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SPENT FUEL FORENSICS

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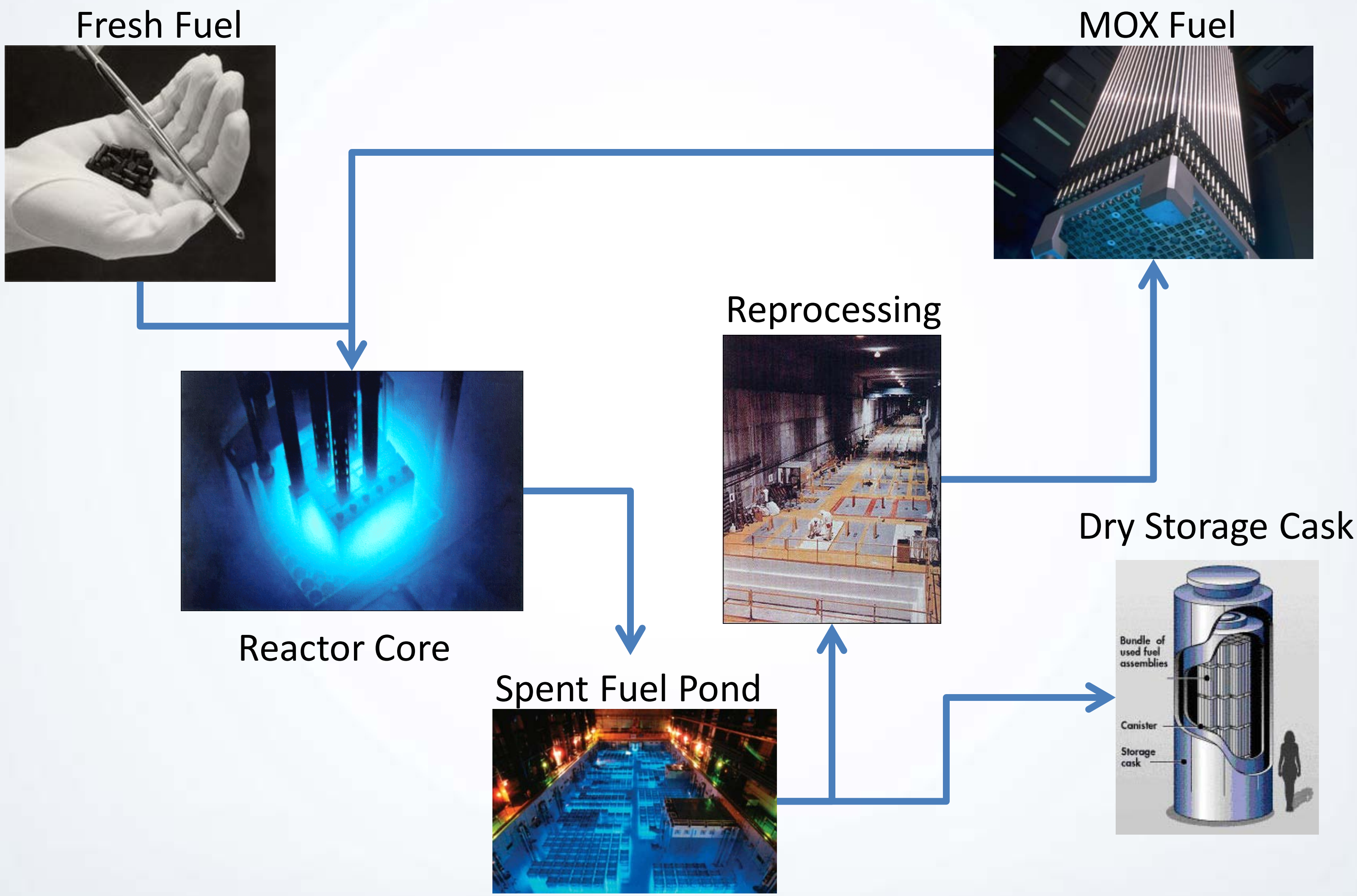
Abstract

Reactor modeling codes have been developed over several decades and are in a mature state. A few common uses for these forward reactor modeling codes are to design reactor cores, plan for refueling and characterize spent fuel. Inverse reactor modeling software to aid nuclear forensics is still in its infancy and lacks the mature development of the forward reactor modeling codes. Recent progress has been made and a few innovative techniques have been developed to characterize unknown spent fuel samples by analyzing measured actinides and fission products. Inverse modeling can be used to determine the fuel burnup, initial fuel enrichment, time of fuel discharge, and other reactor and fuel parameters. These techniques require the use of forward reactor modeling codes in conjunction with newly developed algorithms for inverse modeling. Knowing characteristics of irradiated fuels can contribute towards spent fuel forensics and the validation of reactor operational histories.

