





Radiochemistry Webinars Chronometry





Idaho National Laboratory



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Meet the Presenter...

Dr. Amy Gaffney is a staff scientist in the Nuclear and **Chemical Sciences Division at Lawrence Livermore** National Laboratory. Her research focuses on the use of radiochronometry as a signature of the origin and history of nuclear material. Dr. Gaffney is engaged in collaborations on the development of analytical methods for radiochronometry with several international partners. She also mentors graduate student interns through the Glenn T. Seaborg Institute at LLNL. Through her service at LLNL, she has earned a Department of Energy Secretarial Honor Award and a DOE Office of Science Outstanding Mentor Award. Dr. Gaffney's research is presented in over 30 peer-reviewed scientific publications. Dr. Gaffney received her Bachelor's degree in Geology from The Colorado College, and her M.S. and Ph.D. in Geological Sciences from the University of Washington. She held postdoctoral appointments at the University of New Mexico and LLNL prior to joining LLNL as a staff member in 2009.

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Dr. Amy Gaffney





Radiochronometry for Nuclear Forensics

NAMP Webinar

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National Laboratory

1: now at Los Alamos National Laboratory

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Radiochronometry and signatures

- The model age of nuclear material is a powerful signature for nuclear forensics
- *Comparative signature:* no assumptions are required about sample purification or production history
- *Predictive signature:* model sample history is assumed
 - sample was completely purified from decay products at some time in the past
 - sample has remained a closed system since that time







Uranium fuel cycle

- Uranium processing may purify decay products *or* add contaminants to bulk uranium
- How do decay products behave during uranium production processes?



Uranium-series chronometers



Radioactive decay equations

• Amount of parent present at time = t

$$N_1(t) = N_1(0)e^{-\lambda_1 t}$$

Amount of daughter present at time = t

$$N_{2}(t) = \frac{\lambda_{1}}{(\lambda_{2} - \lambda_{1})} N_{1}(0)(e^{-\lambda_{1}t} - e^{-\lambda_{2}t}) + N_{2}(0)e^{-\lambda_{2}t}$$

•Definitions

- Subscripts: 1 = parent, 2 = daughter
- N = number of atoms
- λ = decay constant
- t = time of interest,

positive value measured from t = 0 in the past

Model age for daughter/parent system

• Model assumption: no daughter present at t=0

$$N_2(t) = \frac{\lambda_1}{(\lambda_2 - \lambda_1)} N_1(t) (1 - e^{(\lambda_1 - \lambda_2)t})$$

• Expression for t, the age of the material



Model age definitions



- Case 1: material completely purified from decay product at time of production, model age = sample age
- Case 2: material incompletely purified at time of production *or* material contaminated since time of production, model age > sample age

Assumptions for model ages

$$t = \frac{1}{\lambda_{234} - \lambda_{230}} \ln \left[1 + \frac{\frac{230}{234} Th}{\frac{234}{234} U} \frac{\lambda_{234} - \lambda_{230}}{\lambda_{234}} \right]$$

- The model age is proportional to the measured daughter/parent isotope ratio
- Contamination or incomplete purification results in sample model age that is older than the sample production age



U-series nuclides used for radiochronometry



Mass spectrometry vs. decay counting

Measurements in practice

Decay-counting methods

High-Resolution HPGE Gamma Spectrometry

Decay-counting methods can be non-destructive or destructive

High-Resolution Solid-State and Gasproportional Alpha Spectrometry

Isotope dilution requires isotope tracers (spikes)

- Purchased from metrology institute (e.g. NIST, IRMM) or calibrated against traceable concentration standard
 - ²³³U, ²²⁹Th, ²³³Pa
- ²²⁹Th calibration example:
 - Calibrated with NIST SRM 4342A ²³⁰Th radioactivity standard
 - Requires ²³⁰Th half-life to calculate ²³⁰Th atoms/g in SRM 4342A

Calibration Uncertainty

- SE (n=6): 0.058%
- NIST 4342A: 0.24%

²³³Pa spike for ²³¹Pa analyses

 ²³³Pa (t_{1/2} = 27 days) is milked from ²³⁷Np and calibrated using geologic standards, assuming secular equilibrium

Eppich *et al.,* J. Anal. At. Spectrom., 28, 666-674, 2013.

