Chapter 8

Depletion Calculations

An important calculation that complements transport solves is the computations of the change in the composition of the reactor due to fissioning of fuel into fission products, transmutation of nuclides from neutron absorption, and radioactive decay (among other more exotic processes such as photofission). Such calculations are usually called depletion calculations because the context for such an analysis is ofter the design of reactors and computing the amount of fissile material "burned" or depleted between refueling.

The typical depletion calculation is done with the following assumptions:

The first assumption is true if we look a particular homogenized region or a small enough mesh zone. The second approximation is reasonable if the time step (usually called a depletion step) is small enough *and* the flux-changing fission products are at or near equilibrium values in the transport calculation for the scalar flux. This is particularly important for xenon (and to a lesser extent samarium) because the difference between startup and equilibrium concentrations of these poisons can greatly affect the scalar flux and power profile.

The depletion problem can be stated succinctly by first defining a vector of N nuclides with densities (in atoms per volume) as

(8.1)

and writing the net change in nuclide i as

(8.2)

The initial condition is

$$n_i(0) = nI0.$$

The form of this equation has all the loss of nuclide n_i couched as an effective decay given by

(8.3)

with the decay constant given by

(8.4)

The cross-sections here are microscopic cross-sections where σ_{ij} is the cross-section for absorption of neutrons in nuclide i producing nuclide j. In the case of fission products the cross-section to produce a fission product would be the product of a the fission cross-section and the fission yield. The production of nuclide i is represented as an effective decay into nuclide i from all other nuclides by defining an effective branching ratio as

(8.5)

Equation (8.2) is a linear, first-order system of equations that we can write the system as

(8.6)

Where the matrix entries are given by

(8.7)

An example problem

Consider a simple depletion problem where a sytem of depleted uranium is bombarded with neutrons from an accelerator. We only concern ourselves with plutonium production and destruction. Therefore, we only follow isotopes of uranium, neptunium, and plutonium and a single americium isotope (^{241}Am). We use the convention of referring to nuclides by the shorthand $n_{\alpha\beta}$ where α is the last digit in the atomic number (Z) of the nuclide and β is the last number in the mass number of the nuclide A. For example, the density of $^{239}_{94}$Pu is n_{49} The equations for ^{238}U and heavier uranium isotopes are

$$\begin{aligned}
\frac{d}{dt}n_{28} &= -\phi\sigma_{\mathrm{a}}^{28}n_{28} \\
\frac{d}{dt}n_{29} &= -(\lambda_{29} + \phi\sigma_{\mathrm{a}}^{29})n_{29} + \phi\sigma_{\gamma}^{28}n_{28} \\
\frac{d}{dt}n_{20} &= -\lambda_{20}n_{20} + \phi\sigma_{\gamma}^{29}n_{29}.
\end{aligned}$$

The neptunium equations are

$$\frac{d}{dt}n_{39} = -(\lambda_{39} + \phi\sigma_{\mathrm{a}}^{39})n_{39} + \lambda_{29}n_{29}$$
$$\frac{d}{dt}n_{30} = -\lambda_{30}n_{30} + \lambda_{20}n_{20} + \phi\sigma_{\gamma}^{39}n_{39}$$

The neptunium equations are

$$\frac{d}{dt}n_{49} = -\phi\sigma_{\mathrm{a}}^{49}n_{49} + \lambda_{39}n_{39}$$
$$\frac{d}{dt}n_{40} = -\phi\sigma_{\mathrm{a}}^{40}n_{40} + \lambda_{30}n_{30} + \phi\sigma_{\gamma}^{49}n_{49}$$
$$\frac{d}{dt}n_{41} = -(\lambda_{41} + \phi\sigma_{\mathrm{a}}^{41})n_{41} + \phi\sigma_{\gamma}^{40}n_{40},$$

we neglect the higher isotopes of plutonium. The ^{241}Am equation is

$$\frac{d}{dt}n_{51} = -\phi\sigma_{\mathrm{a}}^{51}n_{51} + \lambda_{41}n_{41}.$$

We can write the system as

(8.8)

$$\mathbf{A} = \begin{pmatrix}
-\phi\sigma_{\mathrm{a}}^{28} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\phi\sigma_{\gamma}^{28} & -(\lambda_{29} + \phi\sigma_{\mathrm{a}}^{29}) & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \phi\sigma_{\gamma}^{29} & -\lambda_{20} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \lambda_{29} & 0 & -(\lambda_{39} + \phi\sigma_{\mathrm{a}}^{39}) & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \lambda_{20} & \phi\sigma_{\mathrm{a}}^{39} & -\lambda_{30} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \lambda_{39} & 0 & -\phi\sigma_{\mathrm{a}}^{49} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \lambda_{30} & \phi\sigma_{\gamma}^{49} & -\phi\sigma_{\mathrm{a}}^{40} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & \phi\sigma_{\gamma}^{40} & -(\lambda_{41} + \phi\sigma_{\mathrm{a}}^{41}) & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \lambda_{41} & -\phi\sigma_{\mathrm{a}}^{51}
\end{pmatrix} \tag{8.9}$$

Note that **A** has units of inverse time. If we plug in the values of these quantities (using thermal cross-sections) and assume the scalar flux is 10^{14} per $\text{cm}^2 \cdot \text{s}$, for units of inverse days, we get

$$\mathbf{A} = \begin{pmatrix} -0.000023328 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0.000023328 & -42.47389008 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0.00019008 & -1.17982 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 42.4737 & 0 & -0.2954744 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1.17982 & 0.0005184 & -138.629 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.294956 & 0 & -0.00839808 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 138.629 & 0.00236736 & -0.00273888 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0.00228096 & -0.011052877 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0.000131877 & -0.0046224 \end{pmatrix} about \tag{8.10}$$

Notice the dynamic range of entries in this matrix: some processes happen very quickly, e.g., ^{239}U decays to ^{239}Np with a half-life of 23.5 minutes whereas ^{241}Pu decays to ^{241}Am with a half-life 14.4 years. Another way to look at it, there ratio of the largest value in the matrix to the smallest is about 1.05×10^6. Moreover, the matrix is clearly not diagonally dominant in that the diagonal elements are not larger in magnitude than the off-diagonal terms in each row.