Fuel Characterization



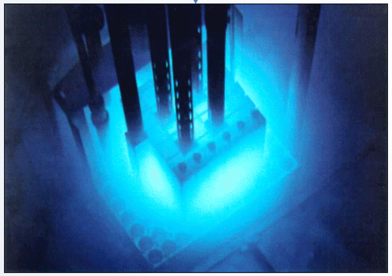
Reactor Depletion Codes

- ORIGEN 2
- SCALE 5 (ORIGEN-S, TRITON)
- MCNPX w/ CINDER90
- Monteburns (MCNP and ORIGEN2)
- Attila
- WIMS

Nuclear Forensics

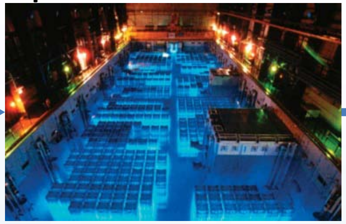
Fresh Fuel

?

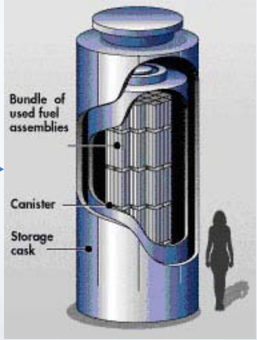


Reactor Core

Spent Fuel Pond



Dry Storage Cask



Inverse Model

Nuclide Monitor Measurements

Burnup

Enrichment

Reactor Type

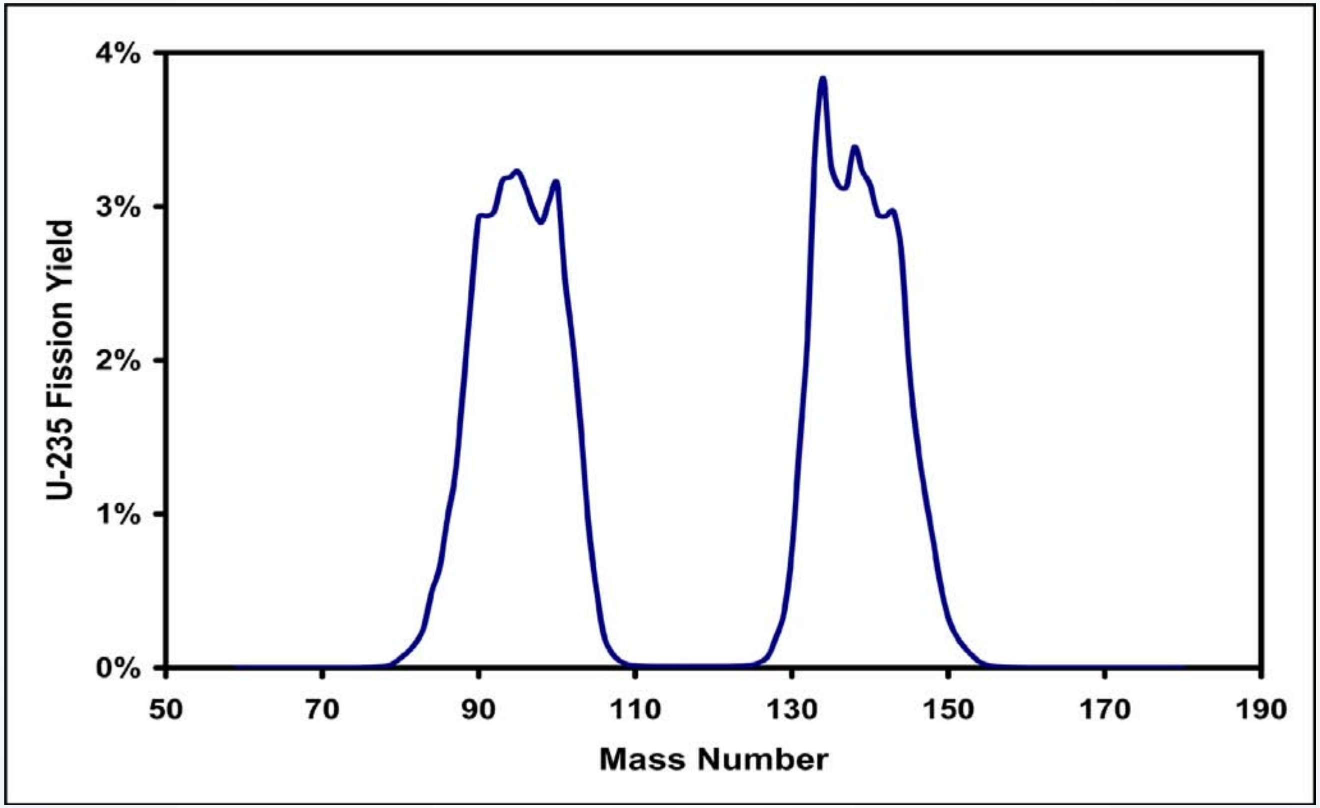
Cooling Time

Suspect Source Material

Burnup

The amount of power produced per kg of fuel

Usually measured in MWd/MTU (megawatt days / metric ton of U)

