that is not of interest may be retained at the expense of much more valuable results.

As suggested above, the best rule to start with is one that is able to predictively estimate the production of an isotope from an initial isotope[2][3][4]. This can be done by simplifying the decay scheme between the initial isotope and the isotope of interest to a linear chain and using an analytical solution to estimate the production using a reference flux and period. With the development of new mathematical methods for solving linear chains with straightened loops, this

calculation is possible for all chains as they are being created[8]. Even better is to calculate this relative production exactly for a reference flux and the exact problem history. While this may take longer than the other methods, it produces more accurate results.

After all this, however, the problem of conservation of atoms in the model has not been addressed. If the relative production of an isotope is low enough that the scheme is truncated immediately following that isotope, atoms will transmute or decay out of the model from that isotope throughout the operation and, if it is radioactive, throughout the shutdown history. This effect can manifest itself in two significant ways. First, during operation, if an isotope's concentration peaks early in the history, a reference calculation of its concentration may reveal an amount less than the tolerance and it will be truncated. However, such a result may indicate that significant quantities of material have decayed/transmuted "through" this isotope and out of the model. Second, after shutdown, if the reference calculation is only carried out for the operation period and the chain is truncated on a radioactive isotope, it may decay significantly out of the model. This issue does not affect the accuracy of results for included isotopes as much as it may result in grossly under-calculated activity or toxicity of the system if the excluded isotopes are radioactive or toxic. It results not just in lost accuracy, but completely missing information. These "lost" products could be important isotopes radiologically or toxicologically, leaving the safety analysis incomplete, or important for material composition, leaving an analysis of the material response with uncertainties.

The effect during operation can easily be eliminated. During the reference calculation, if the bottom isotope in the chain is prohibited from decaying/transmuting the result is no longer the concentration of that isotope, but the sum concentration of all isotopes in a sub-tree below this isotope, i.e., the total amount of material that was converted to this isotope, regardless of whether or not it has been converted again to something else.

After shutdown, the only way that atoms can be converted out of the model is by decay. Thus a simple way to prevent this is to disallow truncation with a radioactive isotope. This restriction would also allow the truncation of the transmutation branches of radioactive isotopes once below the tolerance because it is impossible for these pathways to be followed during decay periods. Another approach would be to use a reference irradiation period for the relative production calculation that includes the after-shutdown time. This allows for a determination of the number of atoms which will be converted out of the model throughout the problem, giving rise to three rules:

1. If the relative production of a sub-tree is greater than the specified threshold at shutdown, do not truncate.
2. If the relative production of a sub-tree is less than the specified threshold at shutdown, but greater at some after-shutdown time,
 - (a) if is is stable, truncate completely.
 - (b) if is is radioactive, truncate only subsequent transmutation branches.
3. If the relative production of a sub-tree is less than the specified threshold at shutdown and at all after-shutdown times, truncate completely.

These rules can be summarized in Table 1 where 'C' indicates to continue the chain without truncation, 'T' indicates to truncate the chain completely, and 'T*' indicates to truncate all subsequent transmutation branches.

As a final measure to increase the accuracy of the solution the engineer or scientist using the code could simply lower the tolerance. This does, however, introduce the danger of solving a problem

		Relative Production at Shutdown	
		> tolerance	< tolerance
Relative Production After Shutdown	> tolerance	Stable: C Radioactive: C	Stable: T Radioactive: T*
	< tolerance	Stable: T Radioactive: T	Stable: T Radioactive: T

Table 1. Outcome Chart for Truncation Rules

which in most cases is far too large and too accurate for the sake of achieving a high accuracy in one part of the problem. It is important to note that cases may arise where an engineer might be interested in the production of a particular isotope in very small quantities, much less than the production of other isotopes in the system. In such a case, it may be more fruitful to implement a code that operates in reverse, performing an “adjoint” calculation, starting with the isotope in question and finding all the sources of that isotope[9].

