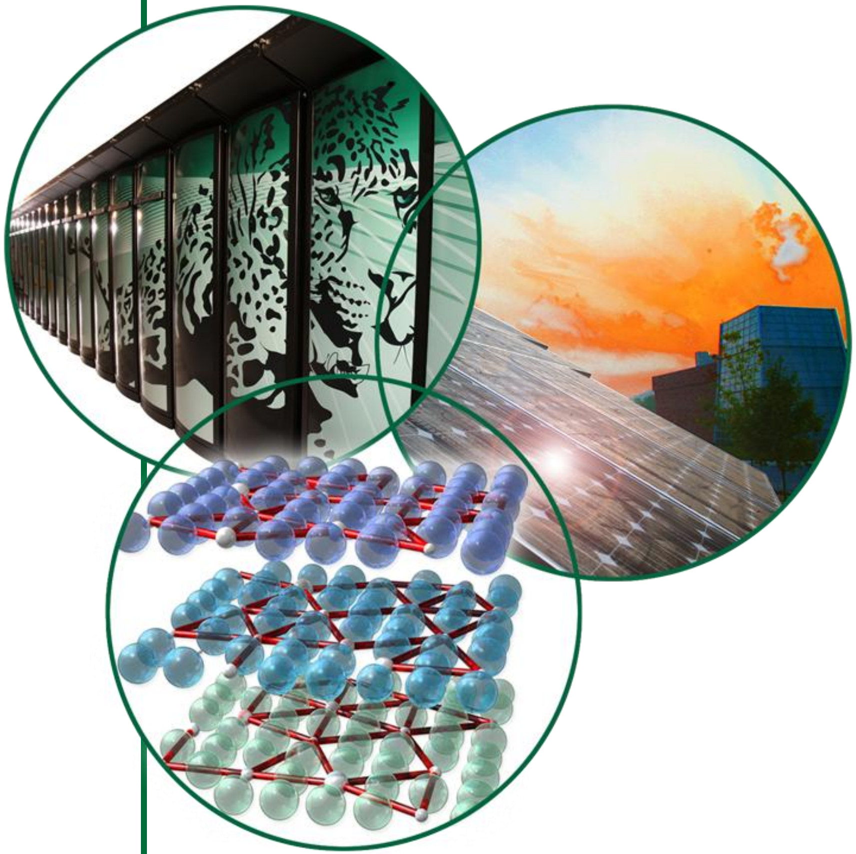
Transactinide research at Oak Ridge National Laboratory: Capabilities and priorities

Presented to
4th International Conference on the Chemistry and Physics of the Transactinide Elements

J. B. Roberto, J. L. Binder, and
K. P. Rykaczewski
Oak Ridge National Laboratory
Oak Ridge, Tennessee, USA

Sochi, Russia
September 5-11, 2011



U.S. DEPARTMENT OF ENERGY

OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

Outline

- Historical perspective
- Actinide production and processing at ORNL
- ORNL's role in transactinide research
- Research plans and priorities

Mission of Oak Ridge National Laboratory in 1943:

Produce gram quantities of plutonium for chemical and engineering research

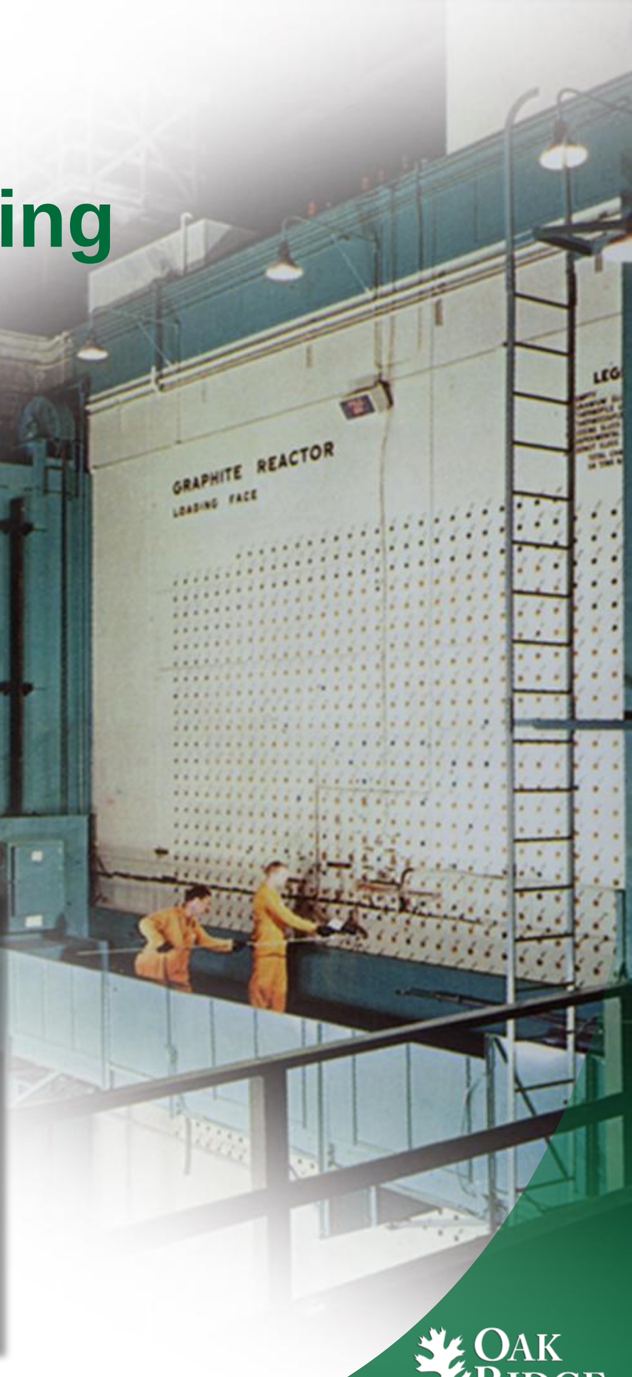
- **Construct the world's first operational nuclear reactor**
- **Develop chemical processing to separate plutonium from irradiated fuel**