Radiochronometry analyses - standards

New Brunswick Laboratory U.S. Department of Energy								EUROPEAN COMMISSION JOINT RESEARCH CENTRE Institute for Reference Materials and Measurements (Geel)					
Certificate of Analysis													
CRM 125-A					CERTIFIED REFERENCE MATERIAL								
Uranium (U Radio	JO ₂) Pelle ochronon	et Assay, Iso netric Stand	otopic, and lard						IRMM-1000a				
Amount Content :	0.88129	g U•g ⁻¹ pellet			CERTIFICATE OF ANALYSIS								
Uncertainty:	0.00014	g U•g ⁻¹ pellet	"(²³⁶ T)/"(²³⁸ T)	····									
Isotope-Amount Ratio: 0.00039130 0.042301 0.0000040754													
Uncertainty:	0.0000038	0.000025	0.000000047						Production date based or	the $a(^{230}\text{Th})/a(^{234}\text{U})$ radiochronometer			
Testers Amount Trestler (100)	n(²³⁴ U)/n(U)	n(²³⁸ U)/n(U)	n(²³⁶ U)/n(U)	n(²³⁸ U)/n(U)					Certified value	Uncertainty ⁴⁾			
Uncertainty:	0.037528	0.0023	0.00039085	0.0023						[day]			
	m(²³⁴ U)/m(U)	m(²³⁵ U)/m(U)	m(²³⁶ U)/m(U)	m(²³⁸ U)/m(U)				Production date	e ¹⁾ 09/07/2012 (UTC) ²⁾	13			
Isotope Mass Fraction (•100):	0.036915	4.0077	0.00038776	95.9550				1) The production date is	2012-07-09Z ³⁷ s the date of the last chemical separation of ²³⁰	Th from ²³⁴ U in the uranium reference material.			
Uncertainty: Molar Mass: 237 927291	0.000036	0.0023 Model Purificati	0.00000045	0.0023 August 18, 1994				 The certified value is 	expressed as a date dd/mm/www_relative to th	e universal coordinated time (UTC)			
Uncertainty: 0.000071	g•mol ⁻¹	Unc	ertainty:	116 days					is expressed as vvvv-mm-ddZ according to I	SO 8601.			
New . U.S. De				New Bri U.S. Departu	Brunswick Laboratory partment of Energy				expanded uncertainty with a coverage factor $k = 2$ corresponding to a level of confidence of in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in 5), ISO, 2008. It is based on the confirmation measurements and calculations using the raceable to the International System of units (SI).				
			С	Certificate of Analysis									
					CRM	1 U630							
			Uran	ium (U ₃ O ₈) Iso	otopic and	l Radiochroi	nometric S	Standard					
					n(²³⁴ U)/n(²³⁵ U	$n(^{236}U)/n(^{235}U)$	n(²³⁸ U)/n(²³⁵ U))	Radioch	ronometrv			
			I	sotope-Amount Ratio:	0.0097698	0.0151895	0.55351			i en en en y			
				Uncertainty:	0.0000062	0.0000097	0.00049		standards	are required			
					n(²³⁴ U)/n(U)	n(²³⁵ U)/n(U)	n(²³⁶ U)/n(U)	n(²³⁸ U)/n(U)		are required			
			Isotope-Am	nount Fraction (•100):	0.61894	63.353	0.96230	35.066	formatha	d validation			
				Uncertainty:	0.00043	0.020	0.00067	0.020	ior metho	validation			
					m(²³⁴ U)/m(U)	$m(^{235}U)/m(U)$	m(²³⁶ U)/m(U)	m(²³⁸ U)/m(U)					
			Isotope	Mass Fraction (•100):	0.61354	63.069	0.96207	35.356					
			Mola	r Mass: 236,10175	0.00043	0.020 Model Purificatio	on Date:	June 6, 1989					
					_								

Uncertainty:

190 days

0.00061 g•mol⁻¹

Uncertainty:

Sample preparation and analysis methods

- Mass spectrometry is a destructive analytical method
 - element of interest is purified from bulk sample
 - eliminate isobaric interferences, matrix effects
- Sample is dissolved
- Purification utilizes ion exchange, selective extraction and other methods

Mass spectrometers

- High-resolution single-collector ICP-MS, e.g., Thermo Scientific Element
- **Multi-collector ICP-MS**, e.g., Nu Plasma, Thermo Scientific Neptune
- Thermal ionization mass spectrometer (TIMS), e.g., Thermo Scientific Triton, Isotopix Phoenix

Mass spectrometry

- Ion Source
 - Plasma ionization
 - Thermal ionization
- Magnet
- Focusing, energy filters
- Collectors
 - Faraday cups
 - Ion counters

Example: ²³⁰Th-²³⁴U model ages of NBL CRMs

- Most CRMs produced in late-1950's to early-1960's
- U030A and U005A produced in 1981
- In general, model ages are slightly biased old
 - Duplicate analyses are in agreement
- Interpreted to represent incomplete purification of ²³⁰Th during material production
- Bias greater in younger CRMs
 - Are more recently produced CRM's less pure?

Williams and Gaffney (2011) Proc. Radiochim. Acta 1: 31-35.

Concordant and discordant radiochronometers

- Concordant model ages from two or more radiochronometers provides a high degree of confidence that the model age represents the actual purification age of the sample.
- Discordant model ages provide information on the relative fractionation between daughter isotopes and parent during sample production.
 - The degree of purification, or contamination, of different daughter isotopes may help to identify the process.
- Regardless of concordance or discordance, if the system remains closed, the model dates are constant, high-value signatures.

Example: ITWG-RR3 (2010)

- Exercise scenario two pieces of metal seized separately at a border crossing
 - are the two samples related, and if so, how?
- Samples are pieces of two HEU 'storage logs'
 - Logs consist of scrap uranium, unknown age
 - Casting dates known: May 2003, January 2004

Example: ITWG-RR3

- ²³⁰Th-²³⁴U model dates are close to known casting dates
 - Th is effectively purified during U metal casting
- ²³¹Pa-²³⁵U model dates are 1974-1976
 - Pa is not purified from U during metal casting
- Two samples are *not* from the same batch of material
- Metal casting has different effects on Th/U and Pa/U

²³⁰Th-²³⁴U model dates

Th migrates to 'hot top'

Example: ITWG-RR3

•

- Grand-daughter chronometry reveals additional information on behavior of Ra and Ac during U metal casting
- ²²⁶Ra-²³⁸U model ages < 2 years older than known casting date
 Ra not as efficiently purified from U as Th during metal casting
 - ²²⁷Ac-²³⁵U_{corr} model ages nearly concordant with casting date: Ac
 - supported by ²³¹Pa since casting

Efficient Ac segregation during metal casting

Example: ITWG-CMX4 (2014)

