

The Discovery of Element 117

Presented to

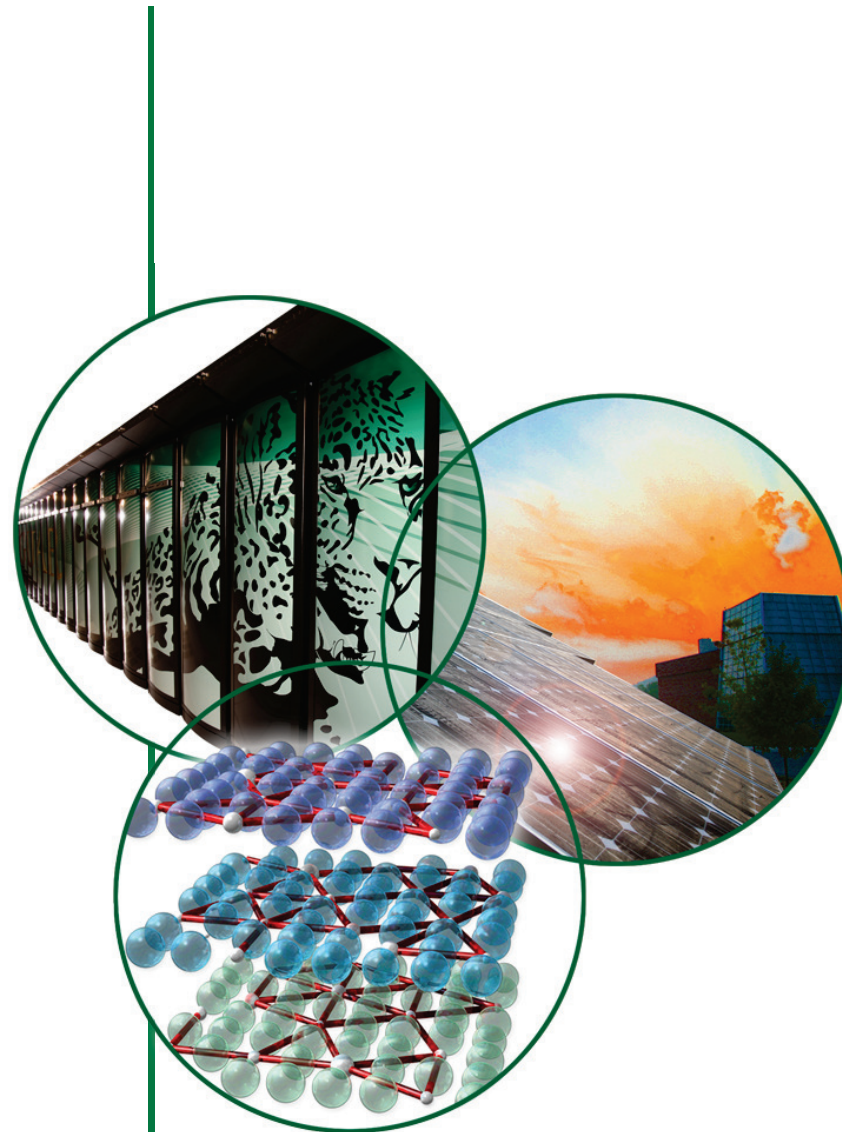
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Jim Roberto

Oak Ridge National Laboratory

Oak Ridge, Tennessee

July 21, 2010

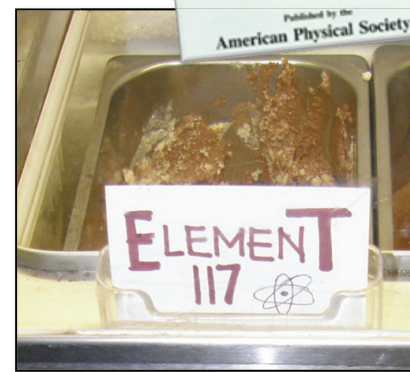
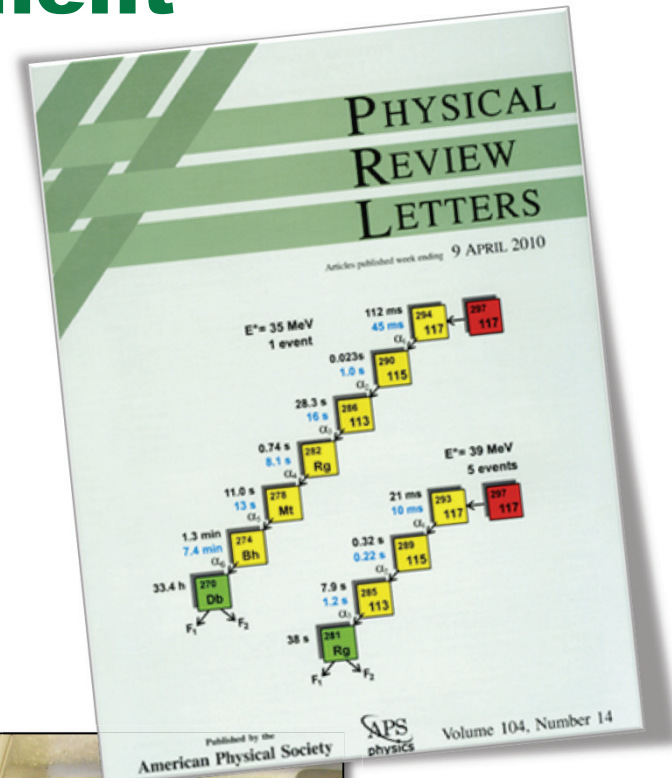


