

# **Burnup matrices**

**Things discovered while implementing depletion capability to Serpent**

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**November 8th, 2013**

## Outline

- Burnup equations and matrix exponential solution
- Characteristics of burnup matrices
  - Known issues and recent discoveries
- Chebyshev Rational Approximation Method (CRAM)
- Some numerical results

## Burnup equations

- Form a system of ordinary differential equations:

$$\mathbf{n}' = \mathbf{A}\mathbf{n} , \quad \mathbf{n}(0) = \mathbf{n}_0 , \quad (1)$$

- Matrix exponential solution

$$\mathbf{n} = e^{\mathbf{A}t} \mathbf{n}_0 \quad (2)$$

- There are various numerical algorithms for computing the matrix exponential but many of them are computationally expensive and of dubious numerical quality [1]

[1] C. MOLER and C. VAN LOAN, *Nineteen Dubious Ways to Compute the Exponential of a Matrix, Twenty-Five Years Later*, *SIAM Rev.*, **45** (2003).

## Known issues about burnup matrices

- Contain both positive (off-diagonal) and negative (diagonal) elements
  - Time steps vary from a few hours to several months
  - Size  $\sim 1700 \times 1700$
  - Magnitudes of eigenvalues vary dramatically
    - Short-lived nuclides inducing eigenvalues with largest magnitudes are the most problematic
- ⇒ Matrix exponential previously not computed for a full system
- In ORIGEN short-lived nuclides are removed from the burnup matrix before computing the matrix exponential solution



## Elements of burnup matrices

- Diagonal elements  $\leq 0$ , off-diagonal elements  $\geq 0$

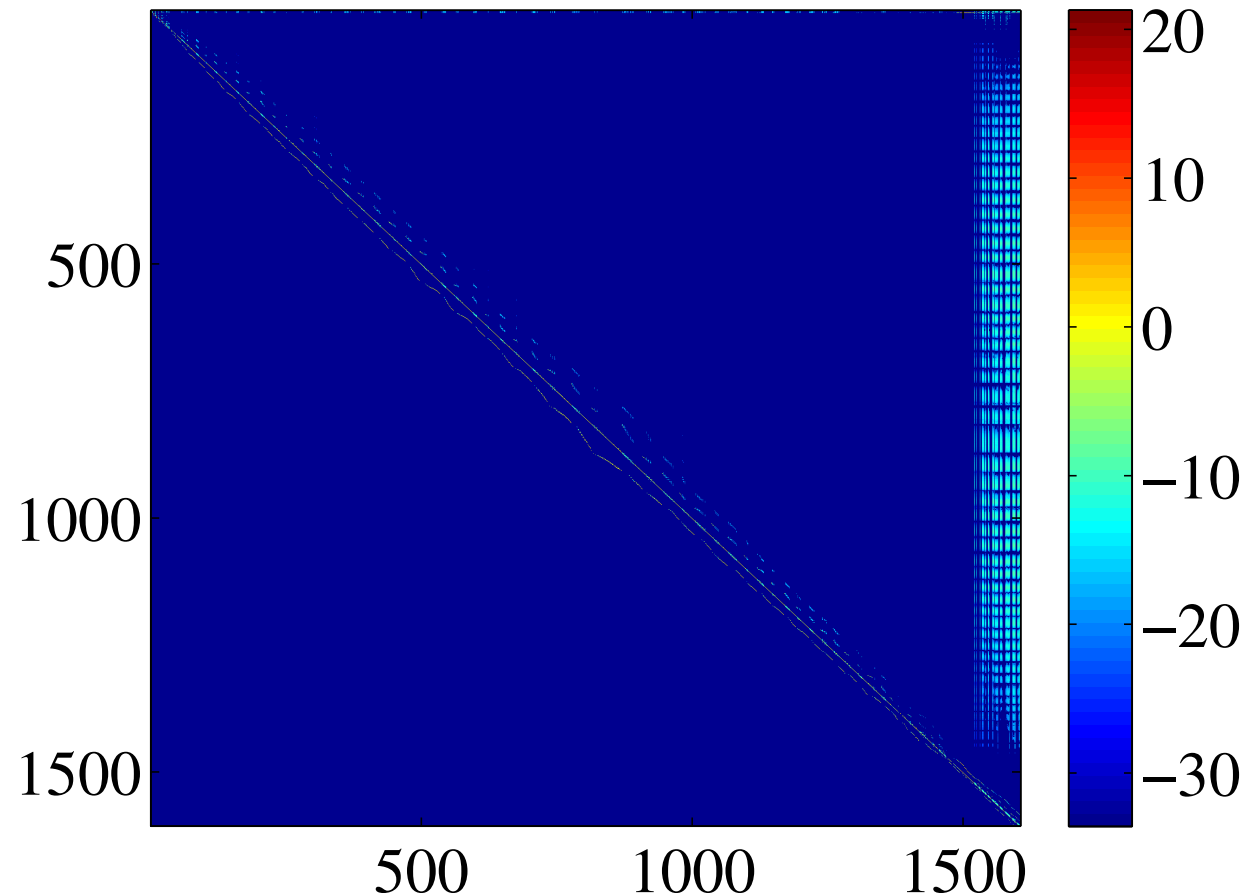


Figure 1: Logarithmic variations in the absolute values of burnup matrix elements.