8.1 Analytic Solution

A first-order linear system has a very simple solution. In particular we can write the solution using the matrix exponential:

(8.11)

The matrix exponential is defined my appealing to the Taylor series expansion of a typical exponential around 0

(8.12)

to write

(8.13)

We can use this formula to calculate the analytic solution to our example problem and see how the solution behaves as a function of time. At this time it is worth pointing out that depletion time steps can be on the order of days, months, or even years. For a small system, such as our example, the matrix exponential is possible to calculate. For example, `Mathematica` has a matrix exponential function one can use.

Nevertheless, many reactor problems track hundreds or thousands of nuclides. For such a situation, with the large range of time scales in the problem, the power series approximation is numerically dubious and unstable.

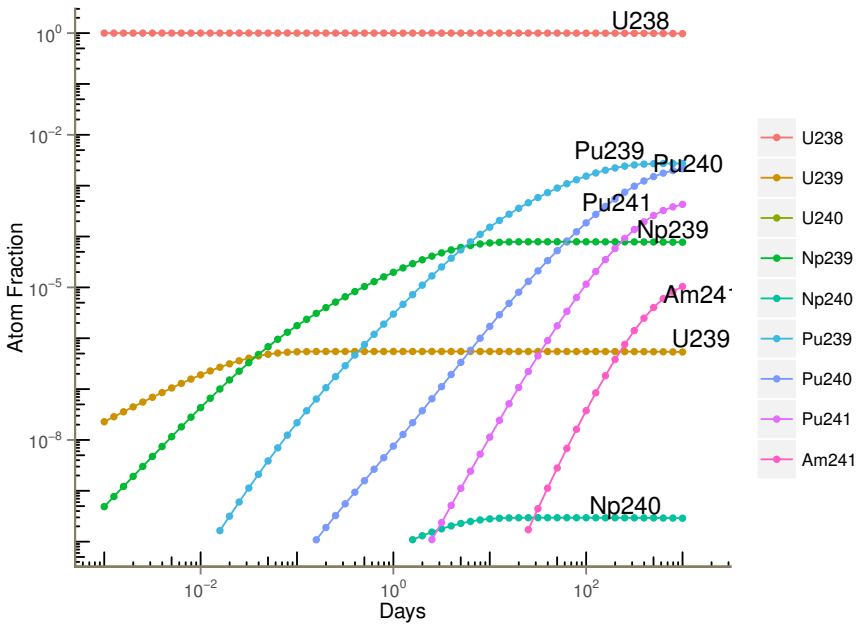

Figure 8.1: Matrix exponential solution for our simple system with a variety of depletion steps.

8.2 Backward Euler Solution

We have already covered the backward Euler method for time integration. This method is rarely used in depletion calculations, but it is useful to demonstrate how these calculations work. Using the backward Euler method, the approximate solution to the depletion equations is

(8.14)

The astute reader will of course notice that this solution is the first-order Padé approximate to the matrix exponential. The first-order Padé approximation to a simple exponential is a rational function that is constant in the numerator and linear in the denominator given by

(8.15)

Notice that the backward Euler solution could be unstable if t is large enough to make the matrix $(\mathbf{I} - \mathbf{A}t)$ singular.

8.3 Rational Approximation and Quadrature Formulas (with a minimum of complex integration theory)

From the Cauchy integral formula the value of any analytic function, that is a function without singularities, be related to the value of the function via the integral in the complex plane

(8.16)

where Γ is any closed-contour in the complex plane that encloses a and moves counter-clockwise. An related statement is the residue calculus which gives

(8.17)

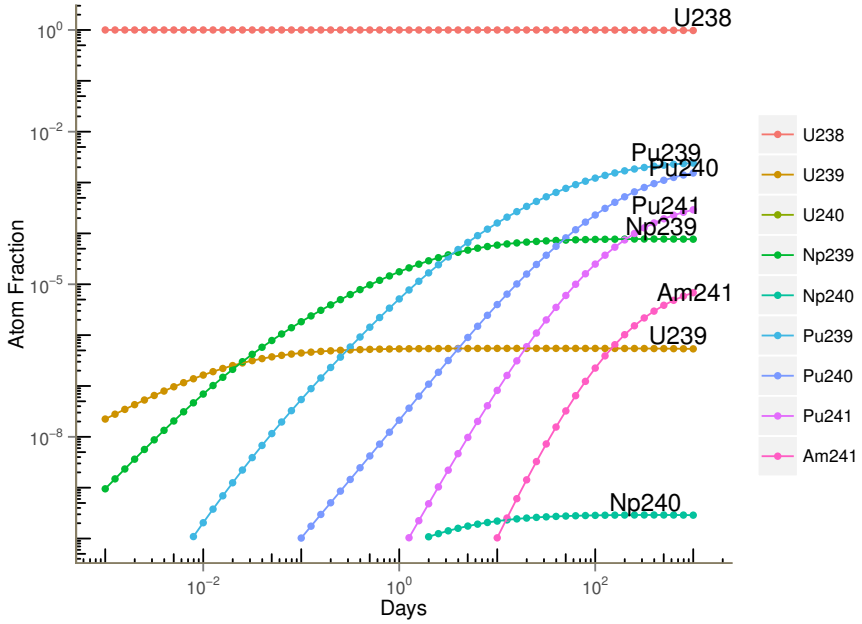

Figure 8.2: Backward Euler solution for our simple system with a variety of depletion steps.

