Radiochemical Diagnostics: A Cornerstone of Stockpile Stewardship

Hugh D. Selby

hds@lanl.gov

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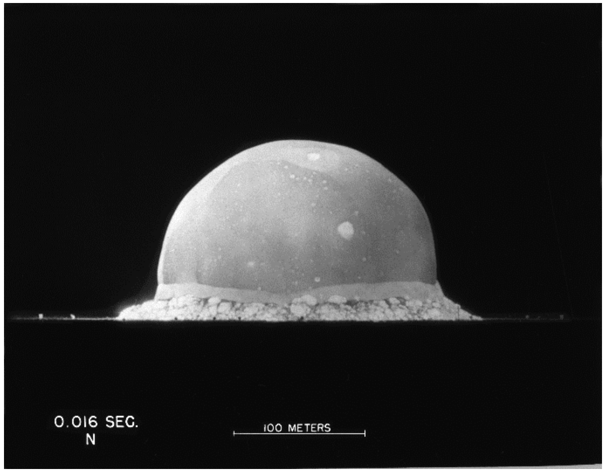
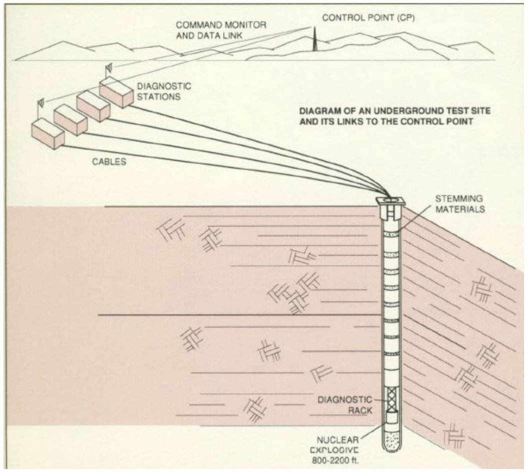
The Significance of Radiochemistry

The Trinity Fireball



Operation Crossroads B-17

Underground Testing



Photos Courtesy R.A Meade

LALP-88-21

- ***Radiochemistry established the yield of every sampled test, from Trinity to Divider***
- Radiochemistry is the gold standard for yield in today's Stockpile Stewardship Program (SSP)
- Radiochemistry plays an integral role in our ever-advancing understanding of weapons physics
- Significant R&D and programmatic investment in radiochemistry continues today
  - Basic nuclear physics
  - Sophisticated chemistry and counting methods
  - Mass spectrometry
  - Modeling and simulation

What is Radiochemical Debris Diagnostics?

Photos courtesy of W. Oldham, S. Hanson

Sample Collection

Dissolution and separation

Counting

- **Assessment of events in support of the AAR, LEPs and experimental science campaign milestones**
- **Interface between radiochemical measurement and design communities**
  - Integrate data streams (e.g MS, counting)
  - Account for chemical and physical phenomenology present in raw sample data
  - Employ computational tools to determine performance from sample data
  - Provide detailed assessments regarding impact of design on total performance
  - The product of this work *is the Gold Standard*

How is Weapon Performance Determined?

Blink!

BOOM!

Nuclear test debris is the neutron analog to photon exposed photographic film

$$Y_x = M_x \times E_x;\ E \equiv \frac{F_{total}}{Fuel_x^{\,0}}$$

For plutonium:

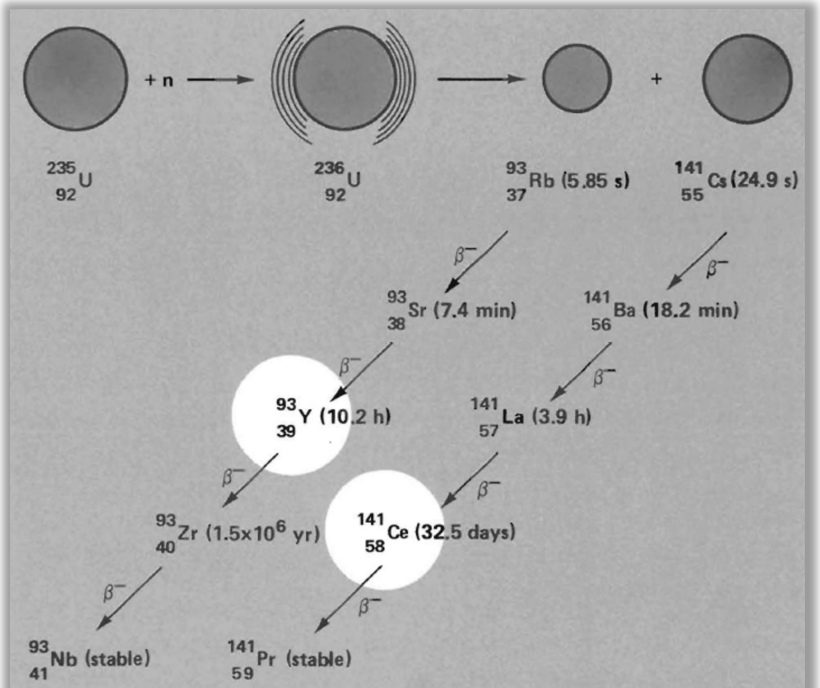
$$E_{pu} = \frac{F_{total}}{Pu^0};\ \xrightarrow{\text{sample}} \frac{f_s}{f_s + pu^{res}}$$

- “Inventory” equations
- Sample interferences must be accounted for
  - Chemical fractionation
  - Uranium blank
- Note: it is unnecessary to know how much of the device was collected

Images from: Alexkunztaipei.wordpress.com, Phys.org, Time.com, emojipedia.com, S.K. Hanson, R.A. Meade

Each Inventory Term is a *Measurement* – Consider Sample Fissions

$$f_s = N_{fp,s} \div CY_{fp}$$

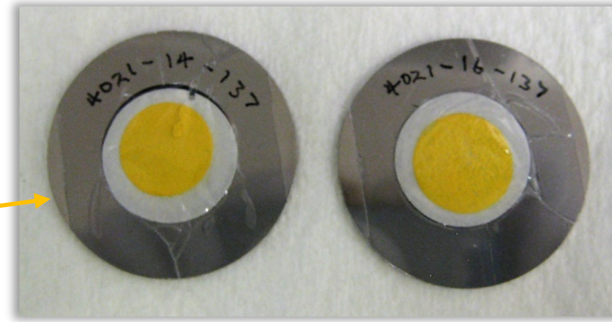


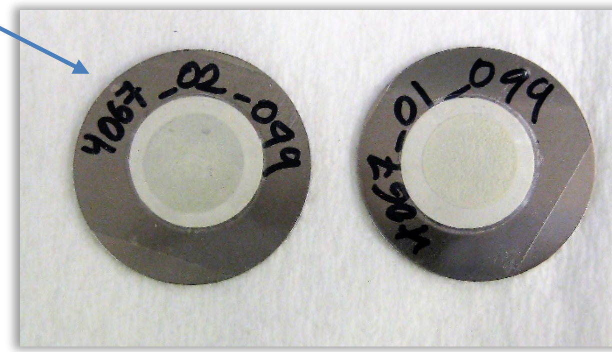
Knight and Sattizahn, Los Alamos Science, 1983

Each sample is yielded gravimetrically and mounted

Example mount forms:

$SrCO_3$
Y_2O_3
Cs_2PtCl_6
MoO_3
$BaCrO_4$
Nd_2O_3
Ag metal
Cd metal
Tb_4O_7



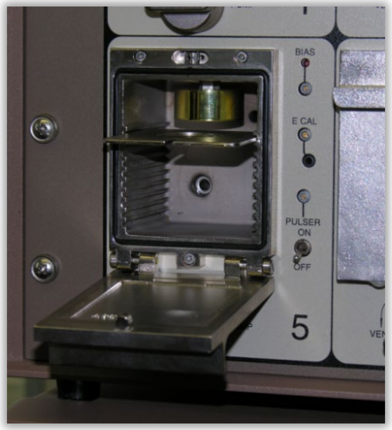


Photos courtesy of W. Oldham, S. Hanson

Measurement of Actinide Fuels

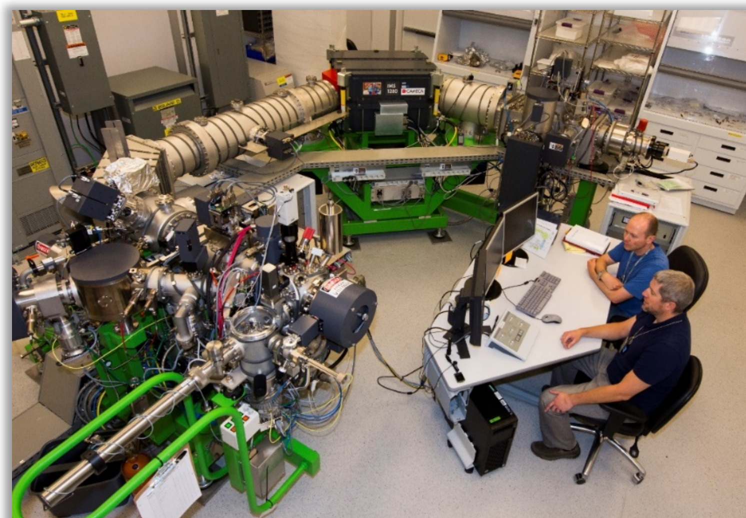
- Actinide fuels are the first term in *y = mass * efficiency*
- LANL point of excellence
  - World-class scientists and instrumentation
  - Alpha spectroscopy and mass spectrometry are primary tools

Photos courtesy of D. Dry, F. Taw, R. Steiner



High resolution α-counting ($^{238\text{-}240}$Pu, ^{241}Am, ^{242}Cm)

Thermal Ionization MS (long-lived U, Pu)



Secondary Ion MS (Spatially resolved actinide isotopic analysis)

So, About Those Nuclear Tests…

- **Nuclear testing ended in 1992**
- **The stockpile did not**
  - How do we ensure the function of our weapons with no testing?
  - How does radiochemistry remain relevant?
- **Scientific Stockpile Stewardship was born in the mid-1990s**
  - Based on advanced computing and simulation of tests
- **Requires more and higher quality radiochemical assessments with much better nuclear data and uncertainty than was possible during testing**
  - Radiochemical assessment has advanced far beyond testing and its requirements have advanced nuclear science
  - Radiochemistry underpins several missions in addition to Stewardship

Radiochemistry at Los Alamos Today: Chemistry Division's Nuclear and Radiochemistry Group