## Elements of burnup matrices

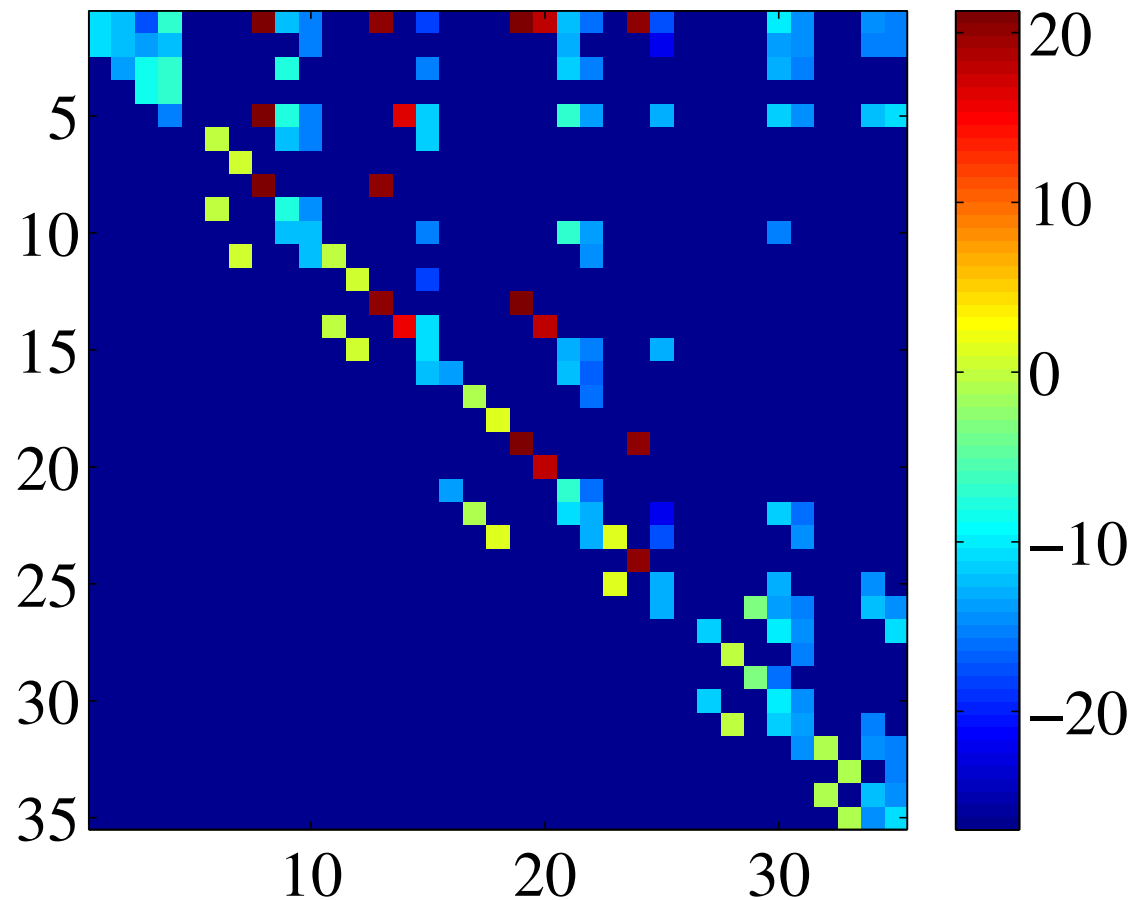
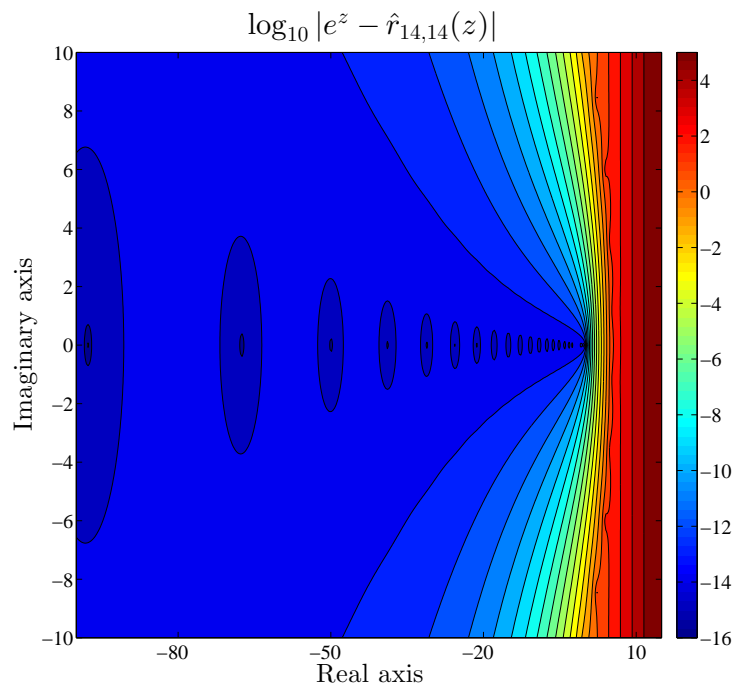


