Experimental Validation of a Nuclear Forensics Methodology for Reactor-Type Attribution of Chemically Separated Plutonium

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Introduction, Experimental Irradiation Campaigns, Maximum Likelihood Calculation, & Reactor Model Library

A nuclear forensics methodology was developed which is capable of reactor-type attribution of separated weapons-grade plutonium from measurements of intra-element isotope ratios. The methodology has the ability to predict reactor parameters such as burnup and time since irradiation, in addition to the reactor-type. The novelty of the developed nuclear forensics methodology arises from the focus on chemically separated weapons-grade plutonium, and therefore dependence on ten intra-element isotope ratios as forensic signatures: $^{137}Cs/^{133}Cs$, $^{134}Cs/^{137}Cs$, $^{135}Cs/^{137}Cs$, $^{154}Eu/^{153}Eu$, $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$, and $^{242}Pu/^{239}Pu$.

The developed nuclear forensics methodology was previously verified through testing with simulation data. The objective of the research presented here was to perform a validation study of the developed methodology. In order to allow a validation study of the methodology, two experimental irradiation campaigns were performed, resulting in two distinct fuel samples containing weapons usable plutonium. To replicate plutonium produced in the blanket of a fast breeder reactor, depleted uranium dioxide fuel samples were irradiated in a gadolinium sheath within the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. To represent weapons-grade plutonium produced in a natural uranium fueled thermal reactor, natural uranium dioxide fuel samples were irradiated in a thermal neutron spectrum at the University of Missouri Research Reactor (MURR). The irradiated samples were subjected to nondestructive and destructive analyses to measure the plutonium and fission product isotope ratios.

For each reactor in the library, a maximum likelihood calculation was utilized to compare the measured and simulated intra-element isotope ratios, producing a likelihood value which is proportional to the probability of the reactor being the source of the measured material. The likelihood and log-likelihood of reactor model, M, matching the measured suite of intra-element ratios, r_{mes}, is given by the following equations:

$$L(M|r_{mes}) = L(r_{mes}|M) \propto \prod_{j=1}^{n} \frac{1}{\sigma_{j,sim}\sqrt{2\pi}} exp\left\{-\frac{(r_{j,mes}-r_{j,sim})^2}{2\sigma_{j,sim}^2}\right\} \quad (1)$$

$$Log\ L(M|r_{mes}) = \sum_{j=1}^{n}\left[log\left(\frac{1}{\sigma_{j,sim}\sqrt{2\pi}}\right) - \frac{(r_{j,mes}-r_{j,sim})^2}{2\sigma_{j,sim}^2}\right] \quad (2)$$

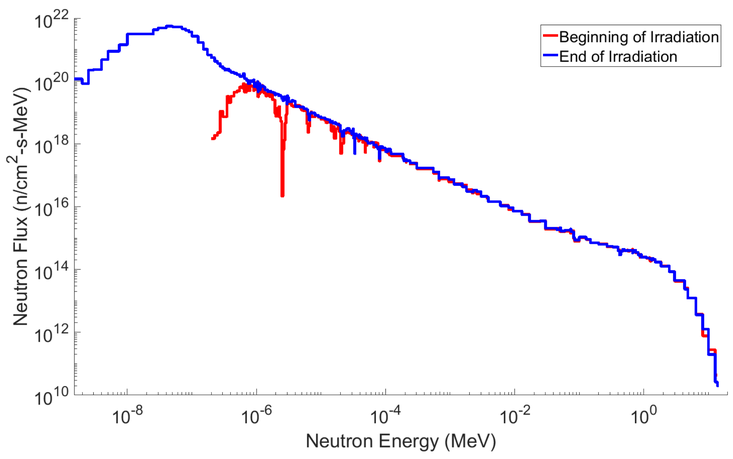
where, $r_{j,mes}$ is the j^{th} intra-element isotope ratio (for example $^{137}Cs/^{133}Cs$) in a suite of n measured intra-element ratios, $r_{j,sim}$ is the j^{th} intra-element isotope ratio in a suite of n simulated intra-element ratios from model, M, and $\sigma_{j,sim}$ is the simulation uncertainty associated with the j^{th} intra-element ratio value from the suite, r_{sim}. The variance in the log-likelihood is given by the following equation:

$$\sigma_{Log\ L}^2 = \sum_{j=1}^{n}\left(\frac{(r_{j,mes}-r_{j,sim})}{\sigma_{j,sim}^2}\right)^2 \times (\sigma_{j,mes}^2 + \sigma_{j,sim}^2) \quad (3)$$

where, $\sigma_{j,mes}$ is the measurement uncertainty associated with the j^{th} intra-element ratio value from the suite, r_{mes}.