Oak Ridge National Laboratory

Success of the wartime effort led to an expanded mission for ORNL:

Basic and applied research utilizing the Graphite Reactor

- Science and engineering of the nuclear fuel cycle
  - Materials and fuels
  - Separations chemistry
  - Reactor technology
- Development of neutron scattering, neutron activation analysis, carbon-14 tracer analysis, etc.
- **Development, production, and distribution of radioisotopes**



Oak Ridge National Laboratory

Early development, production, and distribution of radioisotopes at ORNL

Wigner envisions radioisotopes as a major activity for a postwar laboratory devoted to nuclear research

1944

June 1946

Manhattan Project announces availability of radioisotopes for scientific and medical use through Clinton Laboratories

SCIENCE

Vol. 103, No. 2685 — Friday, June 14, 1946

Availability of Radioactive Isotopes

Announcement From Headquarters, Manhattan Project, Washington, D.C.

PRODUCTION OF TRACER AND THERAPEUTIC RADIOISOTOPES has been heralded as one of the great peacetime contributions of the uranium chain-reacting pile. This use of the pile will unquestionably be rich in scientific, medical, and technological applications.

Manhattan Project scientific, technical, and administrative personnel have, since the inception of the pile, been cognizant of its ...

... absorbed in the introduced material that the chain reaction ceases even though the control rods are withdrawn as far as feasible. With available pile facilities, this limit does not permit the production of a sufficient quantity and quality of many radioisotopes to meet anticipated ...

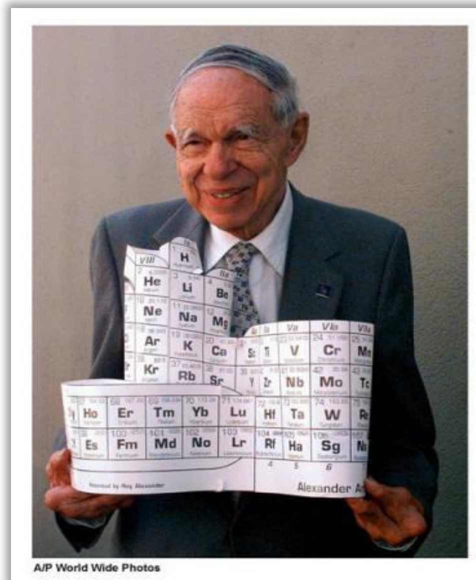
August 1946

First isotope shipment from Clinton Laboratories (carbon-14 to Barnard Hospital in St. Louis)

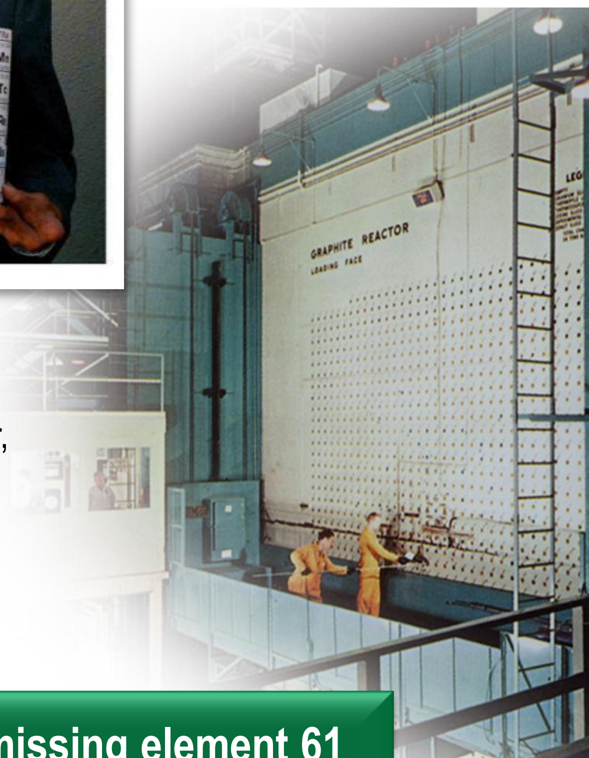
Discovery of the actinides and element 61

- Transuranium 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 advocated construction of HFIR/REDC at ORNL**
- Seaborg proposed a new row (the actinides) to accommodate these elements in the periodic table

* With the exception of trace amounts of Np and Pu



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)