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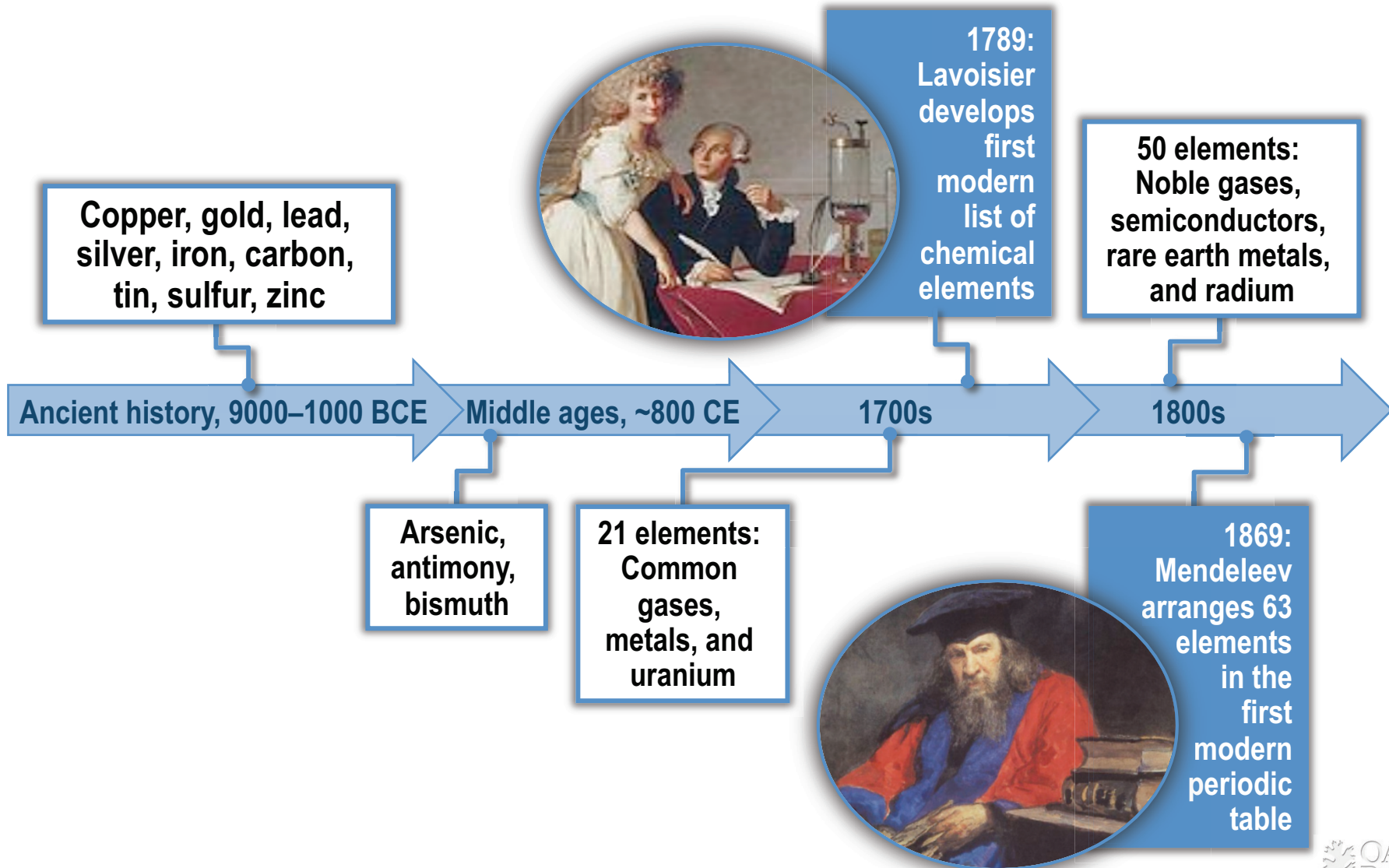
The discovery of element 117 is a historic accomplishment

- 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 discovery of chemical elements began more than 10,000 years ago



Benefits of Mendeleev's table

- Demonstrated trends/similarities by arranging elements in a two-dimensional network
- Reordered elements despite their accepted masses (early indication that atomic mass was not the determining factor)
- Allowed gaps in the table, predicting 10 new elements (7 actually exist)

In 1906, Mendeleev came within one vote of winning the Nobel Prize

ОПЫТЪ СИСТЕМЫ ЭЛЕМЕНТОВЪ.
ОСНОВАННОЙ НА ИХЪ АТОМНОМЪ ВѢСѢ И ХИМИЧЕСКОМЪ СХОДСТВѢ.

	Ti = 50	Zr = 90	? = 180.		
	V = 51	Nb = 94	Ta = 182.		
	Cr = 52	Mo = 96	W = 186.		
	Mn = 55	Rh = 104,4	Pt = 197,1.		
	Fe = 56	Ru = 104,4	Ir = 198.		
	Ni = Co = 59	Pt = 106,8	O = 199.		
H = 1	Cu = 63,4	Ag = 108	Hg = 200.		
Be = 9,1	Mg = 24	Zn = 65,2	Cd = 112		
B = 11	Al = 27,1	? = 68	Ur = 116	Lu = 197?	
C = 12	Si = 28	? = 70	Sn = 118		
N = 14	P = 31	As = 75	Sb = 122	Bl = 210?	
O = 16	S = 32	Se = 79,4	Te = 128?		
F = 19	Cl = 35,5	Br = 80	I = 127		
Li = 7	Na = 23	K = 39	Rb = 85,4	Cs = 133	Tl = 204.
		Ca = 40	Sr = 87,6	Ba = 137	Pb = 207.
		? = 45	Ce = 92		
		?Er = 56	La = 94		
		?Yt = 60	Di = 95		
		?In = 75,6	Th = 118?		

Д. Менделѣевъ

Mendeleev's periodic table (1869)

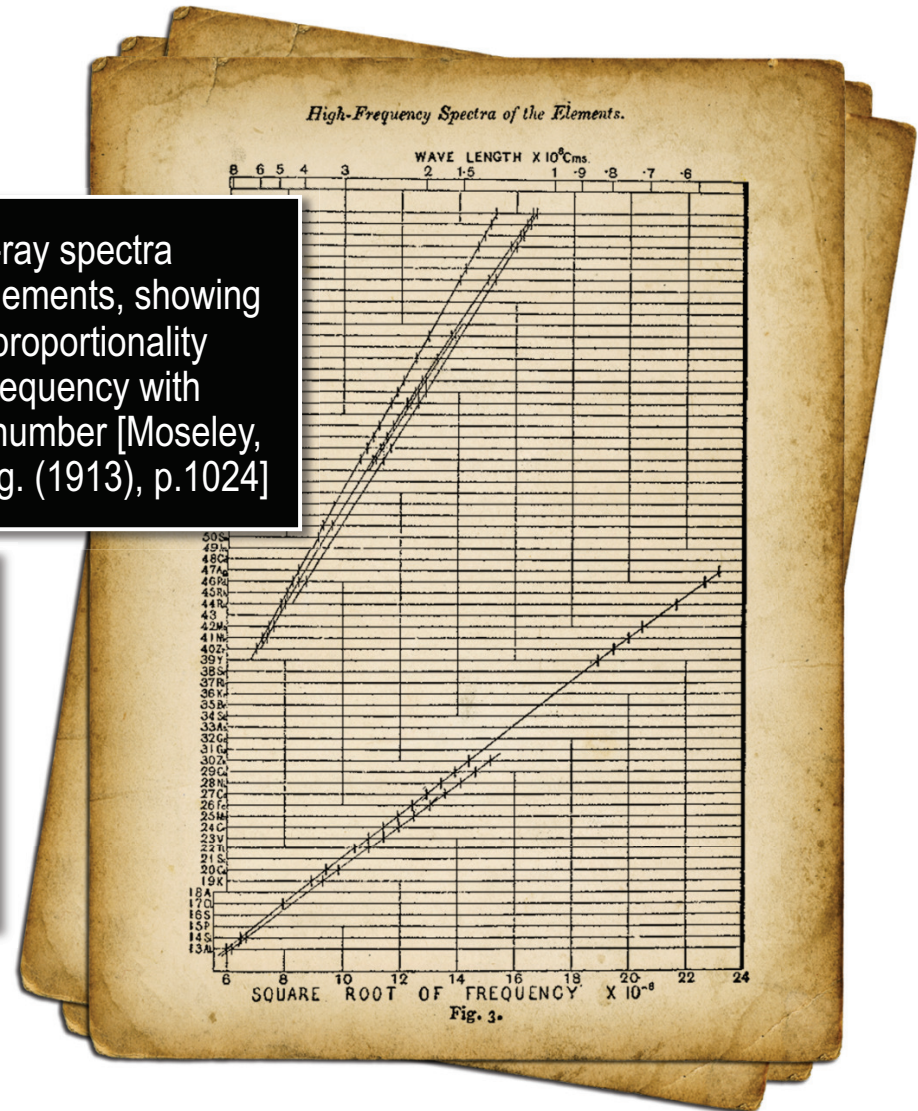
The importance of atomic number

- In 1913, Moseley showed that atomic number Z (the number of protons in the nucleus) is the determining factor in the periodic law
- Moseley's work revealed gaps at atomic numbers 43 and 61 (later filled by the discovery of technetium and promethium)
- By 1939, there were 91 known elements with uranium ($Z = 92$) having the highest atomic number