2.3 Pulsed/Intermittent Operation

Current and future research fusion reactors are not designed to operate continuously; inertial confinement systems are intrinsically pulsed, and physics considerations are suggesting that magnetically confined power reactors may have to operate on some sort of pulsing scheme (see Figure 4). The physical model used to represent this pulsed/intermittent operation has important implications both in the final result and the mathematical method used to solve the equations. In addition, many research and power reactors would have pulsing schemes that would change with time, periodically or otherwise, representing shutdown periods for weekends or maintenance.

History

There is a rich history of models used for this aspect of the problem, originating from the study of activation in inertial confinement fusion, pulsed fission reactor, or accelerator systems. The simplest of these models just approximates the pulsed history with a single steady state flux

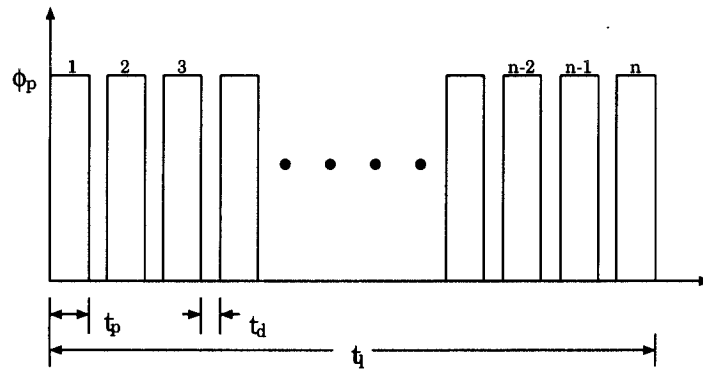


Figure 4. Sample pulsed irradiation history.

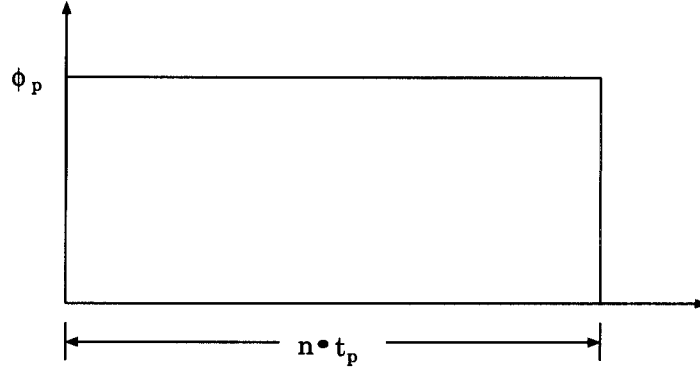


Figure 5. First steady state approximation.

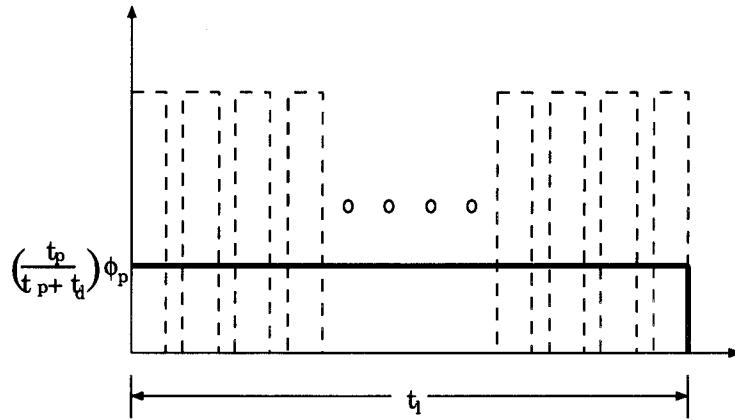


Figure 6. Second steady state approximation.

period by one of two primary methods[5][6]. First (see Figure 5), by conservation of fluence and total operation time, a steady state flux equal in magnitude to the pulse flux would operate for a time equal to the total time the reactor would be operating at power. Otherwise (see Figure 6), by conservation of fluence and total reactor lifetime, a steady state flux lower than the pulse flux would operate for a time equal to the total lifetime of the reactor.