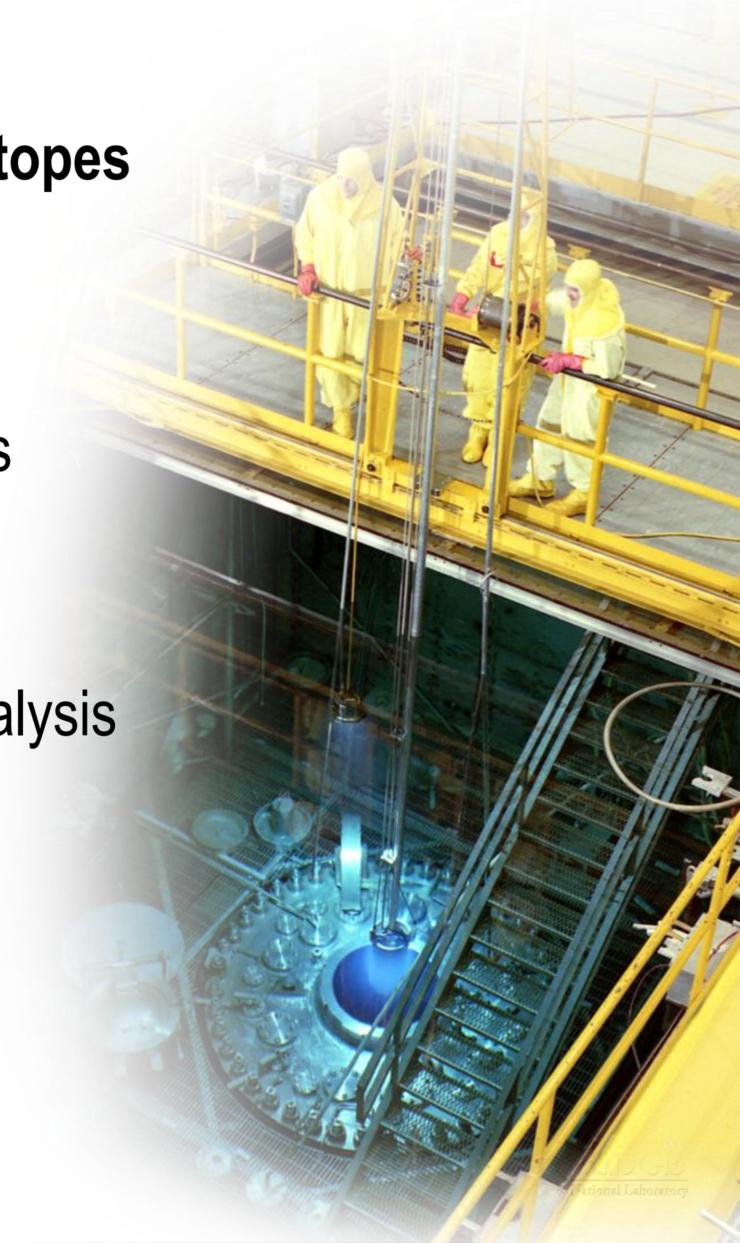
Scientists at Oak Ridge National Laboratory separated the missing element 61 from fission products in 1945, completing the lanthanide series

Isotope research and production facilities at ORNL

HFIR/REDC is a key source of isotopes for a wide range of customers

HFIR/REDC is a leading source for many isotopes

- Science: Heavy element production
  - Cf, Es, Bk, Fm
- Medicine: Diagnostic and therapeutic isotopes
  - Alpha emitters (e.g., Ac-225/Bi-213, Ra-223
  - Beta emitters (e.g., W-188/Re-188, Lu-177)
- Industry: Energy production and materials analysis
- Security
  - Detection of explosives and narcotics
  - HEU downblending monitoring systems

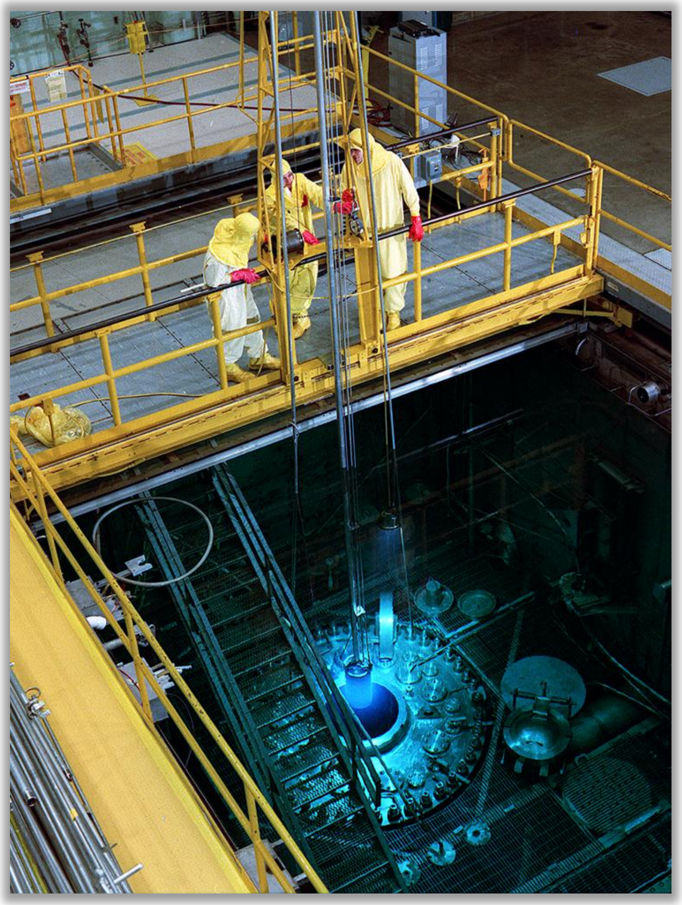


Actinide production by irradiation Am/Cm targets in HFIR

- Targets specially designed for reactor conditions:
  - Composition controls fission and gamma heating
  - Targets remain in the reactor for 8-9 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 ^{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)