Primary capabilities: | **Major program thrusts:**

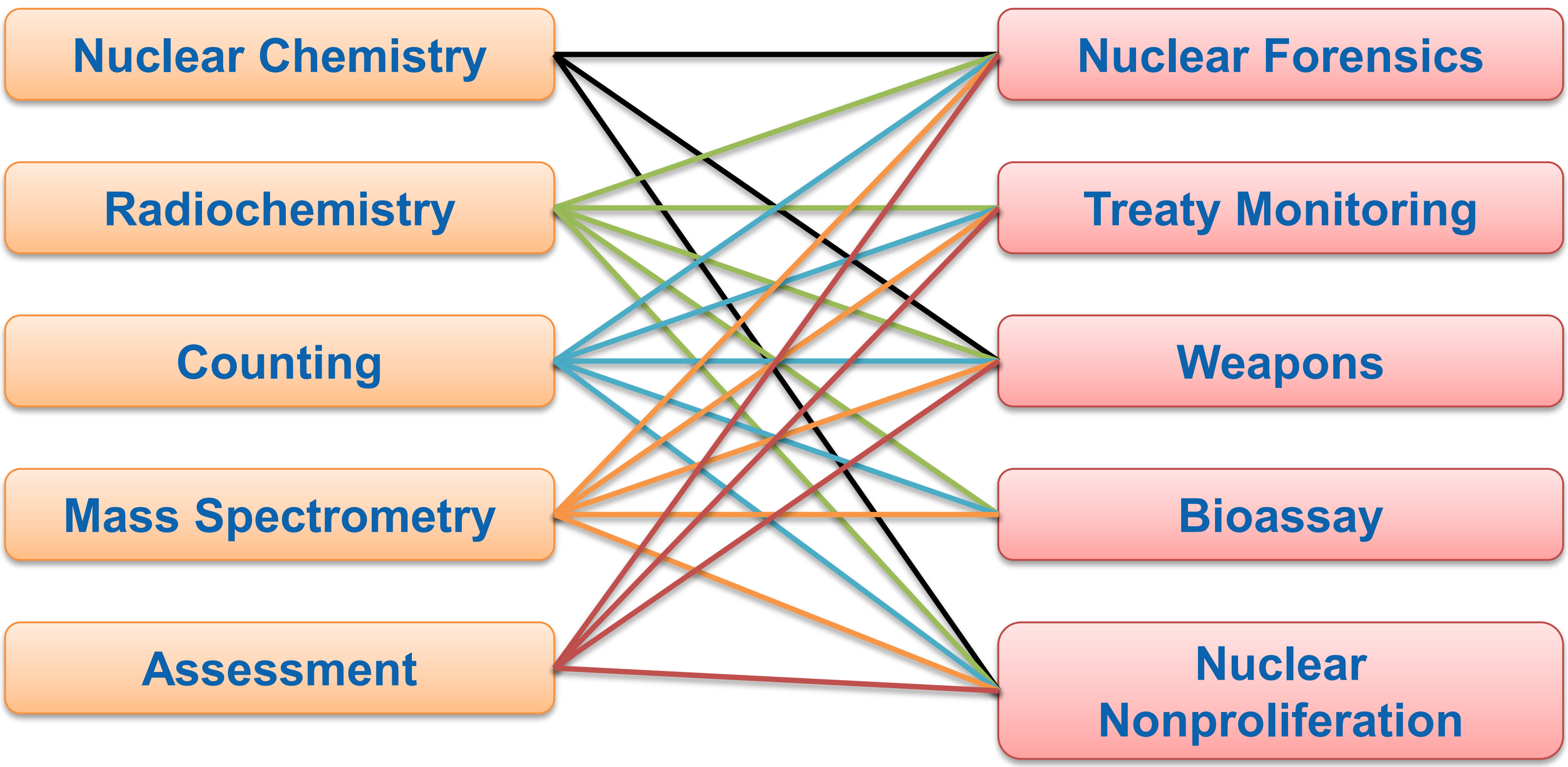


Figure courtesy of F. Taw

Over 100 Personnel Executing Mission and R&D

Administration
JoRonda Abeyta
Jessica Segovia

Felicia Taw, Group Leader
Warren Oldham, Deputy Group Leader

Project/Program Management
Stephen Lamont
David Podlesak
Richard Oldenborg*

February 2020

Nuclear Chemistry

Todd Bredeweg, TL

Evelyn Bond
Kevin Glennon+
Matthew Gooden
Marian Jandel*
Robert Rundberg*
Gencho Rusev
Jenifer Shafer*
David Vieira*
Jerry Wilhelmy*
Xinxin Zhao

Figure courtesy of F. Taw

Radioanalytical Chemistry

Susan Hanson, TL

Mark Boggs
Mateusz Dembowski^
William Kinman
Gabrielle Lee
Cheriece Margiotta
Iain May
Daniel Meininger
Jeffrey Miller
Sean Reilly
Nathan Smythe
Melinda Wren

Countroom

Donald Dry, TL

Melissa Boswell
Donald Cox
Malcolm Fowler*
Thomas Gamble*
Lisa Hudston
Michael James
Camilla Lance
Randy Rendon
Jennifer Romero
Dillon Soenke+
Joshua Williams
Ben Zimmerman*

Assessment

Hugh Selby, TL

Kevin Bennett
Scott Bowen*
Alexander Gancarz*
Corey Keith
Gus Keksis
Amy Lee
Bret Lockhart
Michael MacInnes*
Roger Meade*
John Musgrave*
Reg Rocha
Serena Rodriguez
Christopher Waidmann

Clean Chemistry

Robert Steiner, TL
Jeffrey Roach, PM

Michael Harris
Hannah Holman
Zsuzsanna Macsik
Danielle Roybal
Gilbert Sosaya*
Joseph Sullivan

Trace Radiochemistry
Theresa Kayzar-Boggs, FLM
Ronald Amato
Jennifer Berger
Annelise Cardon
Joanna Denton
Mark Edwards
Rebecca Foley
Steven Goldstein
Kimberly Hinrichs
Allison Price