Figure 2: Close-up of the matrix, corresponding to the 36 lightest nuclides ranging from  $^1\text{H}$  to  $^{18}\text{O}$ .

## Eigenvalues near the negative real axis



- It was discovered that the eigenvalues of burnup matrices are bounded near the negative real axis [2]
  - Chebyshev Rational Approximation Method (CRAM) can be characterized as the best rational approximation on the negative real axis
- ⇒ CRAM can provide accurate solution to burnup equations without excluding any nuclides

[2] M. PUSA and J. LEPPÄNEN, *Computing the Matrix Exponential in Burnup Calculations*, *Nucl. Sci. Eng.*, **164**, 2, 140–150 (2010)

## Sparsity Pattern

- Nuclides sorted according to their ZAI index
- Production of by-product nuclides ( $^1\text{H}$ ,  $^2\text{H}$ ,  $^3\text{H}$ ,  $^3\text{He}$ ,  $^4\text{He}$ ) has been taken into account

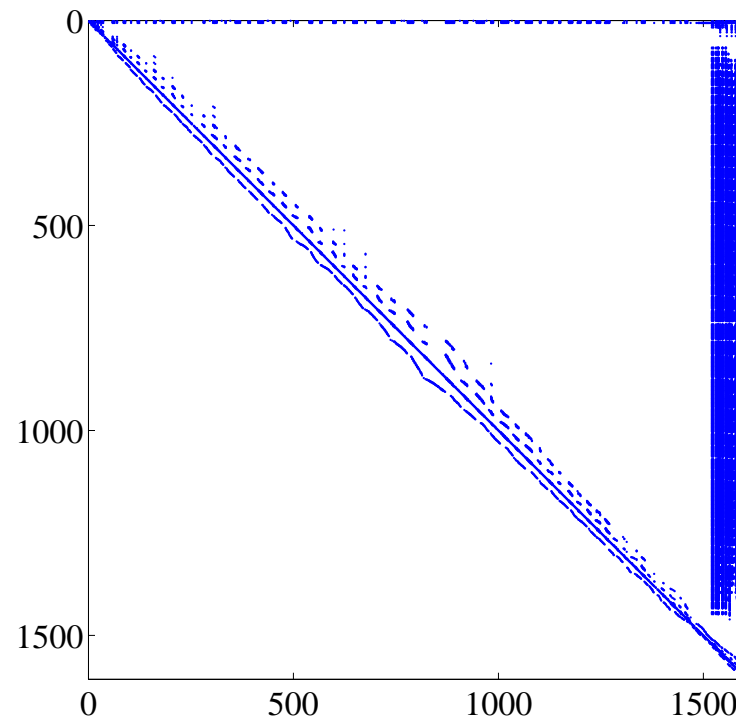


Figure 3: Burnup matrix sparsity pattern

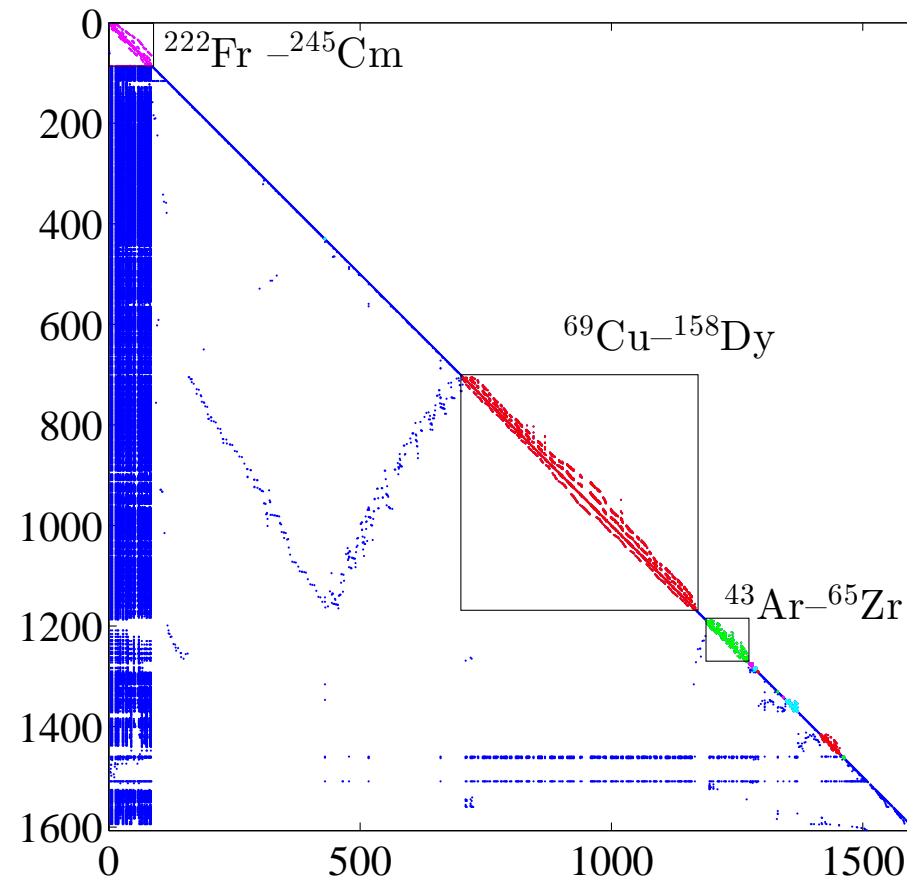
## Sign pattern

- A matrix  $Z \in \mathbb{R}^{n \times n}$  is called a  $Z$ -matrix if its off-diagonal elements are non-positive, i.e.  $z_{ij} \leq 0$  for  $i \neq j$ .  
 $\Rightarrow$  Negatives of burnup matrices are  $Z$ -matrices
- **Theorem:**  
 $e^{At} \geq 0$  for all  $t \geq 0$  if and only if  $-A$  is a  $Z$ -matrix.
- $Z$ -matrices connect burnup matrices with the theory of non-negative matrices.

## Graph-theoretical Approach

- Consider the graph related to burnup matrices
- Set of *strongly connected components* (SCCs) corresponds to a set of nuclides such that there is a transmutation path from every nuclide to every other nuclide in the set.