- Scenario
 - Passenger on Dallas-to-Frankfurt flight found to possess uranium powder
 - Search of passenger's home reveals pellet
 - Similar pellet found previously in Frankfurt warehouse
- Do the materials share an origin?
 - facility
 - process
 - batch

Example: ITWG-CMX4

- ²³⁰Th-²³⁴U and ²³¹Pa-²³⁵U samples are concordant for each sample
- ES2 model ages are concordant with enrichment date, not pellet production date
- ES1 and ES3 are similar in both chronometers, likely from same batch; consistent with known production in 2004

Example: historical plutonium from Hanford

- In 2004, jug containing Pu uncovered in waste trench at Hanford
- 'Low-burn' Pu could represent early US production
- Material has unknown history evaluate model age assumptions

Schwantes (2009) Anal. Chem. 81:1297-1306

Plutonium chronometry: daughter-parent

- ²³⁸Pu \rightarrow ²³⁴U
- ²³⁹Pu \rightarrow ²³⁵U
- ²⁴⁰Pu \rightarrow ²³⁶U
- ²⁴¹Pu \rightarrow ²⁴¹Am

Am232 79 s	Am233 3 m	Am234 2.3 m	Am235 5/- 10. m	(1) Am236 (5) 2.9 m 3.6 m	Am237 ^{5/(-)} 1.22 h	Am238 1+	Am239 (5/)- 11.9 h	Am240 (3) 2.12 d	Δm	⁵⁻ Am242 1- 141 a 16.02 h
ε α (SF) ? ω	ε α 6.78	ε α 6.46 ?	ε γ 290.6,… α 6.46	ε ε ε 319.5, γ 582.8, γ 319.5, α 6.2 ω α 6.4 ω	γ 280.2,… α 6.04 ω	β ⁺ ω γ 962.8, 918.7, 561.0,	^ε γ 277.6, 228.2, […] α 5.774 (ω), 5.734, […] γ 49.3 ω	γ 987.7, 888.8,… α 5.378 νω,…	AIII-	α 5.207 ω, β = 0.63, 0.67 γ 49.2 ω, γ 42.2 e SF vω ε, γ 44.5 e σγ ~17E2, σ _f 21E2
E 5.0	E 3.1 233.0463	E 4.2	E 2.5	E 3.3	E 1.5	E 2.3	E 0.802	E 1.38	241	70E2, E 0.6645 18E2 E ⁺ 0.751
Pu231 8.6 m	Pu232 34 m	Pu233 20.9 m	Pu234 8.8 h	Pu235 (5/+) 25.3 m	Pu236 2.87 a	Pu237 7/- 0.18 s 45.64 d	Pu-	Рн-	Pu-	PII-
ε α 6.72	ε α 6.60, 6.54	ε γ 235.3, 534.7,… α 6.30 ω	ά 6.200, 6.149,…	γ 49.2, 756.4,… α 5.85 ω	γ 47.6 – 643.7 ω SF ννω	x 280.4(vω), 289.9,			10	
E 2.7	E 1.0	5.0.4	5.0.20	E 4 44	σ _f 16E1, 1E3	320.8,··· σ _f 24E2 Ε 0.220	238	239	240	241
231.04110 Np230	Np231 (5/)	Np232 (4+)	Np233 (5/+)	Np234 (0+)	Np235 5/+	1(-) Np236 (%)	Np237	Np238	Np239 5/+	¹⁽⁺⁾ Np240 (5+)
4.6 m ε	48.8 m	14.7 m ε	36.2 m ε	4.4 d ε	1.085 a ε	22.5 h 1.57.5 a ε, β ε, 160.3,	2.14E6 α 4.788, 4 7 1,…	2.103 d β 0.263, 1,148,···	2.356 d β ⁺ 0.438, 0.341,···	7.22 m 1.032 h β 2.18, β 0.89
α 6.66	γ 370.3, 347.5, 262.9,… α 6.26	γ 326.8, 819.2, 866.8, 864.3,…	γ 312.0, 298.9, 546.5,… α 5.53 ? ω	β ⁺ 0.79 ω γ 1558.7, 1527.2, 1602.2,…	α 5.021 (νω), 5.004,… γ 25.6 – 188.8 νω	0.54, γ 44.6 γ 642.3 e 687.7 o ₁ 2.5E3, σ, 2.7 1E3	γ 29.4, 8 σ _γ 169, 5551 σ _f 9, 2, 7	γ 984.5, 728.5, σ _γ 48Ε σ _f 27.2, 9E2	γ 106.1, 277.6, 228.2,… σ _γ (3E1 + 3E1)	1.60, γ 554.6, 597.4, 600. 6 ,
E 3.6	E 1.8	E 2.8	E 1.0	o _f 9∈2 E 1.81	^σ γ (15E1 + ?) E 0.124	E+ 0.9 E- 0.5	237.048173	E 1.2915		IT E 2.19
U229 (3/+)	U230	U231 (5/-)	U232	U233 5/+	<u> </u>			U237 1/+	U238	U239 5/+
58 m	20.8 d α 5.888, 5.818,…	4.2 d	69.8 a α 5.3203, 5.2635,…	1.592E5 a α 4.824, 4.783,…	U- 🗖	U-	U- 🗖	6.752 d β ⁻ 0.24, 0.25,···	UI 99.2742 4.468E9 a	23.47 m β 1.21, 1.28,····
γ 122.5,… α 6.360, 6.332,	γ 72.2 ω (e), 154.2, 230.4,…		γ 57.8 ω(e),129.1,··· SF ω σ. 73, 28F1	97 42.5 00, 97 03, 54.65, SF νω σ _V 46, 14E1	224	0.05	226	γ 59.5, 208.0,··· σ _γ 4E2, 12E2	γ 49.6 φ (e), SF vo	σ _γ 22
