# The Discovery of Element 117

Presented to

**The National Organization of Test, Research, and Training Reactors**

and

**The International Group on Research Reactors**

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Knoxville, Tennessee
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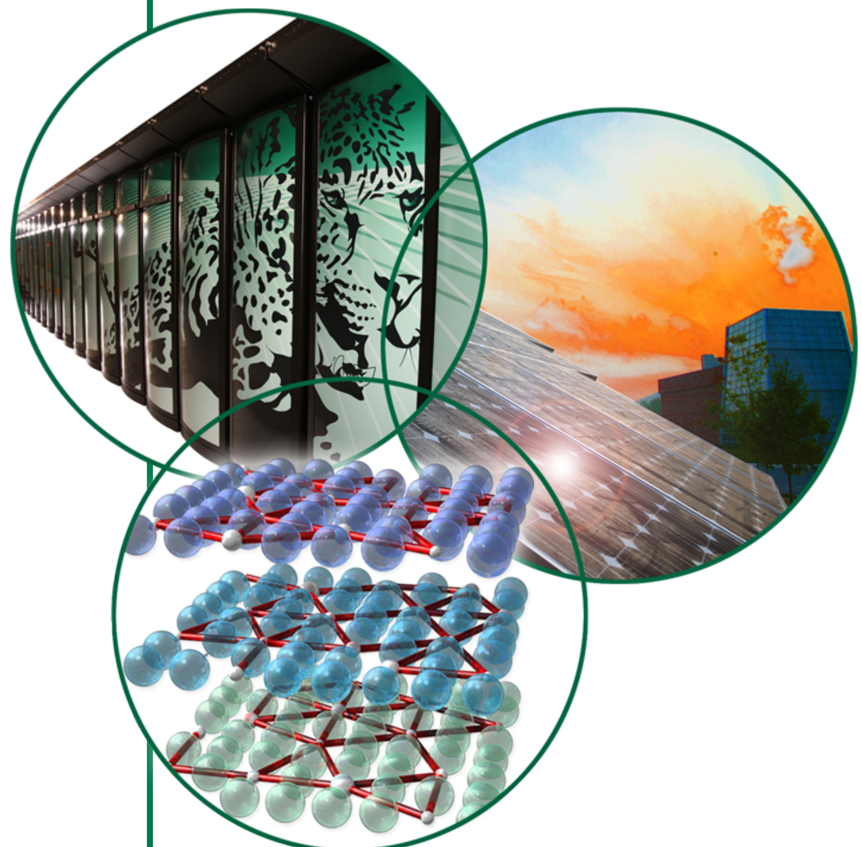

U.S. DEPARTMENT OF ENERGY

OAK RIDGE NATIONAL LABORATORY
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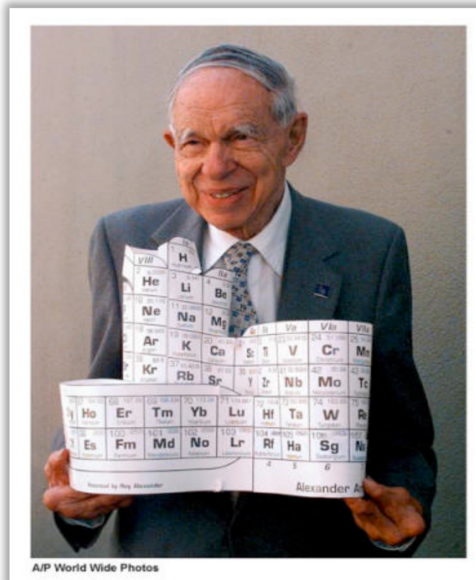
# The discovery of element 117

- An important advance in nuclear structure, confirming the “island of stability” for super-heavy elements
- Includes the discovery of 11 new heaviest known isotopes with atomic numbers 105–117
- Featured on the cover of *Physical Review Letters*, more than 250 news articles (*New York Times, Science*, etc.)
- Changes the periodic table (and eventually every high school chemistry book)
- Even a new ice cream!

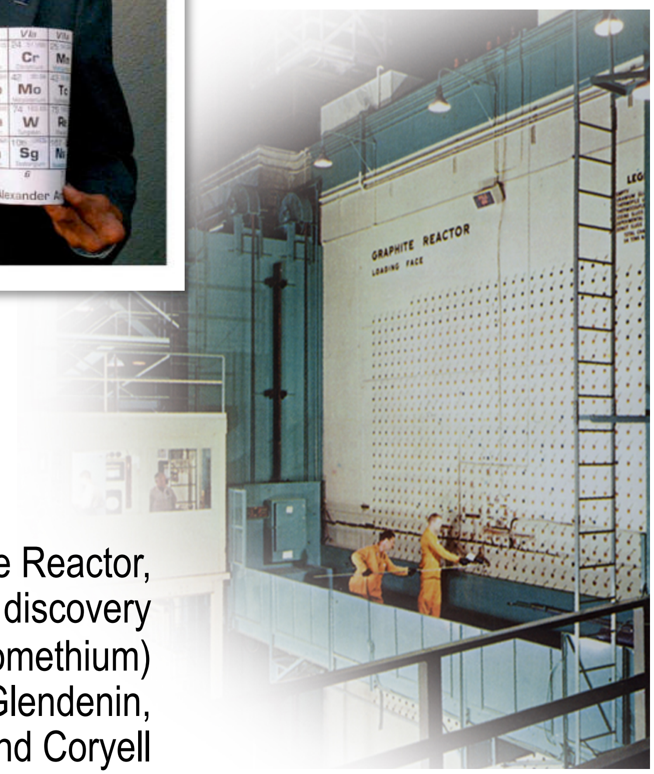

Featured flavor at Razzleberry Ice Cream Lab

# The synthesis of transuranics and element 61

- Transuranic elements (atomic numbers greater than 92, uranium) do not naturally exist on Earth
- Fermi proposed creating heavier elements by irradiating uranium with neutrons (he missed the discovery of fission)
- Seaborg and co-workers synthesized elements 93–103 in the 1940s and 1950s
  - To further this research, Seaborg was a major advocate for construction of HFIR
- Scientists at ORNL separated the missing element 61 from fission products in 1945, completing the lanthanide series