- Example: successive  $(n,\gamma)$  and  $(n,2n)$  reactions  $\Rightarrow$  closed cycle of size two
- After computing the SCCs and sorting them, the corresponding linear systems can be solved independently in this order
- Matrix eigenvalues are union of the eigenvalues corresponding the SCCs

## Strongly Connected Components



## Conclusions about sccs of Burnup Matrices

- Fissile nuclides and fission product nuclide belong to different SCCs
- By-product nuclides  $^1\text{H}$ ,  $^2\text{H}$ ,  $^3\text{H}$ ,  $^3\text{He}$  and  $^4\text{He}$  form a *sink* meaning no nuclides outside this group are produced from these nuclides
- Eigenvalues of a burnup matrix = eigenvalues related to its SCCs.
  - ⇒ Most of burnup matrix eigenvalues coincide with its diagonal elements (non-positive)
  - ⇒ Eigenvalues related to each closed-cycle-system can be considered separately
  - ⇒ Especially the eigenvalues related to by-product nuclides can be considered separately

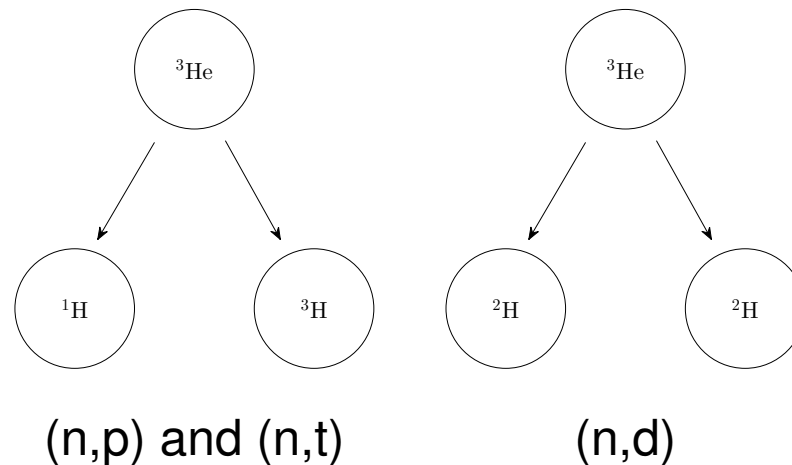


## Real Parts of Eigenvalues (1)

- Production of by-product nuclides **not taken** into account  $\Rightarrow$  number of nuclides increases in fission.
- Production of by-product nuclides **taken** into account  $\Rightarrow$  number of nuclides increases in fission and all reactions producing a by-product nuclide in addition to daughter nuclide.
- **Theorem:** Every solution  $n$  of system  $n' = An$  remains bounded as  $t \rightarrow \infty$  if and only if the following holds
  - (i)  $\text{Re}(\lambda) \leq 0 \quad \forall \lambda \in \Lambda(A)$
  - (ii) Every  $\lambda \in \Lambda(A)$  with  $\text{Re}(\lambda) = 0$  is a semisimple eigenvalue, i.e. the geometric and algebraic multiplicities agree.