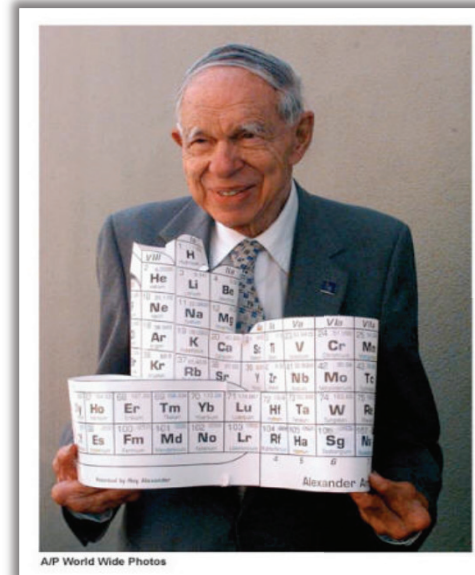
Henry Gwyn
Jeffreys Moseley
(November 1887–
August 1915)

X-ray spectra
of the elements, showing
the proportionality
of frequency with
atomic number [Moseley,
Phil. Mag. (1913), p.1024]



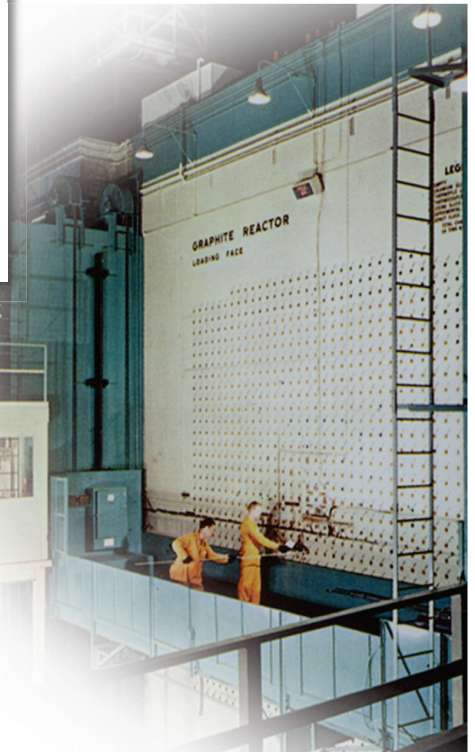
The synthesis of transuranics and element 61

- 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

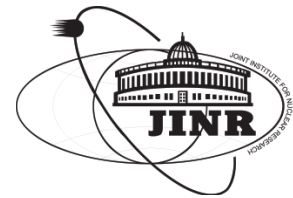
ORNL's Graphite Reactor, site of the discovery of element 61 (promethium) by Marinsky, Glendenin, and Coryell



Super-heavy elements, $Z > 103$

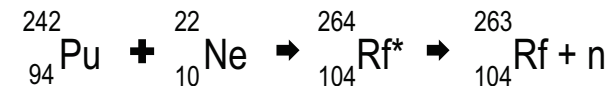
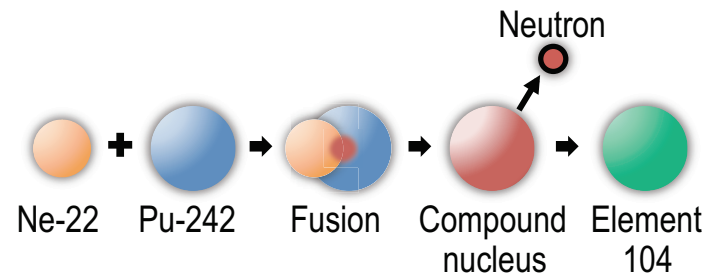
- 1960s: Existence of transactinides (super-heavy elements) first proposed by Seaborg
 - Half-lives ranging from hours ($Z = 104, 105$) to milliseconds ($Z = 115, 116, 118$)
 - Expected to be analogous in chemical properties to Hf, Ta, etc., and placed immediately beneath these elements in the periodic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

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



1	1																	2
	H																	He
2	3	4											5	6	7	8	9	10
	Li	Be											B	C	N	O	F	Ne
3	11	12											13	14	15	16	17	18
	Na	Mg											Al	Si	P	S	Cl	Ar
4	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
5	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54
	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
6	55	56	57	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86
	Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
7	87	88	89	Transactinides (super-heavy elements)														
	Fr	Ra	Ac~															