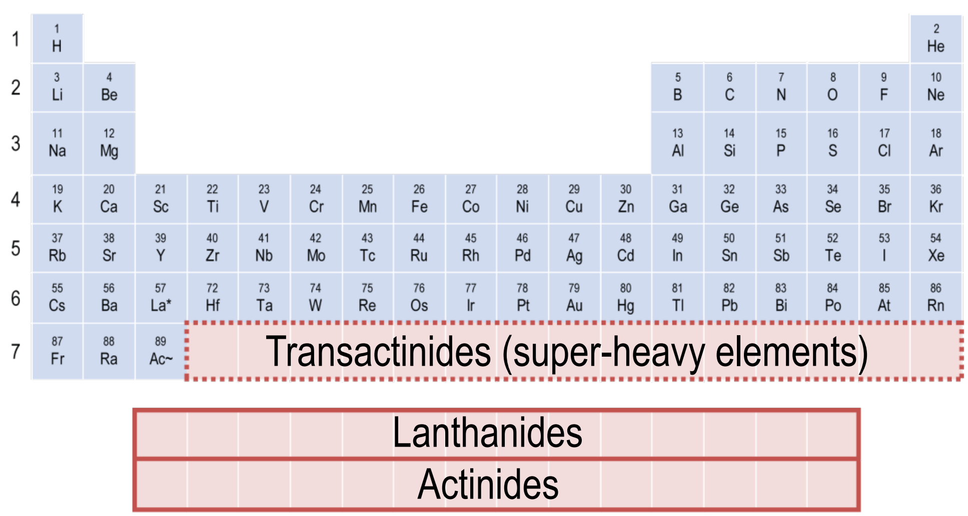

Seaborg's discovery of the actinides led him to modify the periodic table



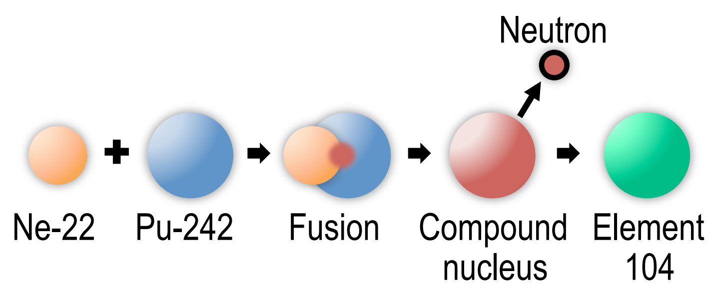

ORNL's Graphite Reactor, site of the discovery of element 61 (promethium) by Marinsky, Glendenin, and Coryell

# Super-heavy elements, Z > 103

- 1960s: Existence of transactinides (super-heavy elements) first proposed by Seaborg
  - Half-lives ranging from hours (Z = 104, 105) to milliseconds (Z = 115, 116, 118)
  - Expected to be analogous in chemical properties to Hf, Ta, etc., and placed immediately beneath these elements in the periodic table




- 1964: First transactinide (element 104, rutherfordium) synthesized at the Joint Institute for Nuclear Research (JINR), Dubna, Russia

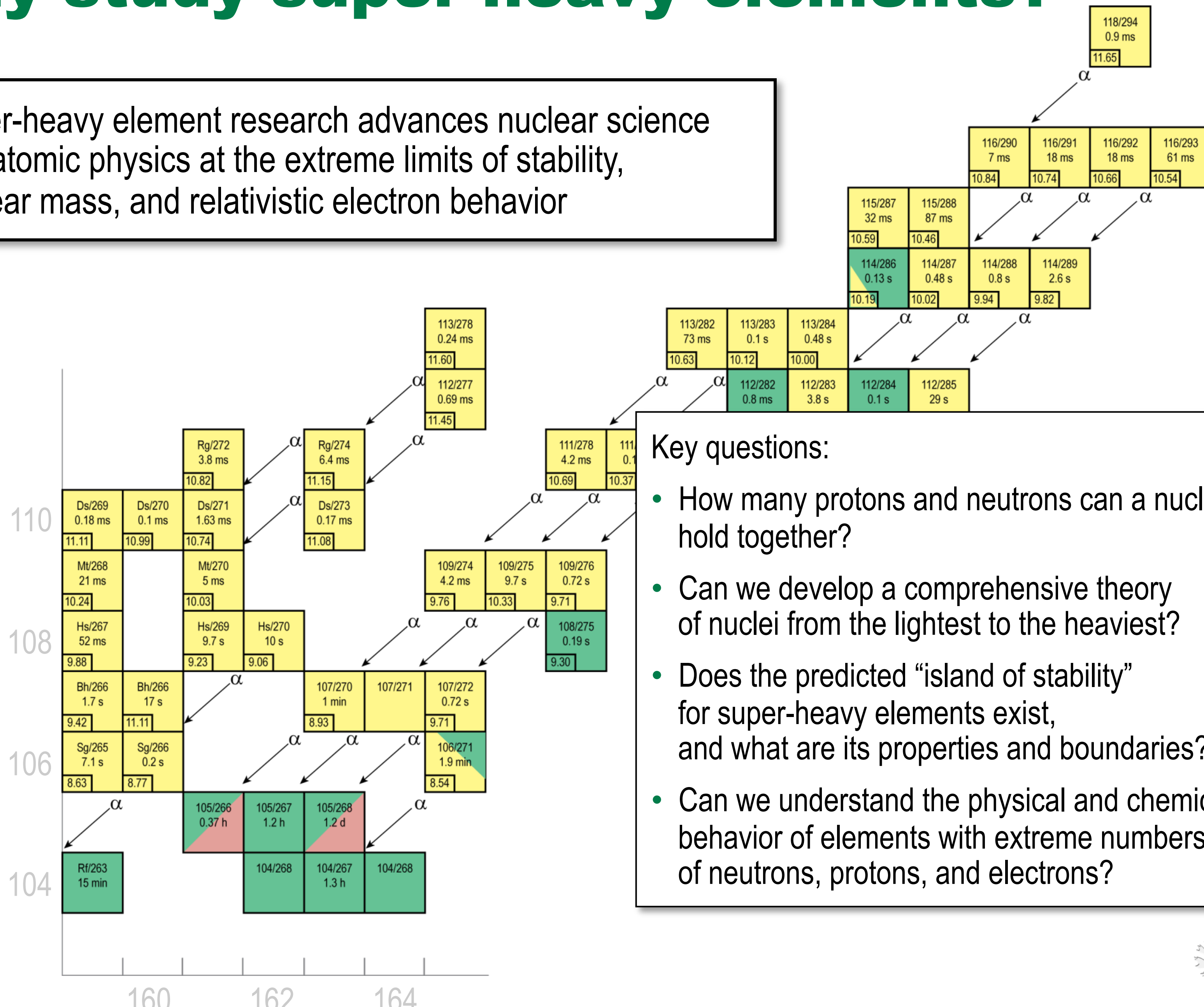



$$^{242}_{94}Pu + ^{22}_{10}Ne \rightarrow ^{264}_{104}Rf^* \rightarrow ^{263}_{104}Rf + n$$

Bombarding Pu (94 protons) with Ne (10 protons) results in a compound nucleus, Z = 104 protons

# Why study super-heavy elements?

Super-heavy element research advances nuclear science and atomic physics at the extreme limits of stability, nuclear mass, and relativistic electron behavior