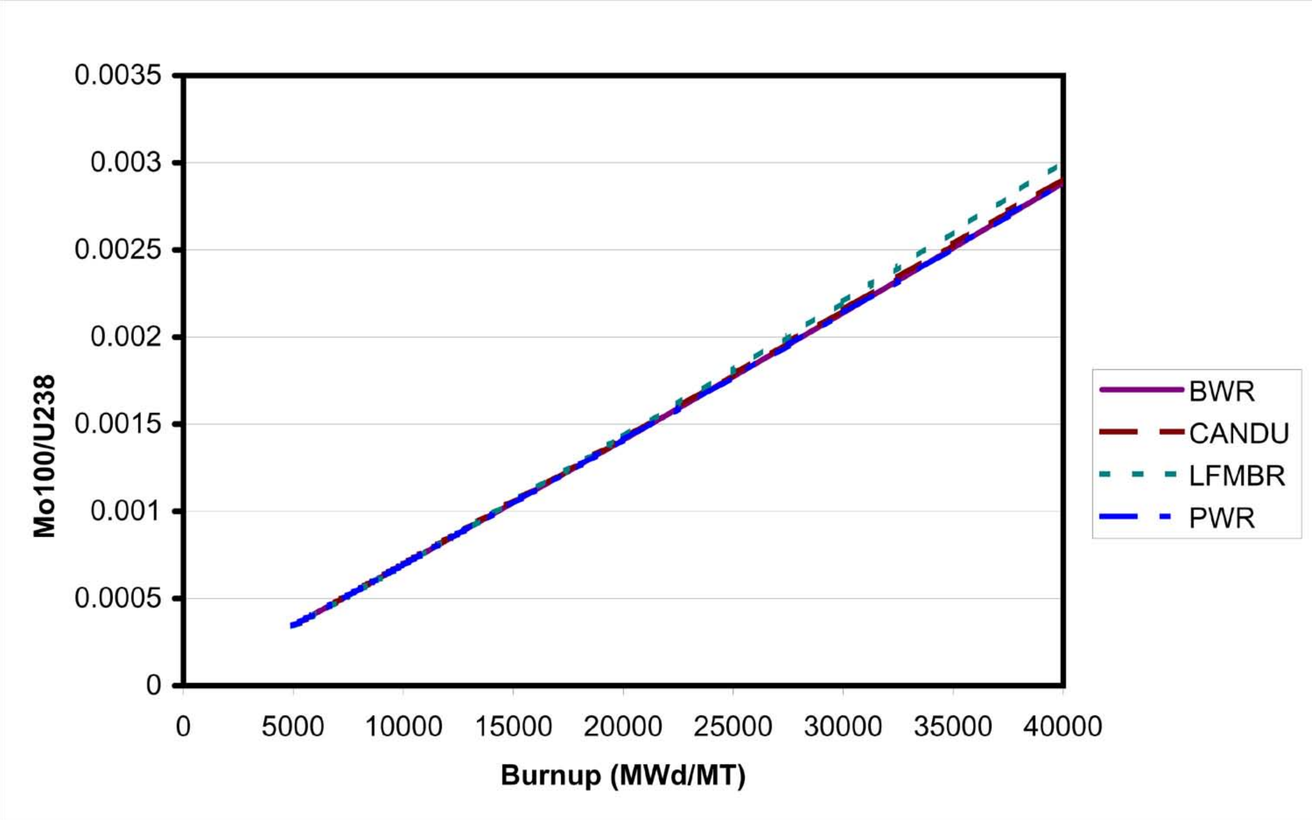


Burnup → **Enrichment** → **Reactor Type** → **Cooling Time**

Burnup

Monitor Requirements:

➢ ^{235}U fission yield is the same for all reactor types



Burnup → **Enrichment** → **Reactor Type** → **Cooling Time**

Burnup

$$BU(T) = \left[\frac{N_B(T)}{N^{U238}(T)}\right]\left[\frac{N^{U238}(T)}{N_o^U}\right]\frac{N_a E_R}{Y_B}$$

