Isotopic Inventory Calculations in the 21st Century:

ALARA and Beyond

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Overview

- Background on Isotopic Inventory
- Fusion Activation: ALARA
- Inventory Analysis of Future Systems
- Summary

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What is Isotopic Inventory Analysis?

A detailed accounting of the isotopic composition of materials used in nuclear systems and fuel cycles during and after their lifetime in the system.

Transmutation products | **Fission products** | **Actinides**

Applications of Isotopic Inventory Analysis

- Facility Analysis
  - Fusion reactors
  - Accelerator-driven neutron sources
  - Fission reactors (power, research, medical)
- Process Simulation
  - Neutron activation analysis
  - Isotope production
  - Nuclear fuel cycles

Two Worlds of Inventory Analysis

- Burnup/Depletion Simulations
  - Traditional fission systems
  - *Time scales:* Slowly varying
  - *Focus:* actinides
  - *Energy range:* < few MeV
  - Significant coupling between inventory & neutron flux

- Activation Calculations
  - Fusion and accelerator systems
  - *Time scales:* Slowly varying or repeating
  - *Focus:* transmutation products of low- & mid-Z elements
  - *Energy range:* <20MeV (fusion) or few GeV (ADS)
  - Inventory changes have little effect on neutron flux

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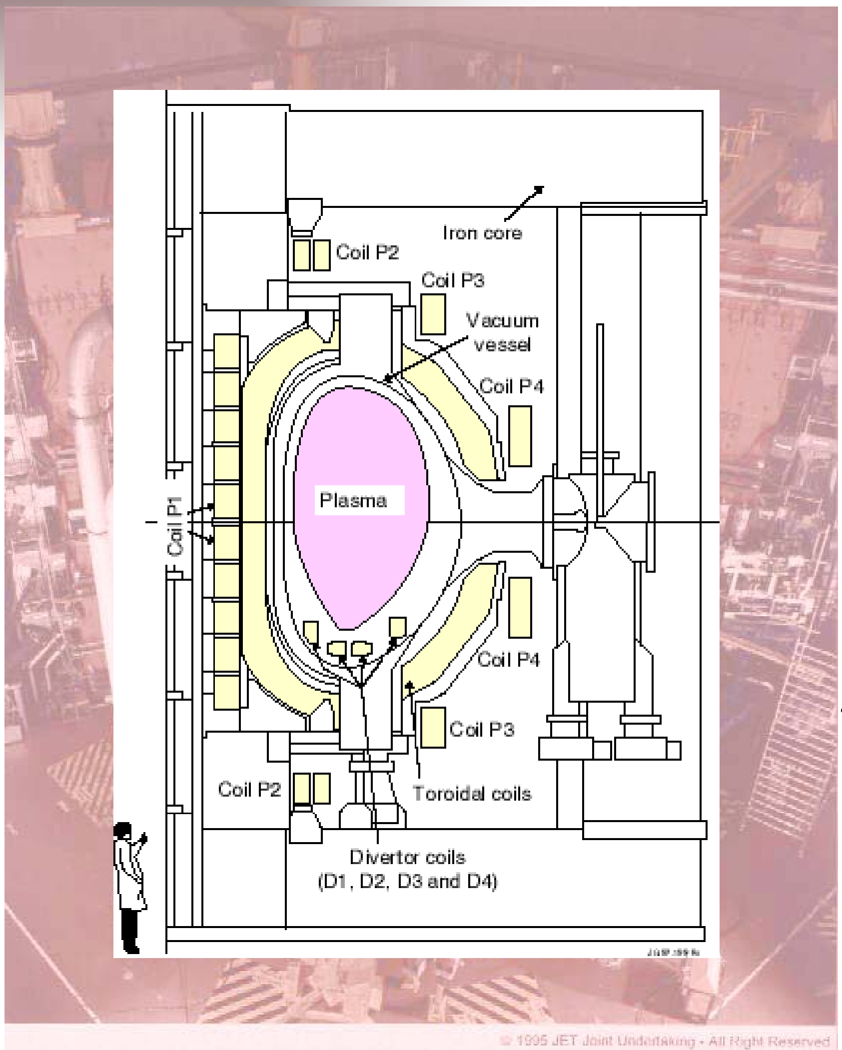
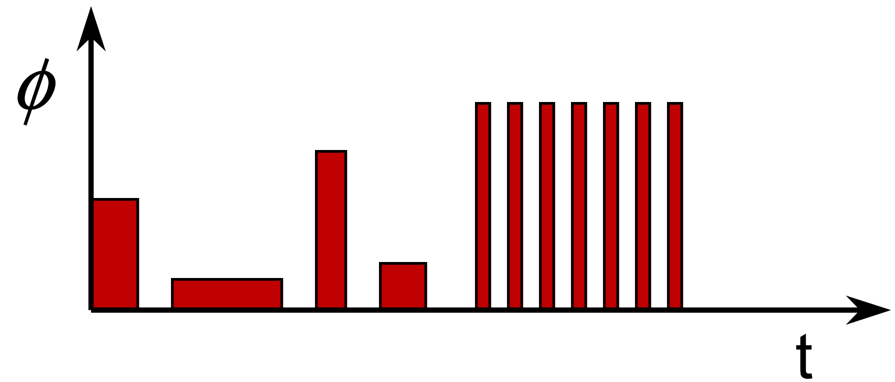
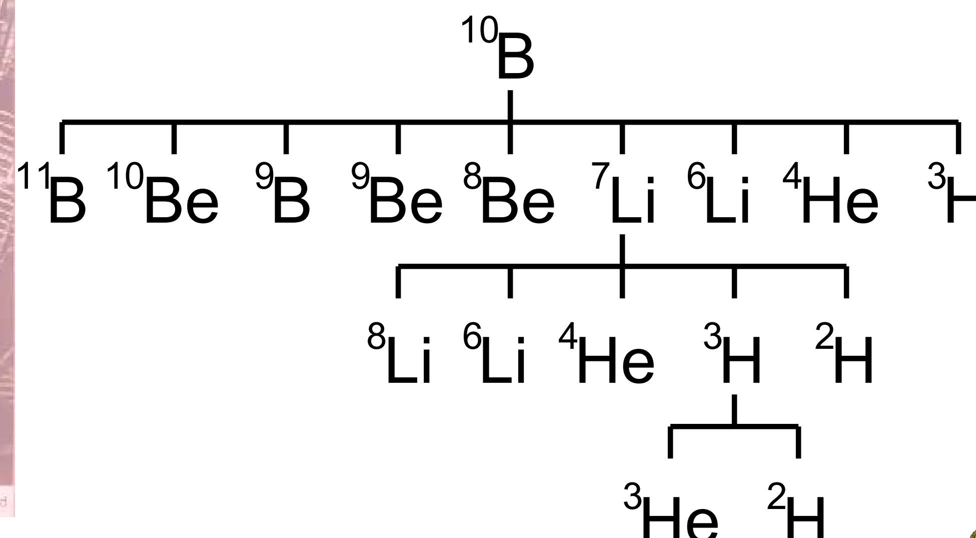
What Is Activation?







Mathematical Representation

$$\dot{N}_i(t) = \sum_{j \neq i} P_{ij} N_j(t) - d_i N_i(t)$$

$$\dot{\vec{N}}(t) = \begin{bmatrix} -d_1 & P_{12} & P_{13} & \cdots & P_{1N} \\ P_{21} & -d_2 & P_{23} & \cdots & P_{2N} \\ P_{31} & P_{32} & -d_3 & \cdots & P_{3N} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ P_{N1} & P_{N2} & P_{N3} & \cdots & -d_N \end{bmatrix} \bullet \vec{N}(t)$$

$$= \mathbf{A} \bullet \vec{N}(t)$$

$$\vec{N}(t) = e^{\mathbf{A}t} \vec{N}(0)$$

History of Fusion Activation Codes

- DKR Family
  - ✔ mathematically exact
  - ✔ multi-dimensional
  - ✘ inefficient data handling
  - ✔ exact pulsing
  - ✘ unable to handle loops
- FISPACT
  - ✘ numerical solution
  - ✘ 0-D
  - ✘ inefficient data handling
  - ✘ steady-state approximation
  - ✔ exact loop solution
- RACC
  - ✘ numerical solution
  - ✔ multi-dimensional
  - ✘ inefficient data handling
  - ✔ recent pulsing modifications
  - ✔ exact loop solution

Desired Activation Code Features

Basic Features

- 3-D (multi-point)
- User-defined precision
- Exact multi-level pulsing
- Accurate loop handling
- Light ion accumulation
- User-friendly input

Advanced Features

- Exact modeling of arbitrary operation schedules
- Adaptive selection of mathematical method to optimize solution
- Reverse solution for detailed studies

Software Design Philosophy

- Accuracy
  - Minimize physical approximations
  - Optimally accurate mathematical method
- Speed
  - Matrix methods for efficient solution of pulsed schedules
  - Efficient data handling and algorithms
- Simplicity/Versatility
  - Modular code with modern practices
  - Ability to solve variety of problems with simple and versatile input file

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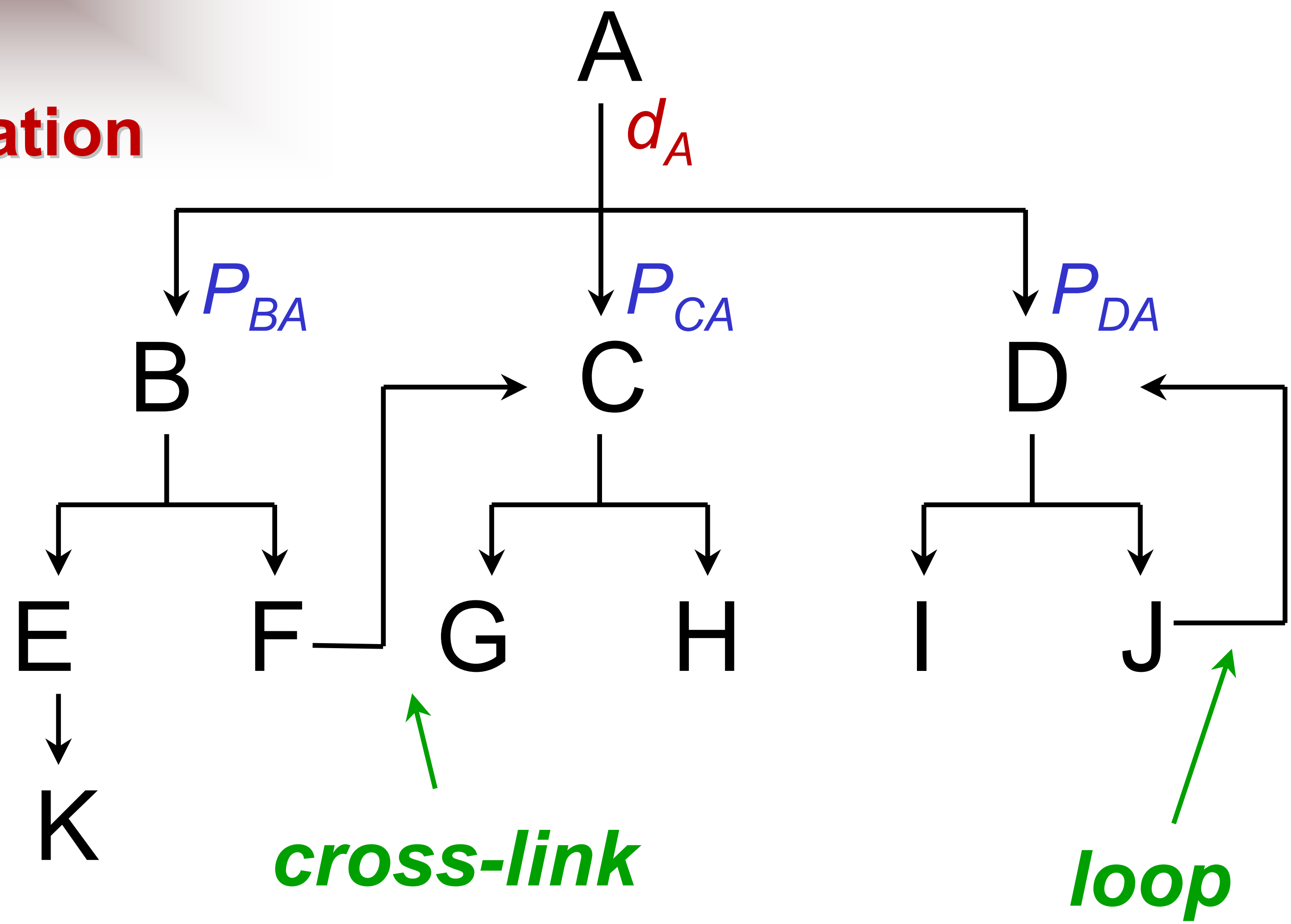
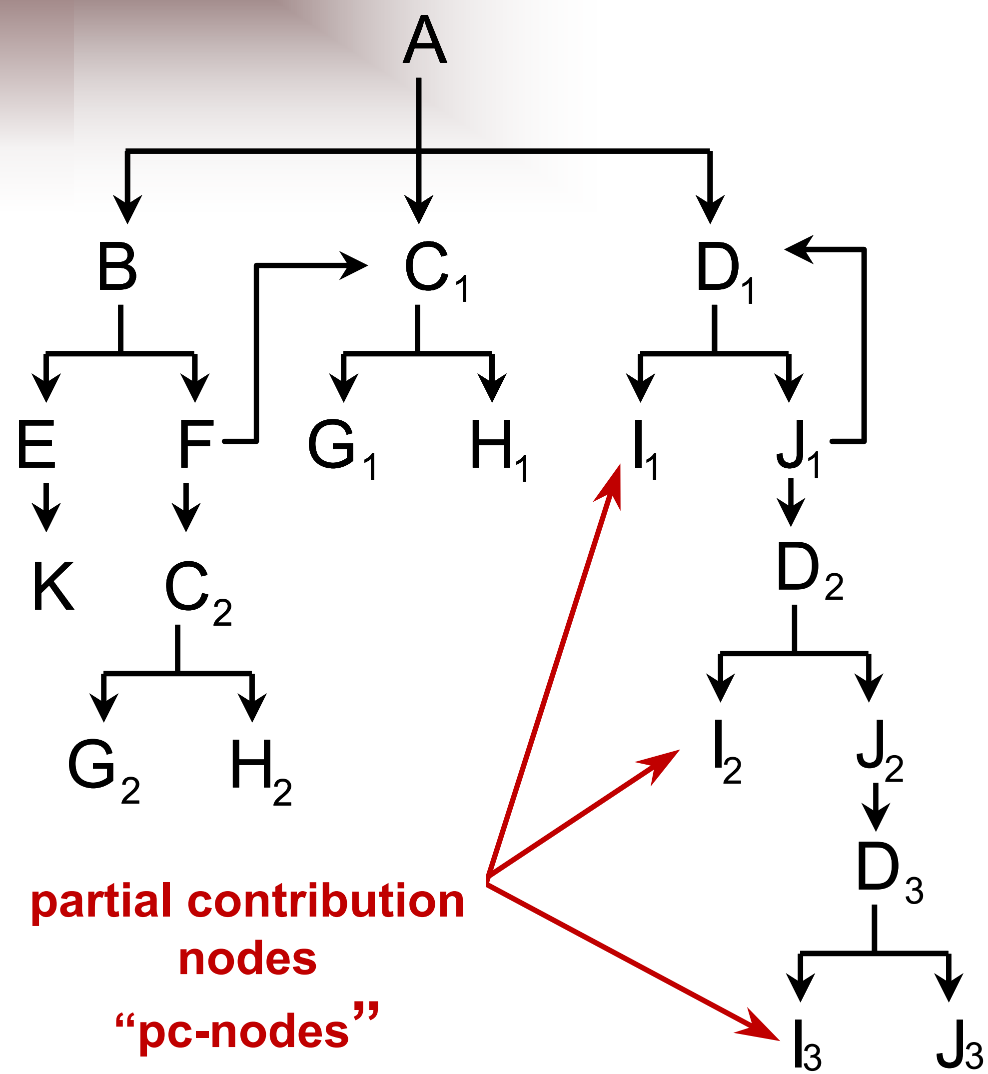
Physical Modeling: Introduction