6.297,… E 1.31			σ _f 75, 38E1	$\sigma_f 531, 76E1$ $\sigma_\alpha < 0.3 \text{ mb}$	234	235	236	E 0.540		
229.03351 Pa228 (3+)	230.03394 Pa229 (5/+)	Pa230 2 ⁻	Pa231 3/-	Pa232 (2-)	Pa233 3/-	(0-) Pa234 4+	Pa235 (3/-)	Pa236 1-	Pa237 (1/+)	Pa238 3-
22 h	1.5 d	17.4 d	Pa 100	1.32 d	26.967 d	UX2 UZ	24.4 m	9.1 m	8.7 m	2.3 m
ε γ 911.20, 463.02,	ε γ 42.4 e	ε, γ 952.0,… β 0.51,…	3.28E4 a		β 0.256, 0.15,··· γ 311.90,···	β ⁻ 2.29, γ 1001.0, β ⁻ 0.48, 0.65	γ 30.1 – 658.9	γ 642.3, 687.5,	2.25,	γ 1015, 635,
968.98,… α 5.71 – 6.12 ω	α 5.41 – 5.693 ω γ 24.8 – 180.2 ω									1.10.000
γ 95 – 345 ω					α _γ (21 + 38), (46E1 + 44E1)	766.4, ω IT < 10 γ 73.9 ω e 34 - 1938		1762.7,… SF νω	γ 853.6, 865.0, 529.3, 540.7,…	448, 680.,…
E 2.152	E 0.311	σ _f 15E2 E ⁺ 1.311 E ⁻ 0.560	^γ 27.4, 300.1, ⁴ σ _γ 20E1, 5E2 σ _f 0.020, 0.05 231.035884	σ _γ 5E2, 3E2 of 7E2 E ⁺ 0.50 E ⁻ 1.34	σ _γ (21 + 38), (46E1 + 44E1) σ _f < 0.1 E 0.570	766.4, ··· ω IT < 10 γ 73.9 ω e ⁻ 7131.3, 34 - 1938 E 2.195	E 1.41	1762.7,… SF νω E 2.9	γ 853.6, 865.0, 529.3, 540.7,… E 2.3	448, 680.,··· E 3.5
E 2.152 Th227 (3/+)	E 0.311 Th228	α 4.766 - 5.345 va σ _f 15E2 E+ 1.311 E- 0.560 Th229 5/+	7 201.52 37 2021.52 231.035884 Th230	⁶ γ 5E2, 3E2 ⁶ γ 7E2 <u>E⁺ 0.50</u> E ⁻ 1.34 Th231 5/(+)	$\sigma_{\gamma} (21 + 38),$ (46E1 + 44E1) $\sigma_{f} < 0.1$ E 0.570 Th232	766.4,ω IT < 10 γ 73.9 ω e ⁻ Th233 (1/+) 24 93 - 5 Th233 (1/+)	E 1.41 Th234	1762.7, SF νω E 2.9 Th235 1/+	γ 853.6, 865.0, 529.3, 540.7,… E 2.3 Th236	448, 680., E 3.5 Th237
E 2.152 Th227 (3/+) RdAc 18.68 d α 6.038, 5.978,	E 0.311 Th228 RdTh 1.912 a α 5.423, 5.340, 84.4 cc - 216.0	$\begin{array}{c} a 4.766 - 5.345 \ \text{W}}{_{\text{Gf}} 15E2} \\ \hline e^+ 1.311 \ e^- 0.560 \\ \hline \textbf{Th229} \ 5/^+ \\ \hline 7.4E3 \ a \\ \alpha \ 4.845, \ 4.901, \end{array}$	⁷ 21:4, 300.1 ^α ₇ 20:1:5:2: ^α ₇ 0.020, 0.05 231.035884 Th230 0 7.56E4 a α 4.688, 4.621	$\begin{array}{c} \sigma_{\gamma} \ 5E2, \ 3E2 \\ \sigma_{f} \ 7E2 \\ E^{\pm} \ 0.50 \\ \end{array} \xrightarrow{E^{\pm} \ 0.50} E^{\pm} \ 1.34 \\ \hline \begin{array}{c} \textbf{Th231} \ 5/(+) \\ \textbf{UY} \\ \textbf{1.063 d} \\ \beta^{\pm} \ 0.305, \cdots \end{array}$	$\begin{array}{c} \sigma_{f}(21+36),\\ (46E1+44E1)\\ \sigma_{f}<0.1\\ E \ 0.570\\ \hline \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ $	$\begin{array}{c} 766.4, \cdots \\ T < 10 \\ 773.9 \\ \infty e^{-} \end{array} \begin{array}{c} \gamma 131.3, \\ 34 - 1938 \\ E 2.195 \\ \hline \textbf{Th233}(1/+) \\ \textbf{21.83 m} \\ \beta^{-} 1.245, \cdots \end{array}$	E 1.41 Th234 UX1 24.10 d β ^{= 0.198}	1762.7, SF νω E 2.9 Th235 1/+ 7.2 m	γ 853.6, 865.0, 529.3, 540.7, Ε 2.3 Th236 37.5 m	448, 680., E 3.5 Th237 4.8 m