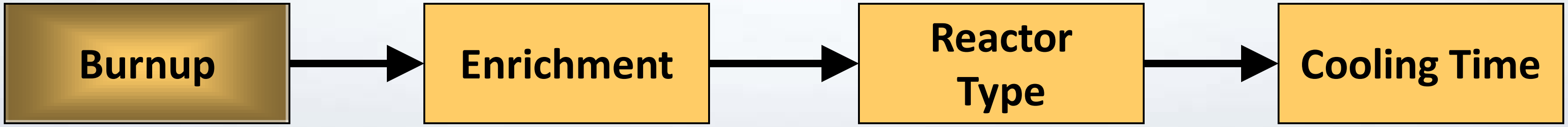
$$\left[\frac{N_o^U}{N^{U238}(T)}\right] = \frac{\left[\frac{N^U(T)}{N^{U238}(T)}\right] + \left[\frac{N^{Pu239}(T)}{N^{U238}(T)}\right] + \left[\frac{N^{Pu240}(T)}{N^{U238}(T)}\right] + \ldots}{1 - \frac{M_o^U}{N_a E_R} BU(T)}$$

Burnup → **Enrichment** → **Reactor Type** → **Cooling Time**

Burnup

Initial Uranium Atom Density per ^{238}U Atom Density Calculation After Using an Iteration Scheme with Burnup

Reactor Type	Burnup (MWd/MT)	Exact $\left[\frac{N_o^U}{N^{U238}(T)}\right]$	Predicted $\left[\frac{N_o^U}{N^{U238}(T)}\right]$	Percent Error
PWR	40,000	.9375	.9386	**0.109**
BWR	40,000	.9378	.9387	**0.097**
CANDU	15,000	.9769	.9770	**0.007**
LMFBR	70,000	.8291	.8284	**0.079**

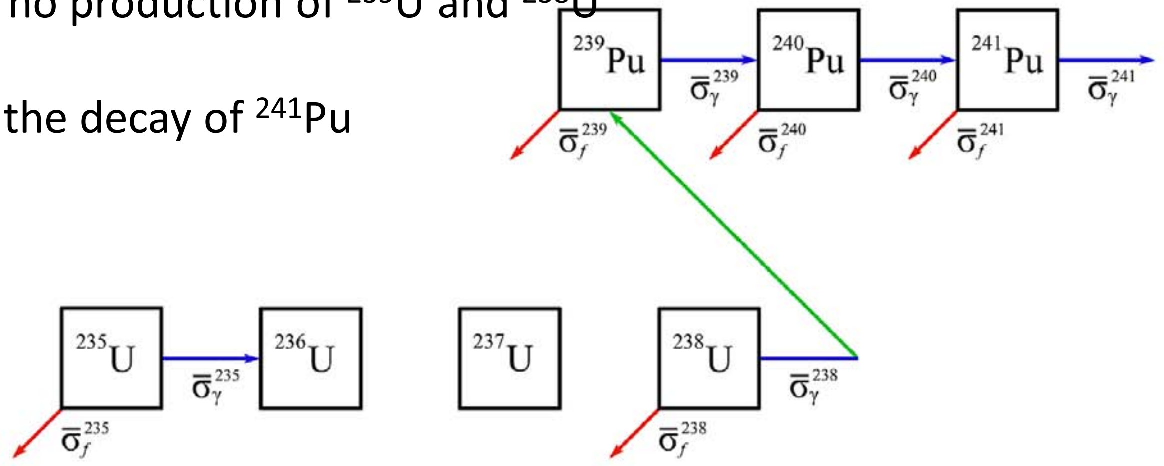


Enrichment

The ratio of ^{235}U to U in the fuel before irradiation

Assumptions on fuel characteristics during irradiation:

- The only isotopes that fission are ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu
- ^{239}Np and ^{239}U decay instantaneously to ^{239}Pu
- There is no production of ^{235}U and ^{238}U
- Neglect the decay of ^{241}Pu



Burnup → **Enrichment** → **Reactor Type** → **Cooling Time**

Enrichment Equation

$$H1 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U238}}{N_o^U}\right]\left(\left[\frac{N^{U235}}{N^{U238}}\right] + \left[\frac{N^{U236}}{N^{U238}}\right]\right)$$

$$H2 = -M_o^U BU(T) \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241}$$

$$H3 = N_a E_R \bar{\sigma}_f^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H4 = -N_a E_R \bar{\sigma}_f^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U234}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H5 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U239}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H6 = -N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H7 = -N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U234}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H8 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U240}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H9 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U239}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H10 = -N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U234}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H11 = N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H12 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_f^{Pu241} \left[\frac{N^{U241}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H13 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241} \left[\frac{N^{U240}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H14 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241} \left[\frac{N^{U239}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H15 = N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H16 = \bar{\sigma}_f^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} - \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241}$$