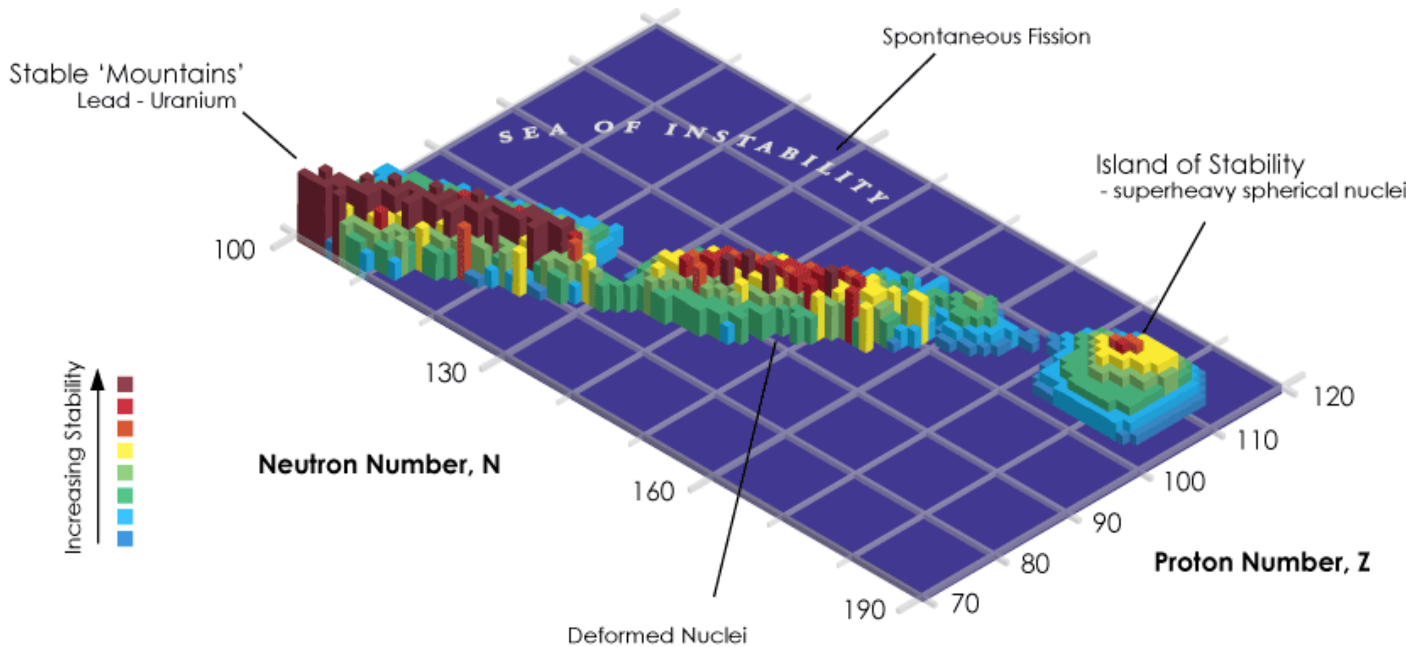



Key questions:

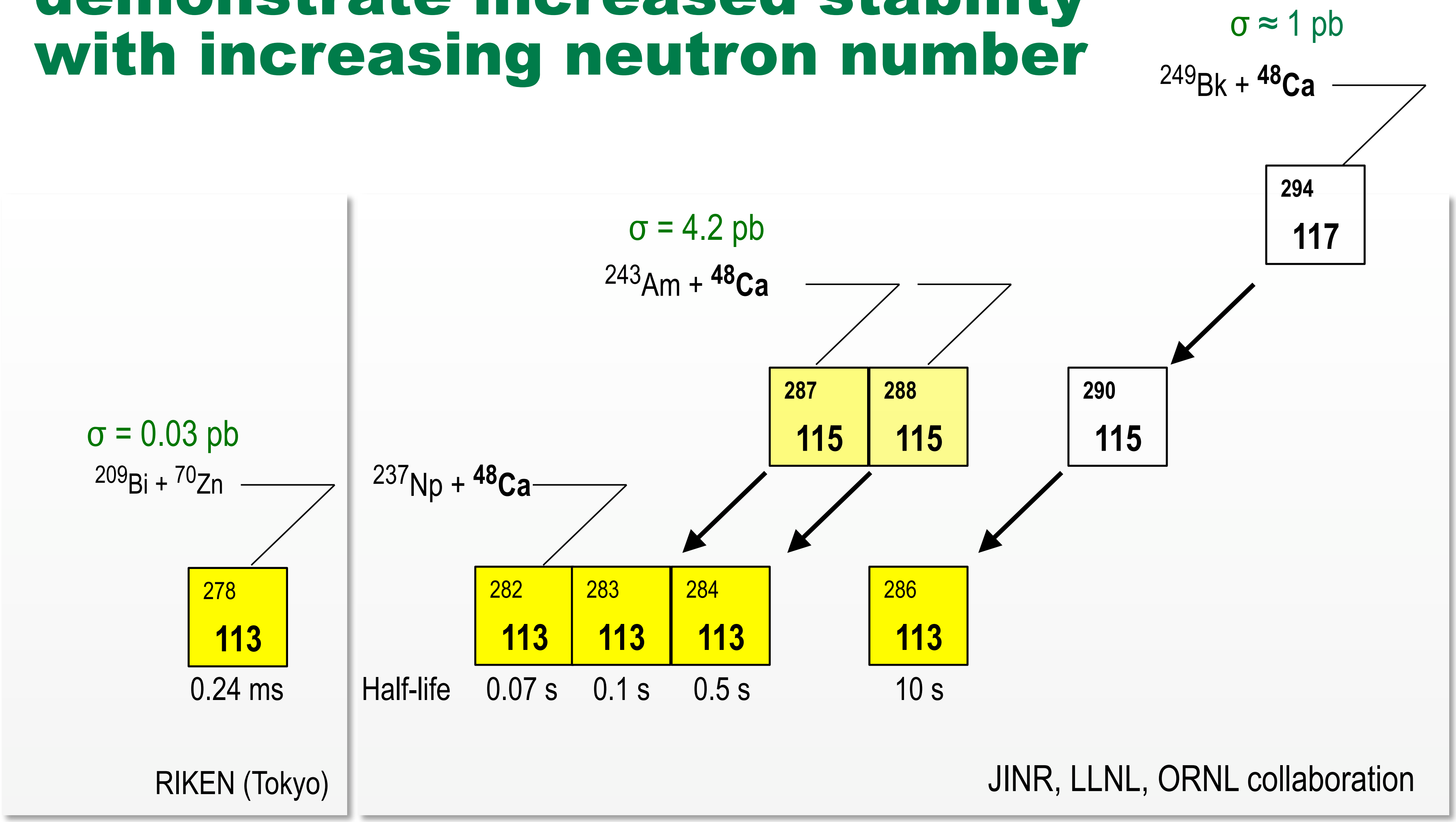
- How many protons and neutrons can a nucleus hold together?
- Can we develop a comprehensive theory of nuclei from the lightest to the heaviest?
- Does the predicted "island of stability" for super-heavy elements exist, and what are its properties and boundaries?
- Can we understand the physical and chemical behavior of elements with extreme numbers of neutrons, protons, and electrons?