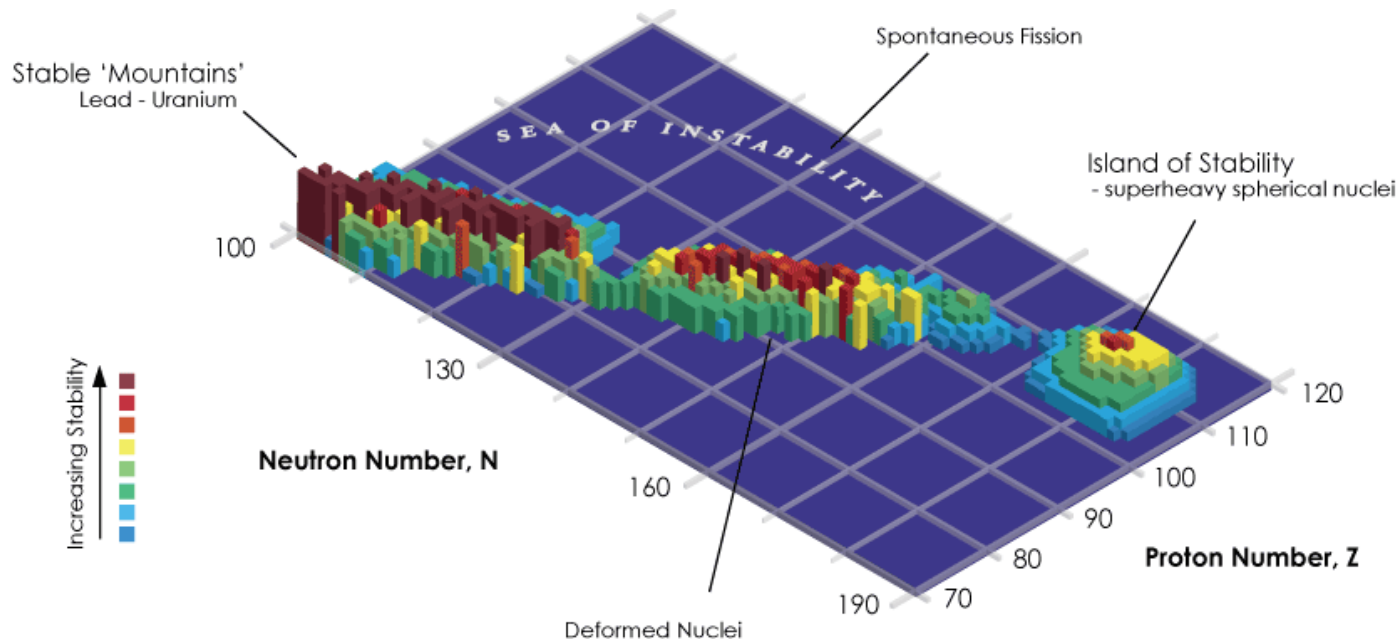
Lanthanides													
Actinides													



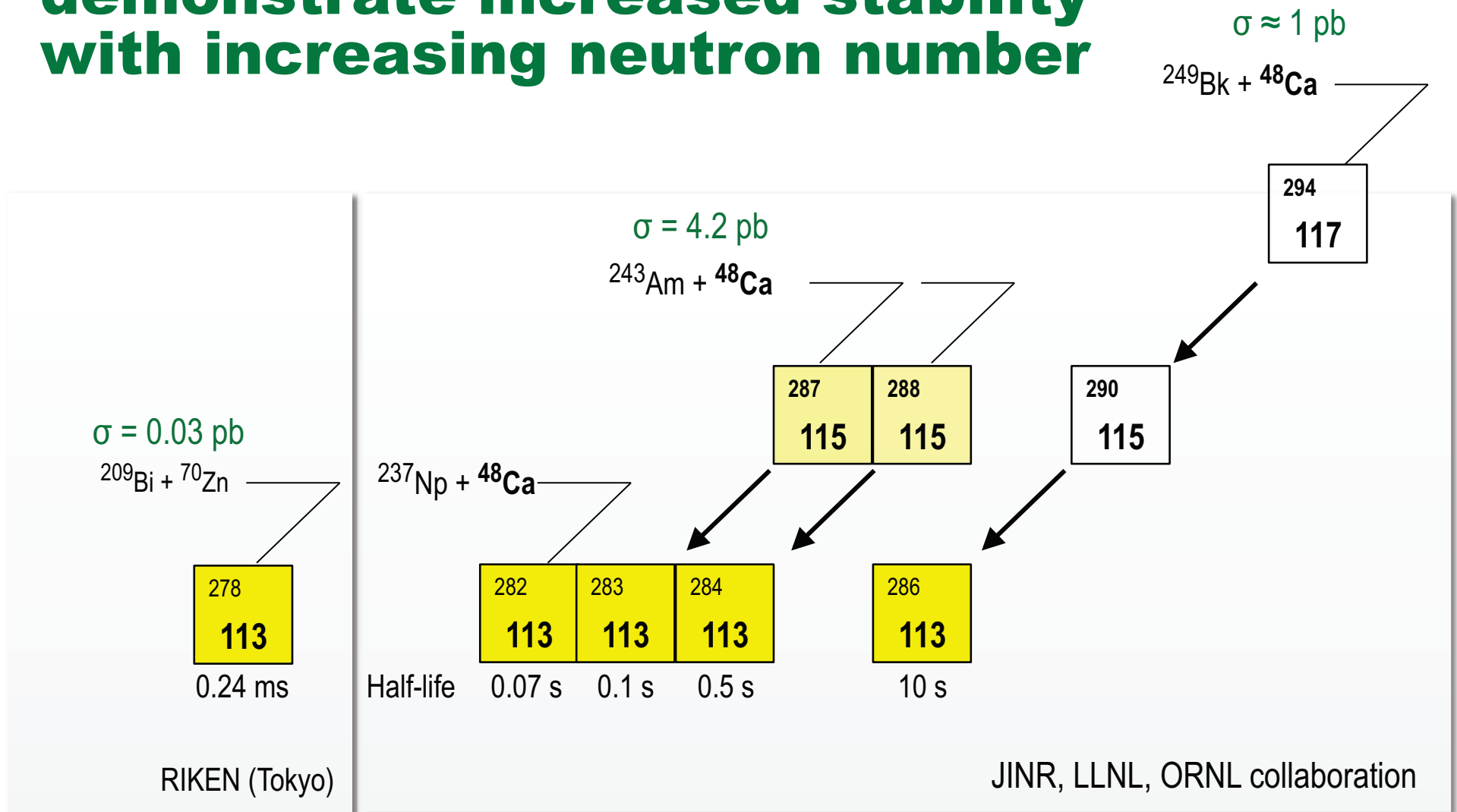
Bombarding Pu (94 protons) with Ne (10 protons) results in a compound nucleus, $Z = 104$ protons