Radiobioassay Sample Preparation
Fred Roensch*, FLM
Julianna Baca
Andrew Hyde
Deanna Lopez
Lynn Miller
Joel Zazueta
Madison Zuniga

ICP-MS
Jeremy Inglis, FLM
Jacob Buettner
April Cardon
Matthew Sanborn
Allison Wende

TIMS
Jeremy Inglis, FLM
Susan Eaton
Daniel Gurganus
James Louis Jean+
Azim Kara
Dennis Lopez
Joel Maassen
Andrew Nunn
Andrew Reinhard^

SIMS
Robert Steiner, FLM
Chloe Bonamici*
Richard Hervig*
Travis Tenner

Design and Fabrication
Abran Chavez, FLM
Connell Lane*
Dale Melton
Gerald Montoya

*Affiliation, Contractor, Guest Scientist, Form B, etc.
+Students
^Postdocs

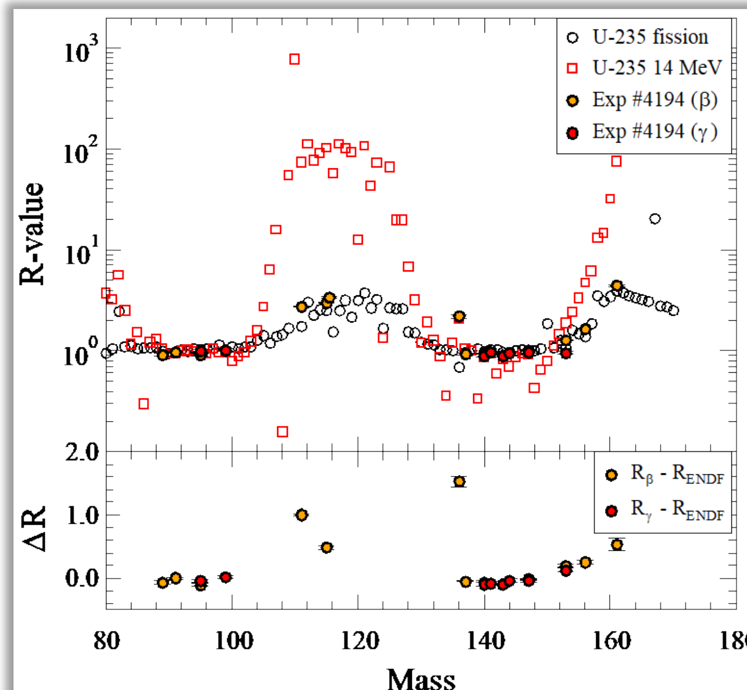
Research Highlights, CY_{fp} (Cumulative Fission Product Yields)

Objective: Measure integral cumulative fission product yields, $R_i^{j,k}$ and $Y_i^{j,k}$, in relevant neutron fields for several major and minor actinides.

Relevance: Fission product yields represent important nuclear fission observables for basic science as well as numerous nuclear applications. This and related work provides experimental data to validate and improve differential nuclear data, nuclear physics modeling and operational tools.

Approach: Make use of critical assemblies and other neutron sources to perform fission chamber measurements in parallel with historical radiochemical analysis on select actinides to determine relative ($R_i^{j,k}$) and absolute ($Y_i^{j,k}$) fission product yields.

Collaborations: Most of these experiments have included participation from LLNL and/or PNNL.



Figures courtesy of T. Bredeweg

Accomplishments/Results: Executed 1-2 irradiations per year since 2012 on various actinide samples including ^{233}U, ^{235}U, ^{238}U, ^{237}Np and ^{239}Pu. For each actinide sample we extracted relative fission product yields, $R_i^{j,k}$, by radiochemical analysis.

Tested and fielded prototype fission chambers with ^{235}U and ^{237}Np reference and macro-foils on the Flattop assembly to extract absolute fission product yields, $Y_i^{j,k}$.