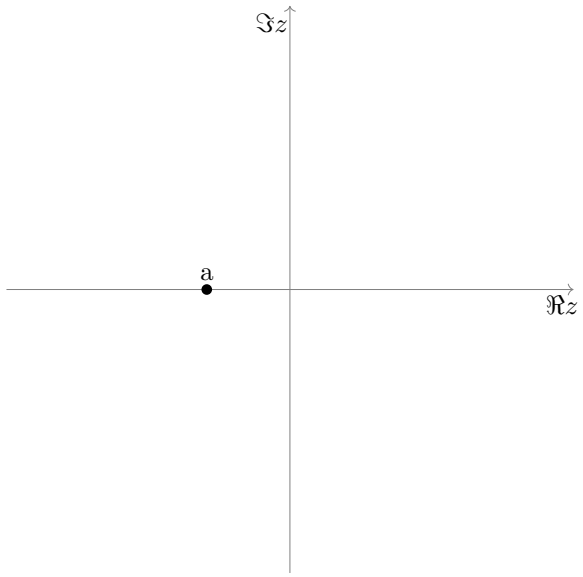

Figure 8.3: Example contours, Γ, in the complex plane to compute e^a.

We can use the Cauchy integral formula to evaluate an exponential function at any negative exponent by writing

(8.18)

where now we choose the contour Γ to cross the real axis somewhere with $\Re z > 0$, and continues out to $\Re z \to -\infty$. One of the beautiful things about contour integration in the complex plane is that you can usually "push-off" some part of the contour in an infinitely far in a given direction and not affect the integral. See, for instance, that in the Cauchy integral formula there are very few restrictions on the actual contour.

Now for the exponential function with a negative exponent, the integration contour must wrap around the negative real axis (because a could be anywhere on there). We could choose an integration contour that is a parabola that opens to the left, has a maximum value somewhere on the positive real axis, and continues to a point d with $\Re d < 0$. At point d the ends of the parabola are connected by a straight, vertical line. In the limit of $d \to -\infty$ the contribution from the straight line also tends to zero because the magnitude of the exponential goes to zero as the real part of the exponent goes to negative infinity. Therefore, we can write the value of the exponential as

(8.19)

where Γ_{para} is the contour just of the parabola, without the connected part.

The next step is to say, what if I don't want to evaluate the integral. Can I use a quadrature rule to estimate the integral? Moreover, you might ask, on the parabolic contour what is the best quadrature rule I could define? These are good questions dear reader because they have very interesting answers. To perform quadrature along a contour in the complex plane, we just need to define a mapping from a known interval to the contour, and map a quadrature rule from that integral to the contour. In particular if the function, $\phi(\theta)$, maps the

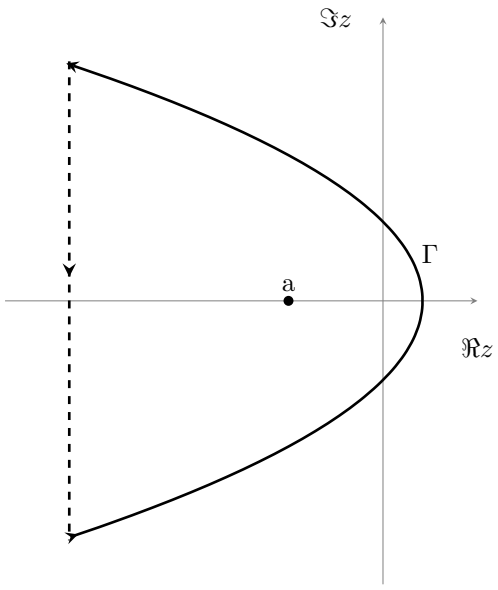

Figure 8.4: An example parabolic contour to compute e^a. The dashed part of the contour can be made arbitrarily small by moving it farther down the negative real axis.

real line onto the contour, then we can write Eq. (8.27) as

(8.20)

For the quadrature rule, we will use the trapezoid rule or its cousin, the midpoint rule, because these are exponentially accurate on a period or unbounded domain provided the integrand is analytic[1]. Moreover, since the integrand decays exponentially, we make an exponentially small error by truncating the domain of integration to a finite domain. For simplicity, we take this domain to be $[-\pi, \pi]$ and use N points creating subintervals of width $2\pi/N$. The N points we label θ_k and the corresponding $z_k = \phi(\theta_k)$ and $w_k = \phi'(\theta_k)$. This makes Eq. (8.20)

(8.21)

where we have written our N point approximation to e^a as E_N^a. By making some substitutions we can write this result as the sum of rational functions:

(8.22)

Based on the residue calculus result in Eq. (8.17)

(8.23)

Therefore, we can write

(8.24)

[1]This is pretty neat and was known to Poisson in the 1820s. Turing also examined this behavior.