Activation Trees

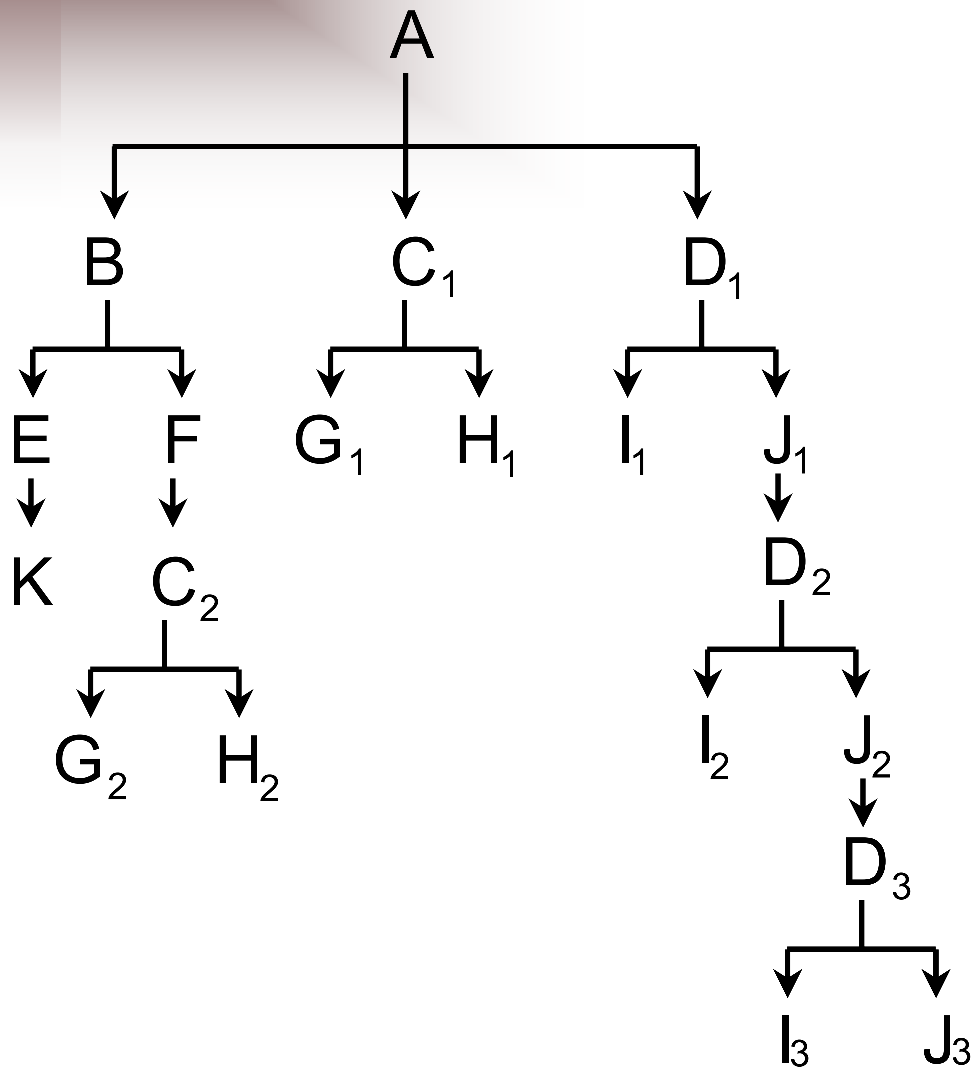


Physical Modeling: Tree Straightening



- Each isotope has only one parent
- Permits simplified mathematical methods
- Permits accurate modeling of loops

Physical Modeling: Linear Chains



- depth-first search

$A \to B \to E \to K$

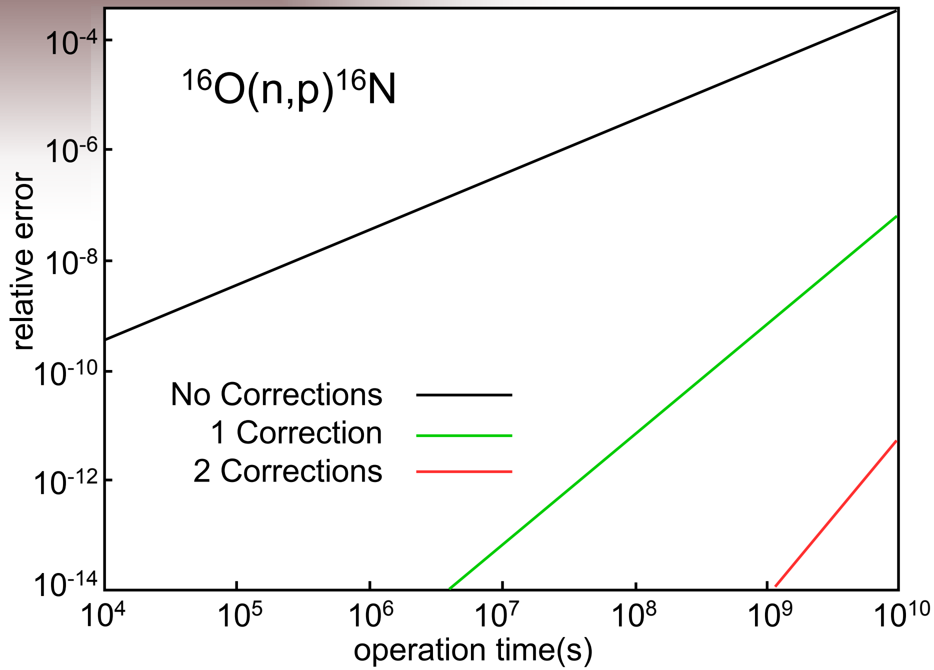
$A \to B \to F \to C_2 \to G_2$

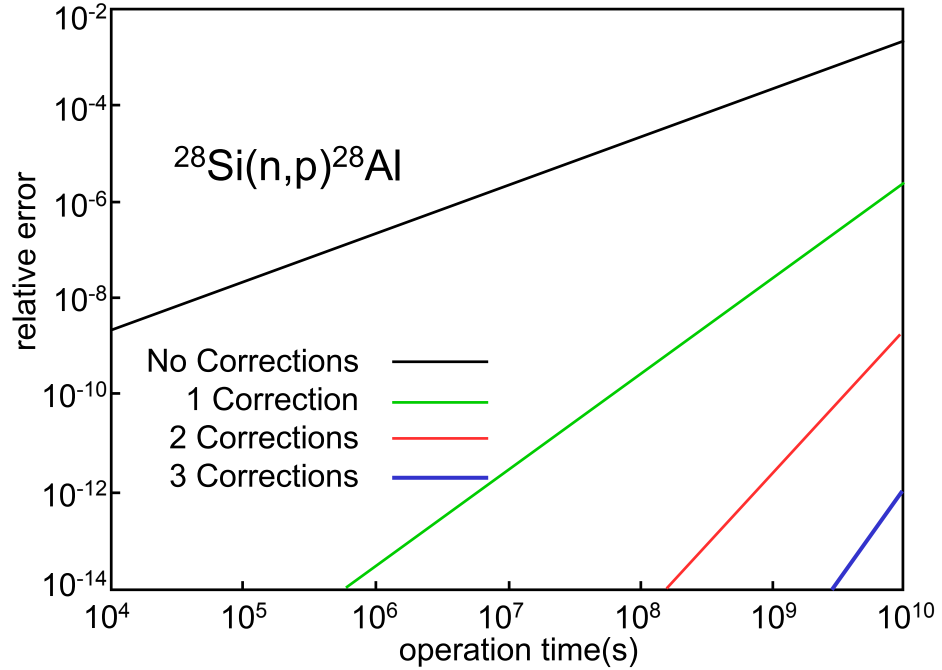
$A \to B \to F \to C_2 \to H_2$

$A \to C_1 \to G_1$

$A \to C_1 \to H_1$

Validity of Loop Straightening





(n,p) Reaction	# of corrections			
	0	1	2	3
^{13}C*(n,p)* ^{13}B	4.06E-05	8.23E-10	1.10E-14	1.12E-16
^{16}O*(n,p)* ^{16}N	0.000351	6.15E-08	7.20E-12	5.62E-16
^{28}Si*(n,p)* ^{28}Al	0.00243	2.95E-06	2.38E-09	1.45E-12
^{49}Ti*(n,p)* ^{49}V	0.000247	3.05E-08	2.51E-12	0
^{56}Fe*(n,p)* ^{56}Mn	0.00103	5.35E-07	1.85E-10	4.77E-14

Physical Modeling: Tree Truncation

- **Isotopes have a finite probability of transmutation**
  - **"infinite" activation trees**
    - **need for truncation of trees**
      - **atoms are lost from model**