E 2.152 Th227 (3/+) RdAc 18.68 d α 6.038, 5.978, 5.757,… γ 236.0, 50.2,…	E 0.311 Th228 RdTh 1.912 a 5.423, 5.340, γ 84.4 e ⁻ , 216.0, 131.6, 166.4, σ ₁ 1.2E2, 1.0E3	$\begin{array}{c} a 4.765 - 5.345 \text{Ve} \\ \sigma_f 15E2 \\ \hline E^+ 1.311 \ E^- 0.560 \\ \hline \textbf{Th229} 5/+ \\ \hline \textbf{7.4E3 a} \\ a 4.845, 4.901, \\ 4.814, \cdots \\ \textbf{7} 193.6, 86.4, \\ 210.9, 31.5, \cdots \end{array}$	¹ 2.2.4,300,1.4 ¹ 0,2021,562 ¹ 0,020,0.05 231.035884 Th230 Io 7,56E4 a α 4.688,4.621, y 67.7 e ⁻ , 110.0−620. α -220.101E1	$\begin{array}{c} {}_{\gamma} 522, 322 \\ {}_{\gamma} 522 \\ {}_{\tau} 722 \\ {}_{\tau} 52 \\ {}_{\tau} 50 \\ {}_{\tau} 50 \\ {}_{\tau} 51 \\ {}_$	$\sigma_{f} < (21 + 38),$ (46E1 + 44E1) $\sigma_{f} < 0.1$ E 0.570 Th 100 1.40E10 a $\alpha + 012, 3.947,$ $\tau_{f} = 63.81 \oplus (\sigma^{-}), 140.88$	$\begin{array}{c} 7664, \cdots \omega \\ 17 < 10 \\ \gamma 73.9 \ \omega \ e^{-} \end{array} \\ \begin{array}{c} \gamma 131.3, \\ 34 - 1938 \\ E \ 2.195 \\ \hline \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \$	E 1.41 Th234 UX1 24.10 d β ^{-0.198,} γ 63.3, 92.4, 92.8,	$\begin{array}{c} 1762.7,\cdots\\ \text{SF }\nu\omega\\ \hline \textbf{E} \ 2.9\\ \hline \textbf{Th235} \ 1/+\\ \textbf{7.2 }m\\ \beta^{-}\\ \gamma \ 417.0, \ 727.2,\\ 696.1, \ 644.9,\cdots \end{array}$	$\begin{array}{c} \gamma \text{ 853.6, 865.0,} \\ 529.3, 540.7, \cdots \\ \hline \text{E 2.3} \\ \hline \textbf{Th236} \\ \textbf{37.5 m} \\ \gamma \text{ 110.8, } \cdots \end{array}$	448, 680., <u>E 3.5</u> Th237 4.8 m β ⁻
E 2.152 Th227 (3/+) RdAc 18.68 d α 6.038, 5.978, 5.757, γ 236.0, 50.2, σ _f 20E1	E 0.311 Th228 RdTh 1.912 a α 5.423, 5.340, γ 84.4 er. 216.0, 131.6, 166.4, σ _γ 1.222, 1.0E3 σ _γ < 0.3	a 4.766 - 5.345 Ve of 15E2 E ⁺ 1.311 E ⁻ 0.560 Th229 5/+ 7.4E3 a a 4.845, 4.901, 4.814, 2109, 315, a ₇ 36.564, 2109, 315, a ₇ 31, 5E2	μ μ <thμ< th=""> μ μ</thμ<>	$\begin{array}{c} c_{\gamma} 5E2, 3E2\\ c_{f} 7E2\\ E^{+} 0.50 \ E^{-} 1.34\\ \hline \mbox{Th231} 5/(^{+})\\ \mbox{UY} \ 1.063 \ d\\ \beta^{-} 0.305, \cdots\\ \gamma \ 25.64, 84.21, \cdots \end{array}$	$\begin{array}{c} \sigma_{T} \ (21+30), \\ (46E1+44E1) \\ \sigma_{f} < 0.1 \\ \hline E \ 0.570 \\ \hline \textbf{L} \ 0.570 \\ \hline \textbf{L}$	$\begin{array}{c} 766.4\\ \pi < 10\\ \gamma 73.9\\ r < 10\\ \gamma 73.9\\ r < 12, 21, 23\\ r < 10, 21, 21, 21, 21, 21, 21, 21, 21, 21, 21$	$\begin{array}{c} {\sf E} \ 1.41 \\ \hline {\sf Th} 234 \\ {\sf UX1} \ 24.10 \ d \\ {\sf F}^- 0.198, \cdots \\ {\sf f}^{23,3,92.4,9} \\ {\sf 92.8, \cdots } \\ {\sf 92.8, \cdots } \\ {\sf 97.2} \\ {\sf \sigma}_f < 0.01 \end{array}$	$\begin{array}{c} 1762.7,\cdots\\ \text{SF }_{V\omega}\\ \hline E 2.9\\ \hline \textbf{Th235} 1/+\\ 7.2 \text{ m}\\ \beta^{-}\\ \gamma 417.0, 727.2,\\ 696.1, 644.9,\cdots\\ \hline \end{array}$	γ 853.6, 865.0, 529.3, 540.7, <u>E</u> 2.3 Th236 37.5 m β ⁻ γ 110.8,	448, 680., <u>E 3.5</u> Th237 4.8 m β ⁻
E 2.152 Th227 (3/+) RdAc 18.68 d a 6.038, 5.978, 5.757, γ 236.0, 50.2, σ _f 20E1 227.027704	E 0.311 Th228 RdTh 1.912 a a 5.423, 5.340, y 84.4 er., 2160, 1316, 166.4, ar 5.423, 28.028741 228.028741 A 2272 3=	a 4,760 - 5.345 vo qr 15E2 E+1.311 E- 0.560 Th229 5/+ 7.453 a 4.845, 4901, 4.814 7.193,6.864, 2109,315,5 qr 6E1,10E3 qr 31,5E2 229.031762	- 2001 - 2000 - 0.00 - 1000 - 0.00 - 0.00 - 2000 - 0.00 - 0.00 - 2000 - 0.00 - 0.00 - 2000 - 0.00 - 0.00 - 2000 - 0.00 - 0.00	C 552, 352 ⁶ / ₁ / ₇₆₂ E ⁺ 0.50 E ⁻ 1.34 Th231 5 ^(/+) U 1.063 d β ⁻ 0.305, x 25,64,84,21, E 