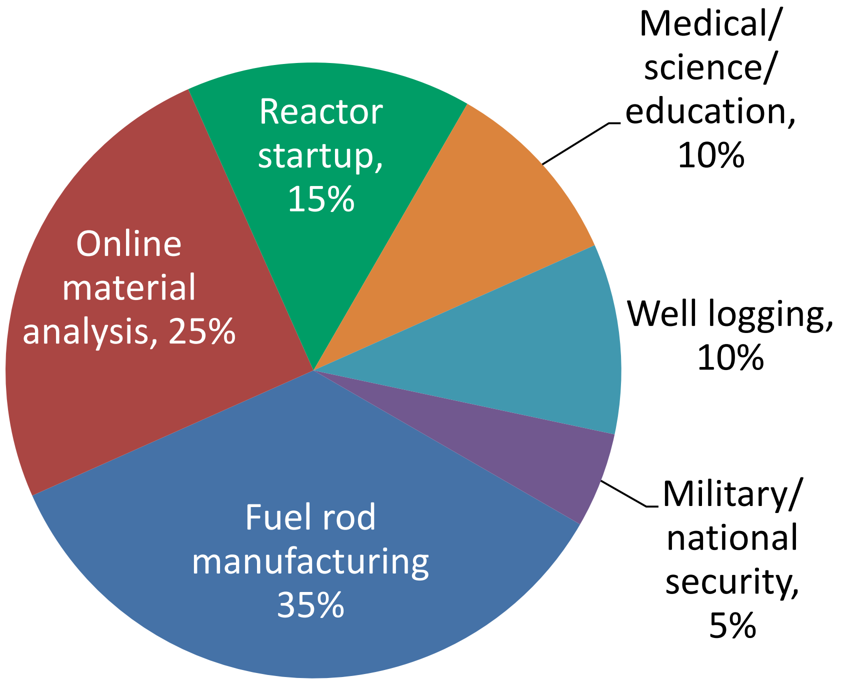
^{252}Cf produced using HFIR is critical to a variety of industries worldwide

Energy	Industrial	Security
• Nuclear fuel quality control • Reactor start-up sources • Coal analyzers • Oil exploration	• Mineral analyzers • Cement analyzers • FHA measurements for corrosion (bridges, highway infrastructure)	• Handheld contraband detectors (CINDI) • Standard for all neutron fission measurements • Monitoring downblending of HEU • Identifying unexploded chemical ordnance and detecting land mines

Demand for ^{252}Cf continues to increase

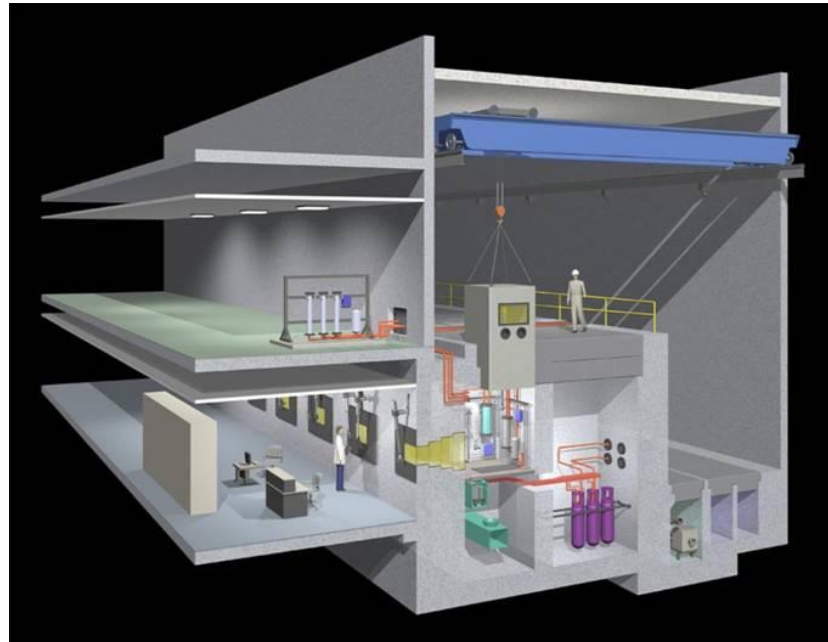
- Cf sales account for ~40% of DOE isotope revenue
- HFIR/REDC supplies the majority of the world's Cf
- Year-to-year fluctuations, but overall trend shows increase in sales
  - Expected growth in material analyzer sales
  - Expected growth in nuclear industry, requiring additional sources
  - Reactor startup sources require ^{252}Cf (no alternative source available)

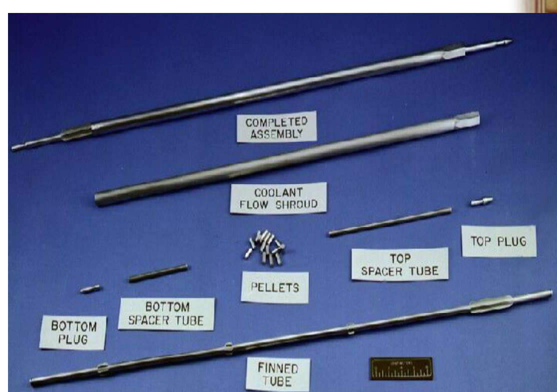
^{252}Cf market segments



REDC performs transuranium target fabrication and processing

- Heavily shielded hot cells
  - Dedicated to pellet production and target fabrication
  - Chemical processing
  - Sample analysis
  - Waste handling
- Shielded caves and glovebox labs for product purification and R&D
- Radiochemical analytical labs