A more sophisticated approximation (see Figure 7) assumes that the material is only subjected to one pulse and removed from the reactor and allowed to decay until the shutdown time, this being repeated for each pulse[3][10][11]. Another method is to use the appropriate steady state model for all but the last few pulses and then use an exact pulsing history for those last pulses (see Figure 8). Finally, a number of codes will perform complete solutions using the exact irradiation history for the entire problem[2][4][7].

Important Considerations and Discussion

Physically, the pulsed nature results in a buildup of transmutation products during the operation pulses and decay of those isotopes during the “dwell” periods between pulses. For isotopes of certain half-lives relative to the ratio of dwell time to operation time, the pulsing representation can have

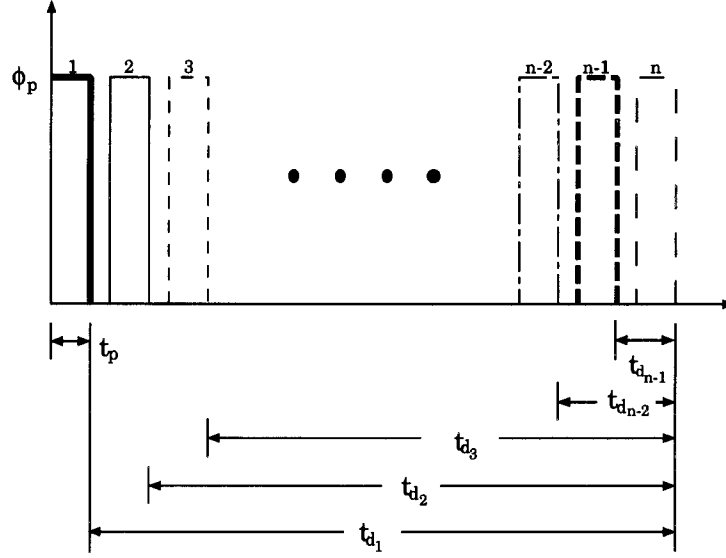


Figure 7. Continuous removal approximation.

a profound impact on the final calculated number density. The most accurate solutions will result from methods which allow for this transient behavior during the dwell times to correctly assess the radioactivity.

Although one approximation is more valid than the other in certain cases, Sisolak et al.[12] showed that both pure steady state approximations above will either overestimate or underestimate the activity for certain medium-lived isotopes by up to 1 order of magnitude. The conserved operation time approximation, used primarily for the analysis of magnetic confinement systems, is reasonable for long-lived isotopes that would not decay appreciably during the dwell times in the real system such that their final density is the result of a continuous build throughout the operation times. Similarly, in the time scales used in magnetic fusion engineering, short-lived isotopes would reach secular equilibrium in each pulse and decay significantly during the dwell time so that the

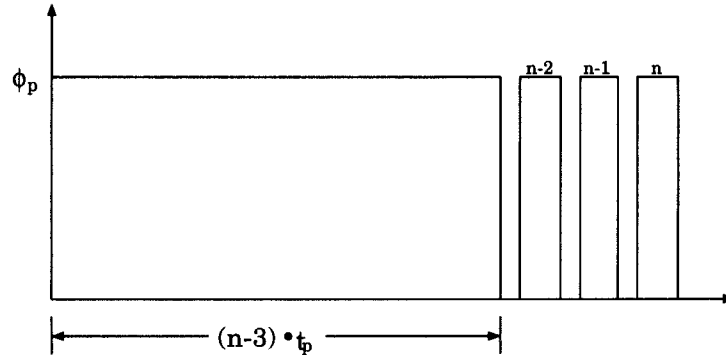


Figure 8. Steady state/pulsed hybrid approximation.

exact result is dominated by the behavior of the last pulse. Thus, the secular equilibrium density predicted by the single long operation would be the same as the density in the real system. On the other hand, for medium-lived isotopes, the unmodelled incomplete buildup and decay in the real system during the pulses and dwell times, respectively, result in significant overcalculation of the activity. For the other steady state approximation, often used for inertial fusion engineering, similar arguments can be used to show why it routinely underestimates the activity of short-lived isotopes.