# The island of enhanced stability

- The possibility of an “island of stability” of superheavy elements with greatly increased lifetimes (perhaps millions of years) was originally postulated by Seaborg
- When neutrons and protons completely fill the energy levels of a given nucleus, the nuclear binding is strongest, leading to longer lifetimes
- Heaviest stable isotope (Pb-208): Closed nuclear shells for both protons (82) and neutrons (126)
- Next “doubly magic” nucleus: Occurs around Z = 114 and N = 184, the presumed center of the island of stability



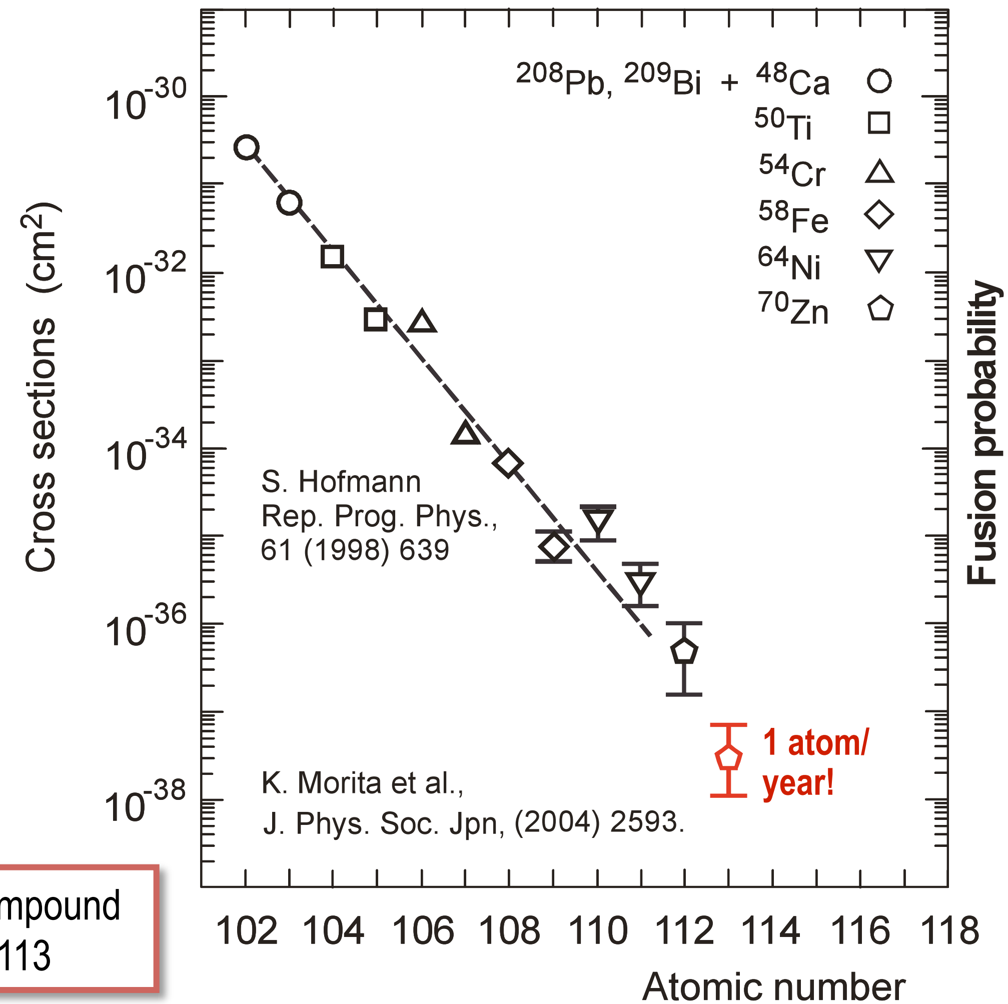
# Isotopes of element 113 demonstrate increased stability with increasing neutron number




Y. Oganessian. Perspectives of JINR–ORNL collaboration in the studies of superheavy elements, JINR Scientific Council, September 24–25. 2009, Dubna

# 1990s: Cold fusion experiments at GSI in Germany extended the periodic table to Z = 112

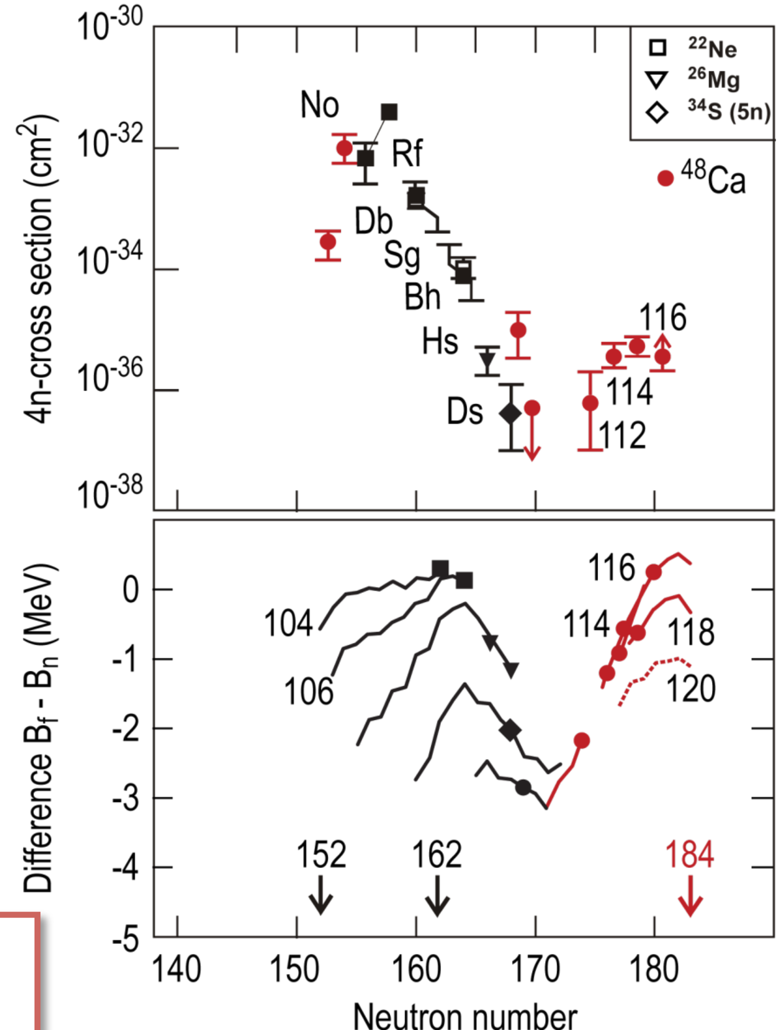
- Bombardment of a heavy target (Pb, Bi) with a heavy ion (Fe, Ni, etc.) at energies just above the Coulomb barrier
- Imparts minimum internal energy to the compound nucleus
- Cross sections decrease steadily with increasing atomic number, making the technique impractical above Z = 112