0.392 Δ 0.220 (141)	$\begin{array}{c} e_{\gamma}\left(2 +30 ,\\ (46E1+44E1)\\ e_{1}(46E1+44E1)\\ e_{1}(570)\\ \hline \\ \hline \\ 1.40E10 a\\ a.4.012, 3947\\ r.6381 a (e^{-1}, 140.88\\ F(vos)\\ e_{\gamma}(7.34, 85, 9(^{-1})\mu)\\ 232.038055\\ \hline \\ \hline \\ \hline \\ 0.2234 (414) \end{array}$	$\begin{array}{c} \hline 766.4, \ast & \omega \\ \pi < 10 \\ \gamma 73.9, \omega < \sigma \\ \hline 2, 10 \\ \gamma 73.9, \omega < \sigma \\ \hline 2, 10 \\ \gamma 73.9, \omega < \sigma \\ \hline 2, 10 \\ \gamma 73.9, \omega < \sigma \\ \hline 2, 10 \\ \gamma 73.9, \omega < \sigma \\ \hline 2, 10 \\ \gamma 73.9, \omega < \sigma \\ \hline 2, 10 \\ \gamma 73.9, \omega < \sigma \\ \gamma 85.5, 29.4, \\ 459.3, \cdots \\ 459.3, \cdots \\ \gamma 75E.2, 4E2 \\ \sigma \\ \gamma 15E.2, 4E2 \\ \sigma $	$\begin{array}{c} E \ 1.41 \\ \hline \ \ \ \ \ \ \ \ \ \ \ \ \$	1762.7,··· SF vo E 2.9 Th235 1/+ 7.2 m β ⁻ 417.0, 727.2, 696.1, 644.9,··· E 1.9 Δ c 234 (1±)	γ 853.6, 865.0, 529.3, 540.7, E 2.3 Th236 37.5 m β ⁻ γ 110.8, E	448, 680, <u>E 3.5</u> Th237 4.8 m β ⁻ <u>E 2.6</u>
E 2.152 Th227 (3/*) RdAc 18.68, 5.978, 5.757, γ 236.0, 50.2, rg 20E1 227.027704 Ac226 (1) 1.224 d	E 0.311 Th228 RdTh 1.912 a a 5.423, 5.340 13.16, 166.4 of 1.222, 1.063 of 2.0.3 228.028741 Ac227 3- Ac 21.772 a	4 4/760 - 5.343 τ00 4 1/562 E+ 1.311 E- 0.560 Th229 5/+ 7.4E3 a α 4.845, 4.901, 4.814- γ 1936, 88.4, γ 109, 31.5 α γ 6E1, 1.063 γ 229,031762 229,031762 Ac228 3(+) MSTh2 6.15 h	$\begin{array}{c} \begin{array}{c} 1 & 2.002 \\ r_{1}^{2} & 0.002 \\ r_{2}^{2} & 0.008 \\ \hline \end{array} \\ \hline \end{array} \\ \begin{array}{c} \begin{array}{c} 1 \\ r_{1}^{2} & 0.05884 \\ \hline \end{array} \\ \hline \end{array} \\ \hline \end{array} \\ \begin{array}{c} \hline \end{array} \\ \begin{array}{c} 1 \\ r_{1}^{2} & 0.035884 \\ \hline \end{array} \\ \hline \end{array} \\ \begin{array}{c} \hline \end{array} \\ \hline \end{array} \\ \begin{array}{c} 1 \\ r_{1}^{2} & 0.57864 \\ r_{2}^{2} & 0.037134 \\ \hline \end{array} \\ \hline \end{array} \\ \begin{array}{c} r_{1}^{2} & r_{2}^{2} & r_{1}^{2} & r_{2}^{2} \\ r_{1}^{2} & r_{2}^{2} & r_{2}^{2} & r_{2}^{2} \\ \hline \end{array} \\ \begin{array}{c} r_{1}^{2} & r_{2}^{2} & r_{2}^{2} & r_{2}^{2} \\ r_{1}^{2} & r_{2}^{2} & r_{2}^{2} \\ \hline \end{array} \\ \begin{array}{c} r_{1}^{2} & r_{1}^{2} & r_{2}^{2} \\ r_{1}^{2} & r_{1}^{2} & r_{1}^{2} \\ \hline \end{array} \\ \begin{array}{c} r_{1}^{2} & r_{1}^{2} & r_{1}^{2} \\ r_{1}^{2} \\ r_{1}^{2} & r_{1}^{2} \\ r_{1}^{2} \\ r_{1}^{2} & r_{1}^{2} \\ r_{1}^{2} & r_{1}^{2} \\ r_{1}^{2} \\ r_{1}^{2} & r_{1}^{2} \\ $	C 5 552, 352 ⁴ /752 E ⁺ 0.50 E ⁻ 1.34 Th231 5 ⁽⁽⁺⁾ UY 1.063 d β ⁻ 0.305, ⁴ /25.64, 84,21, E 0.392 Ac230 (1 ⁺) 2.03 m	$\begin{array}{c} \alpha_{\gamma}\left(21+30\right),\\ (46E1+44E1)\\ \alpha_{f}<0.1\\ E\ 0.570\\ \hline \\ \hline \\ 1.40E10\ a\\ \alpha_{4}.012, 3947,\cdots\\ \gamma_{6}.6341\ a\ (6^{-1}, 140.88\\ 5F\ (vos)\\ \alpha_{7}.734, 85\ \alpha_{f}<1\ \mu b\\ 232.038055\\ \hline \\ \hline \\ Ac231\ (1/^{+})\\ 7.5\ m \end{array}$	$\begin{array}{c} \hline 766.4.*.}{T<10} \gamma 131.3.\\ T<10} \gamma 73.9.o e^{-1}\\ 21.9.0 \gamma 73.9.o e^{-1}\\ 21.83 m e^{-1}\\ 21.95 e^$	$\begin{array}{c} E \ 1.41 \\ \hline \ \ \ \ \ \ \ \ \ \ \ \ \$	1762.7, SF voi E 2.9 Th235 1/+ 7.2 m β ⁻ 417.0, 727.2, 696.1, 644.9, E 1.9 Ac234 (1+) 40 s	γ 853.6, 865.0, 529.3, 540.7, E 2.3 Th236 37.5 m 37.5 m β ⁻ γ 110.8, E Ac235 110.8	448, 680, <u>E 3.5</u> Th237 4.8 m β ⁻ <u>E 2.6</u>