Because the pulse widths are so much smaller than the dwell times in inertial confinement fusion, the continuous removal approximation *can* be a good one for pulsed/intermittent irradiation histories without removal. The burnup of the initial isotope is approximated well, but the burnup of other stable isotopes in the decay scheme, and therefore the buildup of their transmutation products, is grossly inaccurate. Thus, for any history with long pulses and/or short dwell periods, or with many stable isotopes early in the decay scheme, this approximation will be invalid[12][13]. This approximation is only truly valid when used to model a component that is removed after each pulse.

Clearly, the exact model does not introduce any level of approximation whatsoever, but for some solution methods, it makes the problem prohibitively long. For that reason, the steady-state/pulsed hybrid method was developed. In particular, if combined with the conserved operation time steady-state model, the transients of the last few pulses can correct much of the problems encountered as a result of the pure steady state method.

The multi-level pulsing schemes are best represented by the exact model because the change in time scales and patterns further invalidates the other approximations, such as reference flux and period for truncation. One of the problems with determining a reference period is the nature of the history. Since the reference flux is usually a constant level of the same order as the operation flux that will be used in the calculation, it is logical to use the total operation time as the reference period, thus simulating the more conservative of the steady state approaches above. Since this is only used for creating the model and this steady-state approximation is conservative, it is a good choice.

3 Mathematical Approximations

In this section, the inaccuracies due to mathematical approximations will refer to both the approximations made in the development of the method and those computational errors unique to the method. For example, while all methods will be subject to round-off errors, some algorithms will amplify this error while others may attenuate it.

Mathematical approximations are necessary in many cases to facilitate the computational solution of the system of differential equations mentioned in the introduction. Due to the simplicity of the equations themselves, very small systems could be solved by hand without approximation. However, the large systems preclude the manual analytical solution of the problem and, until recently, exact computational solutions for the optimum physical models suggested above have not been identified or implemented[14].

The relationship between the mathematical method and the physical model are very important. In general, as the physical model is made more accurate, the mathematical methods become slower and/or less accurate. First, whether or not the physical model includes loops and how those loops are implemented is crucial to the selection of a method. If loops are implemented explicitly,

the matrix is non-triangular, and a more complicated method must be used to solve the matrix exponential. If loops are implemented by straightening, a simpler matrix method can be employed. Second, the model for pulsed histories is an important factor. In this case, however, the greater accuracy in the physical model implied by the use of the exact pulsed history need not lead to detrimental effects on the efficiency and accuracy of the mathematical solution.

History

Currently implemented mathematical methods can be divided into three types: analytical or expansion matrix methods, non-matrix time-step solvers, and hybrid matrix/time-step methods.

In this context, analytical is used to mean mathematically exact, closed form solutions to the problem. In all cases, these methods are subject to computational error when implemented, although the effect of the method on those errors will vary. The simplest analytical matrix method is a matrix representation of the Bateman equations for linear chain based codes[3][4]. Because of the lower-triangular nondefective nature of the matrix \mathbf{A} , the solution can be calculated by using forms of the standard equations derived by Bateman 80 years ago. Related to this method is eigenvector decomposition[15], in which the matrix \mathbf{A} is decomposed into its eigenvectors and eigenvalues, which are then raised to the exponential, and recombined to get the solution. When the linear chains are extended to include straightened loops, the mathematical method must also be extended. Some such extensions derive from a Laplace transform analysis of the problem and, performing either an exact Laplace inversion or an expansion in the Laplace transform parameter, which is then inverted, inserting the results into a matrix[8]. It is important to note that both Laplace techniques, when applied to chains with no loops, reduce exactly to different formulations of the Bateman solution. Another recently implemented analytical matrix method is the generalized eigenvector expansion method[2]. This method decomposes the matrix \mathbf{A} into its eigenvalues and generalized eigenvectors and recombines them after applying the exponential to form the mathematically exact solution. The Schur decomposition[7][16] produces a triangular matrix, which can be exponentiated using one of the above methods if necessary, and then recombines the matrices to reach a solution. The greatest source of inaccuracy is in the algorithms which perform the decomposition.