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

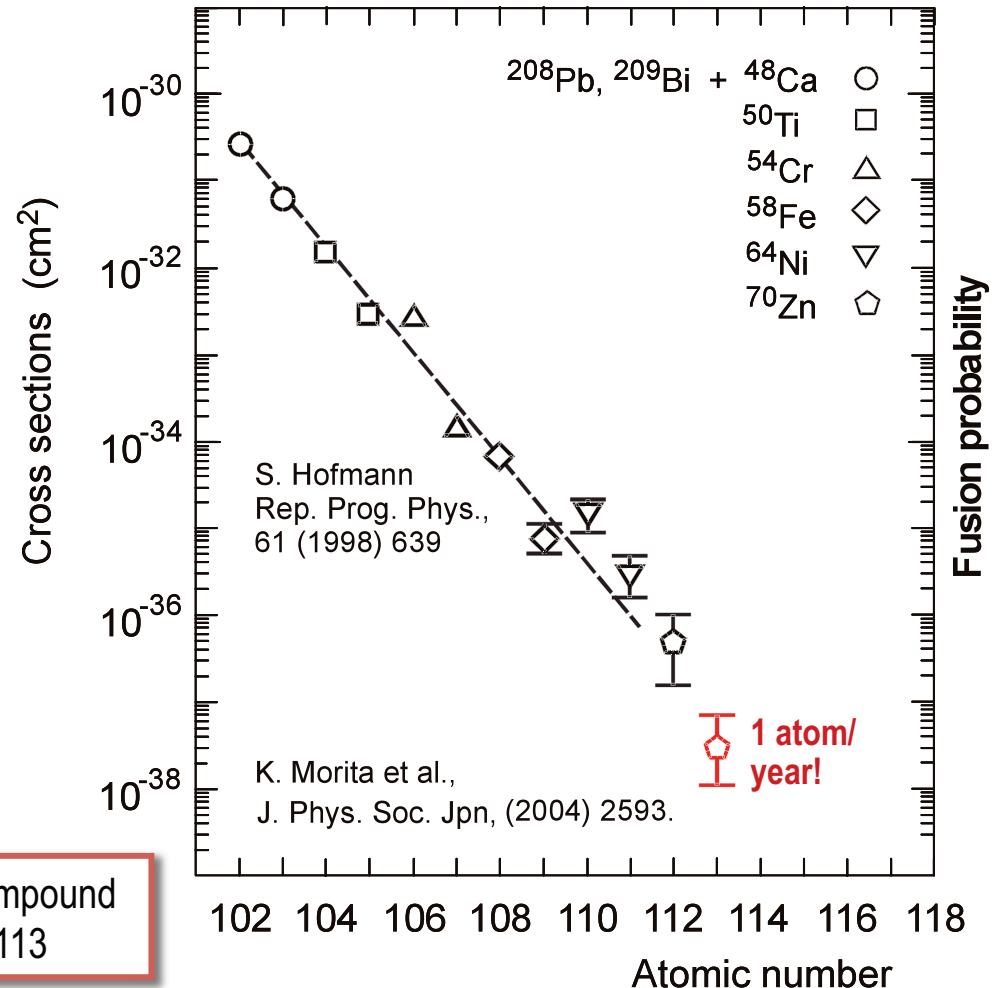


Y. Oganessian. Perspectives of JINR–ORNL collaboration in the studies of superheavy elements, JINR Scientific Council, September 24–25, 2009, Dubna

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 $Z = 112$

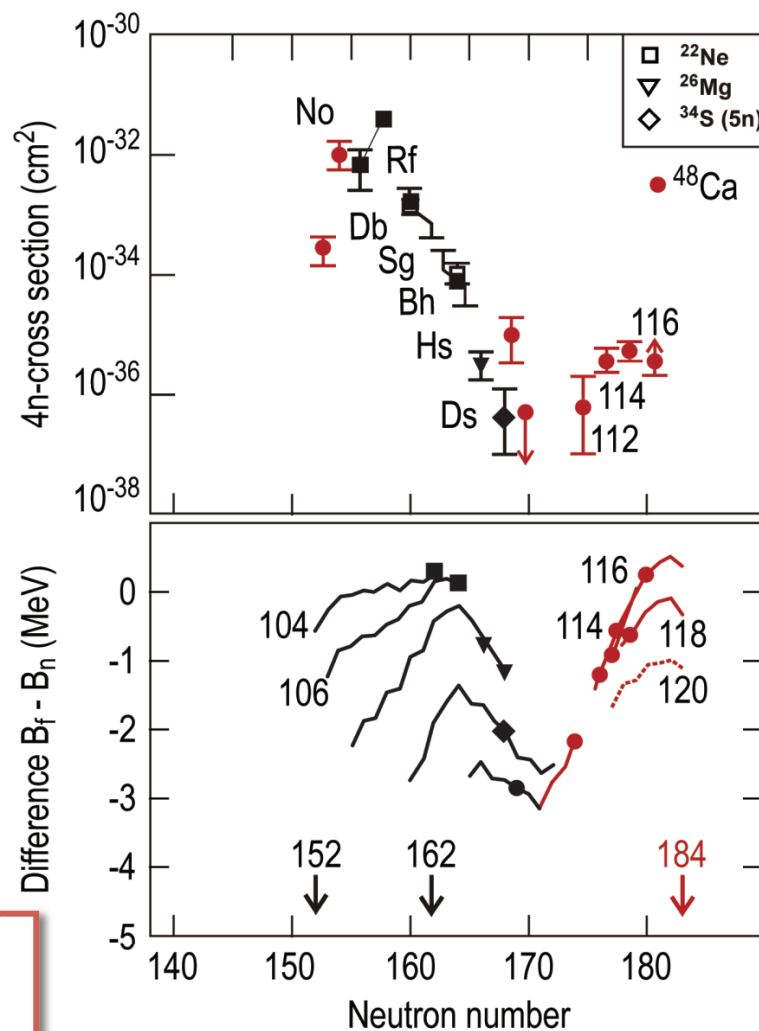
Cold fusion cross sections for compound nucleus formation up to $Z = 113$



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	$^{48}\text{Ca} \rightarrow ^{244}\text{Pu}$ (ORNL)	50 atoms
2004	113	JINR, Russia ¹	Decay product of element 115	8 atoms
2004	115	JINR, Russia ¹	$^{48}\text{Ca} \rightarrow ^{243}\text{Am}$ (ORNL)	30 atoms
2005	116	JINR, Russia ¹	$^{48}\text{Ca} \rightarrow ^{248}\text{Cm}$ (RIAR/ORNL)	30 atoms
2006	118	JINR, Russia ¹	$^{48}\text{Ca} \rightarrow ^{249}\text{Cf}$ (ORNL)	3 – 4 atoms
2010	117	JINR, Russia ²	$^{48}\text{Ca} \rightarrow ^{249}\text{Bk}$ (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
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

Actinide Series

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)**

Yu.Ts. Oganessian, F. Sh. Abdullin, S. N. Dmitriev, M. G. Itkis, Yu. V. Lobanov, A.N. Mezentsev, A. N. Polyakov, R. N. Sagaidak, I. V. Shirokovsky, V. G. Subbotin, A. M. Sukhov, Yu. S. Tsyganov, V. K. Utyonkov, A. A. Voinov, G. K. Vostokin

- **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

ORNL staff were involved in all aspects of the research

Lead-up to the discovery

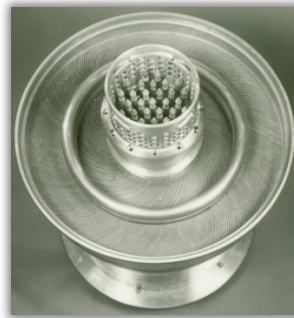
December 2004	<ul style="list-style-type: none">• Yuri Oganessian (JINR) proposes element 117 experiment to ORNL
February 2005	<ul style="list-style-type: none">• Oganessian presents colloquium at ORNL• JINR/ORNL/LLNL/Vanderbilt collaboration established• Experiment awaits next Cf campaign
September 2008	<ul style="list-style-type: none">• Oganessian, Roberto, and Joe Hamilton (Vanderbilt) meet in Nashville to coordinate planning for the experiment
November 2008	<ul style="list-style-type: none">• DOE authorizes Cf separation including research use of Bk
December 2008	<ul style="list-style-type: none">• ORNL and JINR sign Scientific Collaboration and Material Transfer Agreement
January 2009	<ul style="list-style-type: none">• Cf targets removed from HFIR (Bk decay clock starts ticking)
June 15, 2009	<ul style="list-style-type: none">• REDC-separated Bk shipped on schedule to Russia
July 2009	<ul style="list-style-type: none">• Bk target preparation at Dimitrovgrad (30 layers hand-painted on target wheel)
July 28, 2009	<ul style="list-style-type: none">• Experiment begins at Dubna (Ca-48 beam on target)