The function

(8.25)

is an order $(N-1)/N$ rational function. We can look at the difference between the original function e^a and the approximation, by noting that if we reverse the contour in Eq. (8.26) and multiply by a minus sign, we can make the original contour match Γ_N along the parabola and do the usual tricks to make every other part of the contour go to zero. Therefore, we can write

(8.26)

This important result tells us that if our quadrature is accurate, then $r(z)$ is likely to be a good rational approximation to e^z and, conversely, any rational approximation can be thought of as a quadrature rule.

(8.27)

where Γ_{para} is the contour just of the parabola, without the connected part.

We still have not answered the question of how accurate can we make the quadrature. It turns out, that if we make $\phi(\theta)$ a parabola that is a function of N, so that the error in the truncation of the domain to $[-\pi, \pi]$ is smaller than the other errors, we can get a quadrature rule that has an error of order $O(2.85^{-N})$. This is geometric convergence, better than the algebraic convergence typically seen in numerical methods. With this rate of convergence we improve by about 2 orders of magnitude by adding four points. Whoa!

The $\phi(\theta)$ that achieves this convergence is given by

(8.28)

Moreover, since we are interested in a real function, the imaginary part of the integrand will cancel from the evaluations above and below the real axis. This means we only need to consider the points in $[0\pi]$ and multiply the result by 2.

To demonstrate this, let us use the midpoint rule with $N = 4$ to compute e^{-1}. The two points we need to evaluate are $\theta = \frac{\pi}{4}, \frac{3\pi}{4}$. This corresponds to

$$z = 0.22899231 + 0.78539816i, \quad z = -2.12786922 + 2.35619449i,$$

with weights given by

$$w = -0.75021233 + 1i, \qquad w = -2.25063698 + 1i.$$

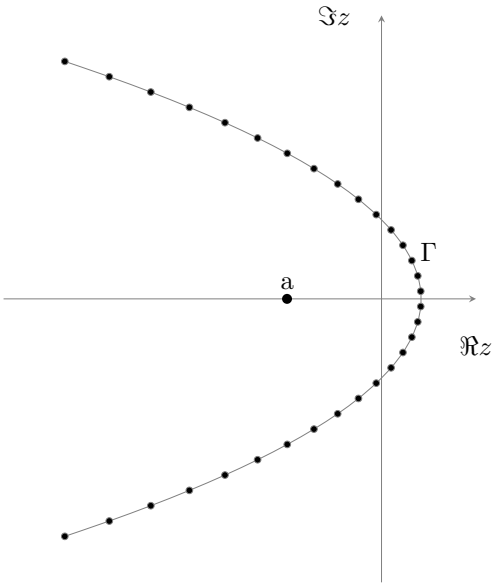

Figure 8.5: Quadrature points for the parabolic contour with $N = 32$.

Evaluating the constants gives

$$c = -0.05551957 - 0.38901455i, \quad c = 0.06843395 + 0.026329i.$$

Putting everything together in Eq. (8.34) gives the result

$$e^{-1} \approx \Re\left[-2 * \frac{-0.05551957 - 0.38901455i}{0.22899231 + 0.78539816i + 1} - 2 * \frac{0.06843395 + 0.026329}{-2.12786922 + 2.35619449i + 1}\right] = 0.355842404052. \quad (8.29)$$

The error in this approximation is about 0.01203704 or about 3.3%. If we were to compute the value of $e^{-0.1}$ we only need to change the denominators:

$$e^{-.1} \approx \Re\left[-2 * \frac{-0.05551957 - 0.38901455i}{0.22899231 + 0.78539816i + 0.1} - 2 * \frac{0.06843395 + 0.026329}{-2.12786922 + 2.35619449i + 0.1}\right] = 0.909007953393. \quad (8.30)$$

This is only a 0.46% error using only two quadrature points. The utility of this approach is that by storing a handful of constants I can evaluate e^a for any negative a to the precision desired.

It turns out that we can do better if we do not use a parabola. For instance, if $\phi(\theta)$ is a cotangent function we can get a convergence rate of $O(3.89^{-N})$. The $\phi(\theta)$ that gives this is

$$\phi(\theta) = N[0.5017\theta \cot(0.6407\theta) - 0.6122 + 0.2645i\theta]. \quad (8.31)$$

where Γ_{para} is the contour just like the parabola, without the connected part.

8.3.1 Matrix Exponential via Quadrature

These results are great if one wants to evaluate the exponential quickly, but the connection to our desired matrix exponentials is not yet clear. Or is it? Nothing about what we did was special for exponentials. All we said is that our integrand was an exponential divided by $z - a$. We could generalize our result to the equation

(8.32)

Therefore, to use our quadrature rules and theory from above we need to assert that the poles of the function $(z\mathbf{I} - \mathbf{A}t)^{-1}$ have only negative real parts and are not too far from the real line. This second requirement is due to the fact that our contours have only a finite distance from the real axis in the complex plane. We can show that the only places where $(z\mathbf{I} - \mathbf{A}t)$ is singular occur at the eigenvalues of $\mathbf{A}t$ using the resolvent formalism, but one probably can surmise this fact because finding the values of z such that $(z\mathbf{I} - \mathbf{A}t)$ will be singular is one definition of the eigenvalues of $\mathbf{A}t$.

One can argue via physical grounds that all the eigenvalues of $\mathbf{A}$ will have real parts that are less than zero because otherwise there would not be a finite steady state to the system. Also, the imaginary parts have been observed to be small. The eigenvalues with a complex part are part of a cyclic chain of nuclides. These observations typically observe imaginary parts of eigenvalues with magnitudes on the order of 10^{-8}.