Approximate non-matrix methods use standard ordinary differential equation [ODE] solving packages or algorithms, break the history into time steps, and sequentially solve the problem. The standard solvers that have been used to date include Runge-Kutta, Euler (and variations [5]) and GEAR[6]. The approximation in these methods is a result of the discretization of time.

Finally, approximate hybrid methods can use a matrix solution to solve a time step problem and continue in such a fashion until reaching the end of the history. A popular example of this is an implementation which combines a series solution to the matrix exponential with short time steps to accelerate convergence of the series[17][18]. Another approximate hybrid method is known as “scaling and squaring” in which the matrix exponential is scaled down by powers of two until it is small enough to solve with a quickly converging series solution, and then the result is successively squared until reaching the final operation time[16].

Important Considerations and Discussion

There are two primary considerations: accuracy and speed.

Being the chief topic of this paper, accuracy deserves special mention here because even mathematically exact methods can be subject to gross errors as a result of various methods of implementation. The basis of inaccuracy is that the matrix \mathbf{A} is ill-conditioned and/or defective.

The system is very stiff having time constants ranging from ms or less to 10^{10} y and is therefore

ill-conditioned. This causes large round-off errors in some analytical methods and the smallest time constant forces the ODE solvers to use short time steps on long histories. The Bateman equations, for example can be implemented as a sum of single exponentials or a sum of differences of exponentials[3][4]. The latter is superior because it can be modified to reduce round-off error in cases when the destruction rates of two isotopes are nearly identical.

The defective nature of the matrix, when loops are handled, is of chief concern when using the matrix decomposition methods mentioned above. In these cases, the standard eigenvector method is not useful at all, while the generalized eigenvector and Schur decomposition are useful depending on what type of loop handling is employed. If the straightened loop handling is used, the generalized eigenvector method is sufficient to solve the problem completely. If explicit loop handling is employed, however, the Schur decomposition is required, but there is the potential that the triangular matrix that must be exponentiated is defective. In such situations, the simple recursive algorithms that have been developed for solving functions of triangular matrices break down and a generalized eigenvector or computationally similar method must be used to solve that sub-problem[19][20].

In general, a study of the accuracy and speed of nineteen ways to compute this solution[16], all dubbed “dubious,” was inconclusive, saying “that some of the methods are preferable to others, but that none are completely satisfactory.” Although not directed at our specific problem, this illuminating study chose three or four methods as “best” candidates, including specialized ODE methods, a series method known to fail, and matrix decomposition methods, citing specifically Schur decomposition and variants thereof.

With the same physical model as the generalized eigenvector method, another alternative is to employ either of the Laplace inversion methods[8]. First, it should be noted, that when these methods are applied to a linear chain without loops, they become two different representations of the Bateman equations, one an exact sum of exponentials, the other an accurate expansion of a sum of differences of exponentials, so organized to minimize round-off error. When loops are included, however, both methods can be employed to get rapid and accurate results. Since they do not solve the system as a matrix exponential, but rather calculate each term independently by taking advantage of the nature of the physical model, they will not be prone to the errors found in many matrix exponential techniques. Thus, the generalized eigenvector technique can be eliminated from contention.

The issue of speed is becoming paramount as the codes are asked to provide results for larger and more complicated systems as a result of more comprehensive and inclusive data libraries. It is also clear that the matrix methods are unique in allowing the rapid solution of systems with exact pulse history models. A transfer matrix, \mathbf{T} , can be found to represent the transient for a single pulse, a decay matrix, \mathbf{D} , can be found to represent the transient for a single dwell time, and the solution for n pulses is[14]:

$$\vec{N}(t) = (\mathbf{T}\mathbf{D})^{n-1}\mathbf{T}\vec{N}_0.$$

A variety of methods can be used to raise the matrix to this power, the most useful of which is a finessed successive multiplication[21]. The drawback of the time-step based methods is that the solution is generally not found as the result of a matrix transform, but rather, the number density vector is found for the end of each time step and used as the initial value for the next time step. As a result, these methods tend to be very slow, having to repeat all the calculations at each time step.