$\begin{array}{c} \textbf{E} 2.152 \\ \textbf{Th227} (3/+) \\ \textbf{RdAc} 18.68 d \\ a 6.038, 5978, \\ 5.757, \\ \gamma 236.0, 50.2, \\ \sigma_{\gamma} 20E1 \\ \hline 227.027704 \\ \textbf{Ac226} (1) \\ \textbf{1.224} d \\ \beta^{\circ} 0.89, 1.11, \\ \gamma 230.3, 181, \\ \dots \\ \gamma 30.3, 181, \\ \dots \\ \gamma 3$	E 0.311 Th228 RdTh 1.912 a a 5423, 5340 137 a fcto, y 1222, 1.023 of 2.03 228.028741 Ac227 3/- Ac 21.772 a β ^{-0.045} 152 m(a-1)	a + 766 - 5.345 vol q-15E2 E+ 1.311 E - 0.5600 Th229 5/+ - 7,4E3 a a 4.845, 4.901, 4.814, - γ 1936, 86.4, 2109, 315, - - γ, 051, 152, 229, 031762 229, 031762 - - Acc283 3(+) - - MSTh2 6.15 h - - 9, 11, 158, 1.731, 9, 112,04, 966, 97. - -	1, 5061 362 γ 0 020.0.6 231.035884 Th230 0 7.5664 a α 4.688,4 621 γ 67.7 er, 110.0-620. αγ -230.10161 γ < 0.5 mb 230.033134 Ac229 (3/+) 1.04 h β ⁻¹ .17 γ 165.254.	$\begin{array}{c} c_{-5} 5 \epsilon_{2} 3 \epsilon_{2} \\ c_{1} 7 \epsilon_{2} \\ c_{1} 7 \epsilon_{2} \\ c_{2} c_{3} \\ c_{1} c_{2} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{2} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{2} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{2} c_{3} c_{3} \\ c_{1} c_{1} c_{3} c_{3} \\ c_{1} c_{1} c_{1} c_{3} \\ c_{1} c_{2} c_{3} \\ c_{1} c_{1} c_{3} \\ c_{1} c_{2} c_{3} \\ c_{1} c_{1} c_{2} c_{3} \\ c_{1} c_{1} c_{2} c_{3} \\ c_{2} c_{3} c_{3} \\ c_{1} c_{2} c_{3} c_{3} \\ c_{1} c_{1} c_{3} c_{3} \\ c_{1} c_{1} c_{1} c_{1} c_{1} \\ c_{1} c_{1} $	$\begin{array}{c} a_{\gamma}(2i+3b),\\ (46E1+44E1)\\ c = 0.1\\ c = 0.570\\ \hline 1.00\\ 1.40E10\\ a = 4.012, 3.947, \cdots\\ E.331\ a (cm), 140.88\\ 5f\ (vos)\\ c = 0, 7.34, 8c\ q < 1\mu b\\ 232,038055\\ \hline Ac231\ (1/^+)\\ 7.5\ m\\ \beta^{-1}, 6, \cdots\\ \gamma \ 282, 5, 307, 0, \end{array}$	$\begin{array}{c} 766.4, \ast & \omega_{\gamma} \gamma 31.3, \\ \pi < 10 \ end{tabular} \\ \gamma 73.9 \ end{tabular} \\ 12.10 \ e$	$\begin{array}{c} {\sf E} 1.41 \\ \hline {\sf Th234} \\ {\sf UX1} & 24.10 \ d\\ {\sf g} - 0.196, \\ {\sf g} 2.8, $	$\begin{array}{c} 1762.7,\cdots\\ \text{SF vol}\\ \hline \textbf{E 2.9}\\ \hline \textbf{Th235 1/+}\\ 7,2m\\ \gamma 417.0,727.2,\\ 696.1,644.9,\cdots\\ \hline \textbf{E 1.9}\\ \hline \textbf{Ac234 (1+)}\\ \textbf{40s}\\ \gamma 1847,1912, \end{array}$	γ 853.6, 865.0, 529.3, 540.7, E 2.3 Th236 37.5 m β ⁻ γ 110.8, E 1.1 Ac235	448, 680, <u>E 3.5</u> Th237 4.8 m β ⁻ <u>E 2.6</u>