Medical isotope processing at REDC: Clinical trials of Ac-225 and Bi-213

Memorial Sloan-Kettering Cancer Center, New York	Acute myeloid leukemia (clinical trial), ovarian cancer
Johns Hopkins School of Medicine, Baltimore	Breast cancer
Albert Einstein College of Medicine, New York	Fungal, bacterial, and viral (HIV) infections
Department of Nuclear Medicine, Technical University Munich	Gastric, ovarian and bladder cancer
University Hospital Düsseldorf	Non-Hodgkin lymphoma
INSERM, Nantes	Multiple myeloma (clinical trial)
University Hospital Basel	Brain tumors (clinical trial), prostate cancer
St. George Hospital and Centre for Experimental Radiation Oncology, Sydney	Malignant melanoma; prostate, pancreatic, breast, and ovarian cancer

July 2011: New cooperative research agreement with Institute for Transuranium Elements (ITU) to supply Ac-225 for cancer therapy trials

Inventories of selected actinide isotopes at ORNL

Isotope	Approximate amount (mg)	Isotopic %	Notes
Pu 242	5500	>99%	
Pu 244	2750	Various	650 mg at 8.7%, 2100 mg at 17.8%
Am 241	3500	>99%	
Am 243	1000	>99%	
Cm 244	1000	>90%	
Cm 248	2500	Various	90 mg at > 95%, 1700 mg at > 90%
Bk 249	—	>99%	Requires HFIR production and chemical processing (~20 milligrams per campaign)
Cf 249	170	>99%	
Cf 251	90	10-40%	Requires chemical processing and isotopic separation

Using ^{48}Ca beams with actinide targets, JINR 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[3]
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
[3] 22 additional atoms of element 115 observed at JINR in 2011

All of these discoveries used ORNL isotopes

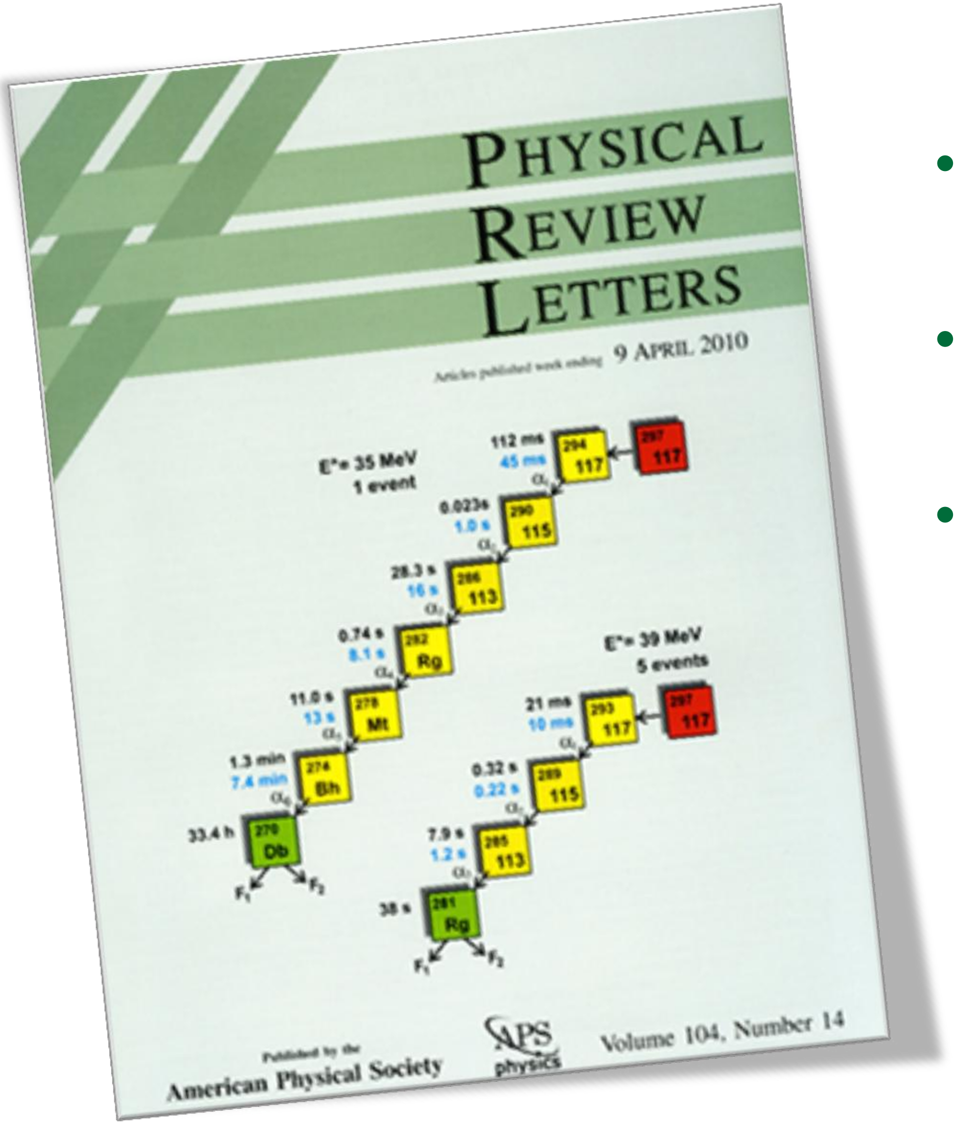
The periodic table in 2009

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

The discovery of element 117



- ^{249}Bk + ^{48}Ca reactions at JINR (Bk from ORNL)
- Confirms the "island of stability" for super-heavy elements
- Includes the discovery of 11 new heaviest known isotopes with atomic numbers 105–117