Research Highlights, Extinct Radionuclides


Measurements of extinct fission products in nuclear bomb debris: Determination of the yield of the Trinity nuclear test 70 y later

Susan K. Hanson[a,1], Anthony D. Pollington[a], Christopher R. Waidmann[a], William S. Kinman[a], Allison M. Wende[a], Jeffrey L. Miller[a], Jennifer A. Berger[a], Warren J. Oldham[a,1], and Hugh D. Selby[a,1]

[a]Nuclear and Radiochemistry Group, Chemistry Division, Los Alamos National Laboratory, Los Alamos, NM 87545

Significance

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Project Goal: Reconstruct the entire suite of diagnostically useful radionuclides by measuring perturbations in stable element isotope ratios and actinide composition.

LANL LDRD-DR
PI: Hugh Selby (C-NR)
Co-PI for modeling: John Scott (XTD-IDA)
Co-PI for measurements: Warren Oldham (C-NR)

^{92}Mo 14.84	^{93}Mo 3.5E3a ε	^{94}Mo 9.25	^{95}Mo 15.92	^{96}Mo 16.68	^{97}Mo 9.55	^{98}Mo 24.13	^{99}Mo 2.75d β	^{100}Mo 9.63	^{101}Mo 14.61m β	^{102}Mo 11.3m β	^{103}Mo 1.13m β	^{104}Mo 1.00m β
^{91}Nb 7E2a ε	^{92}Nb 3.5E7a ε	^{93}Nb 100	^{94}Nb 2.0E4a β	^{95}Nb 34.99d β	^{96}Nb 23.4h β	^{97}Nb [illegible]23h β	^{98}Nb 2.9s β	^{99}Nb [illegible]5.0s β	^{100}Nb [illegible]5s β	^{101}Nb 7.1s β	^{102}Nb 1.3s β	^{103}Nb 1.5s β
^{90}Zr 51.45	^{91}Zr 11.22	^{92}Zr 17.15	^{93}Zr 1.5E6a β	^{94}Zr 17.38	^{95}Zr 64.02d β	^{96}Zr 2.8	^{97}Zr [illegible].8h β	^{98}Zr 30.7s β	^{99}Zr [illegible].2s β	^{100}Zr [illegible].1s β	^{101}Zr 2.4s β	^{102}Zr 2.9s β
^{89}Y 100	^{90}Y 2.67d β	^{91}Y 58.5d β	^{92}Y [illegible]54h β	^{93}Y 10.2h β	^{94}Y [illegible]7m β	^{95}Y [illegible]3m β	^{96}Y [illegible]3s β	^{97}Y [illegible]76s β	^{98}Y 0.59s β	^{99}Y [illegible]47s β	^{100}Y [illegible]73s β	^{101}Y 0.43s β
^{88}Sr 82.58	^{89}Ru 50.52d β	^{90}Sr 28.8a β	^{91}Sr 9.5h β	^{92}Sr [illegible]71h β	^{93}Sr 7.41m β	^{94}Sr [illegible]5m β	^{95}Sr [illegible].1s β	^{96}Sr [illegible]07s β	^{97}Sr [illegible]43s β	^{98}Sr 0.65s β	^{99}Sr [illegible]9ms β	^{100}Sr [illegible]1ms β

Hanson, S.K.; Pollington, A.D.; Waidmann, C.R.; Kinman, W.S.; Wende, A.M.; Miller, J.L.; Berger, J.A.; Oldham, W.J.; Selby, H.D. *Proc. Nat. Acad. Sci.*, **2016**, *113,* 8104-8108.

Why Consider the Weapons Laboratories?

Images from Reuters.com

- **Relevance**
  - The world is not a safe place
  - The Laboratories' primary mission is Science in Defense of the Nation
  - Diverse programs, from Stewardship to Nuclear Forensics, work to this theme
- **Challenge**
  - No one comes out of school trained for this work
  - It is an honor to be trusted with ensuring our deterrent
- **Science**
  - The highest concentration of the best experimental facilities and scientists provide opportunity to explore unlike anywhere else

Acknowledgments

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  - Amy S. Lee, August L. Keksis, Christopher R. Waidmann, Corey C. Keith, Kevin T. Bennett, Reginaldo C. Rocha, Bret L. Lockhart, Elyza J. Ortiz, Serena R. Rodriguez, Roger A. Meade, John A. Musgrave
- **DOE-NNSA Science Campaign 4.1**

Thank You for Your Interest!