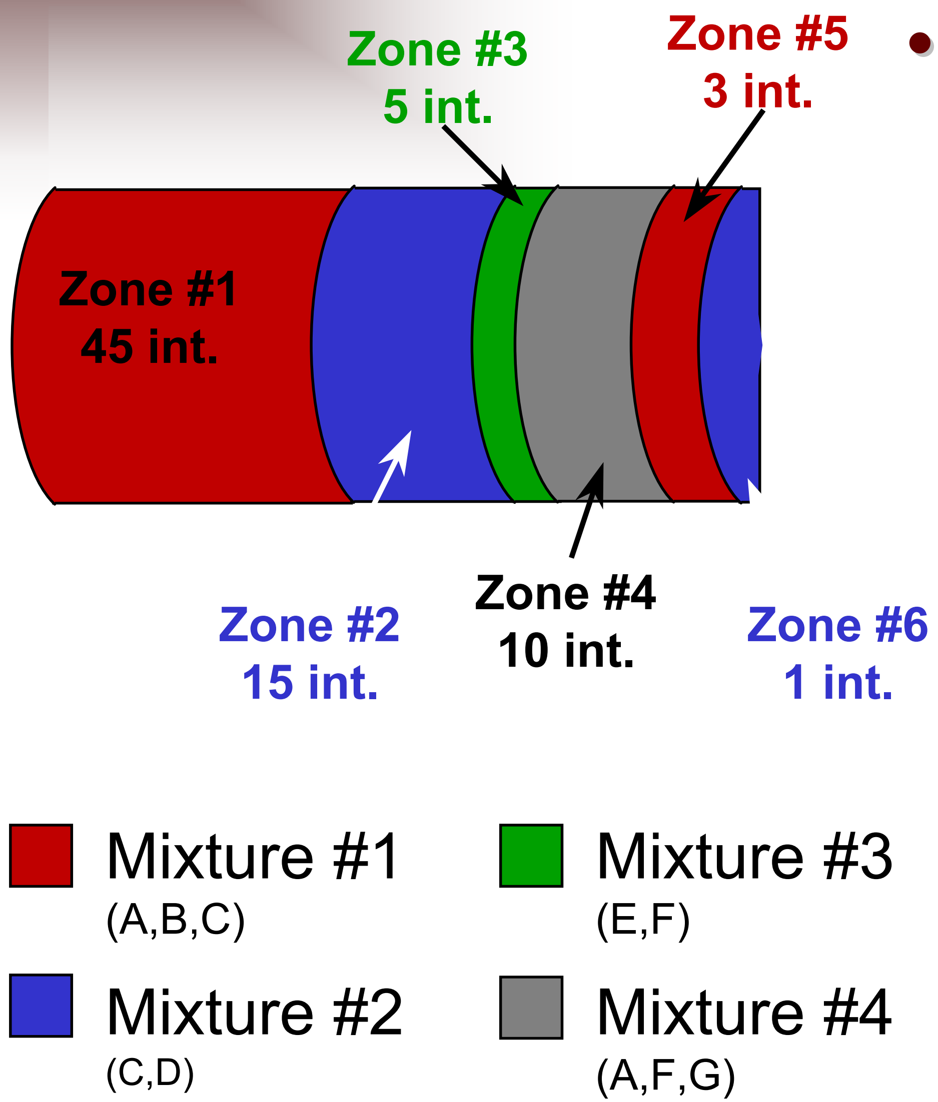
Goal of truncation algorithm

reduce atom loss below user-defined threshold

Truncation Issues

- Atom "pipelines"
  - large decay rates $\Rightarrow$ conduits out of system
  - low inventory $\neq$ low atom loss
  - ⇨ truncation calculation: $d_N = 0$
- After-shutdown build-up
  - low inventory @ shutdown $\neq$ low inventory after shutdown
  - ⇨ test inventory @ all cooling times of interest
- Insignificant solutions
  - nodes with negligible results
  - ⇨ second tolerance to ignore these nodes