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

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

HFIR/REDC reactor/hot cell complex (ORNL)

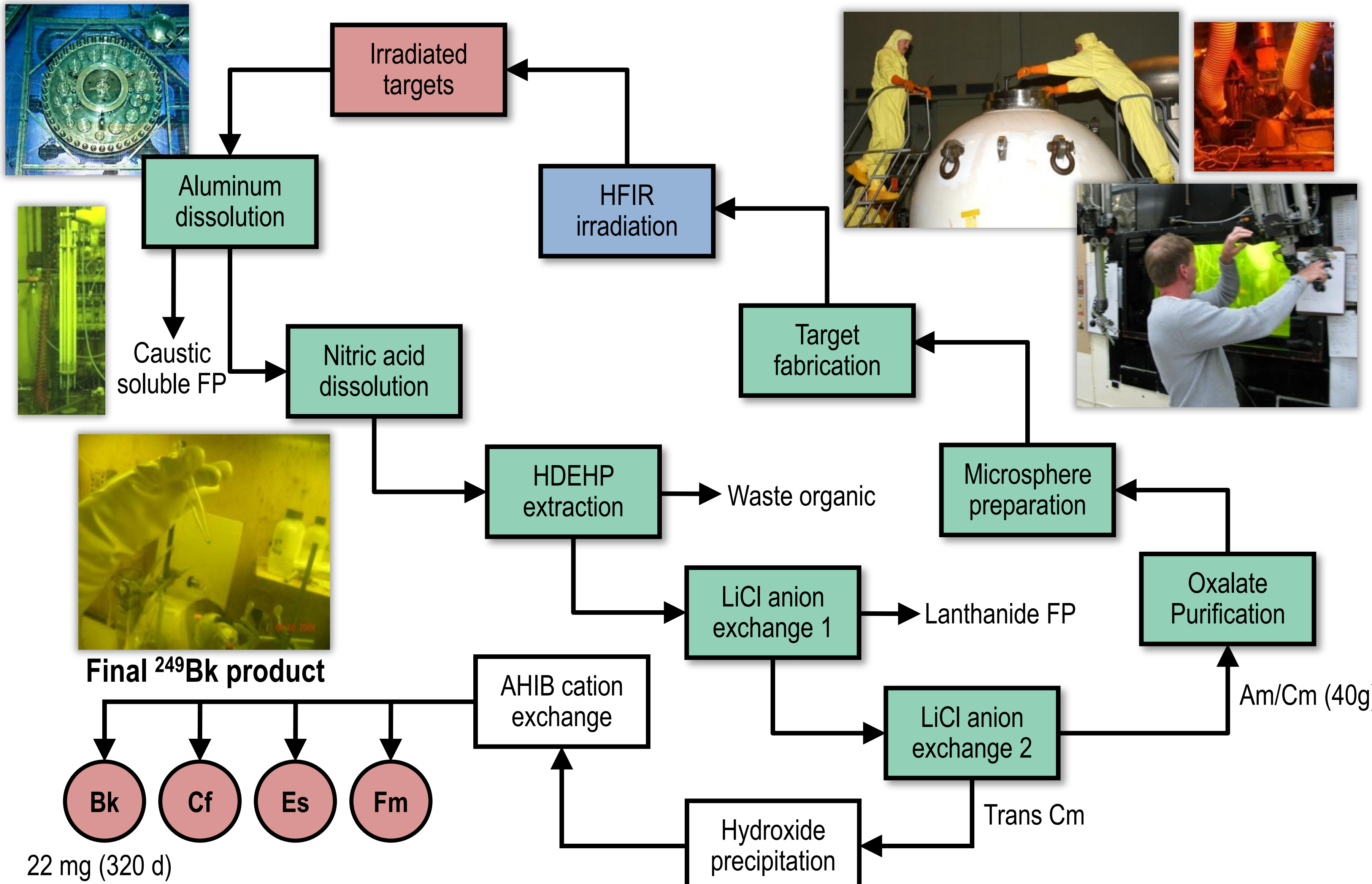
Flerov Laboratory (Dubna)

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

International collaboration was essential

Bk production/separation cycle at HFIR/REDC



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

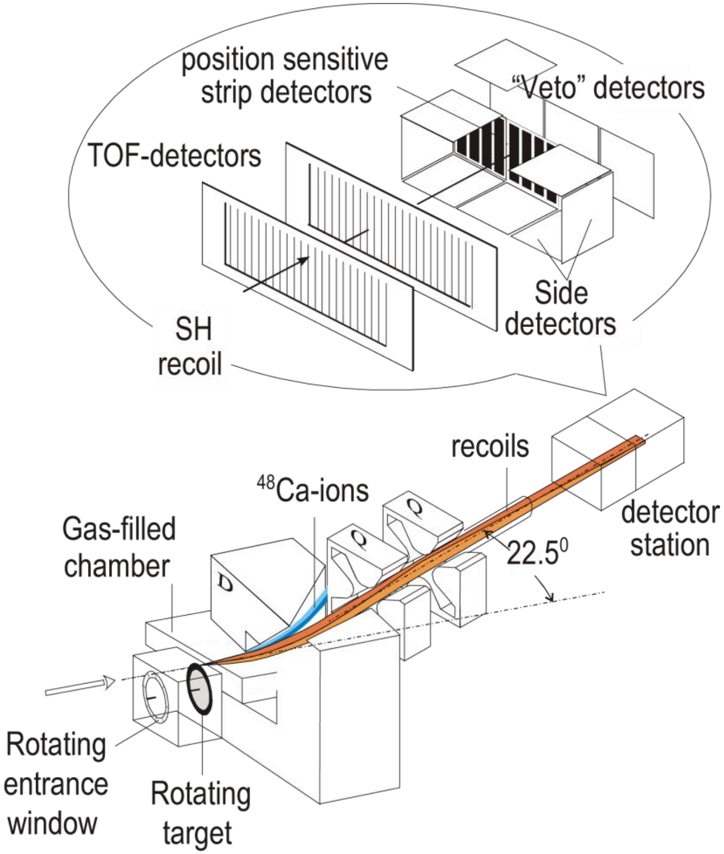
The final product (starting from 40 g of irradiated Am/Cm) is the green speck at the bottom of the glass vial, 22 mg of ultrapure Bk