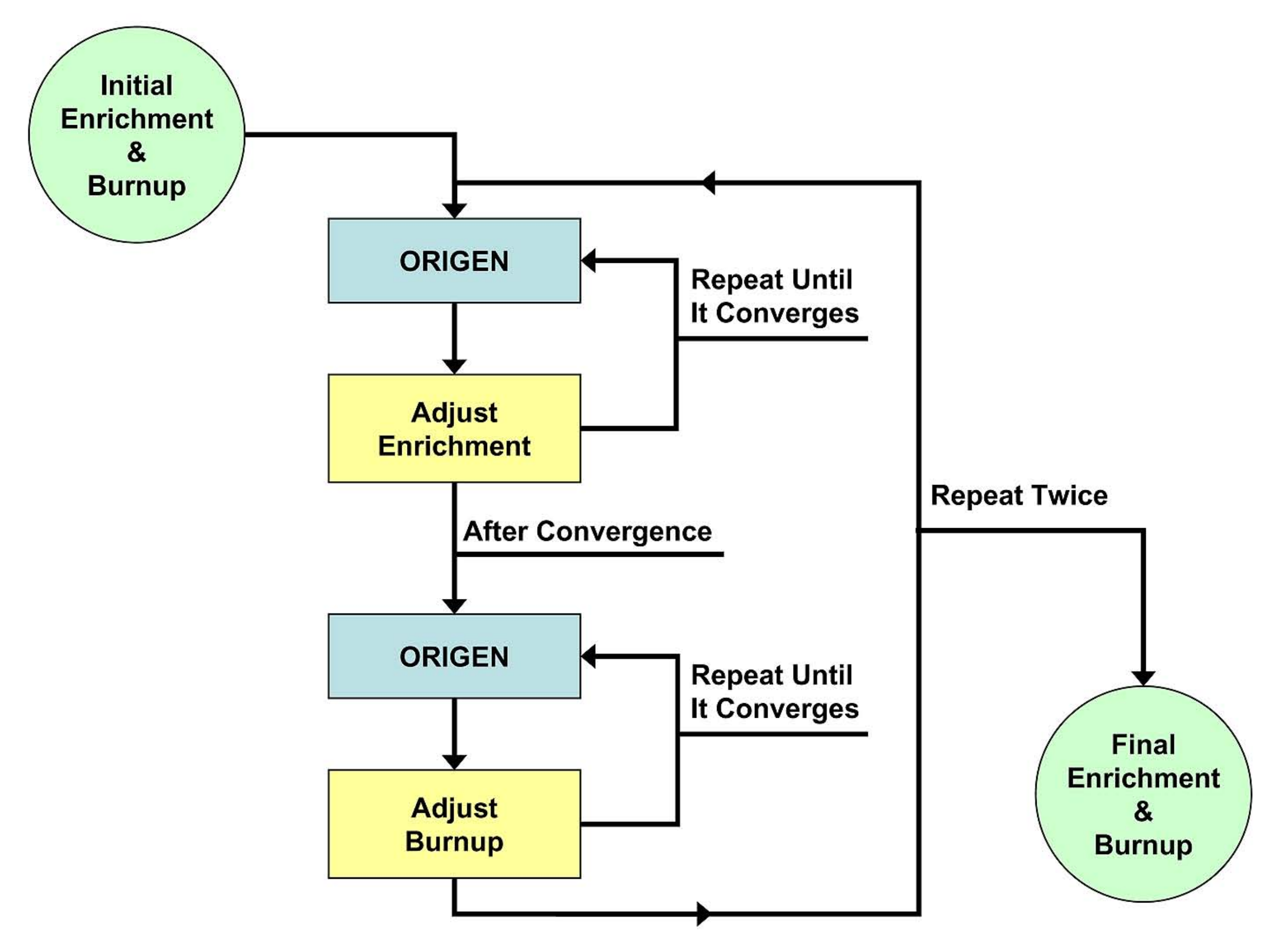
$$H17 = \bar{\sigma}_\gamma^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} - \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241}$$

$$H18 = \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241}$$

$$e_o = \frac{(H1 + H2 + \ldots . H15)}{H16 + H17 + H18}$$

Burnup → **Enrichment** → Reactor Type → Cooling Time

ORIGEN Iteration



Burnup → **Enrichment** → Reactor Type → Cooling Time

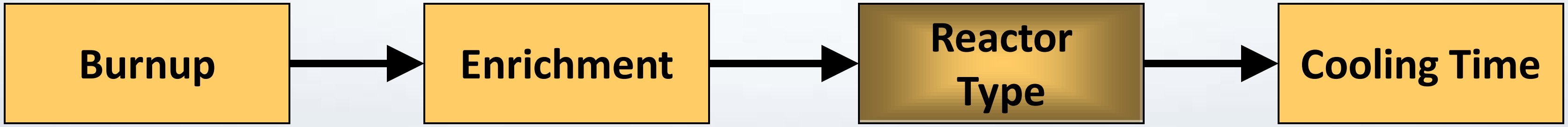
Reactor Type

Reactor Type:

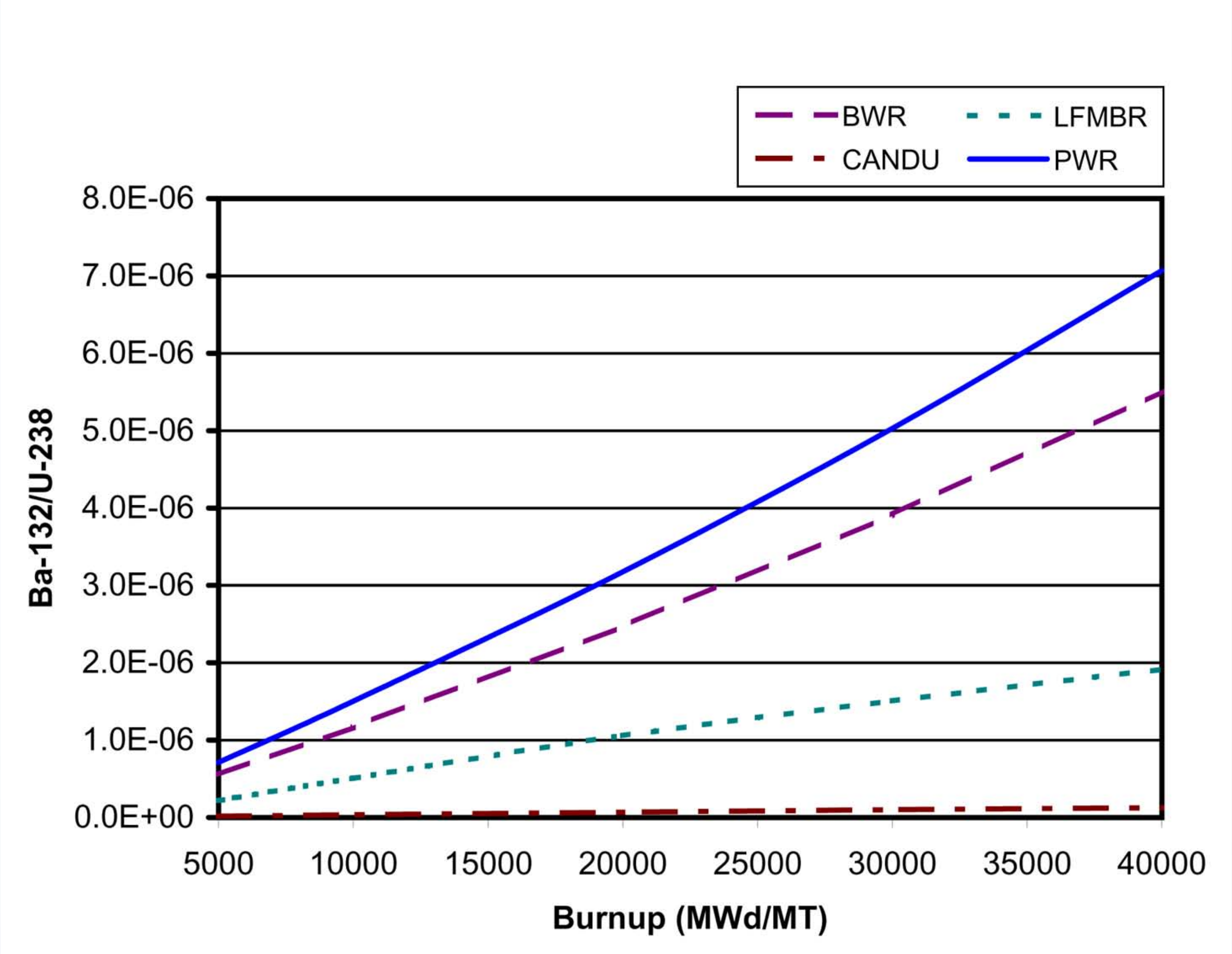
Nuclear reactors that share a common design concept
(e.g. PWR, BWR, CANDU, LFMBR)

Reactor Type Monitor Requirements:

- Fission yield and/or absorption rate changes for each reactor type
- Stable or long-lived nuclides



Reactor Type



Burnup → Enrichment → **Reactor Type** → Cooling Time

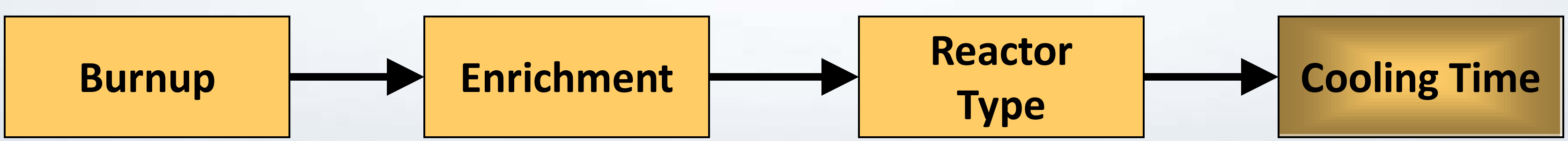
Cooling Time

Cooling Time:

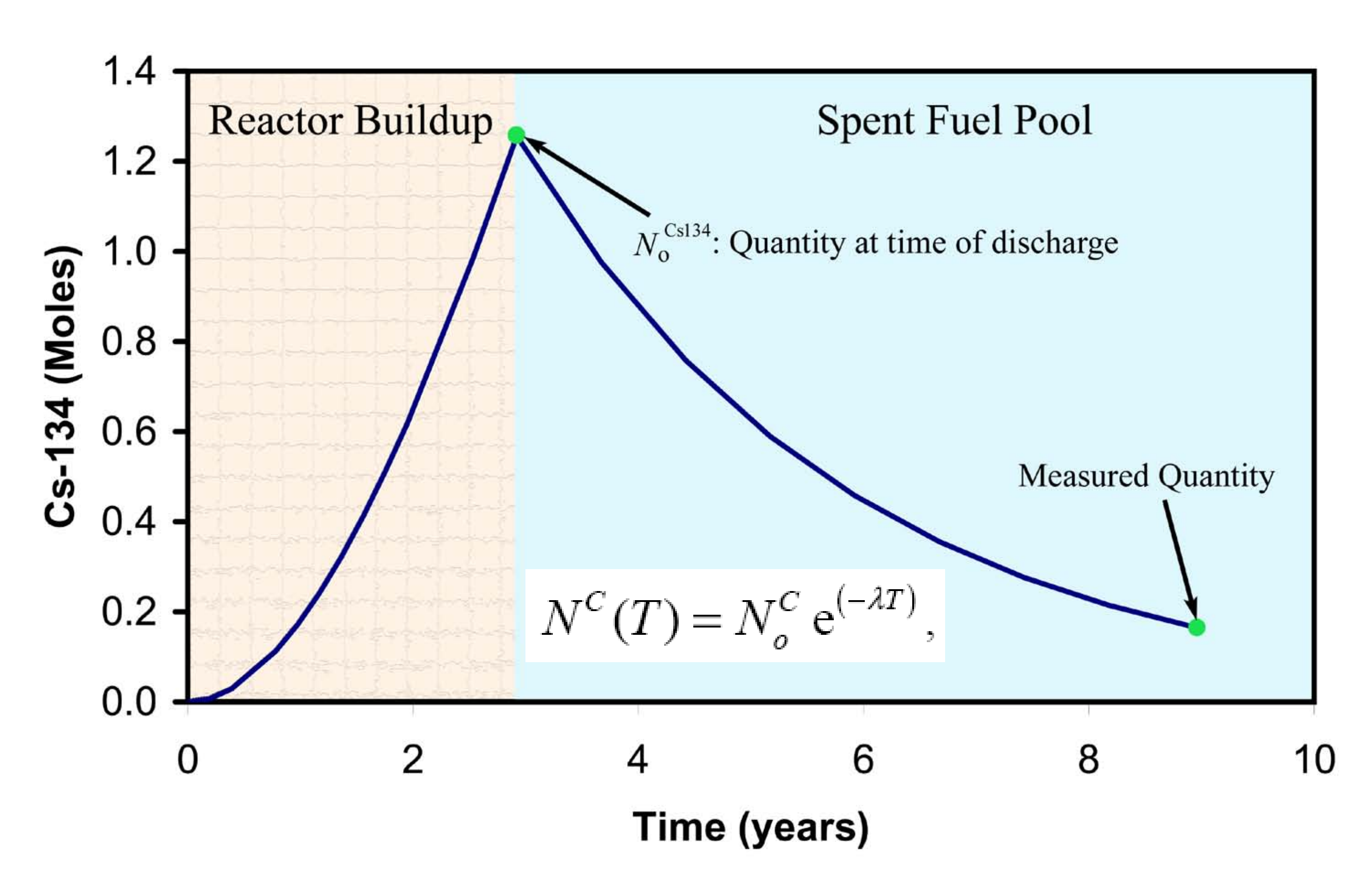
Cooling time is the period from the end of irradiation to the time of measurement

Cooling Time Monitor Requirements:

- Half-life is between 1-30 years
- At least 0.01 moles is produced per MT of fuel