Truncation in Multi-Point Problems

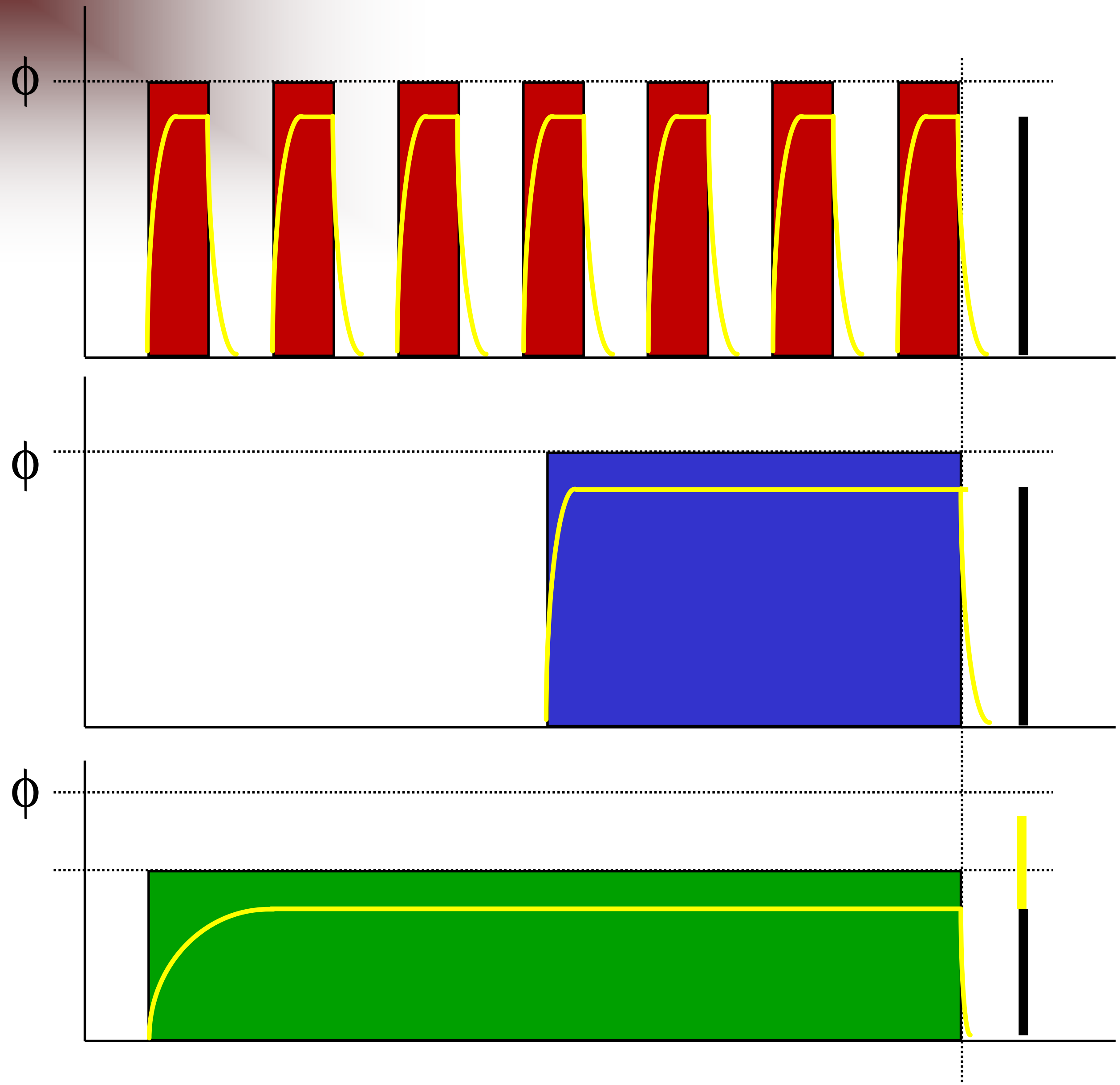


- Varying fluxes result in different trees
  - Computationally expensive

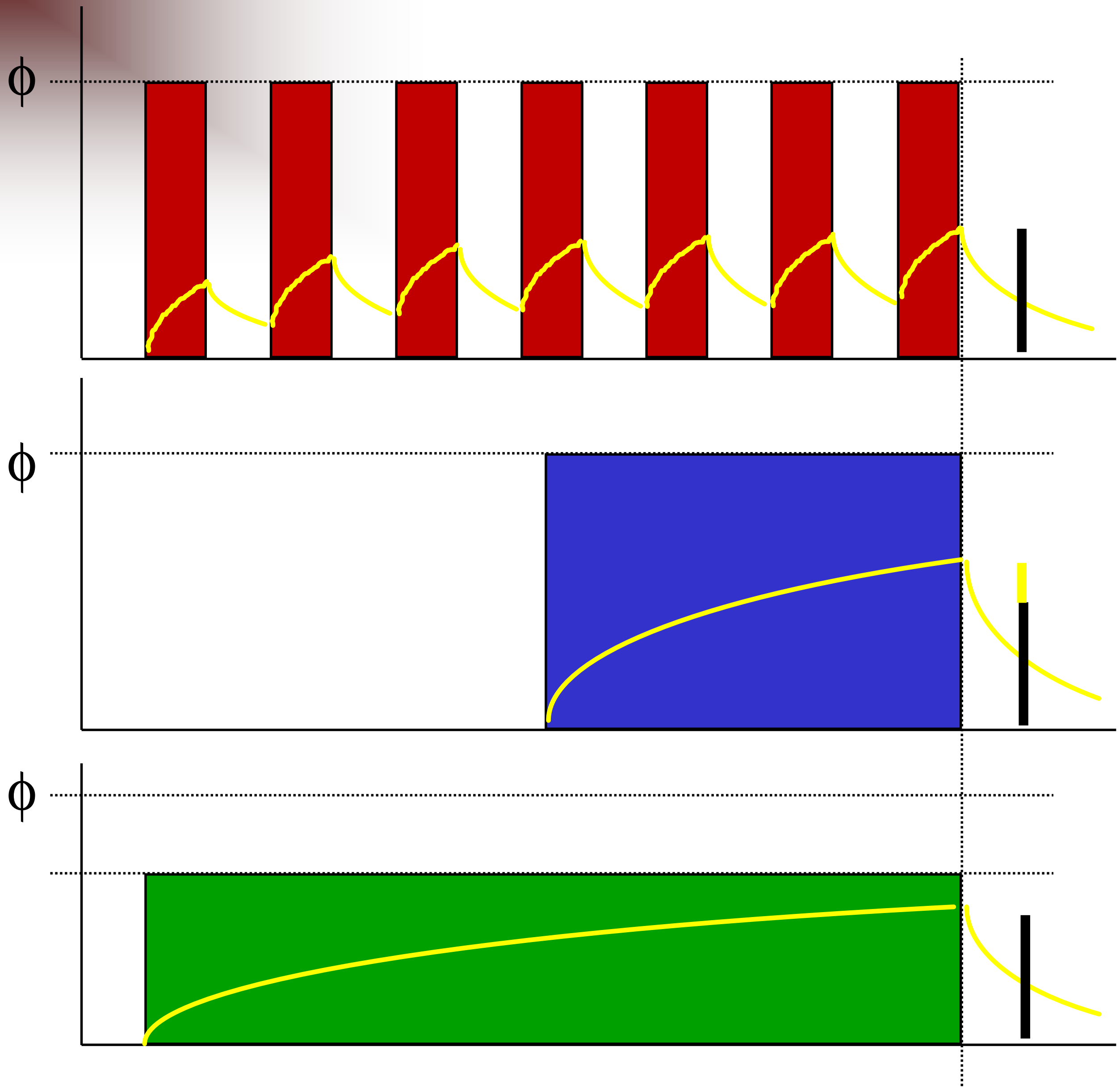
⇨ group-wise maximum reference flux

A	B	C	D	E	F	G
#1	#1	#1				
		#2	#2			
				#3	#3	
#4					#4	#4
#5	#5	#5				
		#6	#6			

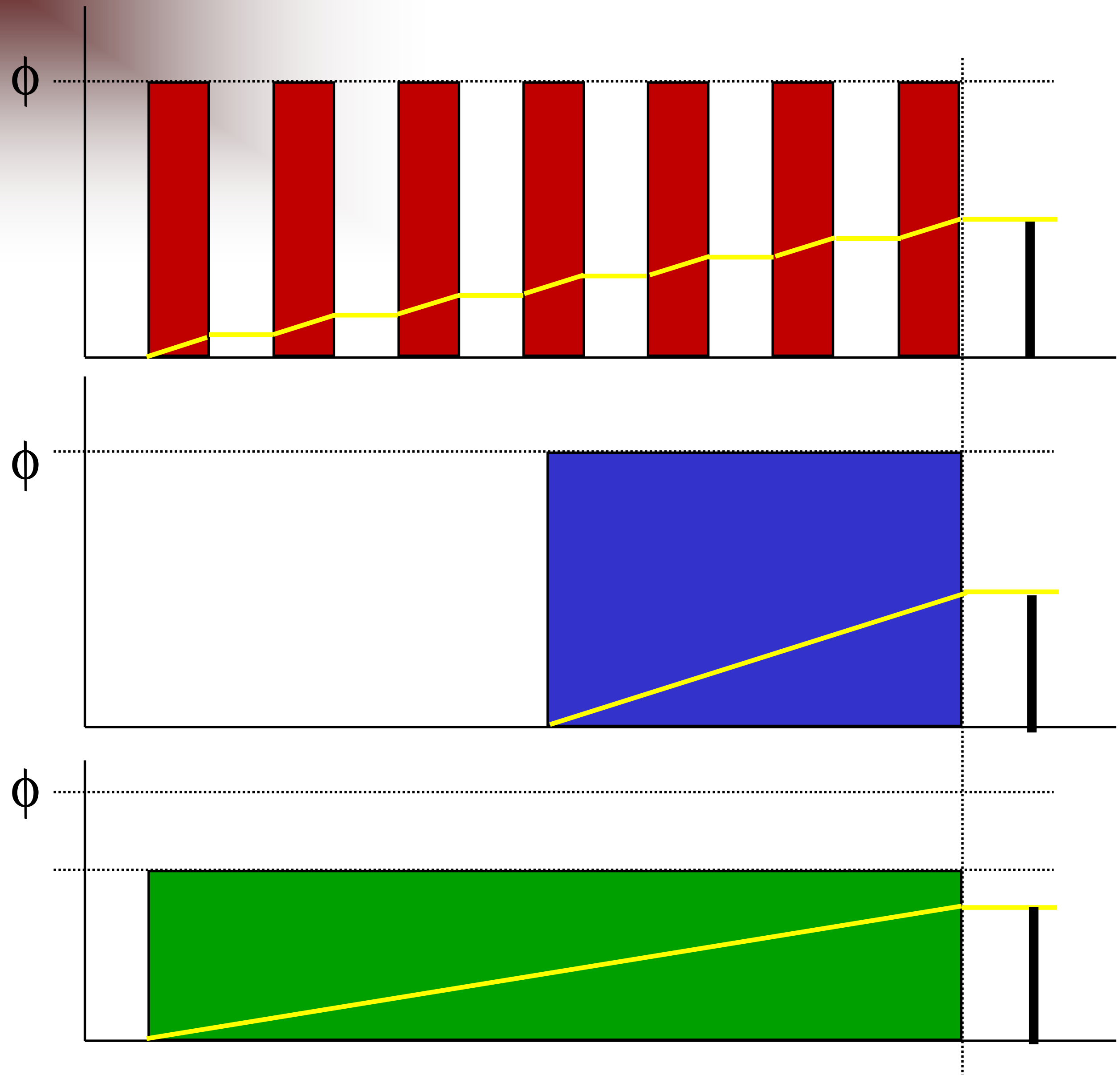
Physical Modeling: Pulsing



Physical Modeling: Pulsing

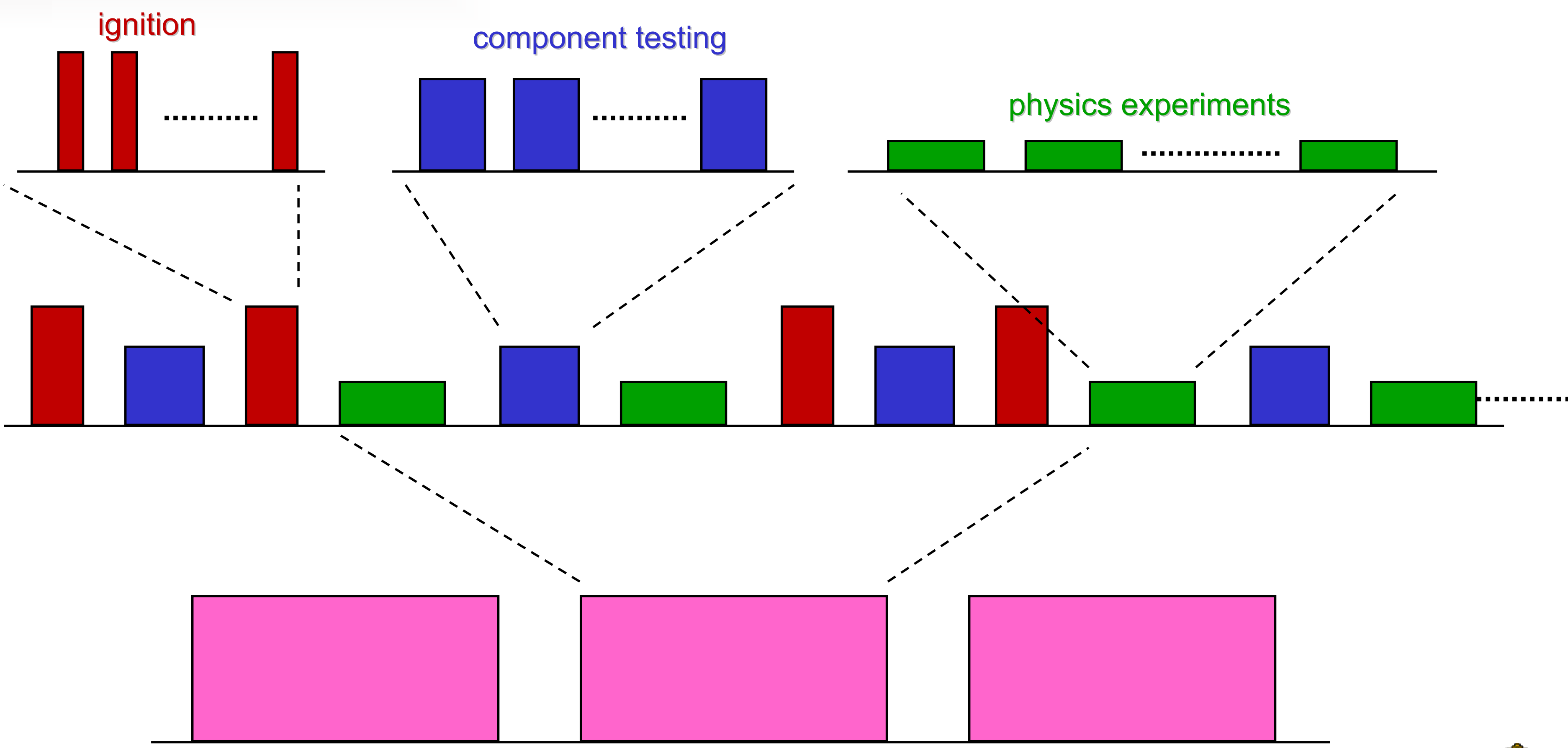


Physical Modeling: Pulsing

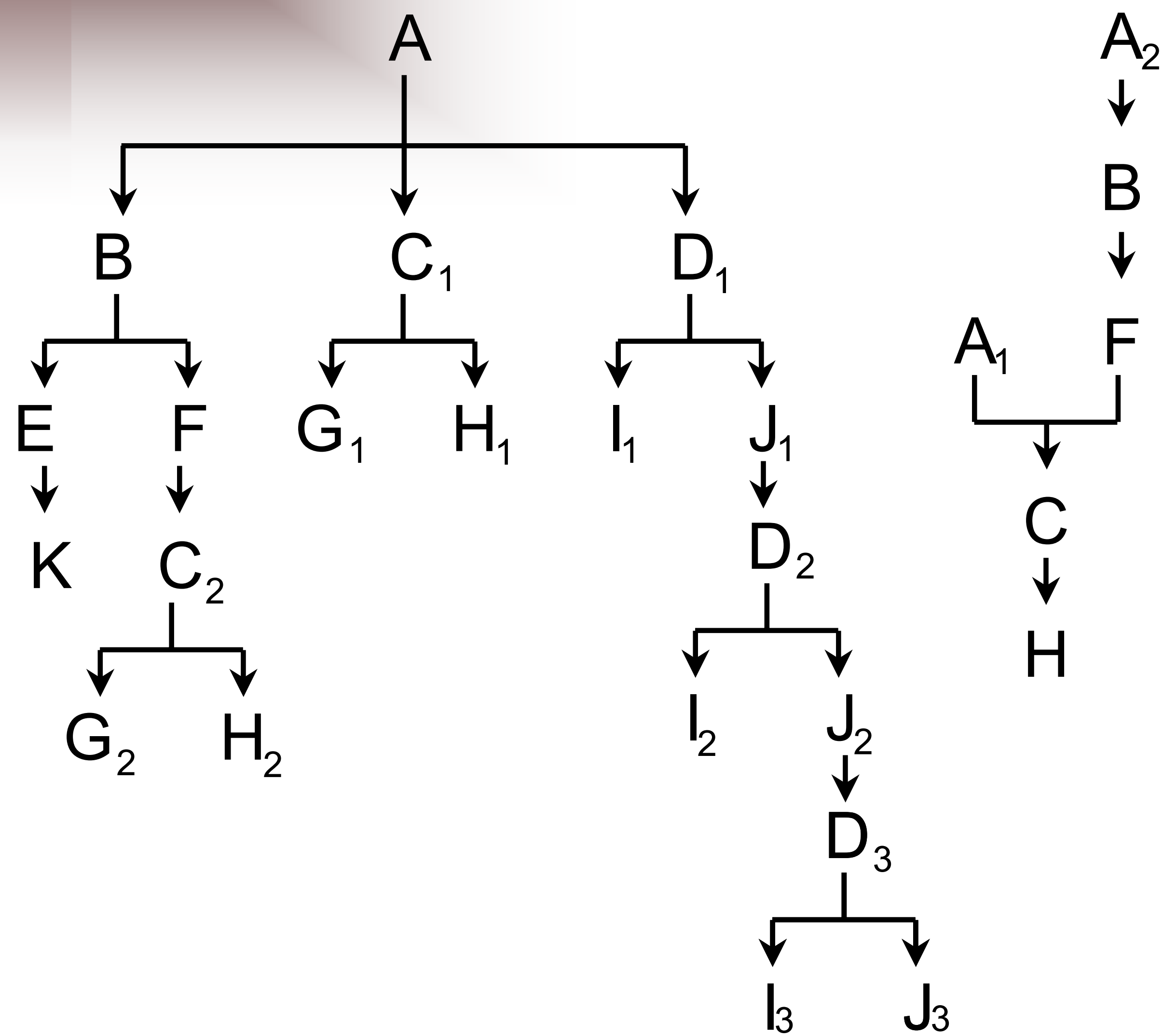


Physical Modeling

Arbitrary Irradiation Schedules



Physical Modeling: Reverse Problem



- Fewer, shorter chains
- Lower truncation tolerances
- More precise solutions

Physical Modeling: Summary

- Sufficiently accurate loop handling
  – Uniform accuracy across problem
- Accurate and precise solutions
  – Both determined by user-defined truncation tolerance
- Exact modeling of arbitrary schedules
  – Based on matrix methods
- Reverse calculation mode allows detailed study of trace products

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Mathematical Methods: Introduction

$$\dot{\vec{N}}(t) = \mathbf{A} \bullet \vec{N}(t) \qquad \vec{N}(t) = e^{\mathbf{A}t}\vec{N}_o(t) = \mathbf{T}\vec{N}_o(t)$$

Explicit Trees

$$\begin{bmatrix} -d_1 & P_{12} & P_{13} & \cdots & P_{1N} \\ P_{21} & -d_2 & P_{23} & \cdots & P_{2N} \\ P_{31} & P_{32} & -d_3 & \cdots & P_{3N} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ P_{N1} & P_{N2} & P_{N3} & \cdots & -d_N \end{bmatrix}$$

Straightened Trees

$$\begin{bmatrix} -d_1 & 0 & 0 & \cdots & 0 \\ P_{21} & -d_2 & 0 & \cdots & 0 \\ P_{31} & P_{32} & -d_3 & \cdots & \vdots \\ \vdots & \vdots & \vdots & \ddots & 0 \\ P_{M1} & P_{M2} & P_{M3} & \cdots & -d_M \end{bmatrix}$$

Single Linear Chains from Straightened Trees

$$\begin{bmatrix} -d_1 & 0 & 0 & \cdots & 0 \\ P_{21} & -d_2 & 0 & \cdots & 0 \\ 0 & P_{32} & -d_3 & \cdots & \vdots \\ \vdots & \vdots & \vdots & \ddots & 0 \\ 0 & 0 & \cdots & P_{L,L-1} & -d_L \end{bmatrix}$$

Laplace Transform

$$\dot{N}_i(t) = P_i N_{i-1}(t) - d_i N_i(t)$$

$$\tilde{N}_i(s) = \frac{P_i}{s+d_i}\tilde{N}_{i-1}(s) + \frac{N_i(0)}{s+d_i}$$

$$\tilde{N}_i(s) = \sum_{j=0}^{i} N_{j_o} \prod_{k=j+1}^{i} P_k \tilde{F}_{ij}(s)$$

$$\tilde{F}_{ij}(s) = \prod_{l=j}^{i} \frac{1}{s+d_l}$$

d_i may be degenerate!!!

