85th Birthday Symposium for Prof. John R. Huizenga

University of Rochester, April 21, 2006

Nuclear Chemistry at the Frontiers: Reminiscences and Future Challenges

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I had just returned from the 1989 conference in Berlin celebrating the 50th anniversary of the discovery of nuclear fission where I had given a talk on our recent research on properties of spontaneous fission in the heaviest elements, especially the fermium (100) isotopes. When I returned I found my graduate students had picked up on the cold fusion" claims which were being highly publicized and were already trying to detect 'excess neutrons" –I assured them that if there were as many excess neutrons generated as these claims would indicate we had better be very careful!

Pres. George H. W. Bush then asked Seaborg to come To Washington, D. C. on April 13, 14 to meet with him and Secretary of Energy James Watkins to ask Seaborg's advice about what action (if any) should be taken and whether to allocate resources to investigate or follow up on these claims

"COLD FUSION"

March 13, 1989: Paper submitted. "Electrochemically Induced Nuclear Fusionof Deuterium", *J. Electroanal. Chem*., 261, 301 (1989). Revised form received March 22. (M. Hawkins was added in one of many 'errata' to original article.)

April 27, 1989: Jones et al. publish on very low levels of neutrons, A Nature 338, 737 (1989)

"More Searches for Cold Fusion", R.A. Henderson, K.R. Czerwinski, H.L. Hall, K.T. Lesko, E.B. Norman, B. Sur, D.C. Hoffman, J. Fusion Energy, 9, 475 (1990).

Seaborg briefing Pres. Geo. H. W. Bush White House, April 14, 1989

We subsequently published a paper entitled,
"More Searches for Cold Fusion",
R. A. Henderson, K.R. Czerwinski, H.L. Hall, K.T. Lesko,
E.B. Norman, B. Sur and D. C. Hoffman.
J. of Fusion Energy, Vol. 9, 1990.

From observed D/Pd ratios in the cathodes,
we set very low limits on the cold fusion reactions
$d(d,n)^3He$ and $d(d,\gamma)^4He$.

Our results were inconsistent with Pons,
Fleischmann &Hawkins results (J. Electroanal. Chem.
261, 301 (1989) and even a factor of 10 below Jones et
al. reported very low level of neutrons generated.
Nature, 388, 737 (1989).

June 5, 1995 Chem. & Eng. News

Cold Fusion Believer Turned Skeptic Crusades For More Rigorous Research

■ ***Physicist Steven Jones, one originator of cold fusion, retracted previous claims and is pushing others to be as diligent***

Ron Dagani, C&EN Washington

Steven E. Jones has come a long way in six years. In 1989, he believed that cold nuclear fusion in an electrolysis cell was possible because he had detected the resultant neutrons in his own laboratory. But over the years, as he looked more and more carefully at the phenomenon, checking and rechecking, his doubts grew. Finally, last year, he retracted some of his original findings.

And just a few weeks ago, Jones, an associate professor of physics at Brigham Young University in Provo, Utah, took

Jones's cold fusion findings came to light at about the same time that electrochemists Martin Fleischmann and Stanley Pons announced their bombshell at a 1989 press conference at the University of Utah, Salt Lake City. The two chemists reported that the electrolysis of heavy water (D_2O) using a palladium cathode produced large amounts of heat—much more than could be accounted for by any chemical process. They also claimed to observe tritium and neutrons at much lower levels, not commensurate with the excess heat. They concluded that they were seeing the telltale signs of a nuclear reaction, perhaps D + D fusion, inside the deuterium-packed palladium electrode.

These incredible results were far more dramatic than what Jones and his coworkers were claiming: Using a similar electrolysis cell and an advanced neutron detector, Jones measured a neutron flux that was just above the neutron background. His neutron intensity was some five orders of magnitude lower than the intensity claimed by Fleischmann and Pons. And it was 13 orders of magnitude lower than the neutron flux that would be expected from Fleischmann and Pons's cells if their excess heat were produced by conventional D + D fusion.

Unlike Fleischmann and Pons, Jones performed no calorimetry and claimed no excess heat. And he didn't proclaim that cold fusion would be a source of cheap, clean, and nearly inexhaustible energy, as the two University of Utah electrochemists did.