Cold fusion cross sections for compound nucleus formation up to Z = 113

# 2000s: Hot fusion techniques using $^{48}Ca$ beams were pioneered in Russia

- $^{48}Ca$ is a "doubly magic" neutron-rich isotope
- Bombardment of actinide targets (Cm, U, Cf, etc.) with $^{48}Ca$ beam creates a highly-excited compound nucleus
- At higher neutron numbers, these nuclei stabilize by emission of 3–4 neutrons
- Overall cross sections increase, reversing the cold fusion trend and extending the range of synthesis experiments to Z = 118 and above
- For element 117, this technique requires a berkelium target




At higher neutron numbers, spontaneous fission is suppressed, allowing excited compound nuclei to stabilize via multiple neutron emissions

# The development of $^{48}$Ca hot fusion has extended the periodic table to Z = 118

| Year | Element | Laboratory | Reaction | Number of atoms synthesized to date |
|---|---|---|---|---|
| 2000 | 114 | JINR, Russia[1] | $^{48}Ca \rightarrow {}^{244}Pu$ (ORNL) | 50 atoms |
| 2004 | 113 | JINR, Russia[1] | Decay product of element 115 | 8 atoms |
| 2004 | 115 | JINR, Russia[1] | $^{48}Ca \rightarrow {}^{243}Am$ (ORNL) | 30 atoms |
| 2005 | 116 | JINR, Russia[1] | $^{48}Ca \rightarrow {}^{248}Cm$ (RIAR/ORNL) | 30 atoms |
| 2006 | 118 | JINR, Russia[1] | $^{48}Ca \rightarrow {}^{249}Cf$ (ORNL) | 3 – 4 atoms |
| 2010 | 117 | JINR, Russia[2] | $^{48}Ca \rightarrow {}^{249}Bk$ (ORNL) | 6 atoms |

[1] In collaboration with LLNL
[2] In collaboration with ORNL, LLNL, Vanderbilt, and UNLV

**All of these discoveries used ORNL isotopes**

# The periodic table in 2009

| Period | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | 1 H | 2 IIA | | | | | | | | | | | 13 IIIA | 14 IVA | 15 VA | 16 VIA | 17 VIIA | 2 He |
| 2 | 3 Li | 4 Be | | | | | | | | | | | 5 B | 6 C | 7 N | 8 O | 9 F | 10 Ne |
| 3 | 11 Na | 12 Mg | | | | | | | | | | | 13 Al | 14 Si | 15 P | 16 S | 17 Cl | 18 Ar |
| 4 | 19 K | 20 Ca | 21 Sc | 22 Ti | 23 V | 24 Cr | 25 Mn | 26 Fe | 27 Co | 28 Ni | 29 Cu | 30 Zn | 31 Ga | 32 Ge | 33 As | 34 Se | 35 Br | 36 Kr |
| 5 | 37 Rb | 38 Sr | 39 Y | 40 Zr | 41 Nb | 42 Mo | 43 Tc | 44 Ru | 45 Rh | 46 Pd | 47 Ag | 48 Cd | 49 In | 50 Sn | 51 Sb | 52 Te | 53 I | 54 Xe |
| 6 | 55 Cs | 56 Ba | 57 La | 72 Hf | 73 Ta | 74 W | 75 Re | 76 Os | 77 Ir | 78 Pt | 79 Au | 80 Hg | 81 Tl | 82 Pb | 83 Bi | 84 Po | 85 At | 86 Rn |
| 7 | 87 Fr | 88 Ra | 89 Ac | 104 Rf | 105 Db | 106 Sg | 107 Bh | 108 Hs | 109 Mt | 110 Ds | 111 Rg | 112 **Cn** | 113 Uut | 114 Uuq | 115 Uup | 116 Uuh | 117 | 118 Uuo |

| | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Lanthanide Series | 58 Ce | 59 Pr | 60 Nd | 61 Pm | 62 Sm | 63 Eu | 64 Gd | 65 Tb | 66 Dy | 67 Ho | 68 Er | 69 Tm | 70 Yb | 71 Lu |
| Actinide Series | 90 Th | 91 Pa | 92 U | 93 Np | 94 Pu | 95 Am | 96 Cm | 97 Bk | 98 Cf | 99 Es | 100 Fm | 101 Md | 102 No | 103 Lr |

# What it takes to produce a few atoms of element 117

| | |
|---|---|
| **3 g of Ca-48** | Natural abundance enriched 500 times at Sverdlovsk-45 |
| **20 mg of Bk-249** | Produced by 250-day neutron irradiation in the world's highest thermal neutron flux at Oak Ridge |
| **Chemical separation of Bk from irradiated targets** | Impurities less than 2 ng (one part in $10^7$), performed at Oak Ridge |
| **Preparation of Bk target foils** | Specially produced at Dimitrovgrad to survive massive ion bombardment |
| **Target irradiation with Ca-48** | 150 days continuous irradiation in the world's most intense Ca-48 beam at Dubna |
| **Detection** | One superheavy atom per $10^{12}$ reaction products at Dubna |
| **Analysis** | Nuclear data analysis of thousands of candidate reactions (Dubna and LLNL) |

HFIR/REDC reactor/hot cell complex (ORNL)

Flerov Laboratory (Dubna)

# The element 117 research team

- **Joint Institute for Nuclear Research (Dubna)**
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- **Oak Ridge National Laboratory**
  P. D. Bailey, D. E. Benker, J. G. Ezold, C. E. Porter, F. D. Riley, J. B. Roberto, K. P. Rykaczewski
- **Lawrence Livermore National Laboratory**
  R. A. Henderson, K. J. Moody, S. L. Nelson, D. A. Shaughnessy, M. A. Stoyer, P A. Wilk
- **Vanderbilt University**
  J. H. Hamilton, A. V. Ramayya
- **University of Nevada, Las Vegas**
  M. E. Bennett, R. Sudowe
- **Research Institute for Advanced Reactors (Dimitrovgrad)**
  M. A. Ryabinin