With this knowledge, we can push ahead with our geometric quadrature rules to compute $e^{\mathbf{A}t}\mathbf{n}_0$. The steps in doing so will be the same as evaluating the scalar exponential, except now instead of dividing by $z_k - a$, we will be solving the system

(8.33)

In other words, each quadrature point requires the solution of a linear system. We then sum up these solutions to get our approximation

(8.34)

```
import numpy as np
import scipy as sp
import scipy.sparse as sparse
import scipy.sparse.linalg as splinalg
N = 32
t = 1
theta = np.pi*np.arange(1,N,2)/N
z = N*(.1309 - 0.1194*theta**2 + .2500j*theta)
w = N*(- 2*0.1194*theta + .2500j)
c = 1.0j/N*np.exp(z)*w
u = np.zeros(I*J*K)
for k in range(int(N/2)):
phi,code = splinalg.gmres(z[k]*sparse.identity(I*J*K)  - A*t,b, tol=1e-12, maxiter=2000)
        if (code):
            print(code)
        u = u- c[k]*phi
```

Figure 8.6: Simple python code to compute matrix exponential $e^{\mathbf{A}t}$.

where we have only $N/2$ terms and the factor of 2 appears because the matrix exponential we seek is real. Therefore, the evaluation of the matrix exponential takes $N/2$ linear solves.

8.4 Best Rational Approximation

Rather than adapting a quadrature rule to estimate the exponential, we could ask what is the function $r(z)$ that minimizes the integrand in Eq. (8.26). To do this we seek the rational function of order (m, n) with $m < n$ that satisfies

(8.35)

If we select $m = N-1$ and $n = N$, the best rational approximation will yield an error that converges as $O(9, 28903^{-N})$, or about double the previous rules. This procedure of finding the best rational approximation is sometimes known as the Chebyshev best rational approximation. To find the best rational approximation we write the generic function

(8.36)

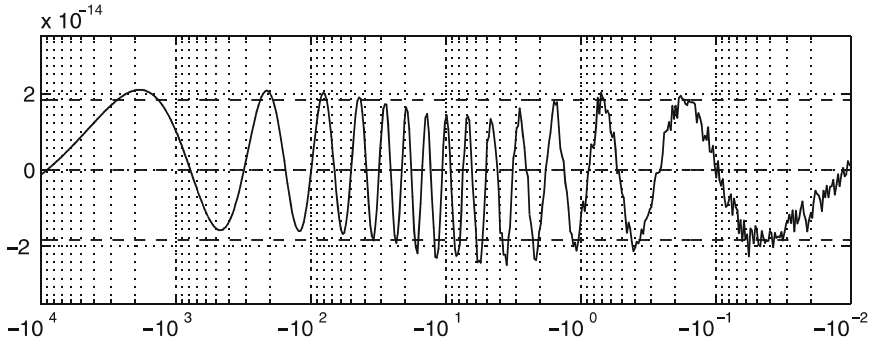

Figure 8.7: Value of $|e^z - r^*(z)|$ with $N = 14$. Figure taken from Ref. [5]

Table 8.1: Poles and Residues for the best rational approximation to the exponential with $N = 14$. For this function, $r^*(\infty) \approx 0$

k	z_k	$c_k/100$
1	5.623151880088747 + 1.194068309420004i	-0.278754565727894 - 1.021482174078080i
3	5.089353593691644 + 3.588821962583661i	0.469337296545605 + 0.456439548888464i
5	3.993376923428209 + 6.004285841369451	-0.234984158551045 - 0.058083433458861i
7	2.269789514323265 + 8.461734043748510i	0.048071353323537 - 0.013210030313639i
9	-0.208754946413353 +10.991254996068200i	-0.003763599179114 + 0.003351864962866i
11	-3.703276086329081 +13.656363257468552i	0.000094388997996 - 0.000171848578807i
13	-8.897786521056833 +16.630973240336562i	-0.000000715408253 + 0.000001436094999i

The form we write this best approximation is

(8.37)

where $r^*(z)$ the best rational approximation of order (N, N). There are algorithms available to compute $r^*(z)$, both with fun names, the Remes algorithm and the Carathéodory-Fejér algorithm[2]. A version of the CF algorithm is given for Matlab in Ref. [5]. This code is used to compute the residues, c_k, and poles, z_k, for the $N = 14$ case.

Using the these points we can compute the matrix exponential as in Figure 8.6, except that now the z_k and c_k are from this table.

8.5 Transmutation Trajectory Analysis

The method known as Transmutation Trajectory Analysis (TTA) solves the decay equations by looking at the decay and transmutation reactions and writes

[2]The CF algorithm is approximate, but the errors are $O(56^{-N})$.

the reaction networks as a set of linear chains. The population of each of these chains can be independently solved and then combined to get the total population of nuclides in the system. The resulting algorithm then only needs to solve linear chains, which can be done analytically in principle. In practice, when a branch in a chain occurs one then treats each branch as an independent linear chain. Difficulties can arise when chains are cyclic, i.e., the same nuclide appears multiple times in a chain as happens when an (n,2n) reaction is followed by radiative capture. As we will see, it is possible to treat this with a simple damping factor to terminate the chain after some number of cycles.