Based on the above comments, the only methods which for this problem combine speed and

accuracy are matrix decomposition methods and the Laplace methods. While the Schur decomposition method is mathematically exact, the algorithms needed to perform the decompositions may amplify the computational errors more than the Laplace techniques. Unfortunately, for both the decomposition method and the exact Laplace inversion technique, the final solution has the same drawbacks as the sum of single exponentials formulation of the analytical Bateman equation method mentioned above, which can cause problems with nearly degenerate eigenvalues. On the other hand, in cases where the expansion can be calculated with enough accuracy, the Laplace expansion technique is perfect for removing these effects.

4 Conclusions and Recommendations

The above arguments all point to certain optimums in the approximations made for the calculation of induced radioactivity and related values.

The first and most obvious recommendation is to use a method that has exact modeling of the pulsed history. Research has shown that the modeling of an arbitrary pulsed system with a steady state flux can and will produce severely erroneous results. There are methods available, in terms of other physical approximations and mathematical solutions, which allow the exact pulsing solution to be as accurate and fast as any other type of model.

Similarly, if efficient solution methods exist, there is little reason not to handle loops. Although this does add complexity to the mathematical solution, the contribution of loops could be significant in some problems. The method by which they are handled is really dependent on which mathematical method is chosen and what the truncation philosophy is. It is important to note, however, that with the existence of methods to solve linear chains with loops, a linear chain representation opens the door to an adaptive calculation, one which uses the standard Bateman solution if no loops exist, but uses one of the extensions, the Laplace techniques, if loops do exist.

A more contentious issue in physical approximations is that of truncation philosophy. The most reasonable first rule is to truncate based on some comparison of the production of the last isotope to the first isotope. Furthermore, the additional information gained from adding the restriction that the decay scheme may only be truncated after a stable isotope easily balances the cost of a larger problem. The other important truncation decision is what reference period to use while performing the truncation calculations. Since extremely fast mathematical methods do exist, the most reasonable approach is to solve the entire problem throughout the operation history and after shutdown. Therefore, a relative production tolerance rule with a stable last isotope modification, as defined in Table 1 is the best truncation philosophy.

Perhaps the subject of most debate is that of mathematical methods, yet a best choice does exist. To allow the solution of the exact pulsed history in a reasonable time, a true matrix method is necessary. To handle loops, as recommended above, neither the linear chain based Bateman analytical method nor the eigenvector decomposition is useful. This leaves the Schur decomposition method and the Laplace techniques. The inverse Laplace method is not subject to the computational errors that might arise in the decomposition. Further, the Schur decomposition results in a matrix that then requires a robust method for the exponentiation of the triangular matrix. Thus, the only advantage to Schur decomposition is that it can be used to handle loops explicitly. The loop straightening method is as accurate as the explicit method within the tolerance being determined in the truncation rules, hence there is no reason to add the additional computational complexity of explicit loop handling. Therefore, the best mathematical methods are the Laplace

techniques, especially if chosen adaptively with the simple Bateman solution.

While these issues are the most important in terms of speed and accuracy, there are many other issues which should be considered when choosing an activation code for *your* problem. For example, even if the code does use the optimum combination of these approximations, it may be important that it allow for the solution of multi-dimensional problems in a variety of geometries. This is just one of the many usability issues which must also be considered.

In conclusion, the optimum calculation uses an exact pulsing history, straightened loop handling with linear chains, a relative production truncation rule with a stable truncation restriction, and an adaptive mathematical method chosen from the Bateman solution and the Laplace techniques. While many of these recommendations were being implemented in the GERAPH[2] code, the complete set of recommendations has been implemented in a new code, ALARA, in the final stages of development at the University of Wisconsin-Madison.

Acknowledgements

This work would have never been started without the the many hours of fruitful discussion and debate with the late Prof. Emeritus Charles Maynard and Jim E. Sisolak of the Nuclear Engineering and Engineering Physics Department of the University of Wisconsin-Madison. Partial support for this work was provided by the Fusion Technology Institute of the University of Wisconsin-Madison under DOE Grant DE-AS08-88DP10754 and the University of Wisconsin-Madison Reactor Safety Analysis Group.