Cooling Time



Burnup → **Enrichment** → **Reactor Type** → **Cooling Time**

Possible Monitors

Burnup Monitors	^{140}Ce, ^{100}Mo, ^{98}Mo, ^{97}Mo, ^{138}Ba, ^{142}Ce, ^{148}Nd
Enrichment Indicators	^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu
Reactor Type Monitors	^{109}Ag, ^{153}Eu, ^{156}Gd, ^{143}Nd, ^{240}Pu, ^{108}Cd, ^{113}Cd, ^{149}Sm, ^{166}Er, ^{132}Ba, ^{98}Tc, ^{115}In, ^{72}Ge, ^{115}Sn
Cooling Time Monitors	^{90}Sr, ^{93}Nb, ^{106}Ru, ^{101}Rh, ^{102}Rh, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{146}Pm, ^{147}Pm

Monitors that can differentiate a PWR and BWR
^{132}Ba, ^{98}Tc, ^{115}In, ^{72}Ge, ^{108}Cd, ^{115}Sn

NEMASYS

Nuclear **E**vent **M**aterial **A**ttribution **SYS**tem

- Written with Microsoft Visual Studio .NET
- GUI based
- Requires Microsoft Windows
- Requires Microsoft’s .NET framework

Benchmark

Mihama Unit 3

U.S.-style PWR Design

9 Samples
from
3 Assemblies

SFCOMPO
Developed by Fuel Cycle Safety Evaluation Laboratory at the JAERI Department of Fuel Cycle Safety Research

http://www.nea.fr/sfcompo/Ver.2/Eng/index.html

JAPAN

Benchmark

Monitors used for attribution of the Mihama-3 unit.

$^{148}Nd/^{238}U$	Burnup
$^{235}U/^{238}U$	Enrichment
$^{236}U/^{238}U$	Enrichment
$^{238}U/U$	Enrichment
$^{239}Pu/^{238}U$	Enrichment
$^{240}Pu/^{238}U$	Enrichment
$^{143}Nd/^{148}Nd$	Reactor Type
$^{240}Pu/^{148}Nd$	Reactor Type
$^{241}Pu/^{238}U$	Age
$^{134}Cs/^{238}U$	Age
$^{137}Cs/^{238}U$	Age
$^{106}Ru/^{238}U$	Age

Benchmark

Burnup results for Mihama-3.

Assembly	Sample No.	Reported Burnup (MWd/MT)	Predicted Burnup (MWd/MT)	Error
JPNNM3SFA1	1	8,300	7,952	4.19%
JPNNM3SFA1	2	6,900	6,678	3.22%
JPNNM3SFA1	3	15,300	14,664	4.16%
JPNNM3SFA2	4	21,200	20,399	3.78%
JPNNM3SFA2	5	14,600	14,043	3.82%
JPNNM3SFA3	6	29,400	28,394	3.42%
JPNNM3SFA3	7	32,300	30,931	4.24%
JPNNM3SFA3	8	33,700	32,371	3.37%
JPNNM3SFA3	9	34,100	32,920	3.46%
Average Error	-	-	-	3.74%

Benchmark

Enrichment Calculations Before and After Using an Iteration Scheme with ORIGEN

	Reported	Initial Prediction		After Iteration	
Mihama-3	Enrichment (a/o)	Enrichment (a/o)	Percent Error	Enrichment (a/o)	Percent Error
Sample 1	3.25	2.93	**9.94**	3.22	**1.08**
Sample 2	3.25	2.98	**8.31**	3.27	**0.62**
Sample 3	3.24	2.81	**13.27**	3.20	**1.33**
Sample 4	3.24	2.91	**10.19**	3.27	**0.93**
Sample 5	3.24	2.77	**14.51**	3.20	**1.23**
Sample 6	3.25	3.16	**2.77**	3.21	**1.23**
Sample 7	3.25	3.32	**2.15**	3.29	**1.23**
Sample 8	3.25	3.36	**3.38**	3.22	**0.92**
Sample 9	3.25	3.38	**4.00**	3.21	**1.23**

Benchmark

Age prediction results for Mihama-3.

Sample	Reported Age	Predicted Age	Error
1	5	4.93	**1.36%**
2	5	4.89	**2.16%**
3	5	4.78	**4.36%**
4	5	4.86	**2.76%**
5	5	4.60	**7.96%**
6	5	5.05	**1.04%**
7	5	4.93	**1.36%**
8	5	5.15	**3.04%**
9	5	5.09	**1.84%**
		Average	**2.87%**

Results Summary

	Error
Burnup	3.74%
Enrichment	1.09%
Reactor Type	*0.00%
Age	2.87%

Assumptions

- The specific power (MW/MT) stayed near constant
- The power level was near full power
- No long shutdown periods (> 30 days) before discharge

In most cases this is only valid for commercial reactors

Future Work

- Need characterization data on spent fuel with known irradiation histories for further benchmarking
- Expand the library of inverse algorithms
- Develop capability to handle reprocessed material
- Uncertainty propagation needs to be implemented
- Identify possible scenarios where our algorithms will not work properly to avoid invalid conclusions
- Developed methods to check for inconsistent data

Questions?