## Real Parts of Eigenvalues (2)

- Concentrations of by-product nuclides grow unboundedly as  $t \rightarrow \infty$ .
  - Neutrons are not part of the system but are assumed to be added to it constantly
  - In  $\beta^-$  decay of  $^3\text{H}$  to  $^3\text{He}$ , neutrons are converted to protons
  - In (n,p), (n,d) and (n,t) reactions of  $^3\text{He}$ , the number of nuclides increases



- SCC corresponding to by-product nuclides can have positive eigenvalues!
- All other burnup matrix eigenvalues have non-positive real parts.

## Imaginary Parts of Eigenvalues (1)

- Let  $-A \in Z_n$ , then:  
–  $A$  is a  $M$ -matrix **if and only if** every eigenvalue of  $A$  has a non-positive real part
  - $A \in L_0^k$  **if and only if**  $A$  is a  $Z$ -matrix and each  $k \times k$  principal sub-matrix of  $A$  is an  $M$ -matrix, but there is at least one  $(k + 1) \times (k + 1)$  principal sub-matrix that is not an  $M$ -matrix.
- $\Rightarrow$  SCC corresponding to by-product nuclides belongs to  $L_0^2$ .

## Recap

- Graph of burnup matrix  $\Rightarrow$  Nuclides can be divided into SCCs
- Burnup matrix eigenvalues = Eigenvalues related to the SCCs
- By-product nuclides form one SCC and the corresponding matrix belongs to  $L_0^2$ .
- All other SCCs correspond to matrices that are  $M$ -matrices

## Eigenvalues of burnup matrices

- **Theorem:** Let  $A \in \mathbb{R}^{n \times n}$  be an augmented burnup matrix. If  $n = 2$ ,

$$\Lambda(A) \subset (-\infty, 0] .$$

Otherwise, if the nuclides  $^1\text{H}$ ,  $^2\text{H}$ ,  $^3\text{H}$ ,  $^3\text{He}$  and  $^4\text{He}$  are included to the burnup system,  $A$  has four eigenvalues corresponding to them.

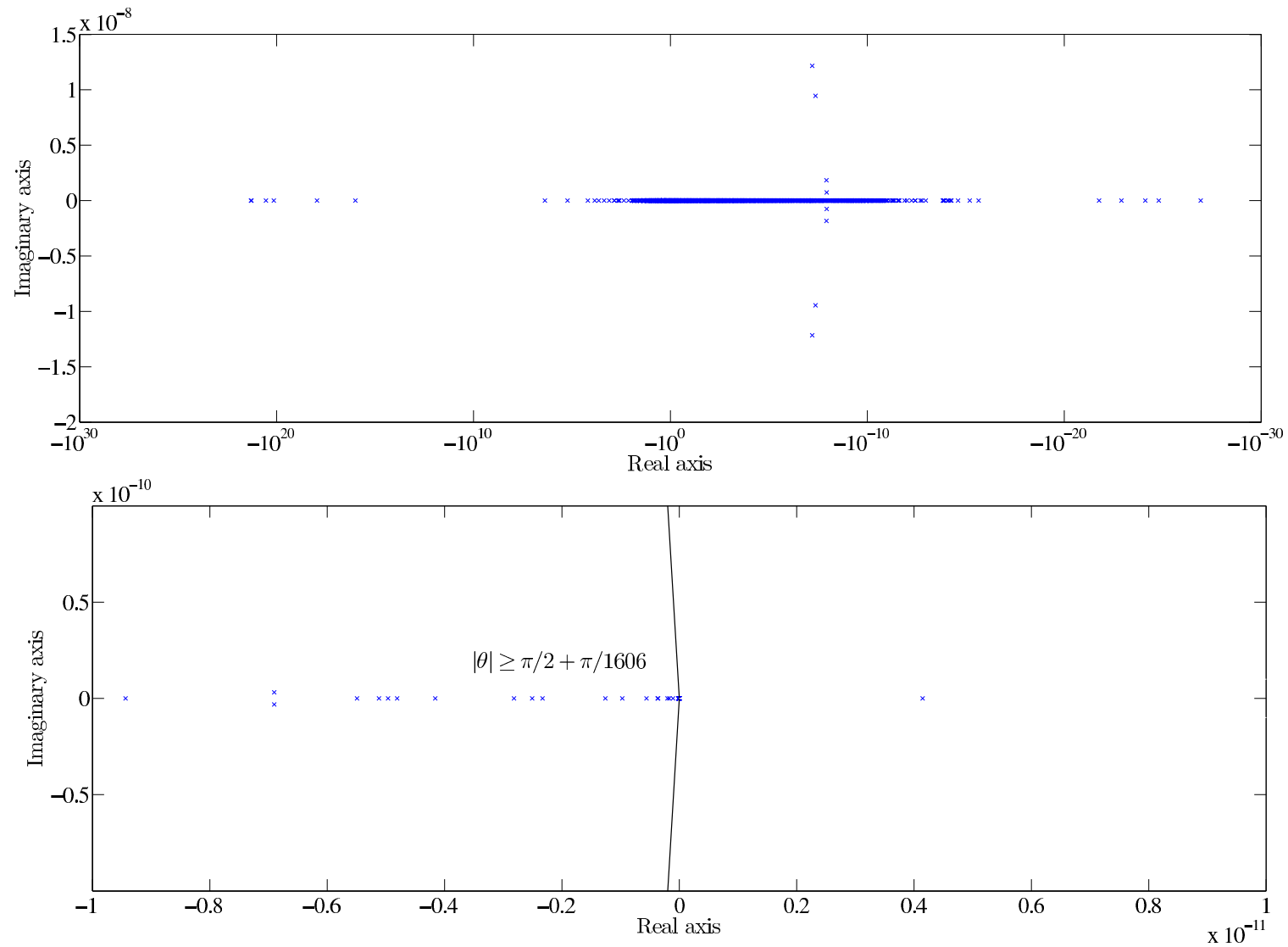
Exactly one of these eigenvalues is real-valued and positive, while the other three eigenvalues have non-positive real parts.