Inverse Laplace Transform

$$\tilde{F}_{ij}(s) = \prod_{k'=j}^{i} \frac{1}{s+d_{k'}} = \prod_{k=j}^{i} \frac{1}{\left(s+d_k\right)^{m_k}} = \sum_{k=j}^{i} \sum_{n=1}^{m_k} \frac{R_{kn}}{\left(s+d_k\right)^{n}}$$

$$R_{kn} = \frac{1}{(m_k-n)!} \lim_{s \to d_k} \frac{d^{(m_k-n)}}{ds^{(m_k-n)}} \left[(s+d_k)^{m_k} \tilde{F}_{ij}(s) \right]$$

$$f_{ij}(t) = \sum_{k=j}^{i} e^{-d_k t} \sum_{n=1}^{m} R_{kn} \frac{t^{n-1}}{(n-1)!}$$

For unique poles

$$R_k = \lim_{s \to d_k} (s+d_k) \tilde{F}_{ij}(s) = \prod_{\substack{l=j \\ l \neq k}}^{i-1} \frac{1}{d_k - d_l}$$

$$f_{ij}(t) = \sum_{k=j}^{i} R_k e^{-d_k t}$$

Bateman Solution (Analytical)

Unique poles/eigenvalues = no loops

$$N_i(t) = N_{i_o} e^{-d_i t} + \sum_{j=1}^{i-1} N_{j_o} \prod_{l=j}^{i-1} \frac{P_{l+1}}{d_i - d_l} \sum_{k=j}^{i-1} \left(e^{-d_k t} - e^{-d_i t}\right)$$

$$N_i(t) = N_{i_o} e^{-d_i t} + \sum_{j=1}^{i-1} N_{j_o} \sum_{k=j}^{i-1} \frac{P_{k+1}\left(e^{-d_k t} - e^{-d_i t}\right)}{d_i - d_k} \prod_{\substack{l=j \\ l \neq k}}^{i-1} \frac{P_{l+1}}{d_i - d_l}$$

$$T_{ii}(t) = e^{-d_i t}$$

$$T_{ij}(t) = \sum_{k=j}^{i-1} \frac{P_{k+1}\left(e^{-d_k t} - e^{-d_i t}\right)}{d_i - d_k} \prod_{\substack{l=j \\ l \neq k}}^{i-1} \frac{P_{l+1}}{d_i - d_l}$$

Laplace Inversion (Analytical)

For arbitrary multiplicity, require derivatives of:

$$\tilde{G}_{ij}^{k}(s) = (s + d_k)^m \tilde{F}_{ij}(s)$$

A recursive method for evaluating these derivatives has been developed:

$$\left[\tilde{G}_{ij}^{k}(s)\right]^{(n)} = \sum_{p=1}^{n} (-1)^p \frac{(n-1)!}{(n-p)!} \left[\tilde{G}_{ij}^{k}(s)\right]^{(n-p)} \sum_{\substack{l=j \\ l \neq k}}^{i} (s + d_l)^{-p}$$

which can be proven inductively.

Laplace Expansion (Numerical)

$$\tilde{F}_{ij}(s) = \prod_{l=j}^{i} \frac{1}{s+d_l}$$

$$= \frac{1}{s^{i-j+1}} \prod_{l=j}^{i} \frac{1}{1+\frac{d_l}{s}}$$

$$= \frac{1}{s^{i-j+1}} \prod_{l=j}^{i} \left(1 - \frac{d_l}{s} + \frac{d_l^2}{s^2} - \frac{d_l^3}{s^3} + \ldots\right)$$

$$= \frac{1}{s^{i-j+1}} \left[1 - \frac{\sum_{l=j}^{i} d_l}{s} + \frac{\sum_{l=j}^{i} d_l \sum_{k=l}^{i} d_k}{s^2} - \frac{\sum_{l=j}^{i} d_l \sum_{k=l}^{i} d_k \sum_{m=k}^{i} d_m}{s^3} + \ldots\right]$$

$$f_{ij}(t) = t^N \left[\frac{1}{N!} - \frac{t}{(N+1)!}\sum_{l=j}^{i} d_l + \frac{t^2}{(N+2)!}\sum_{l=j}^{i} d_l \sum_{k=l}^{i} d_k - \frac{t^3}{(N+3)!}\sum_{l=j}^{i} d_l \sum_{k=l}^{i} d_k \sum_{m=k}^{i} d_m + \ldots\right] \qquad (N = i-j)$$

Adaptive Selection of Methods

- Adaptively chosen for each matrix element
  - $\mathbf{T}_{ij}$ represents transfer on sub-chain between isotopes *j* and *i* inclusive
- If NO loop on sub-chain
  - Use Bateman solution
  - Otherwise use Laplace Expansion
- If Laplace Expansion does not converge
  - Use Laplace Inversion

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Summary: Current Features

- ✔Straightforward input file creation
- ✔Multi-point solutions in a variety of geometries
- ✔Accurate loop solutions in the activation trees
- ✔User-defined calculation precision/accuracy

- ✔Exact modeling of arbitrary hierarchical irradiation schedules
- ✔Full easy-to-read activation tree output
- ✔Flexible output options
- ✔Unlimited number of reaction channels
- ✔Reverse calculation mode

ALARA Status

ALARA is a fully developed and validated alternative to other activation codes.

(standard for ARIES, IFMIF)

Development of ALARA is continuing to include more features, increasing its flexibility and versatility.

(v. 2.5.0 Jan 2002)

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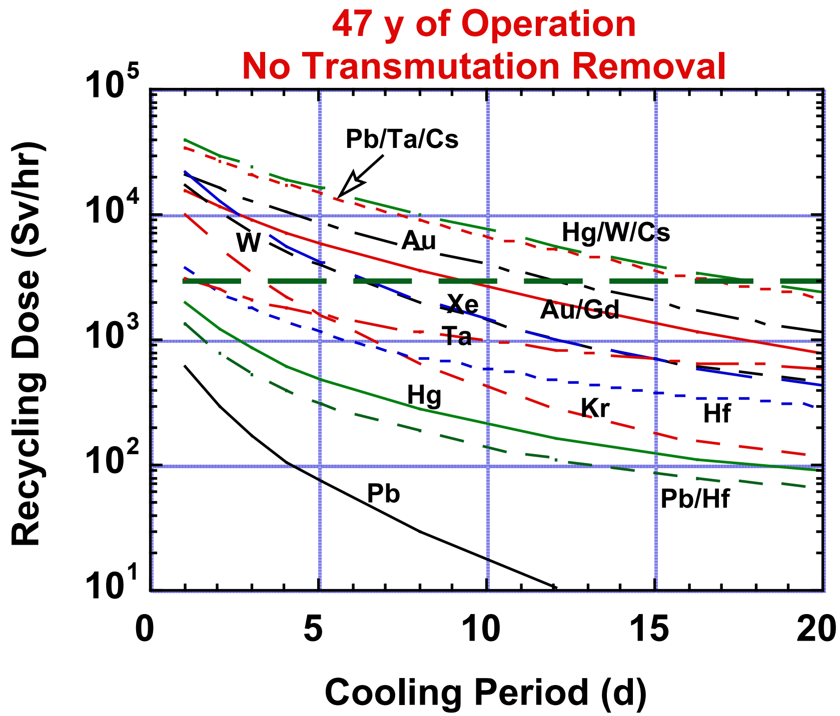
ALARA in ARIES

- User: UW Fusion Technology Institute
- Used exclusively since 1999 (replaced DKR)
- Routine reactor component analysis
- Typical problems perform calculation at over 450 points in ~40 regions filled with ~10 materials (80 isotopes) in <1 hour
- Recently modeling advanced schedules for specialized components

ALARA in ARIES

Recycling of IFE Hohlraum Material*

- Following single pulse, material cools, is recycled and re-fabricated into new capsule
- Recycling equipment has dose limit
- ALARA used to determine minimum cooling time before reprocessing/ refabrication



* L. El-Guebaly, et al, "Feasibility of Recycling Hohlraum Wall Material", ARIES Project Meeting, Madison, WI, April 2002

ALARA in IFMIF

- User: Forschungszentrum Karlsruhe (FZK)
- FZK/Russian collaboration for <150 MeV activation data
  - Large number of activation channels
    - FISPACT (FZK workhorse activation code) has hard-coded reaction table
    - ALARA has library-driven reaction information
  - Newest version IEAF-2001 (NEA Databank) has 679 nuclides
- Various investigations of data importance and data benchmarking

ALARA in IFMIF

Data Benchmarking*

- **Experimental Parameters:**

(U. von Möllendorff, Fus. Eng. & Des. 51(2000)919):

- – Neutron Source: 40 MeV d on thick Li-target (En < 55 MeV)
- – Neutron Flux : 4.3×10^{11} n/cm^2/s
- – Irradiation Time: 2.1 h
- – Sample: Vanadium foil (m%: V-99.87, Al-0.025, O-0.041, Si-0.017, Fe-0.016, N-0.013 …)

* Simakov, et al, "Activation Analyses of Vanadium irradiated by d-Li neutrons using IEAF-2001 cross sections", Workshop on Activation Data – EAF 2003, Prague, 24-26 June 2002

ALARA in IFMIF

Data Benchmarking*

Nuclide ($T_{1/2}$)	C/E	Dominant Pathways (Threshold)
^{47}Ca (4.5 d)	0.81 ± 0.22	^{51}V(n,pα) ≈ 100% (E_{thr} = 11.7 MeV)
^{46}Sc (84 d)	0.77 ± 0.04	^{51}V(n,2nα) = 97.8% (E_{thr} = 21.3 MeV) ^{50}V(n,nα) = 2.2% (E_{thr} = 10.1 MeV)
^{47}Sc (3.4 d)	0.17 ± 0.01	^{51}V(n,nα) ≈ 99.6% (E_{thr} = 10.5 MeV) ^{50}V(n,nα) ≈ 0.4% (E_{thr} = 10.1 MeV)
^{48}Sc (44 d)	1.07 ± 0.06	^{51}V(n,α) ≈ 99.7% (E_{thr} = 2.1 MeV) ^{50}V(n,^{3}He) ≈ 0.3% (E_{thr} = 11.8 MeV)
^{48}V (16 d)	1.07 ± 0.24	^{51}V(n,4n) ≈ 93.3% (E_{thr} = 32.6 MeV) ^{50}V(n,3n) ≈ 6.7% (E_{thr} = 21.3 MeV)
^{51}Cr (28 d)	$(0.41 \pm 0.04)10^{-3}$	^{54}Fe(n,α) ≈ 68.4% (E_{thr} = 0.0 MeV) ^{56}Fe(n,2nα) ≈ 31.6% (E_{thr} = 20.0 MeV)
^{92m}Nb (28 d)	1.32 ± 0.36	^{93}Nb(n,2n) ≈ 94.6% (E_{thr} = 8.9 MeV) ^{92}Mo(n,p) ≈ 4.9% (E_{thr} = 0.0 MeV)

* Simakov, et al, "Activation Analyses of Vanadium irradiated by d-Li neutrons using IEAF-2001 cross sections", Workshop on Activation Data – EAF 2003, Prague, 24-26 June 2002

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Goals for Future Nuclear Systems

- 7 (of 11) Generation IV Requirements (May 2000)
  - Waste Disposition
    - Minimal waste
    - Solutions for all waste streams
    - Public acceptance of waste solutions
  - Proliferation Resistance
    - Minimal attractiveness to potential proliferation
    - Evaluation of Proliferation Resistance
  - Safety
    - No need for offsite response
    - "As Low As Reasonably Achievable" radiation exposure
- 9 (of 21) Generation IV Roadmap Criteria (Jan 2002)

Inventory Analysis: Waste

- Inventory calculations needed to characterize waste
  - activity
  - waste disposal ratings
  - decay heat
  - contact dose
- Some proposed waste solutions are themselves nuclear systems
  - Accelerator Transmutation of Waste [ATW]
- Waste or Product?
  - Possibility for recycling of materials with low levels of radioactivity

Inventory Analysis: Proliferation

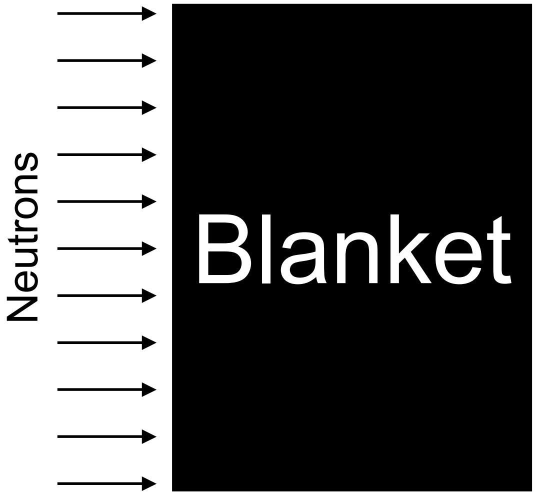
- No comprehensive methodology or metric for quantitative assessment of proliferation resistance
- Accurate inventory calculations are input for other considerations
  - chemical form of fissile inventory
  - accessibility of fissile inventory
  - ability to monitor changes in fissile inventory
- Inventory calculations as part of monitoring procedure?