Academician Yuri Oganessian (right) with JINR staff member Vladimir Utyonkov

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 2.5×10^{15} neutrons/cm²·s (world's highest steady-state neutron flux)
 - 31 target positions (10–13 targets typically irradiated)
 - Produces ~35 mg ²⁵²Cf per target (smaller quantities of Bk, Es, Fm, others)



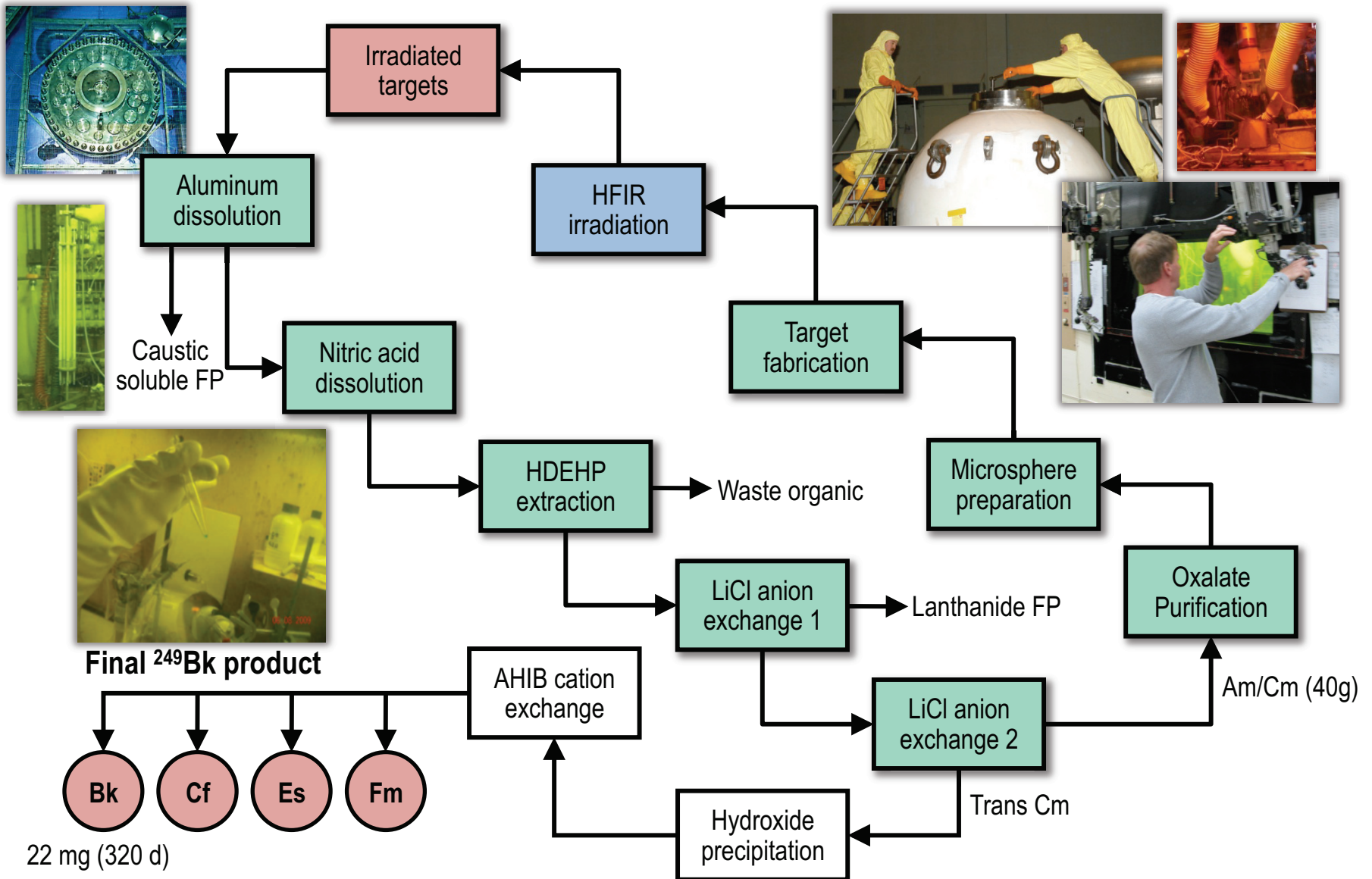
Target positions in the flux trap of a HFIR fuel element



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



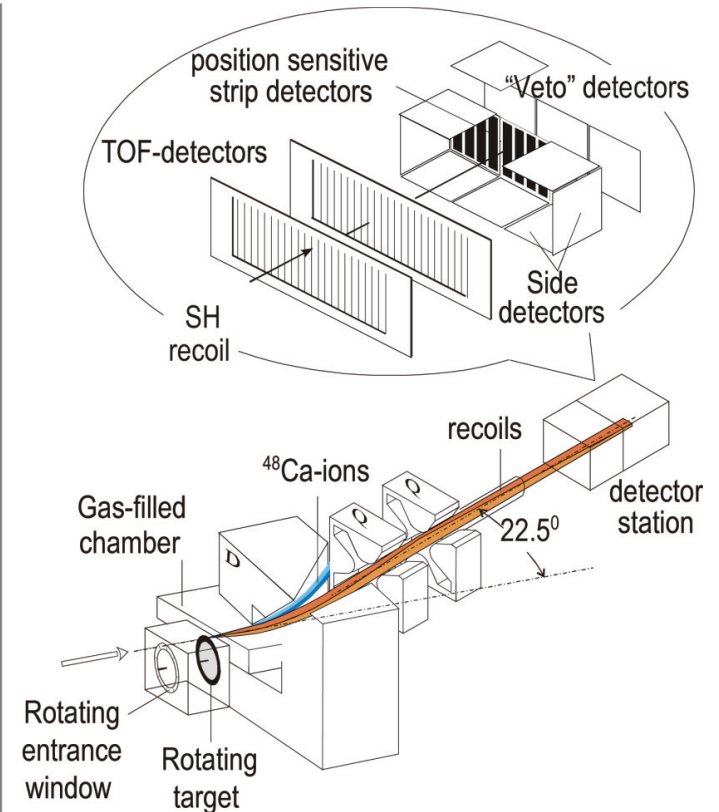
Bk packages being inspected
at Sheremetyevo Airport