**International collaboration was essential**

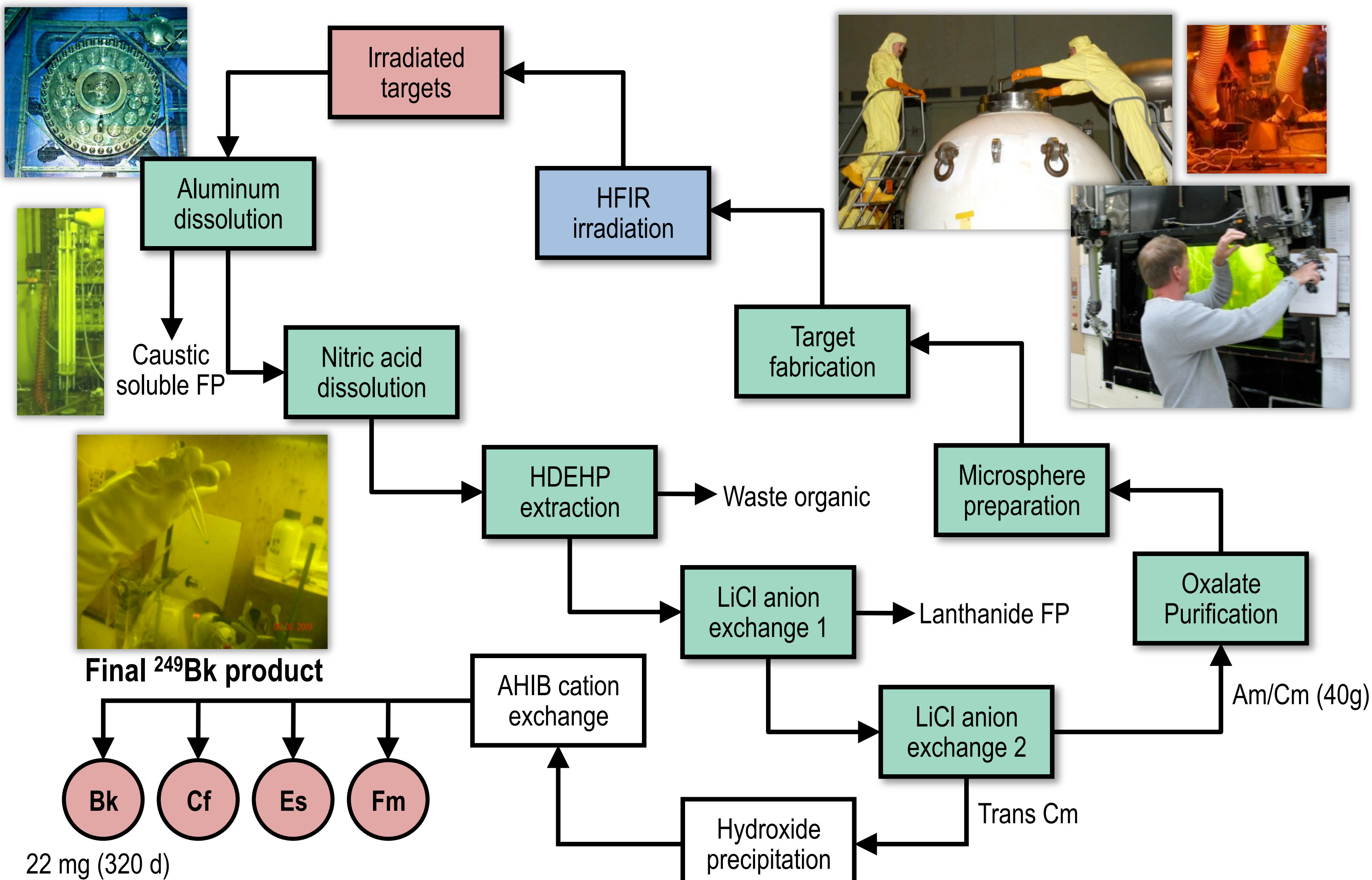
# Reactor irradiation of Am/Cm targets at HFIR

- Targets specially designed for reactor conditions:
  - Composition controls fission and gamma heating
  - Targets remain in the reactor for 11 cycles (approximately 18 months)
- Irradiation in the HFIR flux trap
  - Thermal neutron flux of $2.5 \times 10^{15}$ neutrons/cm²·s (world's highest steady-state neutron flux)
  - 31 target positions (10–13 targets typically irradiated)
  - Produces ~35 mg $^{252}$Cf per target (smaller quantities of Bk, Es, Fm, others)

Target positions in the flux trap of a HFIR fuel element

Fuel change-out at the High Flux Isotope Reactor (ORNL)

# Bk production/separation cycle (18–20 months)
## Radiochemical Engineering Development Center, ORNL



# More than 50 ORNL staff contributed to the Bk production and separation

- Radiochemical Engineering Development Center
- High Flux Isotope Reactor
- Nuclear Science and Technology Division
- Chemical Sciences Division

The final product (starting from 40 g of irradiated Am/Cm) is the green speck at the bottom of the glass vial, 22 mg of ultrapure Bk

# A race against time

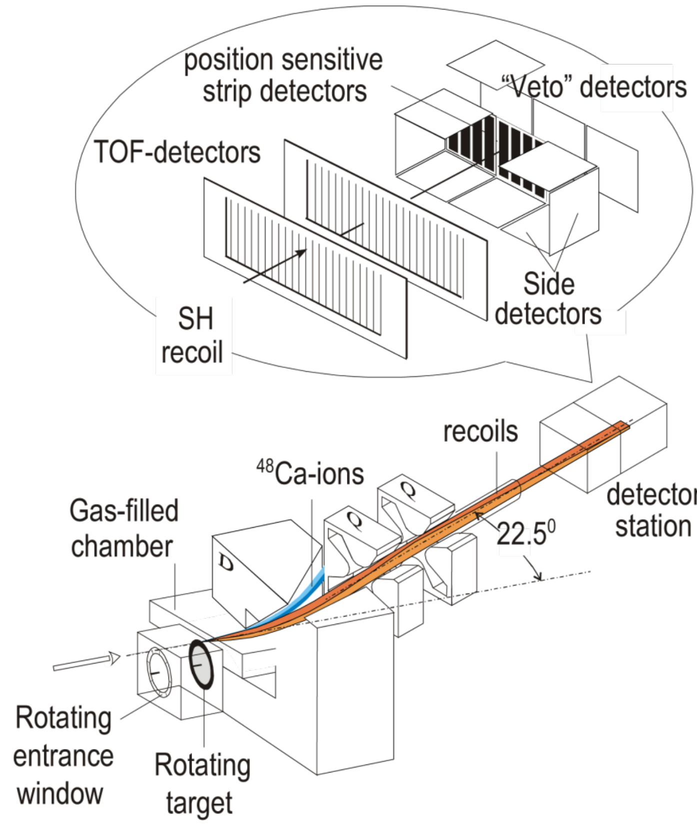
- Bk-249 has a 320-day half-life
  - 40% decay loss over the experiment
- Seamless coordination required
  - HFIR operations (11 on-time cycles)
  - REDC processing
  - Transportation
  - Target preparation
  - Accelerator operations (24/7 for 5 months)
- Customs issues added drama
  - 5 flights across the Atlantic (lots of frequent flyer miles for our Bk)
  - Still the fastest U.S.-Russia actinide transfer in history


Bk packages being inspected at Sheremetyevo Airport

# Heavy element production and detection at the Dubna Gas Filled Recoil Separator

- $^{48}$Ca beam supplied by the U400 cyclotron
- Total beam dose >$10^{19}$ particles
- Rotating target distributes beam heating
- Rapid separation allows detection of nuclei with short half-lives
- Suppression factors are $10^{15}$ for beam particles and $10^4$ for target-like particles