Consider a linear chain where only the first nuclide, n_1, has a nonzero initial concentration. Bateman, in 1910, gave the solution for the lth nuclide in a chain as

(8.38)

where

(8.39)

and

(8.40)

Notice that this solution is undefined when the effective decay constants for two nuclides are equal. This will clearly happen if the chain is cyclic, but can also happen if there is a large number of nuclides. In practice this is treated by making the adjustment

(8.41)

A typical value of ϵ is 10^{-5} and the condition $j > i$ means that we can adjust the decay constants as we progress down the chain.

We decide when to stop a chain by looking at the passage, $P_l(t)$, which is the fraction of the initial concentration of the first nuclide that has passed through

the first l nuclides in the chain. The passage is calculated via the formula

(8.42)

In this equation m is the total number of nuclides in the chain. It is possible that B_i is a small number, so that the second means of calculating the passage in Eq. (8.42) is not numerically stable. It, however, is possible to compute the passage as one moves along the chain by recognizing that the passage is equal to the ratio of the concentration of the next nuclide in the chain assuming that it does not decay to the initial nuclide:

(8.43)

The passage is used to terminate chains if the remaining nuclides are negligibly small. For instance one could cut chains when

(8.44)

for some cutoff value δ. Common values of δ are 10^{-15} and 10^{-20}.

8.5.1 Example of TTA

We will demonstrate the TTA approach and the Bateman solution using the depleted uranium example from before. To begin we look at all the linear chains in the system. In Figure 8.8 the entire transmutation network is shown. Examining this figure one can break the entire network into three chains, shown in Figures 8.9 through 8.11. Each of these chains has the same beginning, ^{238}U to ^{239}U by radiative capture. To compute the solution for this network, we first compute the concentration of ^{238}U and ^{239}U as a function of time using the Bateman solution in Eq. (8.38). Then for each of the three chains we compute the solution for the remaining members of the chain and add the solution together for each nuclide because some nuclides appear in multiple chains. We do not need to consider the passage cutoff for these chains because they are short and not cyclic.

We could also complicate matters by adding an $(n, 2n)$ reaction to the chain to make it cyclic. In Figure 8.12 there is now a cycle between ^{239}U and ^{238}U. Now for the calculation we have an additional chain that is the cycle between these

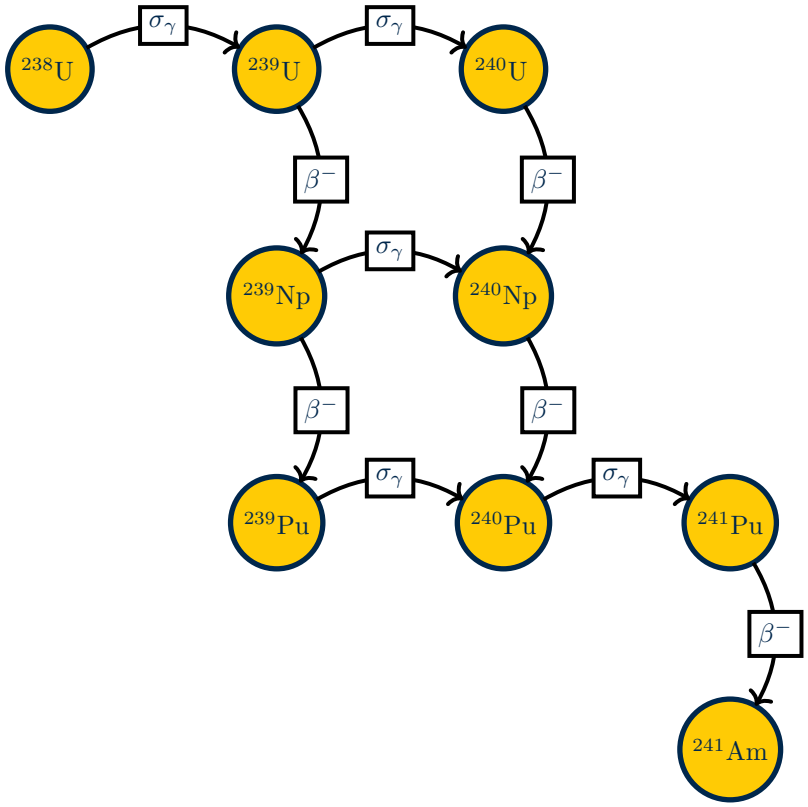

Figure 8.8: A transmutation network for ^{238}U.