Inventory Analysis: Safety

- Based on release of radioactive isotopes
  - To eliminate need for off-site response, need to demonstrate negligible release levels
- Radiation protection policy changes
  - Importance of radioactivity calculations if the Linear Non-Threshold theory is abolished

Future Complications

- Flows and cycles of material with various time scales and processes
  - e.g. fusion blanket



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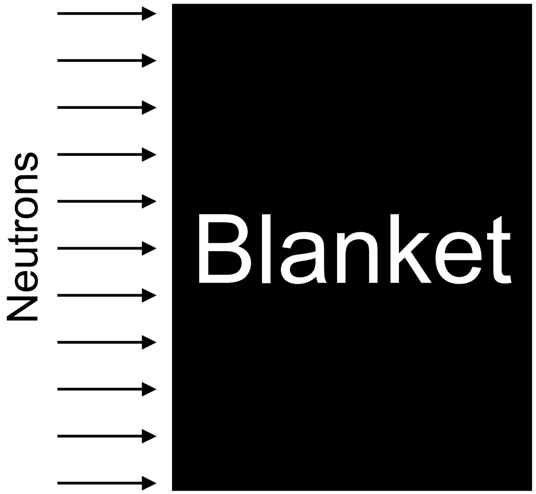

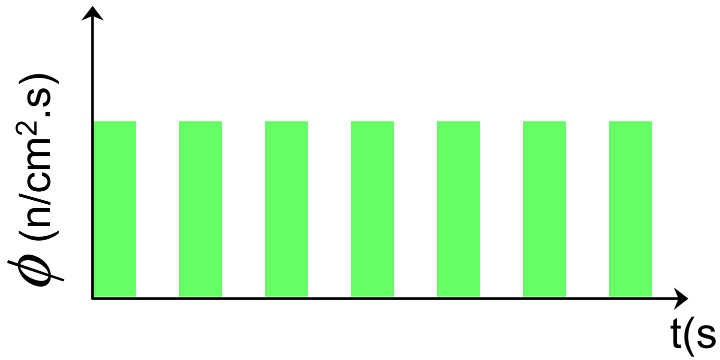

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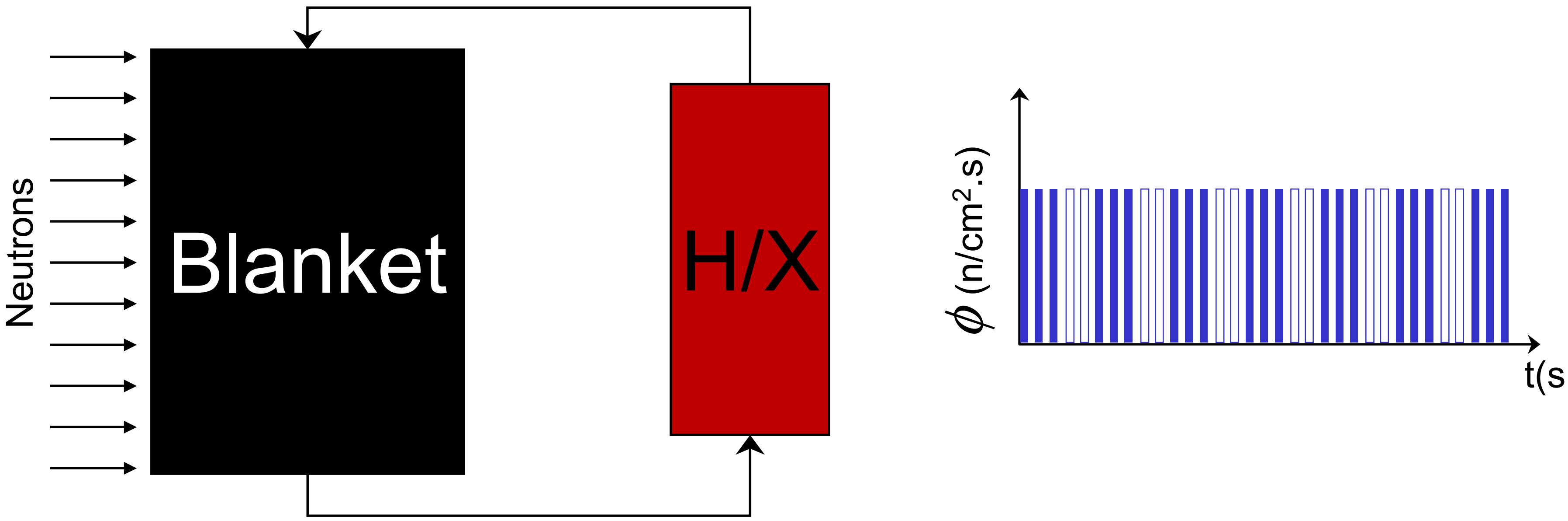
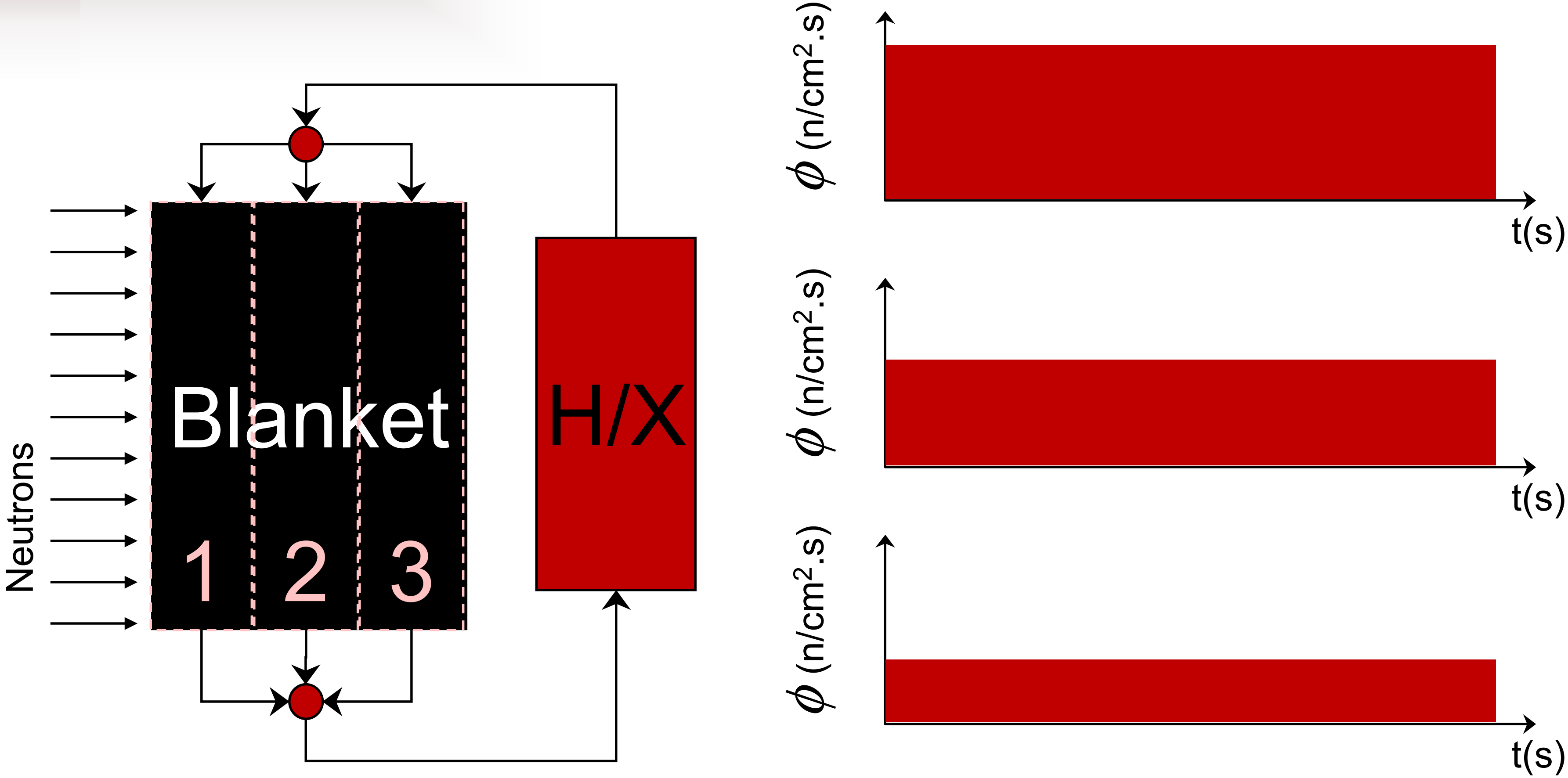
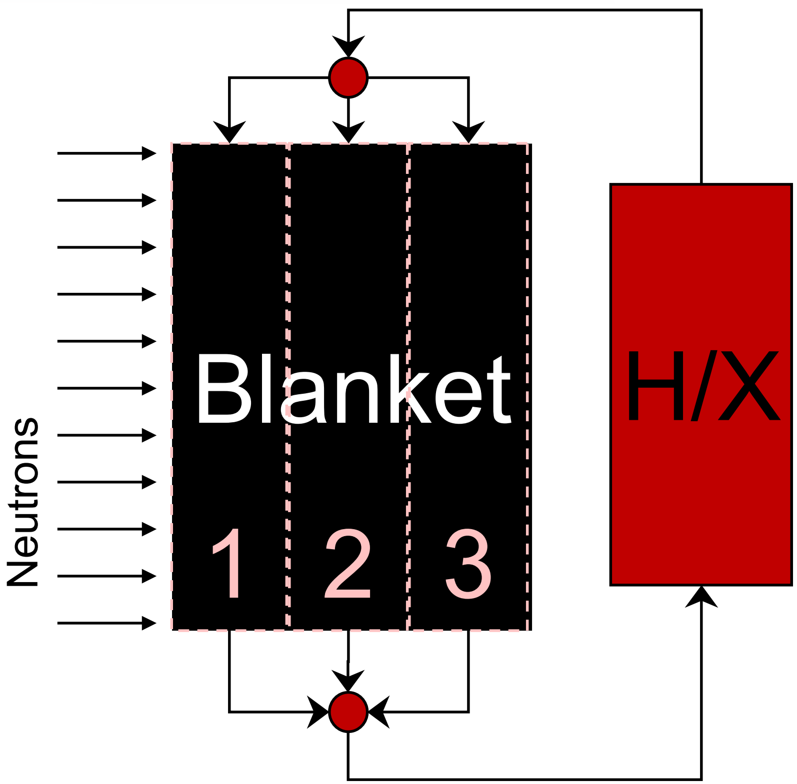
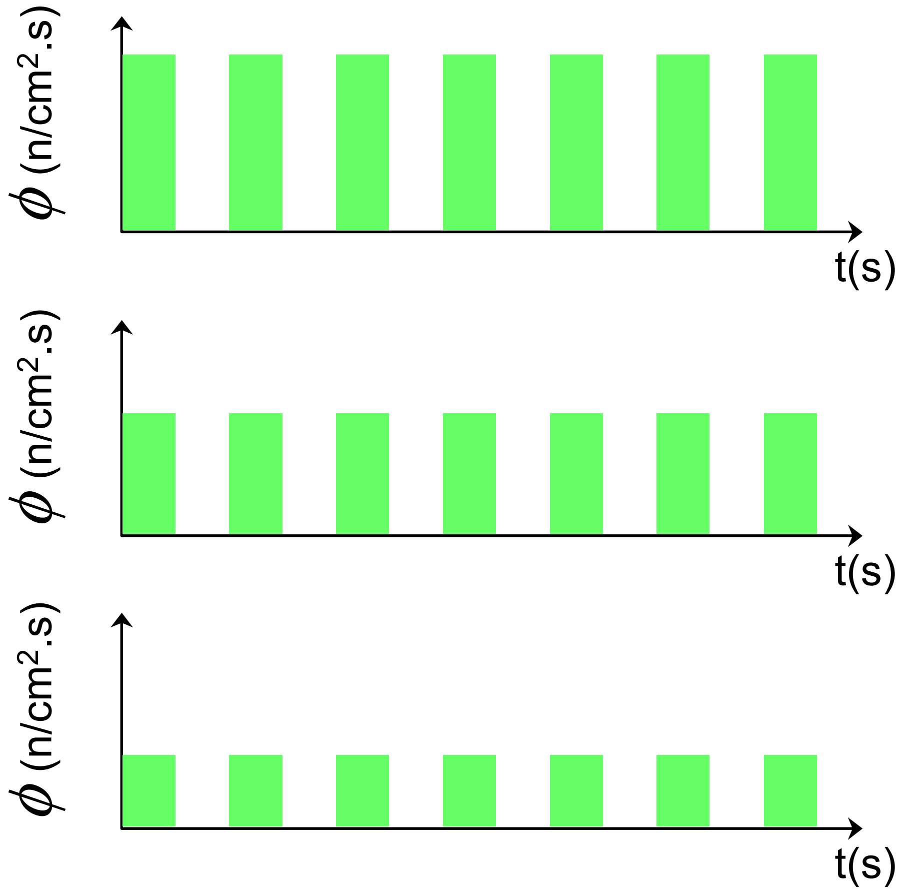