E 2.152 Th227 (3/+) RdAc 18.68 d a.6.038, 5978, 5.757 γ 236.0, 50.2 rg 20E1 227.027704 Ac226 (1) 1.224 d β ⁻ 0.89, 1.11, ε, γ253.7, 186.0 a.540.0 5.41.0 c.541.		a + /60 - 5.345 void q + 15E2 E + 1.311 E - 0.560 Th229 5/+ 7, 4E3 a a 4.845, 4.901, 4.814, 4.814, 2109, 315, q 05E1, 1.0E3 9, 315, q 05E1, 1.0E3 q, 31, 56, 229,031762 AC228 3(+) MST12 6.15 h β ⁻¹ , 1.158, 1.731, 9, 46, 97, 9, 12, 04, 966, 97, 338, 320, 4, 27, vo ?	$\begin{array}{c} & 0.021, 0.022, 0.03\\ \hline & 0.022, 0.03, 0.03, 0.04\\ \hline & 0.022, 0.03, 0.03, 0.04\\ \hline & 0.02, 0.03, 0.04\\ \hline & 0.04, 0.04, 0.04, 0.04\\ \hline & 0.04, 0.04, 0.04, 0.04\\ \hline & 0.04, 0$	$\begin{array}{c} c_{-9} 5 c_{2} 3 c_{2} \\ c_{1} 7 c_{2} \\ c_{1} 7 c_{2} \\ c_{1} c_{2} \\ c_{1} c_{2} \\ c_{1} c_{2} \\ c_{2} c_{3} c_{4} \\ c_{1} c_{3} c_{3} \\ c_{1} c_{2} c_{3} c_{4} \\ c_{1} c_{2} c_{3} c_{3} \\ c_{1} c_{2} c_{3} c_{3} \\ c_{2} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{2} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{1} c_{3} c_{3} \\ c_{1} c_{1} c_{1} c_{3} c_{3} \\ c_{1} c_{1} c_{1} c_{1} c_{3} \\ c_{1} c_{2} c_{3} c_{1} \\ c_{1} c_{1} c_{2} c_{3} c_{3} \\ c_{1} c_{1} c_{1} c_{1} c_{1} c_{1} \\ c_{1} c_{1} $	$\begin{array}{c} \alpha_{\gamma}(2i+30),\\ (46E1+44E1)\\ \alpha_{\gamma}<0.1\\ E\ 0.570\\ \hline 1.40E10\ a\\ a\ 4.012\ 3.987,\cdots\\ \hline 1.40E10\ a\\ s\ 4.012\ 3.987,\cdots\\ \hline 1.40E10\ a\\ s\ 7.34,\ 85\ \sigma_{\gamma}<1,\mu\\ 232.038055\\ \hline Ac231\ (1/^{+})\\ 7.5\ m\\ \beta^{+},6,\cdots\\ \gamma\ 282.5\ 307.0,\\ 221.4,\ 185.7,\\ 368.9,\cdots\\ \end{array}$	$\begin{array}{c} \hline 766.4, \cdots \\ \eta & 73.9 \\ \eta & 73.2 \\ \eta & 73.2 \\ \eta & 74.2 \\ \eta $	$\begin{array}{c} E \ 1.41 \\ \hline \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \$	$\begin{array}{c} 1762.7, \cdots \\ \text{SF vol} \\ \hline \\ \textbf{E} 2.9 \\ \hline \\ \textbf{T}.2 \textbf{ m} \\ \textbf{T}.2 \textbf{ m} \\ \beta^{\top} \\ \textbf{Y} 417.0, 727.2, \\ 696.1, 644.9, \cdots \\ \hline \\ \textbf{E} 1.9 \\ \hline \\ \textbf{Ac234} (1^+) \\ \textbf{\phi}^{-} \\ \textbf{M} \textbf{s} \\ \textbf{Y} 1847, 1912, \\ 668, 1954, \cdots \\ \hline \end{array}$	γ 853.6, 865.0, 529.3, 540.7, <u>E 2.3</u> Th236 37.5 m β ⁻ 110.8, <u>E 1.1</u> Ac235	448, 680, <u>E 3.5</u> Th237 4.8 m β ⁻ <u>E 2.6</u>

Example: U-Pu radiochronometry

- Three U-Pu daughter-parent pairs
 - ²³⁴U-²³⁸Pu
 - ²³⁵U-²³⁹Pu
 - ²³⁶U-²⁴⁰Pu
- Model age: 1946 ± 4.5 years

- Absence of detectable ²⁴¹Pu (²⁴¹Pu t_{1/2} = ~14 years) consistent with this model age
- Reactor modeling and historical records suggest that material was produced in X-10 reactor, Oak Ridge
- Material is second-oldest known sample of Pu

Summary

- Model age is a powerful signature for nuclear forensics
 - comparative: establish or eliminate genetic link
 - predictive: assume sample history
- Model age assumptions
 - material is purified from decay products at time of production
 - material is 'closed system'
- Measuring multiple chronometers for a sample is important for increased confidence in interpretation
 - Concordant ages provide validation for model age assumptions
 - Discordant ages can constrain the processes used in material production or contaminant characteristics

Upcoming Webinars

- Development of Signatures: October 27, 2016
- Statistics in Nuclear Forensics: November 17, 2016
- Source and Route Attribution: December 8, 2016

NAMP website: www.wipp.energy.gov/namp