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



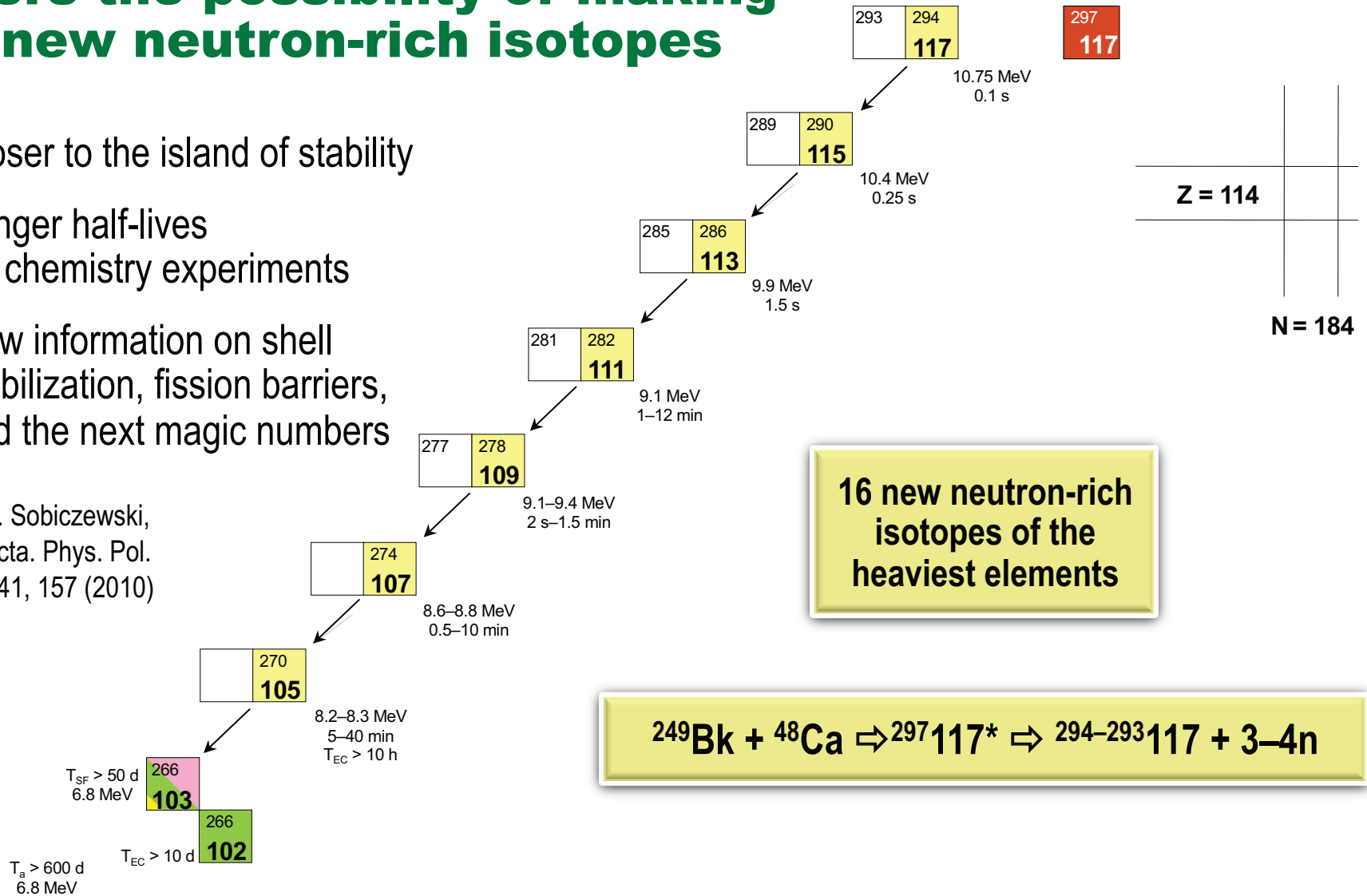
Heavy Ion Cyclotron U-400 at the Flerov Laboratory, JINR (Dubna)



The element 117 experiment offers the possibility of making 16 new neutron-rich isotopes

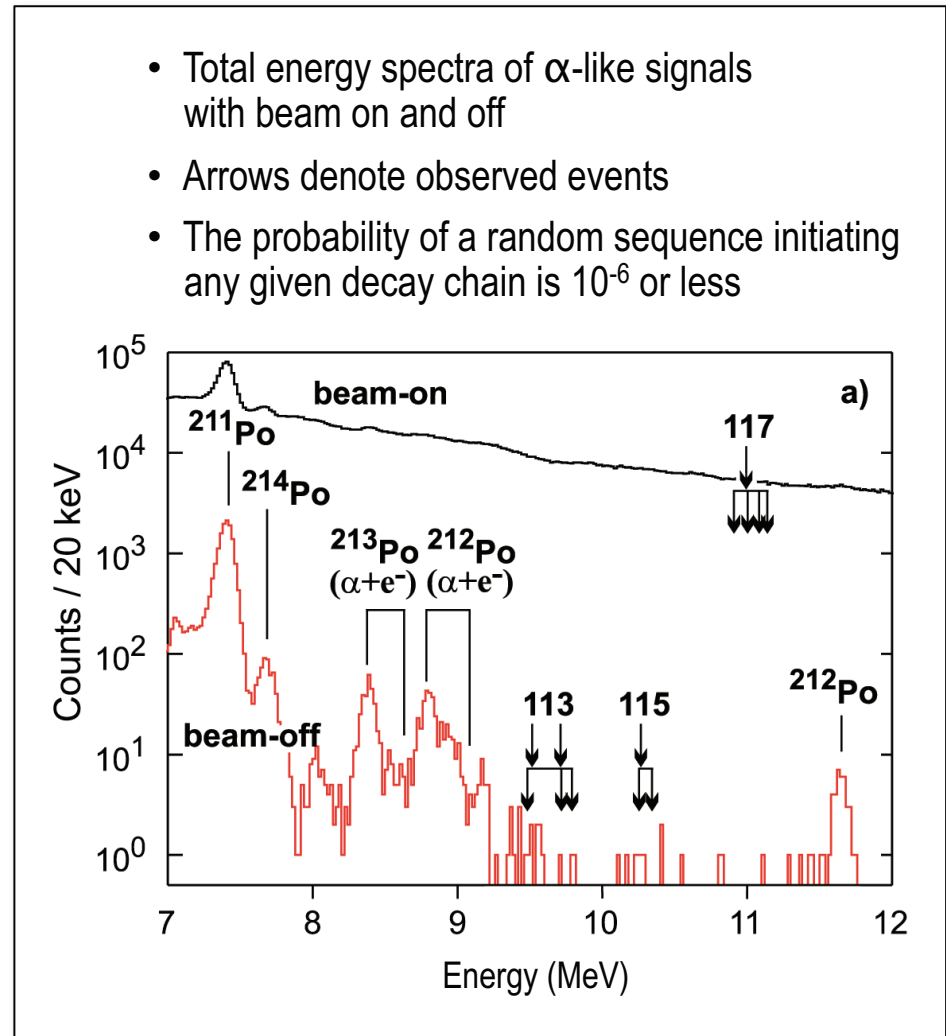
- Closer to the island of stability
- Longer half-lives for chemistry experiments
- New information on shell stabilization, fission barriers, and the next magic numbers