Oak Ridge National Laboratory

Superheavy element synthesis and detection at the Dubna Gas Filled Recoil Separator

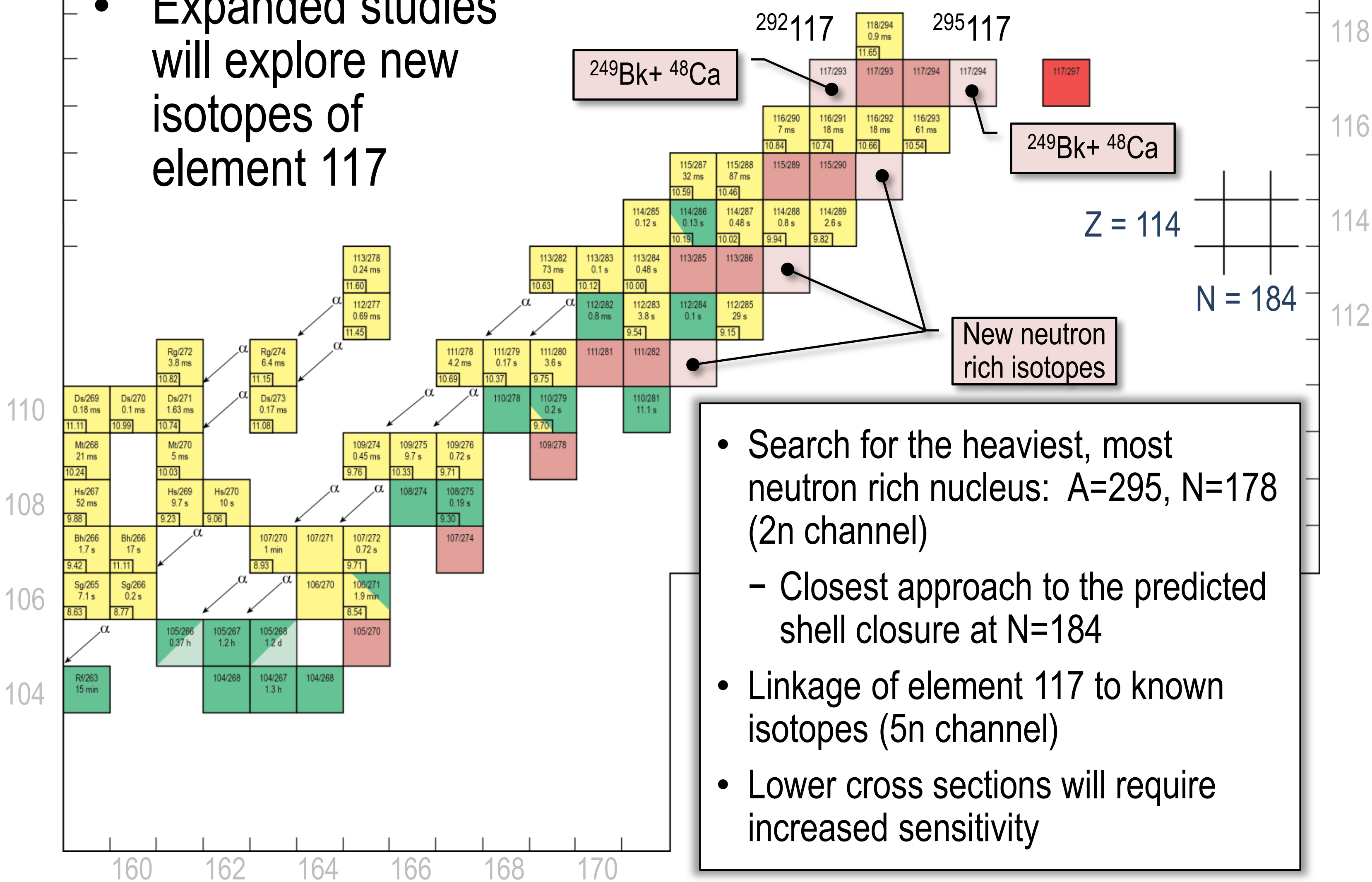
- ^{48}Ca beam supplied by the U400 cyclotron
- Total beam dose >10^{19} particles
- Rotating target distributes beam heating
- Rapid separation allows detection of nuclei with short half-lives
- Suppression factors are 10^{15} for beam particles and 10^{4} for target-like particles