Reactor Library Model Characteristics

Reactor Model	Thermal Power (MWth)	Fuel Type (at.% ^{235}U)	Moderator	Coolant
PWR (2.35%)	3400	UO_2 (2.35)	Light Water	Light Water
PWR (3.4%)	3400	UO_2 (3.4)	Light Water	Light Water
PWR (4.45%)	3400	UO_2 (4.45)	Light Water	Light Water
FBR (blanket)	1250	UO_2 (0.25)	-	Liquid Sodium
PHWR	756	UO_2 (0.72)	Heavy Water	Heavy Water
NRX	40	UO_2 (0.72)	Heavy Water	Heavy Water
MAGNOX	25	U metal w/ 0.5% Al (0.72)	Graphite	Carbon Dioxide
HFIR (irradiation)	85	UO_2 (0.25)	Light Water	Light Water
MURR (irradiation)	10	UO_2 (0.72)	Light Water, Beryllium, Graphite	Light Water



Comparison of 238-energy-group neutron flux per MEV for the experimental irradiation at HFIR at the beginning and end of irradiation, due to depletion of the gadolinium irradiation capsule

HFIR Irradiated Material Measured Intra-Element Isotope Ratios

Mass Spectroscopy Measured and HFIR Simulated Intra-Element Isotope Ratio Values

Ratio	Measured Value	Measurement Error	Simulated Value	S/E
$^{137}Cs/^{133}Cs$	1.30×10^{0}	6.7%	1.07×10^{0}	0.82
$^{134}Cs/^{137}Cs$	3.74×10^{-3}	4.2%	4.44×10^{-3}	1.19
$^{135}Cs/^{137}Cs$	4.25×10^{-1}	10%	4.58×10^{-1}	1.08
$^{154}Eu/^{153}Eu$	4.67×10^{-2}	4.5%	4.88×10^{-2}	1.05
$^{150}Sm/^{149}Sm$	3.23×10^{0}	2.7%	3.38×10^{0}	1.05
$^{152}Sm/^{149}Sm$	2.93×10^{0}	1.3%	2.66×10^{0}	0.91
$^{240}Pu/^{239}Pu$	8.28×10^{-2}	0.59%	9.49×10^{-2}	1.15
$^{241}Pu/^{239}Pu$	3.30×10^{-2}	0.88%	4.44×10^{-2}	1.35
$^{242}Pu/^{239}Pu$	1.88×10^{-3}	0.88%	2.89×10^{-3}	1.54

MURR Irradiated Material Measured Intra-Element Isotope Ratios

Mass Spectroscopy Measured and MURR Simulated Intra-Element Isotope Ratio Values

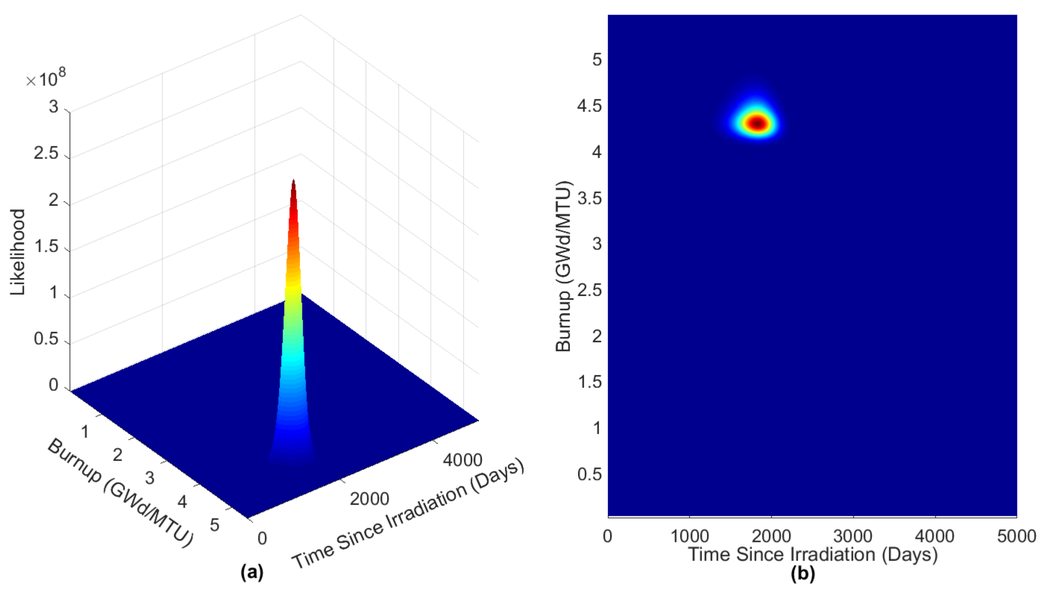
Ratio	Measured Value	Measurement Error	Simulated Value	S/E
$^{137}Cs/^{133}Cs$	9.75×10^{-1}	6.6%	9.48×10^{-1}	0.97
$^{134}Cs/^{137}Cs$	3.84×10^{-3}	7.0%	3.52×10^{-3}	0.92
$^{135}Cs/^{137}Cs$	2.95×10^{-1}	6.8%	2.76×10^{-1}	0.93
$^{150}Sm/^{149}Sm$	9.88×10^{0}	6.7%	9.72×10^{0}	0.98
$^{152}Sm/^{149}Sm$	6.65×10^{0}	5.7%	5.87×10^{0}	0.88
$^{240}Pu/^{239}Pu$	4.77×10^{-2}	5.7%	4.23×10^{-2}	0.89
$^{241}Pu/^{239}Pu$	2.40×10^{-3}	5.8%	2.06×10^{-3}	0.86
$^{242}Pu/^{239}Pu$	5.99×10^{-5}	8.3%	4.93×10^{-5}	0.82