References

- [1] H. Bateman, *Proc. Cambridge Phil. Soc.* **15**: 423 (1910).
- [2] P.P.H. Wilson, J.E. Sisolak, and D.L. Henderson, "GERAPH: A Novel Approach to the General Solution of Pulsed History Activation Problems," *Fusion Tech.* **26**, 1092 (Nov. 1994).
- [3] D.L. Henderson and O. Yasar, "DKRICE: A Radioactivity and Dose Rate Calculation Code Package," UWFD-714, University of Wisconsin Fusion Technology Institute, Vol. 1, Madison, Wisconsin (1986).
- [4] DKR-PULSAR is a new version of the DKRICE code which implements methods from Reference [12] for the exact treatment of pulsed history irradiation. It is being developed by D.L. Henderson and H. Khater at the University of Wisconsin-Madison.
- [5] R.A. Forrest and J-Ch. Sublet, "FISPACT3 - User Manual," AEA/FUS 227, April 1993.
- [6] J. Jung, "RACC: Theory and Use of the Radioactivity Code RACC," ANL Report: ANL/FPP/TM-122, May 1979.
- [7] H. Attaya, "Input Instructions for RACC-P," ANL Report: ANL/FPP/TM-270, September 1994.
- [8] P.P.H. Wilson and D.L. Henderson, "Expanding Towards Excellence: Ironing Out DKR's Wrinkles," UWFD-995, University of Wisconsin Fusion Technology Institute, Madison, Wisconsin (November 1995).
- [9] A. White, "Transmutation and Activation Analysis of Fusion Power Plants," Doctoral Thesis, University of Wisconsin-Madison (May 1985).
- [10] L.-P. Ku and J.G. Kolibal, "Radioactivation Characteristics for the Tokamak Fusion Test Reactor," *Nuc. Tech./Fusion* **4**, 586 (1983).
- [11] H. Etherington, *Nuclear Engineering Handbook*, McGraw-Hill Book Company, Inc., New York, 1958, Chapter 7.
- [12] J.E. Sisolak, S.E. Spangler, and D.L. Henderson, "Pulsed/Intermittent Activation in Fusion Energy Reactor Systems," *Fusion Tech.* **21**, 2145 (1992).
- [13] S.E. Spangler, "A Numerical Method for Calculating Nuclide Densities in Pulsed Activation Studies," Master of Science Thesis, University of Wisconsin-Madison (August 1991).
- [14] S.E. Spangler, J.E. Sisolak, and D.L. Henderson, "Calculational Models for the Treatment of Pulsed/Intermittent Activation Within Fusion Energy Devices" *Fusion Engineering & Design*, **22**, 349 (July 1993).
- [15] H. Fukumoto, "New Approach to Neutron-Induced Transmutation, Radioactivity and After-heat Calculations and Its Application to Fusion Reactors," *Nuc. Sci. & Tech.*, **23**, 97, February 1986.
- [16] C. Moler and C. Van Loan, "Nineteen Dubious Ways to Compute the Exponential of a Matrix," *SIAM Review*, **20**, 801 (Oct. 1978).

- [17] M. Bell, "ORIGEN-The ORNL Isotope Generation and Depletion Code," Report ORNL-4628, May 1973.
- [18] J. A. Blink, "FORIG: A Computer Code for Calculating Radionuclide Generation and Depletion in Fusion and Fission Reactors – Users's Manual," Report UCRL-53633, March 1985.
- [19] B.N. Parlett, "A Recurrence Among the Elements of Functions of Triangular Matrices," *Linear Algebra Appl.*, **14** (1976). pp. 117-121
- [20] B.N. Parlett, "Computations of Functions of Triangular Matrices", Electronics Research Laboratory, University of California Berkeley Memorandum No. ERL-M481 (November 1974).
- [21] E.S. Lee, "Computer Engineering: Computer Algorithms, Data Structures, and Languages," Prepared Notes, University of Toronto, 1989.