Complicated Coolant Flows



Complicated Coolant Flows





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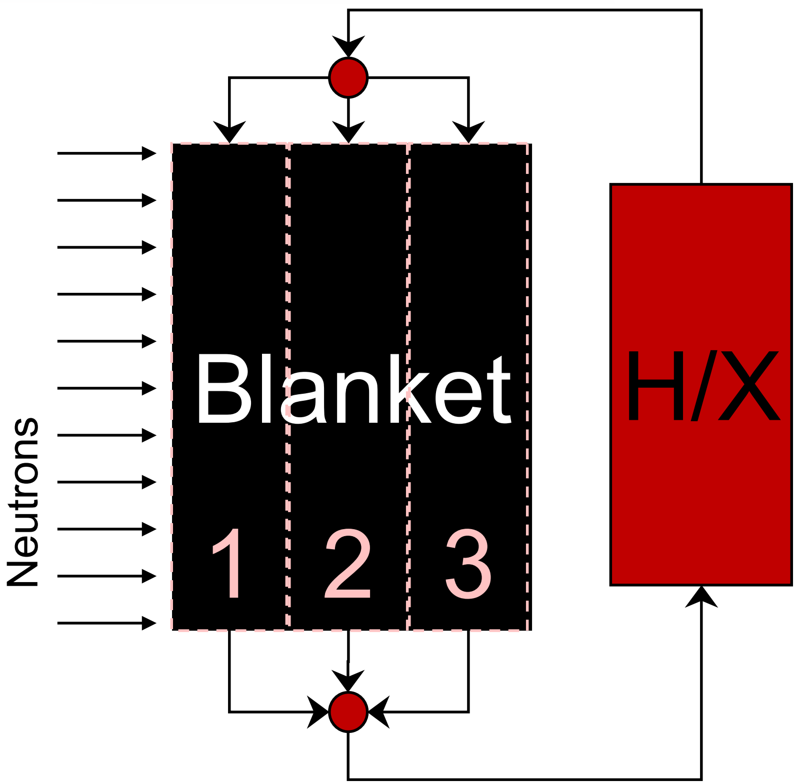
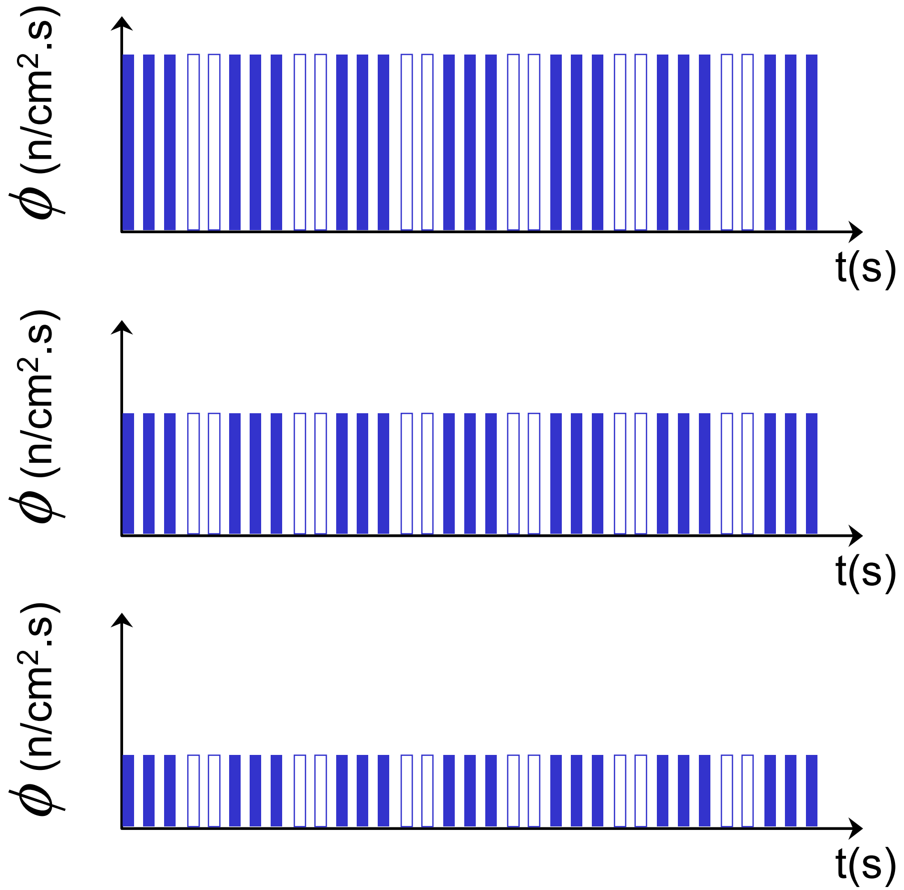
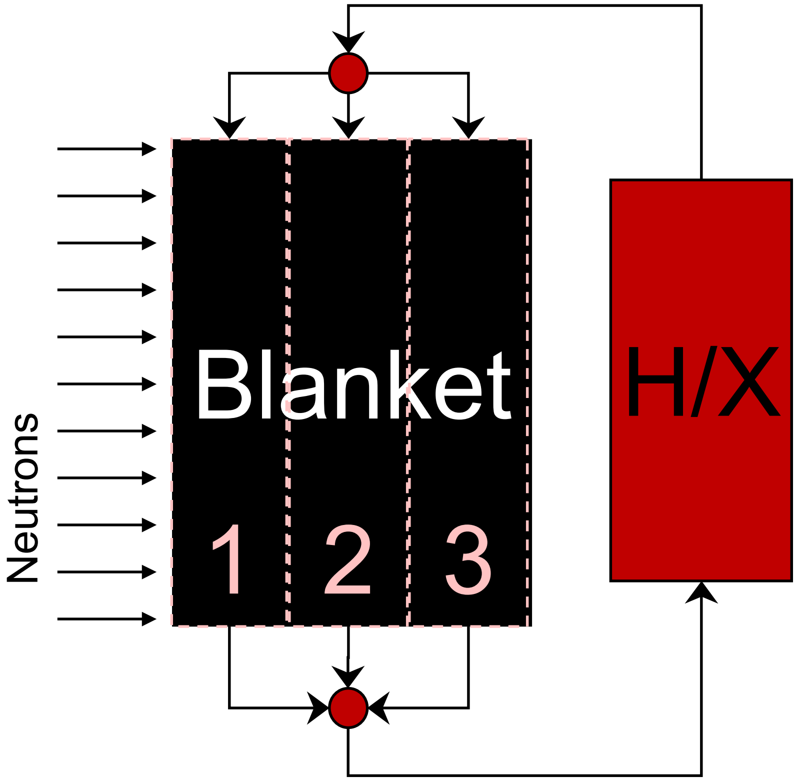
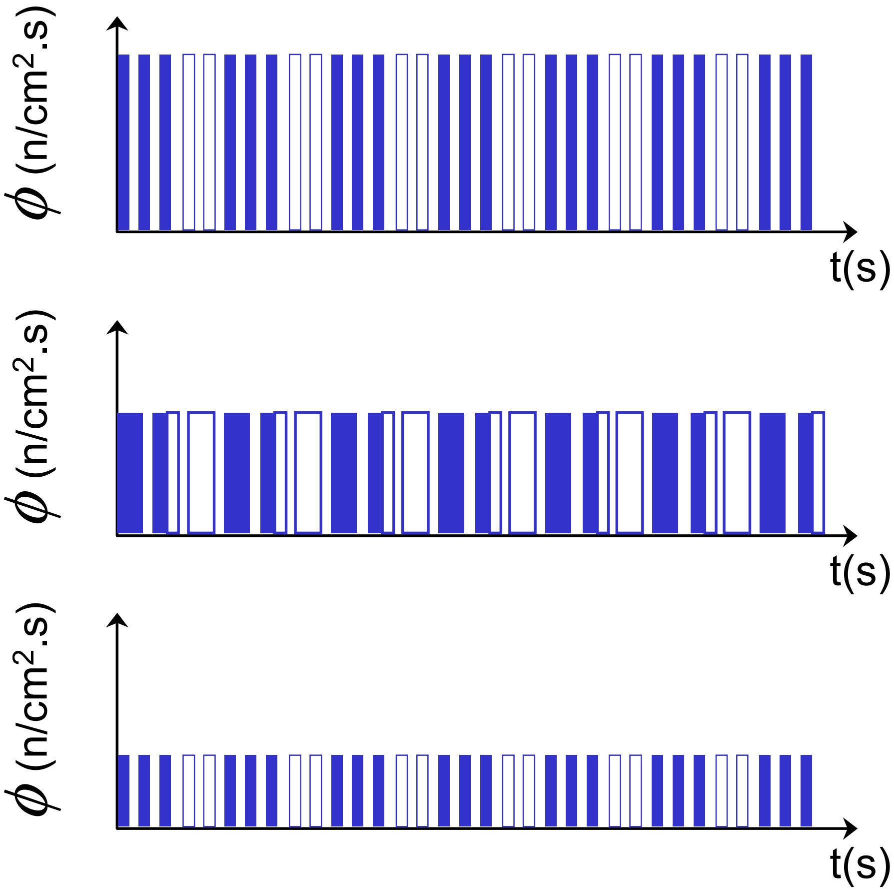