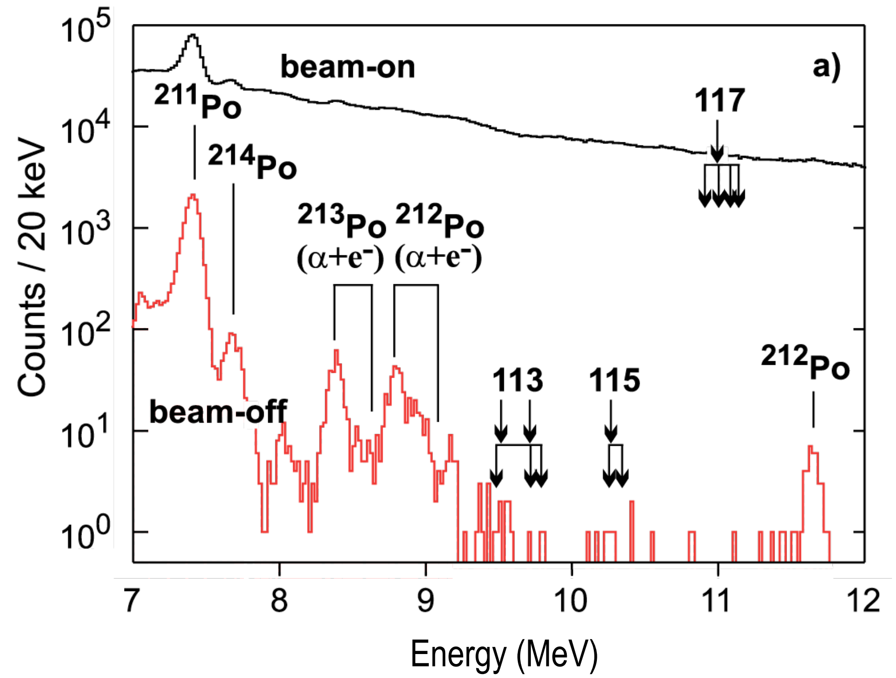
Heavy Ion Cyclotron U-400 at the Flerov Laboratory, JINR (Dubna)



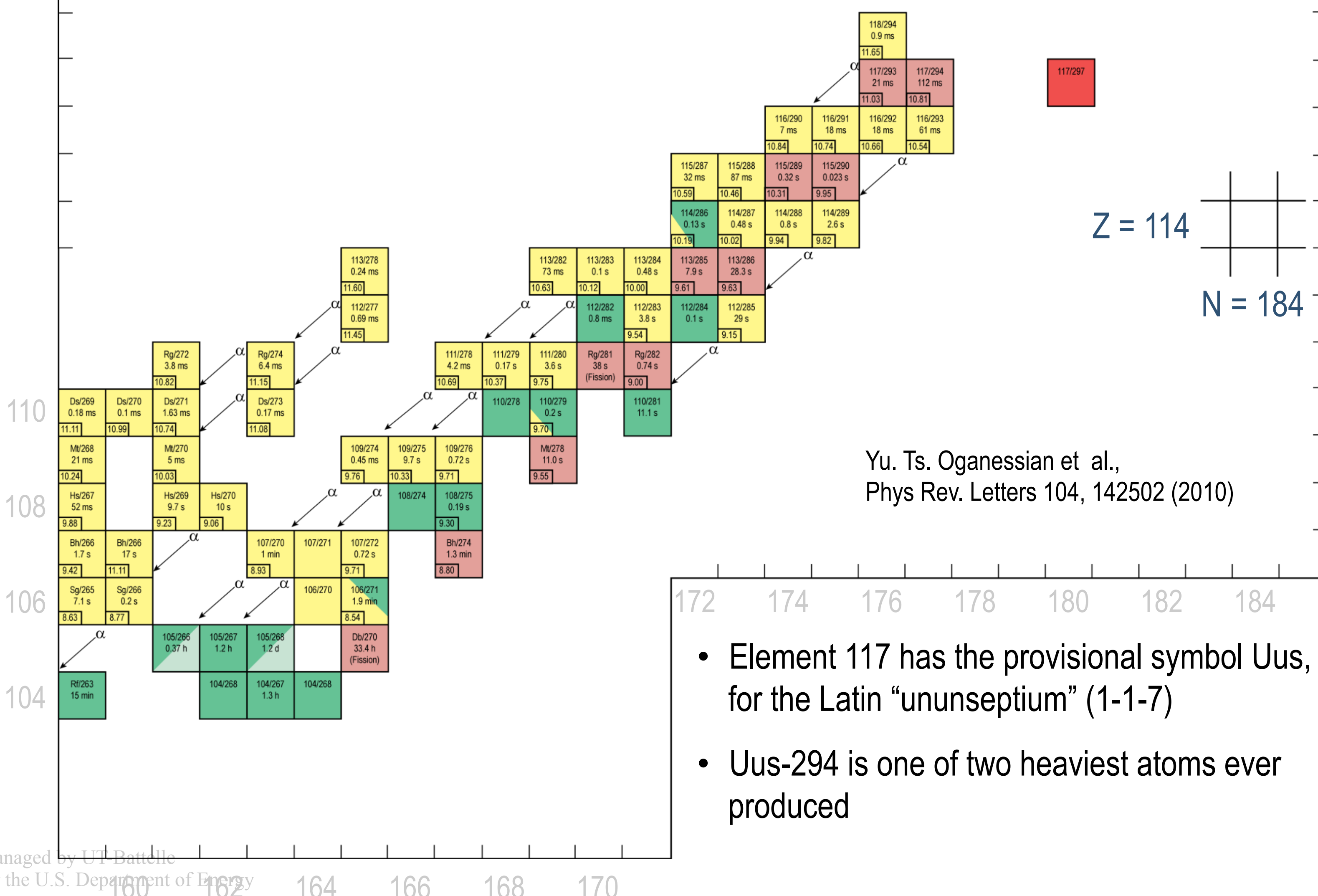
# Energy spectra in the detector

- Evaporation residues (including element 117 nuclei) are implanted into a silicon detector array
- Emitted alpha particles are registered in the implantation detector or side detectors
- The beam is switched off following candidate events (identified ion implantation followed by a fast α-like signal from the same location within the expected energy range for Z = 117)
- With the beam off, detection system is monitored for subsequent decay chain α-signals from the same location (with the decay chain ending in spontaneous fission)
- If no additional α's are detected, beam on target is resumed after 15 minutes

- Total energy spectra of α-like signals with beam on and off
- Arrows denote observed events
- The probability of a random sequence initiating any given decay chain is $10^{-6}$ or less