The remaining eigenvalues of  $A$  are confined to the wedge

$$W_n = \left\{ z = r e^{i\theta} \mid r > 0, |\theta| \geq \frac{\pi}{2} + \frac{\pi}{n} \right\} \quad (3)$$

around the negative real axis.

# Eigenvalues of burnup matrices



## Chebyshev Rational Approximation Method (CRAM)

- Burnup matrix eigenvalues are bounded near the negative real axis
- CRAM approximation of order  $k$  is defined as the unique rational function  $\hat{r}_{k,k}$  such that

$$\sup_{-\infty < x \leq 0} |\hat{r}_{k,k}(x) - e^x| = \inf_{r_{k,k} \in \pi_{k,k}} \left\{ \sup_{-\infty < x \leq 0} |r_{k,k}(x) - e^x| \right\} .$$

- It can be characterized as the best rational approximation on the negative real axis

## Burnup Matrix Exponential

- Fresh PWR pin-cell, 1606 nuclides,  $t \approx 8.64 \times 10^5$  s,  $\|A\| \sim 10^{21}$

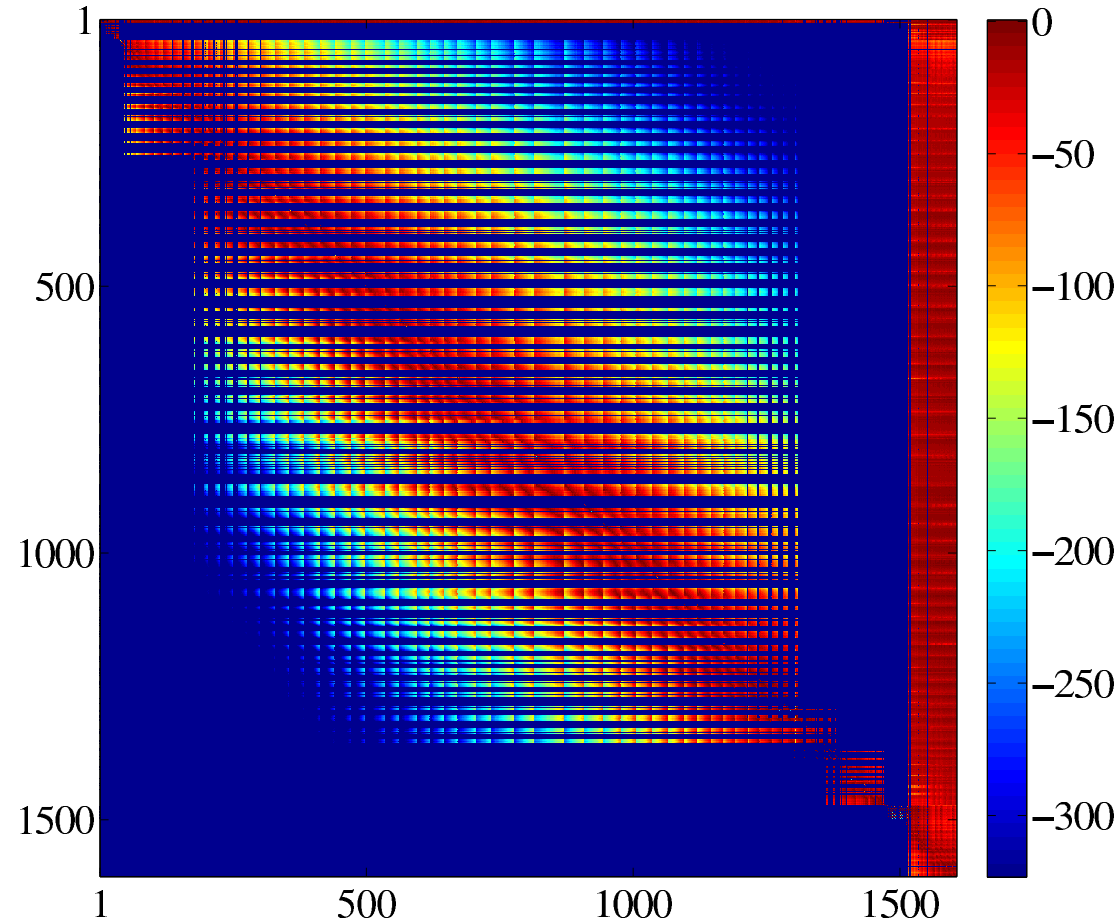
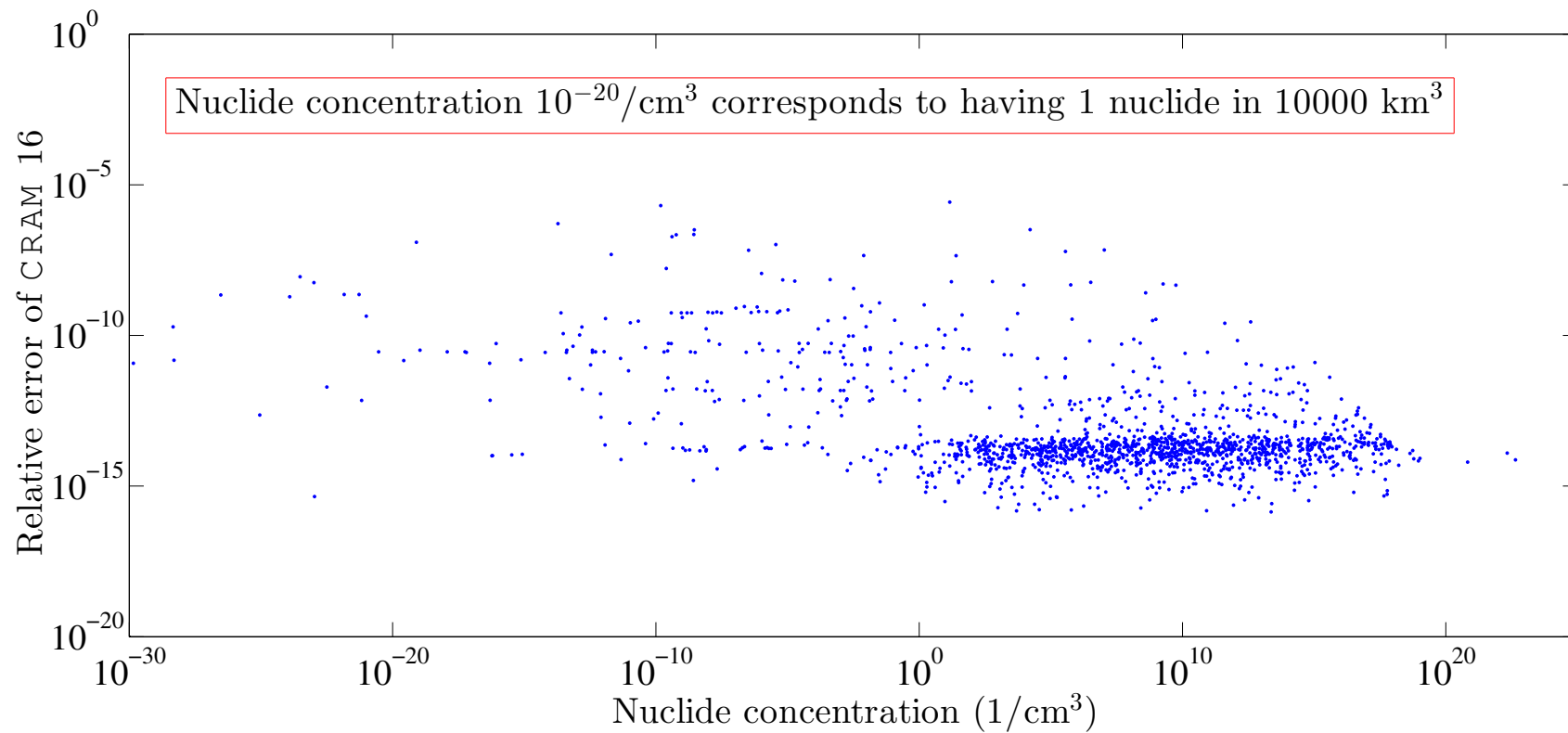