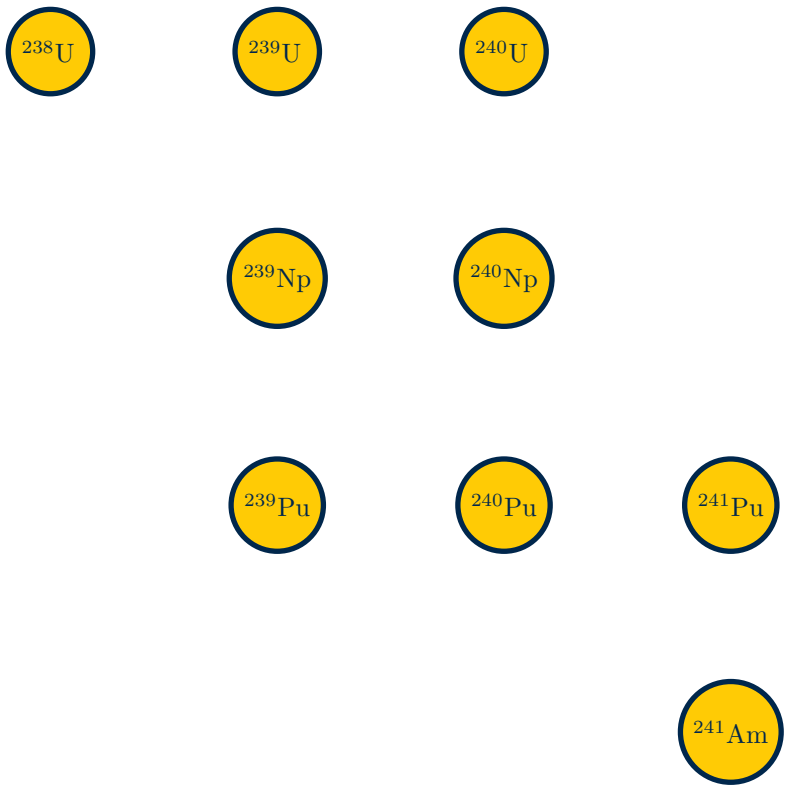

Figure 8.9: First of three linear chains for the ^{238}U transmutation network.

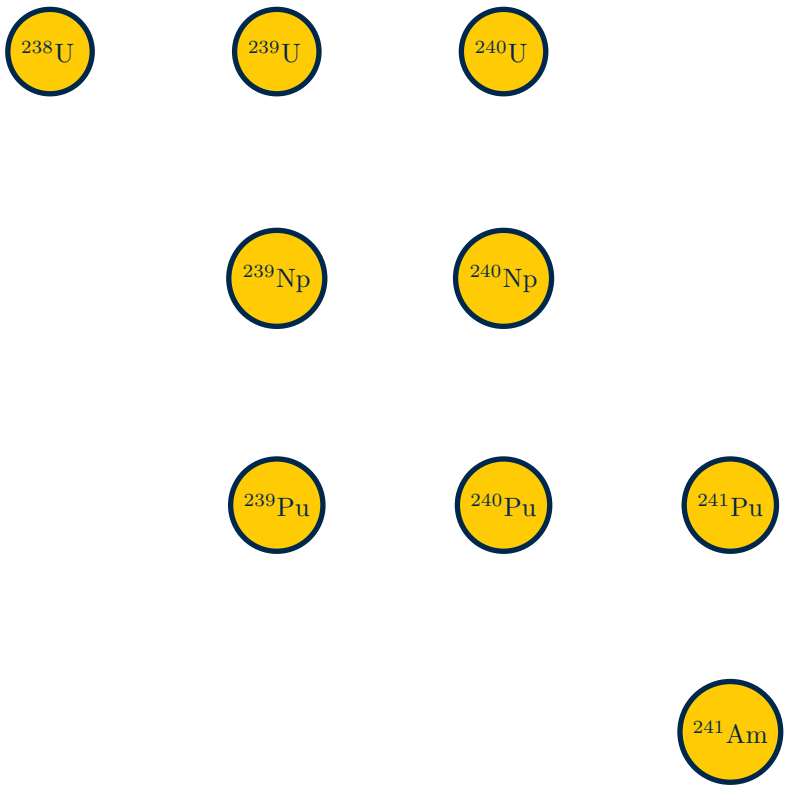

Figure 8.10: Second of three linear chains for the ^{238}U transmutation network.

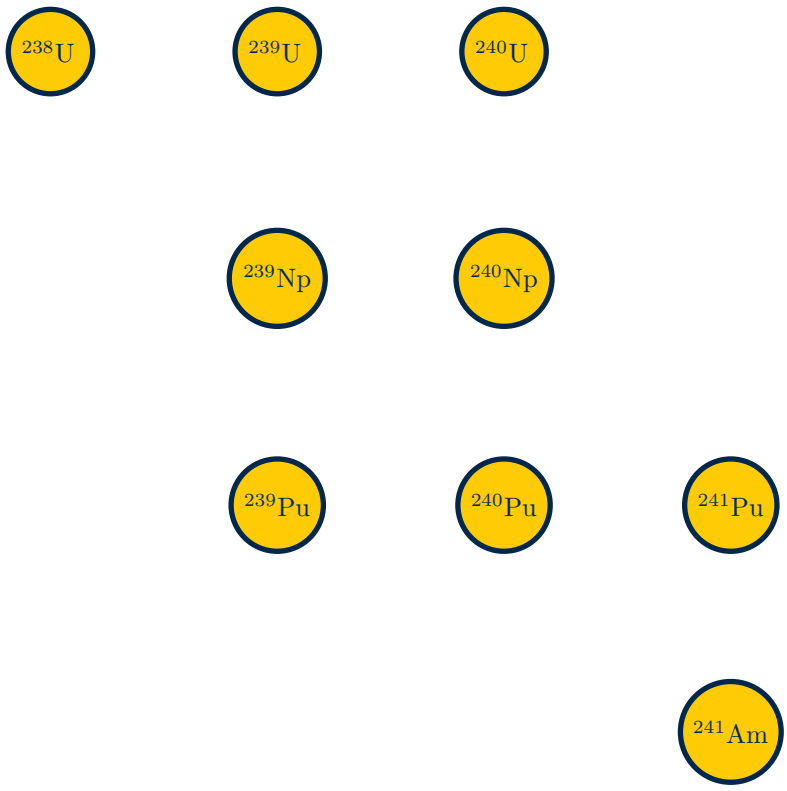

Figure 8.11: Third of three linear chains for the ^{238}U transmutation network.