A. Sobiczewski,
Acta. Phys. Pol.
B41, 157 (2010)



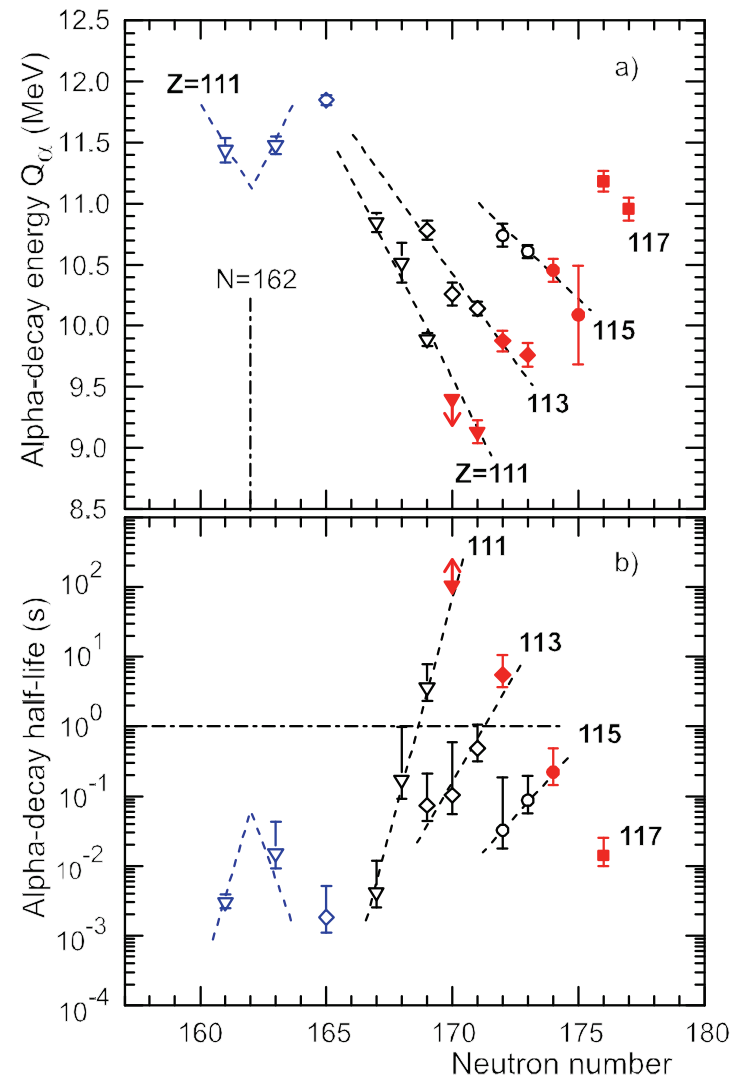
Energy spectra in the detector

- 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

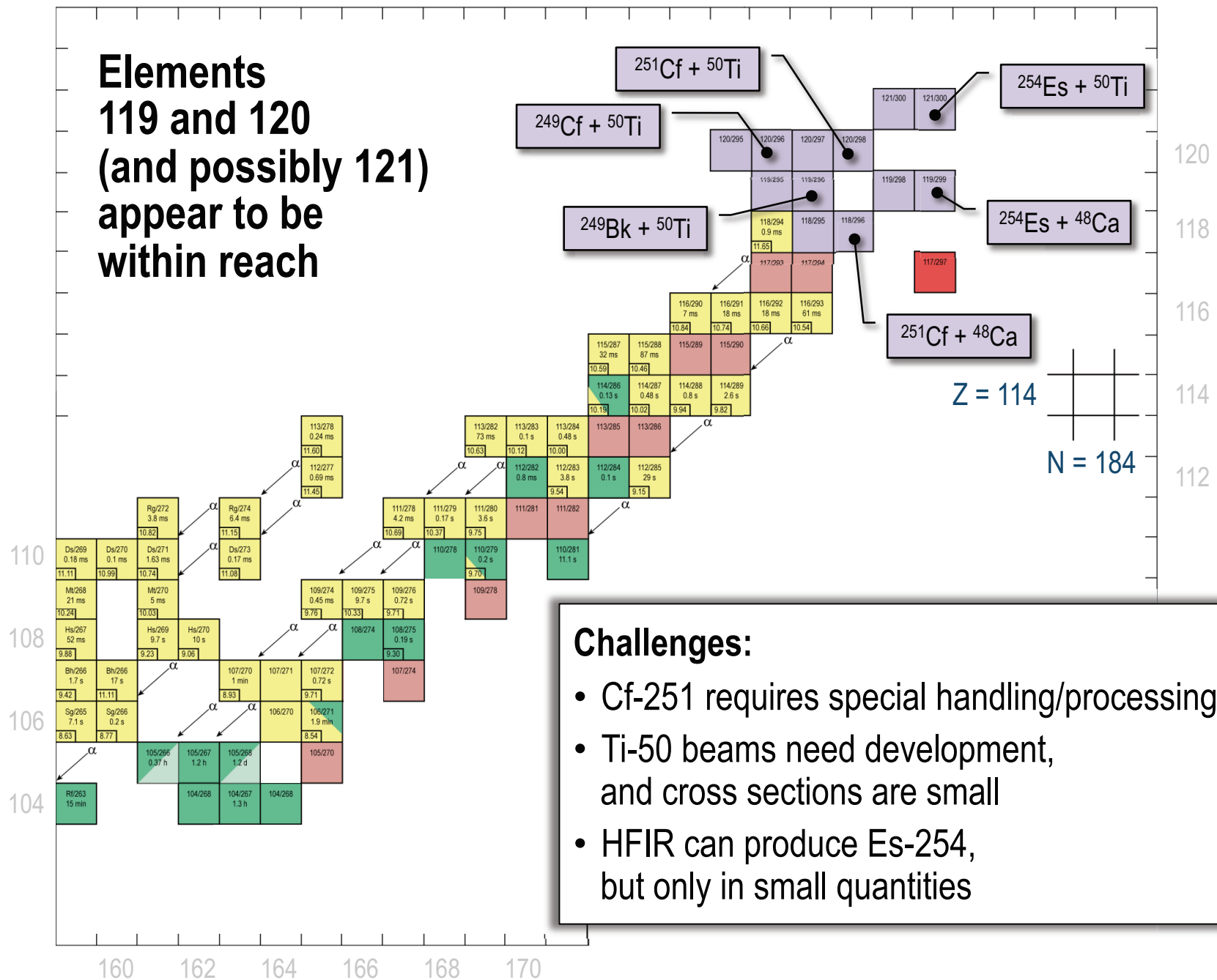


New isotopes from element 117 decay chains expand our knowledge of neutron-rich nuclei

- Closer approach to the shell at $N = 184$ results in a decrease in α -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

A portion of the periodic table showing elements from Lead (Pb) to Ununoctium (Uuo). Element 117, Ununseptium (Uus), is highlighted in green. The elements shown are:

82 Pb Lead 207.2	83 Bi Bismuth 208.98040	84 Po Polonium (208.9824)	85 At Astatine (209.9871)	86 Rn Radon (222.0176)
114 Uuq Ununquadium (289)	115 Uup Ununpentium (288)	116 Uuh Ununhexium (292)	117 Uus Ununseptium	118 Uuo Ununoctium