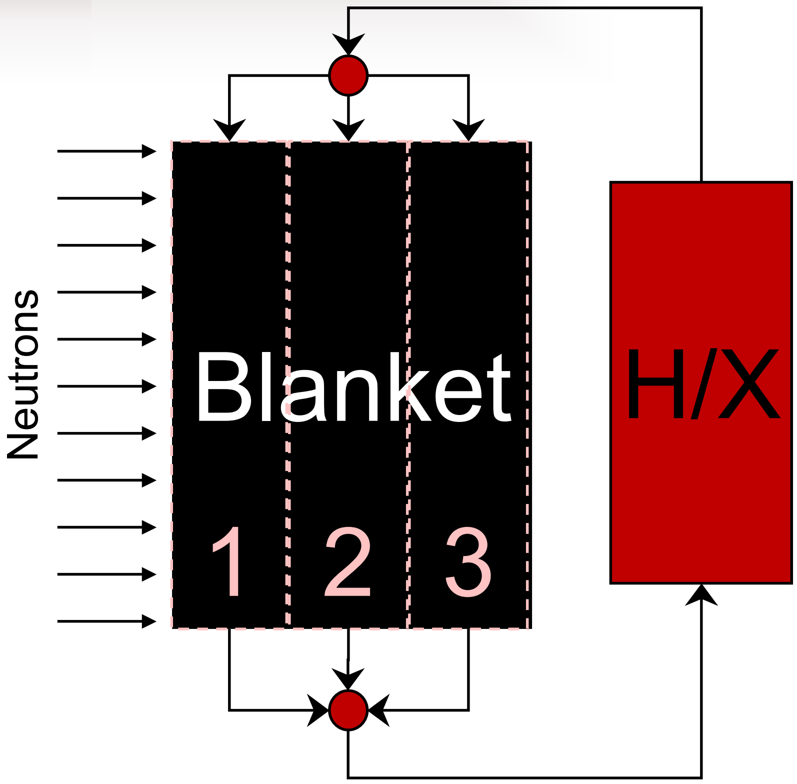


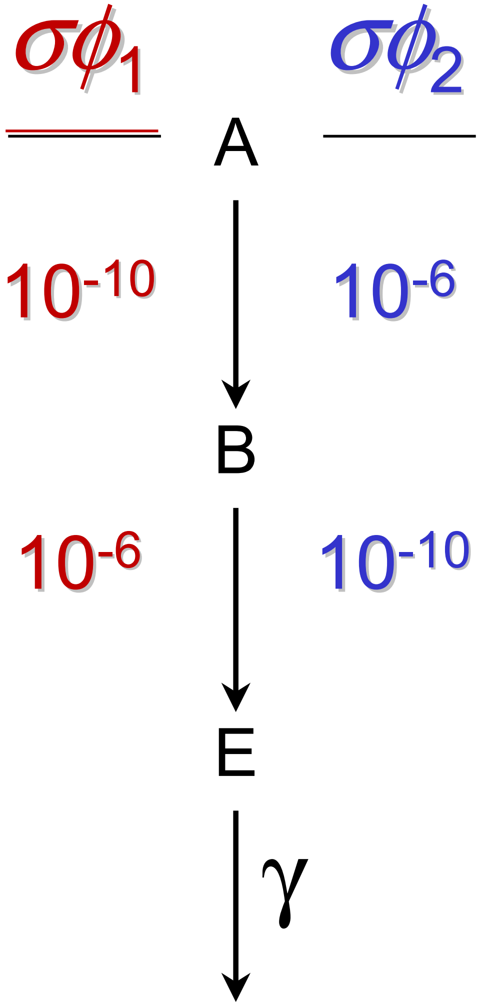
Complicated Coolant Flows



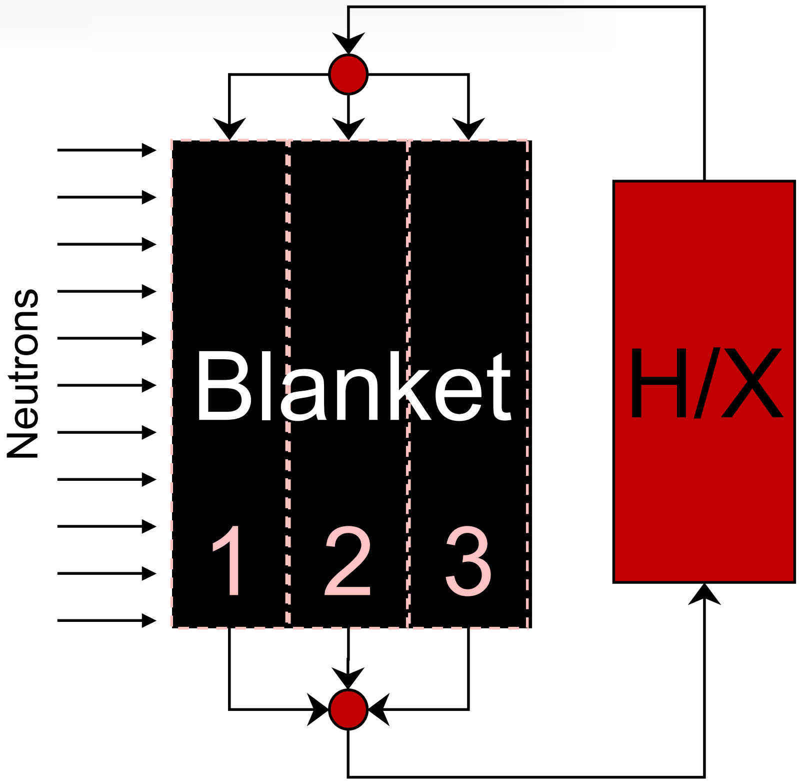


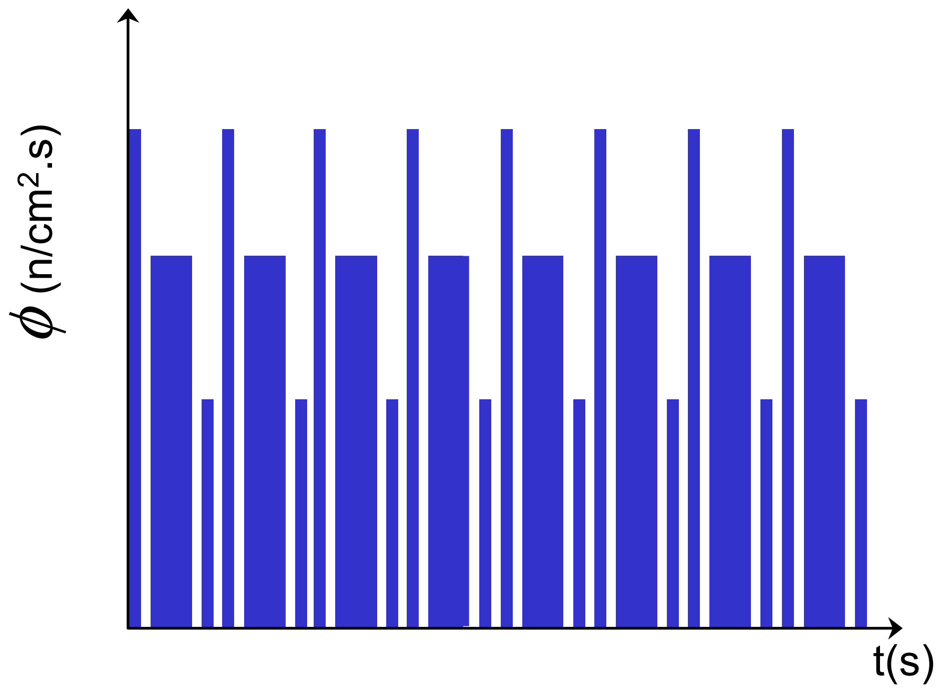
Reaction Rates & Flow Paths



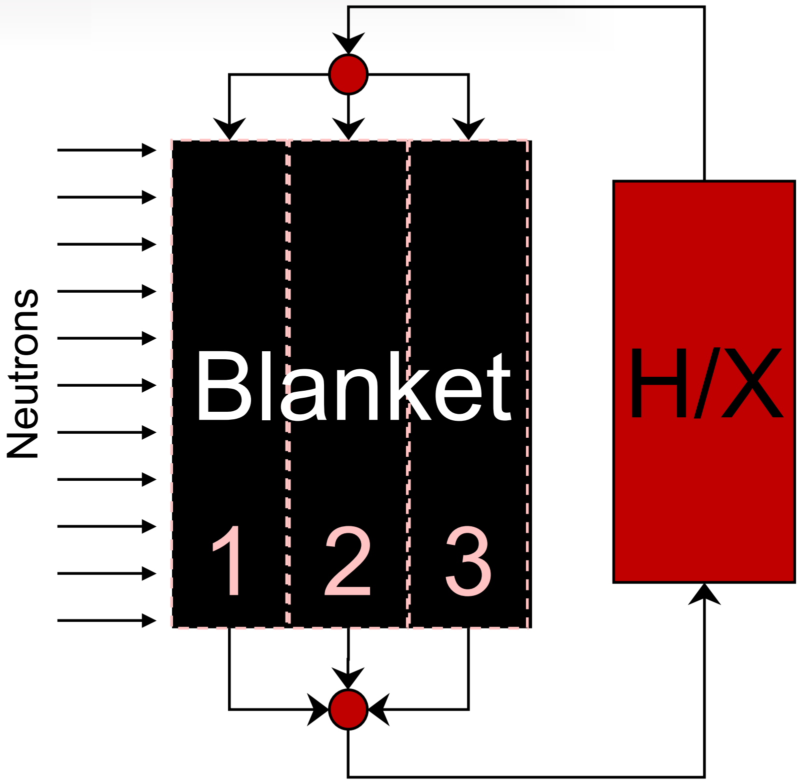


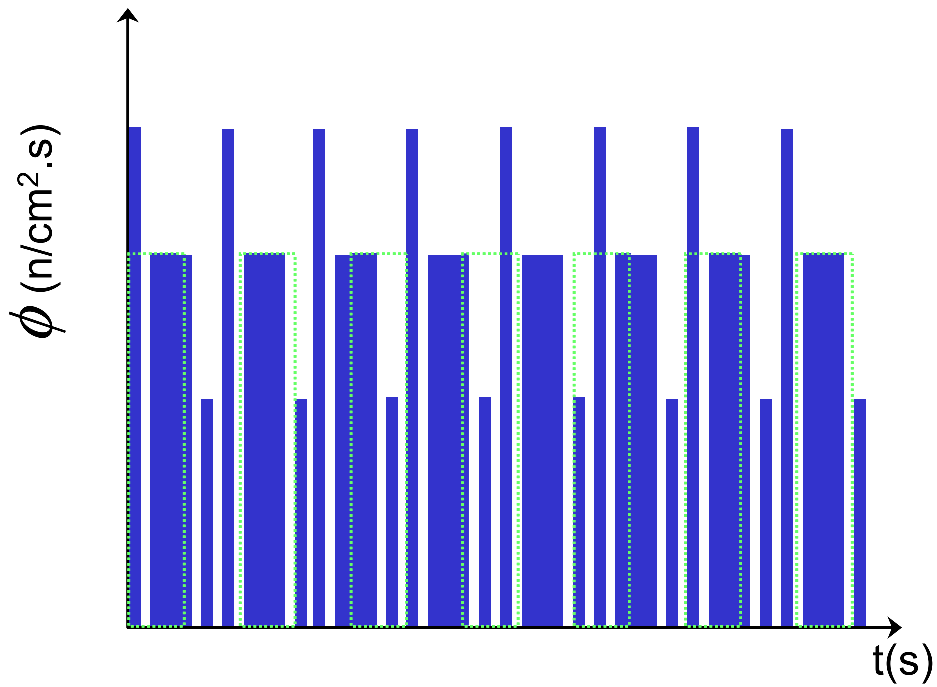
Approximations



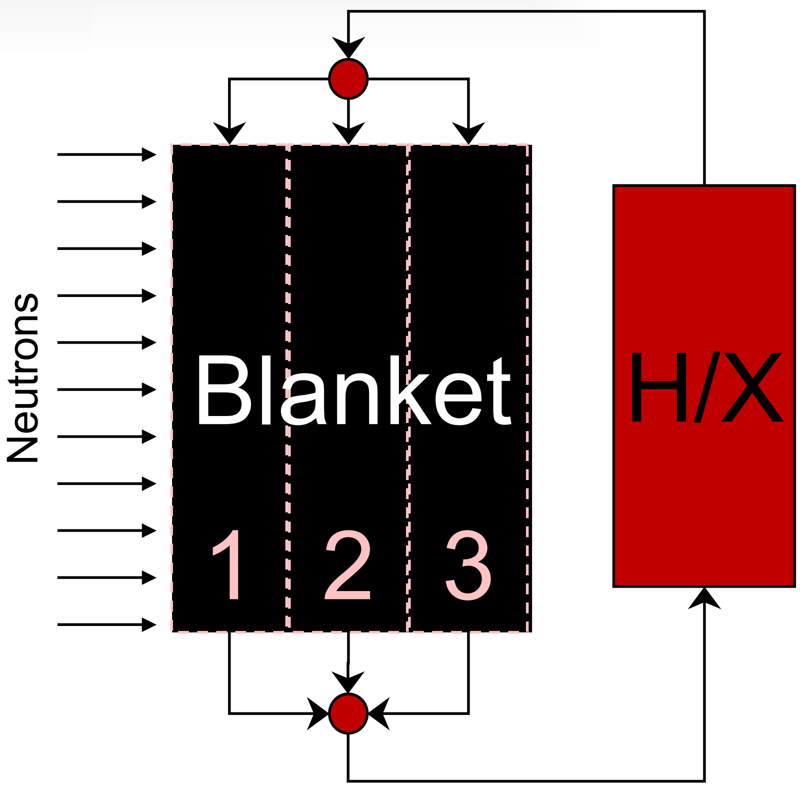


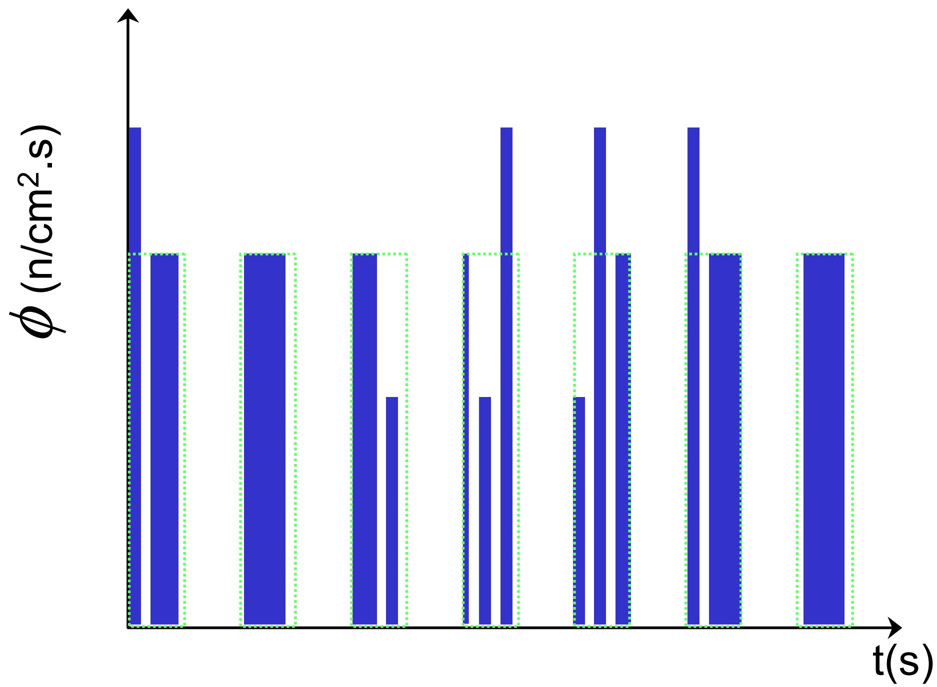
Approximations





Approximations





Future Systems – New Challenges

- ATW/AAA/ADS
  - Liquid accelerator targets – spallation and activation products
  - Process streams with fissile material and fission products
- Symbiotic fuel cycles
  - PWR + CANDU: DUPIC (Korea)
  - LWR + FBR + ADS
    - Various chemical processes in between

Future Systems – New Challenges

- Generation IV (V? VI?)
  - Online chemical processing of flowing fuels
  - Thorium fuel cycles
- Fusion Power Plants
  - Inertial fusion target material recycle
  - Liquid walls
  - Liquid breeders
  - Online chemical processing

Future Developments

- ALARA
  - Support for fission data
  - Depletion feedback with new deterministic methods
- New projects
  - Stochastic methods
  - Fuel cycle analysis
  - Interface with probabilistic analyses of proliferation risk

Overview

- Background on Isotopic Inventory
- Fusion Activation: ALARA
- Inventory Analysis of Future Systems
- Summary

Summary

- Isotopic inventory analysis brings together traditional burnup/depletion analysis and activation analysis
- Inventory analysis methods can benefit from constant improvement
- Renewed interest in advanced nuclear systems gives new and interesting research opportunities