HFIR Irradiated Material – Maximum Likelihood Analysis

Results of the Maximum Likelihood Analysis for the HFIR Irradiated Material [a]

Reactor Model	Log-Likelihood Value [b]	Predicted Burnup (GWd/MTU)	Predicted Time Since Irradiation (days)
HFIR	19.5 ± 4.6	4.29	1827
MURR	-46.6 ± 12.8	4.16	1700
NRX	-52.5 ± 12.5	4.13	1590
MAGNOX	-59.5 ± 13.3	3.00	421
PWR (2.35%)	-86.7 ± 21.0	≥ 5.31	1705
PHWR	-129 ± 32	≥ 4.35	2308
PWR (3.4%)	-284 ± 26	≥ 5.01	0
PWR (4.45%)	$-5.27 \times 10^{3} \pm 1.38 \times 10^{2}$	≥ 3.90	0
FBR	$-6.39 \times 10^{5} \pm 1.05 \times 10^{4}$	≥ 4.73	0

[a] Measured material was HFIR irradiated to a burnup of 4.36 ± 0.28 GWd/MTU with 1601 days decay.
[b] The maximum possible log-likelihood value for the measured material was 28.5.



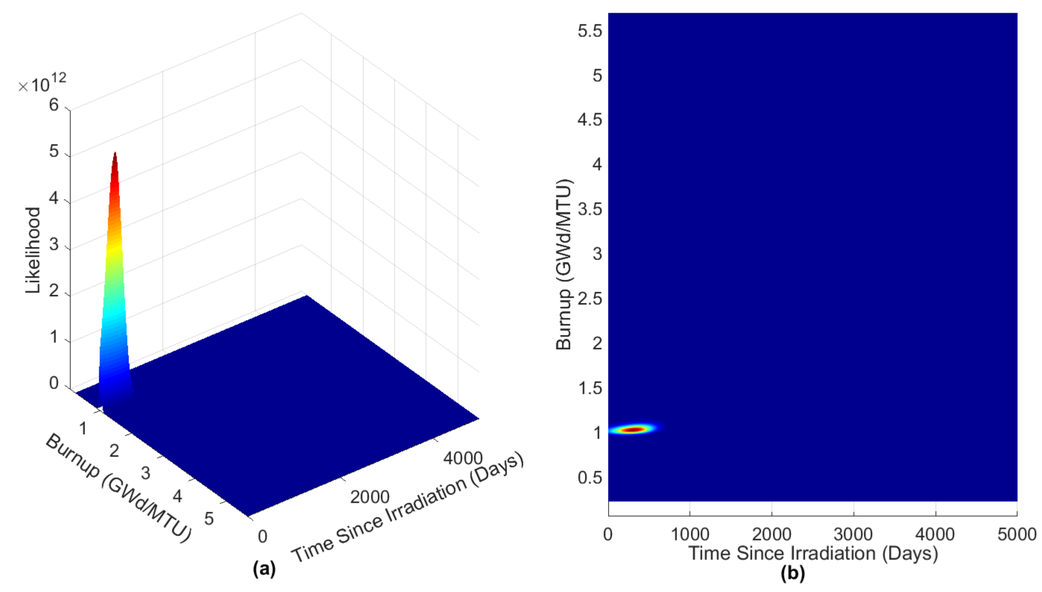
Results of Maximum Likelihood Analysis for the HFIR Irradiated Material (a) 3-D Likelihood Surface Map and (b) 2-D Contour Map for the Most Likely Reactor (HFIR)

MURR Irradiated Material – Maximum Likelihood Analysis

Results of the Maximum Likelihood Analysis for the MURR Irradiated Material [a]

Reactor Model	Log-Likelihood Value [b]	Predicted Burnup (GWd/MTU)	Predicted Time Since Irradiation (days)
MURR	29.5 ± 1.1	1.02	295
NRX	25.3 ± 3.0	1.03	208
MAGNOX	13.2 ± 5.7	0.73	0
PWR (3.4%)	-6.02 ± 8.71	3.91	1381
PWR (4.45%)	-8.88 ± 10.2	≥ 3.90	1196
PWR (2.35%)	-12.7 ± 10.2	3.10	1202
PHWR	-14.7 ± 13.8	1.02	360
HFIR	-166 ± 28	4.40	1790
FBR	$-1.52 \times 10^{5} \pm 2.02 \times 10^{4}$	≥ 4.73	0

[a] Measured material was MURR irradiated to a burnup of 0.973 ± 0.032 GWd/MTU with 318 days decay.
[b] The maximum possible log-likelihood value for the measured material was 29.7.



Results of Maximum Likelihood Analysis for the MURR Irradiated Material (a) 3-D Likelihood Surface Map and (b) 2-D Contour Map for the Most Likely Reactor (MURR)

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