Figure 4: Logarithmic variations in the elements of the matrix exponential

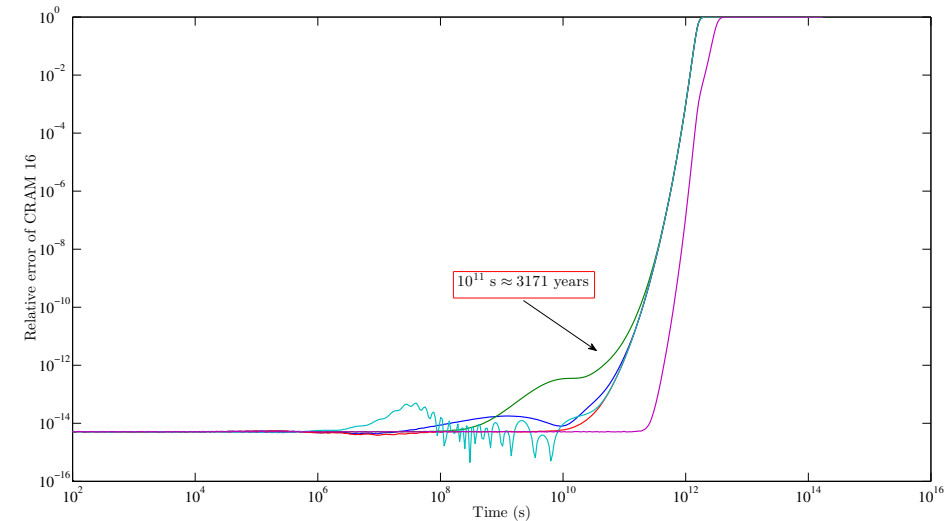
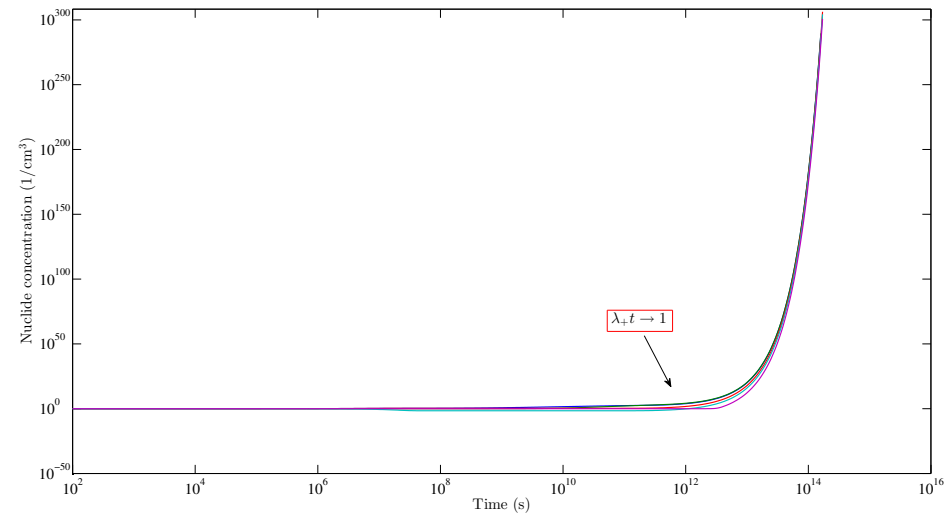


## Accuracy of CRAM of order 16



# What about the positive eigenvalue?

- $\lambda_+ \sim 10^{-12}$
- When  $\lambda_+ t \rightarrow 1$ , nuclide concentrations begin to increase unrealistically



## Summary

- The computation of matrix exponential has been considered challenging in the context of burnup equations
- Burnup matrix eigenvalues were discovered to lie around the negative real axis
- CRAM can be characterized as the best rational approximation on the negative real axis and it can provide very accurate solution to burnup equations without excluding any nuclides
- New knowledge on burnup matrices and their mathematical properties was discovered during this work

## Further reading

- M. PUSA and J. LEPPÄNEN, *Computing the Matrix Exponential in Burnup Calculations*, *Nucl. Sci. Eng.*, **164**, 2, 140–150 (2010)
- M. PUSA, *Rational approximations to the matrix exponential in burnup calculations*, *Nucl. Sci. Eng.*, **169**, 2, 155–167 (2011)
- M. PUSA, *Correction to partial fraction decomposition coefficients for Chebyshev rational approximation on the negative real axis*, arXiv:1206.2880v1 [math.NA] (2012).
- M. PUSA and J. LEPPÄNEN, *Solving linear systems with sparse Gaussian elimination in the Chebyshev rational approximation method (CRAM)*, accepted for publication in *Nucl. Sci. Eng.* (Nov 2013).
- M. PUSA, *Numerical methods for nuclear fuel burnup calculations*, D.Sc. Thesis, VTT Science, **32**,  
<http://montecarlo.vtt.fi/download/S32.pdf>