# Eleven new neutron-rich isotopes were observed in element 117 decay chains

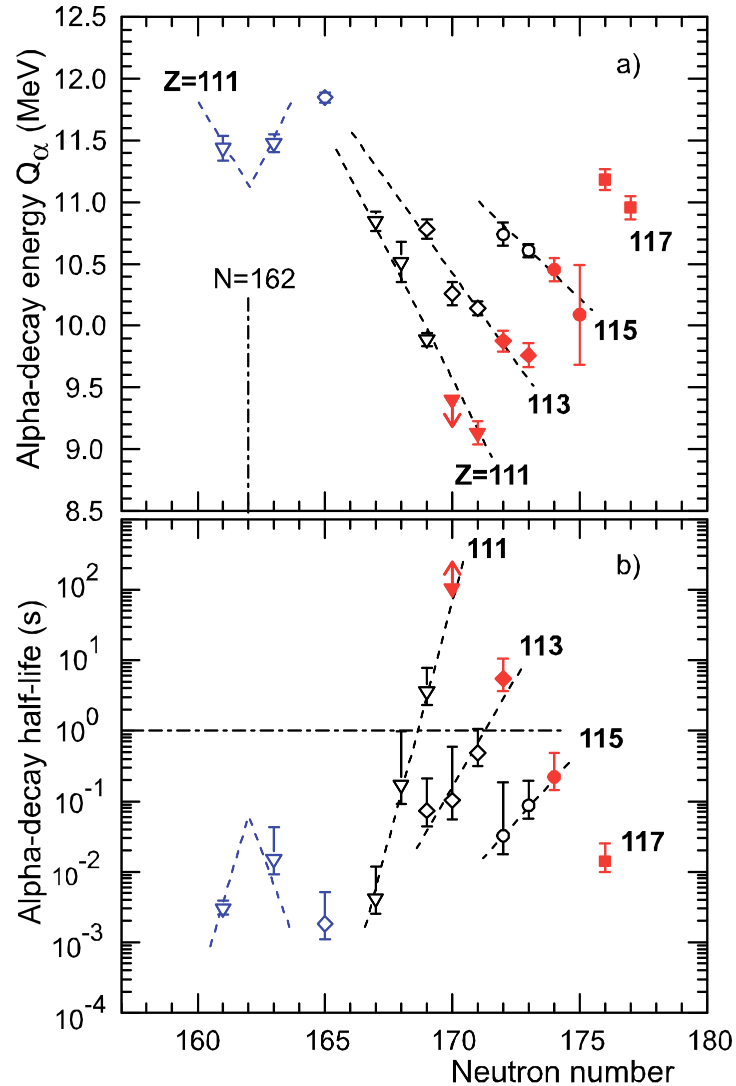



Yu. Ts. Oganessian et al.,
Phys Rev. Letters 104, 142502 (2010)

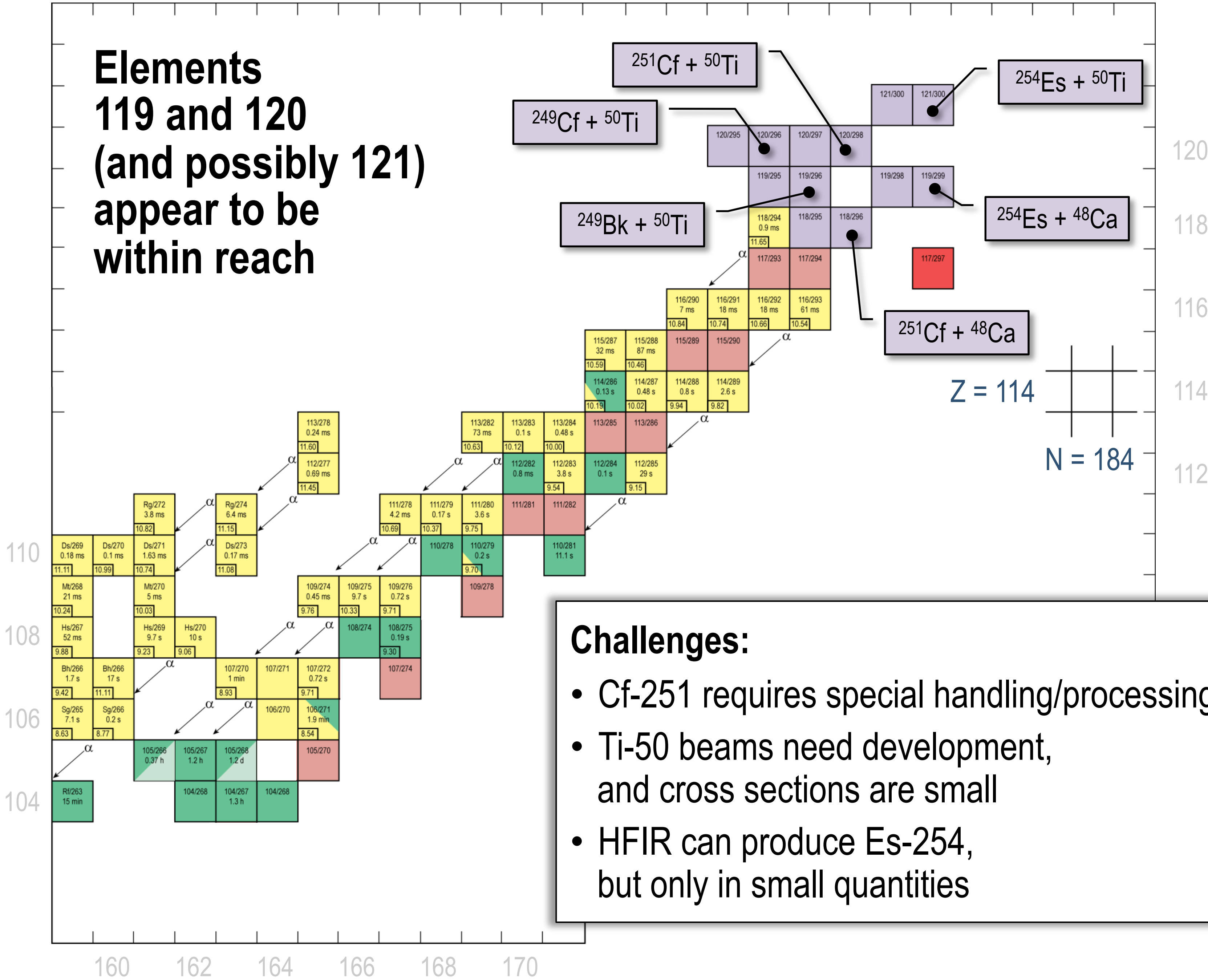
- Element 117 has the provisional symbol Uus, for the Latin "ununseptium" (1-1-7)
- Uus-294 is one of two heaviest atoms ever produced

# New isotopes from element 117 decay chains expand our knowledge of neutron-rich nuclei

- Closer approach to the shell at N = 184 results in a decrease in $\alpha$-decay energy and an increase in lifetime
- Continued trend toward increasing stability for higher neutron numbers



# Elements 119 and 120 (and possibly 121) appear to be within reach




**Challenges:**

- Cf-251 requires special handling/processing
- Ti-50 beams need development, and cross sections are small
- HFIR can produce Es-254, but only in small quantities

# Summary

- A new chemical element with atomic number Z = 117 has been synthesized
  - 2nd element discovered by ORNL, 9th discovery using ORNL isotopes
- 11 new heaviest known isotopes observed in element 117 decay chains
- General trend toward increased stability with increasing neutron number
- Longer half-lives offer the potential for chemistry studies to establish location in periodic table
  - On-line experiments in progress at Dubna
- A consistent picture of nuclear properties of heaviest nuclei is emerging
  - Critical role of nuclear shells
  - Experimental verification of the island of stability
- New targets and beams offer the potential for higher neutron numbers and extending the periodic table to even heavier elements