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



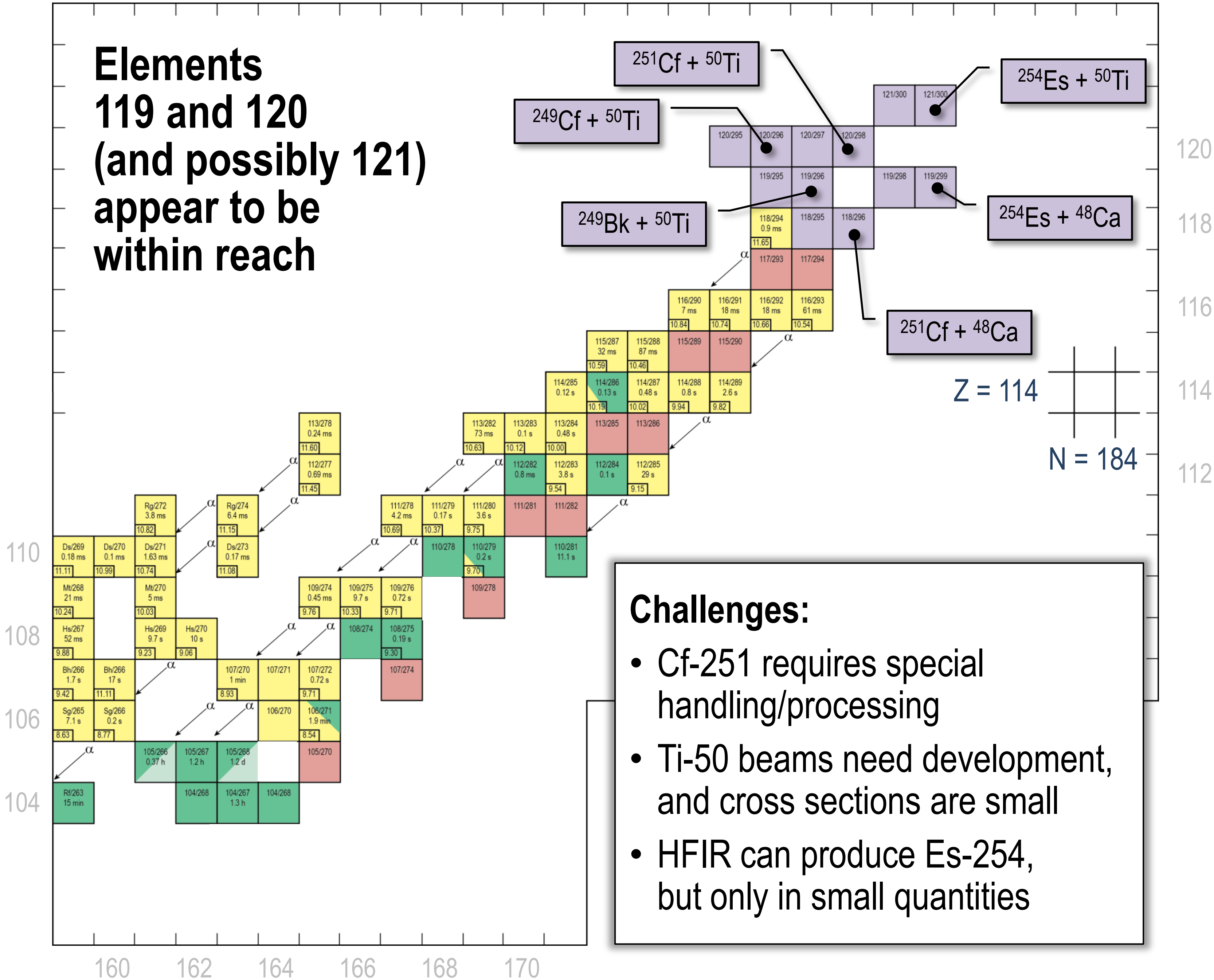
Element 117 results include eleven new heaviest isotopes: $^{293}117$, $^{294}117$, and decay products

- Expanded studies will explore new isotopes of element 117



- Search for the heaviest, most neutron rich nucleus: A=295, N=178 (2n channel)
  - Closest approach to the predicted shell closure at N=184
- Linkage of element 117 to known isotopes (5n channel)
- Lower cross sections will require increased sensitivity

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

Excerpts from the DOE/NSF Nuclear Science Advisory Committee Isotopes Subcommittee (2009)

- Research priority: Create and understand the heaviest elements possible
- Recommended actions include:
  - Make certain actinides in HFIR for accelerator-based experiments to make superheavy elements
  - Perform R&D to prepare for the reestablishment of a domestic source of mass-separated stable and radioactive research isotopes
- ORNL has reestablished actinide production at HFIR and is building a prototype 10 mA stable isotope separator
  - This prototype may lead to an actinide separator

The DOE Office of Nuclear Physics is responsible for research isotopes

- A new program: Isotope Development and Production for Research and Applications
- Includes R&D, infrastructure, and production and distribution of research isotopes
- Based on established DOE Office of Science practices, we anticipate
  - Research and production priorities developed with consideration of NSAC and community input (coordinated by the DOE National Isotope Development Center)
  - Distribution of research isotopes based on competitive, peer-reviewed proposals
  - Financial models are under consideration

ORNL plans/priorities in transactinide R&D

- Work with JINR and GSI partners to confirm element 117, discover new isotopes of element 117, and search for elements 119 and 120
  - Accelerator-bombardment with new Bk targets begins in April 2012
- Work with the DOE Office of Nuclear Physics to ensure:
  - Continued production and availability of actinide materials from HFIR/REDC
  - Development and implementation of an actinide separator for research isotopes
- Future research priorities
  - Collaborative experiments on neutron-rich isotopes of element 118, using Cf-251+Ca-48 reactions
  - Collaborative experiments on element 120, using Cf-251+Ti-50 reactions

Oak Ridge National Laboratory:
Science and technology for the 21st century