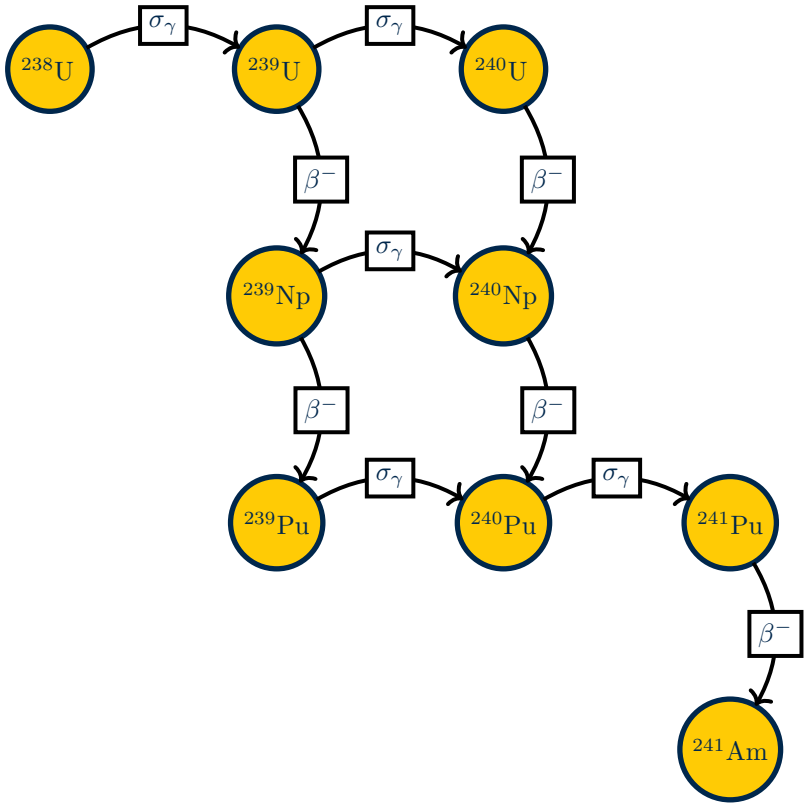

Figure 8.12: A transmutation network for ^{238}U with a cycle.

two. The cycle will be terminated by adjusting the decay constant and having a passage cutoff. For completion we show a reaction network that includes a fission product in Figure 8.13. This adds another chain to the system that will have to be handled. At this point, it is probably clear that developing the reaction networks and linear chains for a full-blown depletion calculation is a straightforward, though not simple, endeavor.

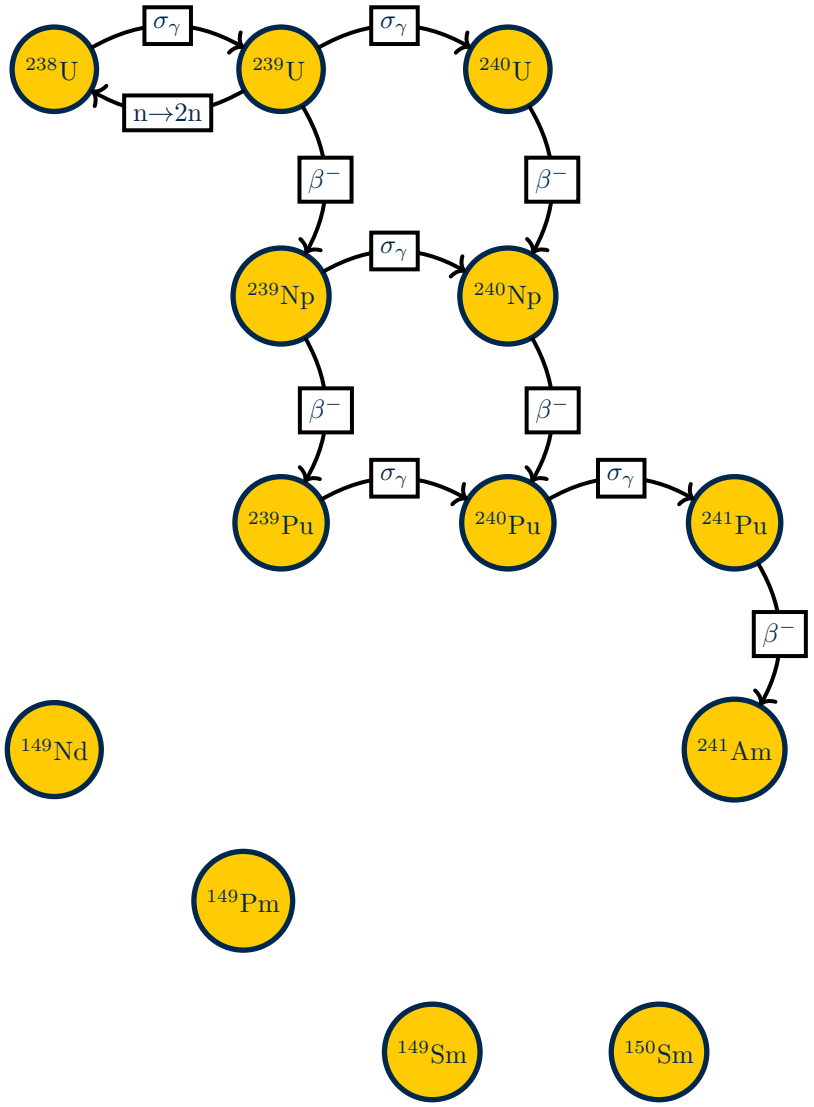

Figure 8.13: A transmutation network for ^{238}U with a fission product.