Nevertheless, Jones's claim was scientifically startling because it suggested an enormous enhancement (by 40 orders of magnitude) of the theoretical fusion rate for deuterium gas when it is loaded into a metal lattice. Even so, many scientists—including Hansen, who was "an outside observer" at the time—enter-

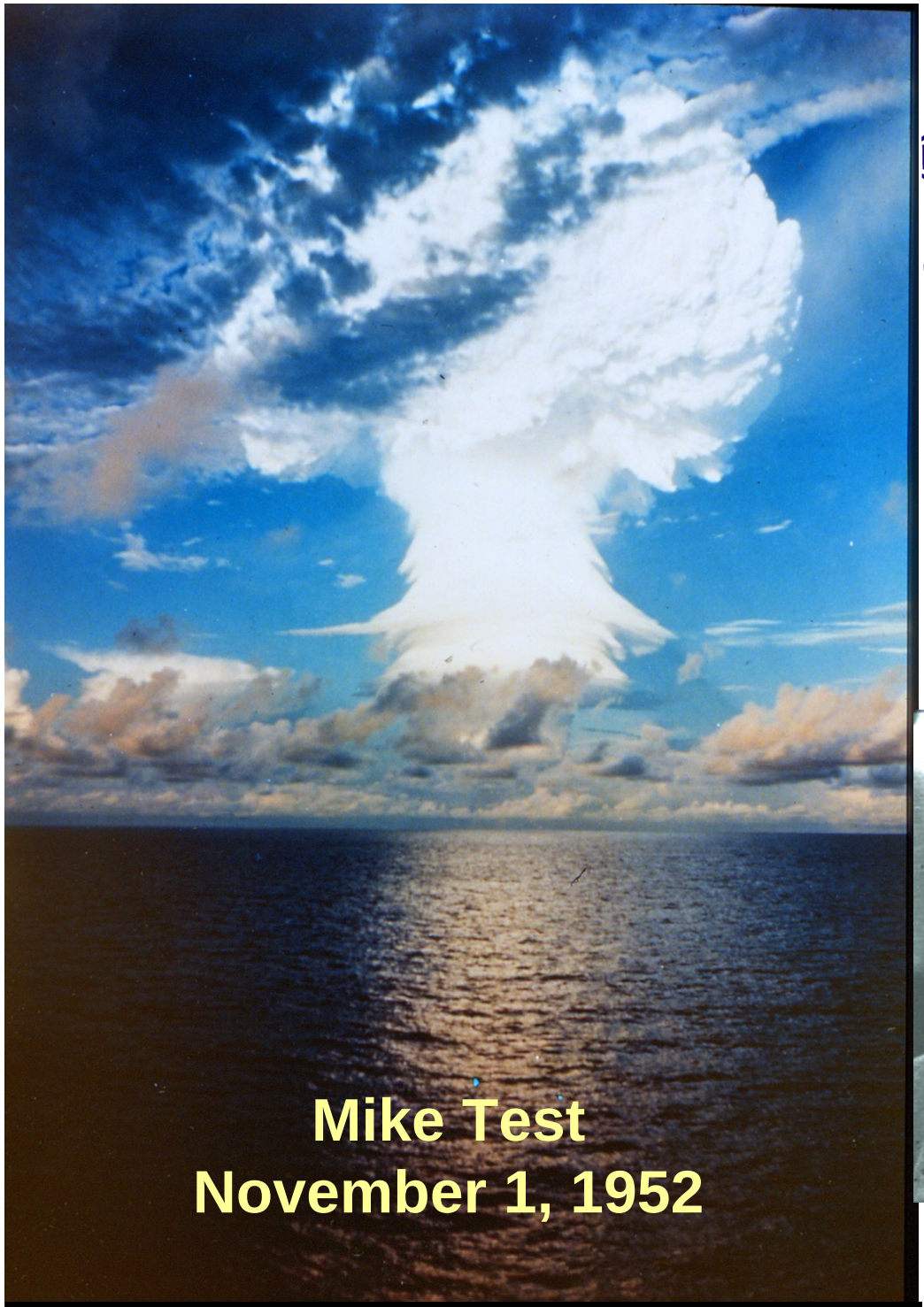
(Con.)

another big step: Collaborating with BYU chemistry professor Lee D. Hansen, he published two papers in the *Journal of Physical Chemistry* (*JPC*) that critically examine other cold fusion claims—only to find them "far from compelling."

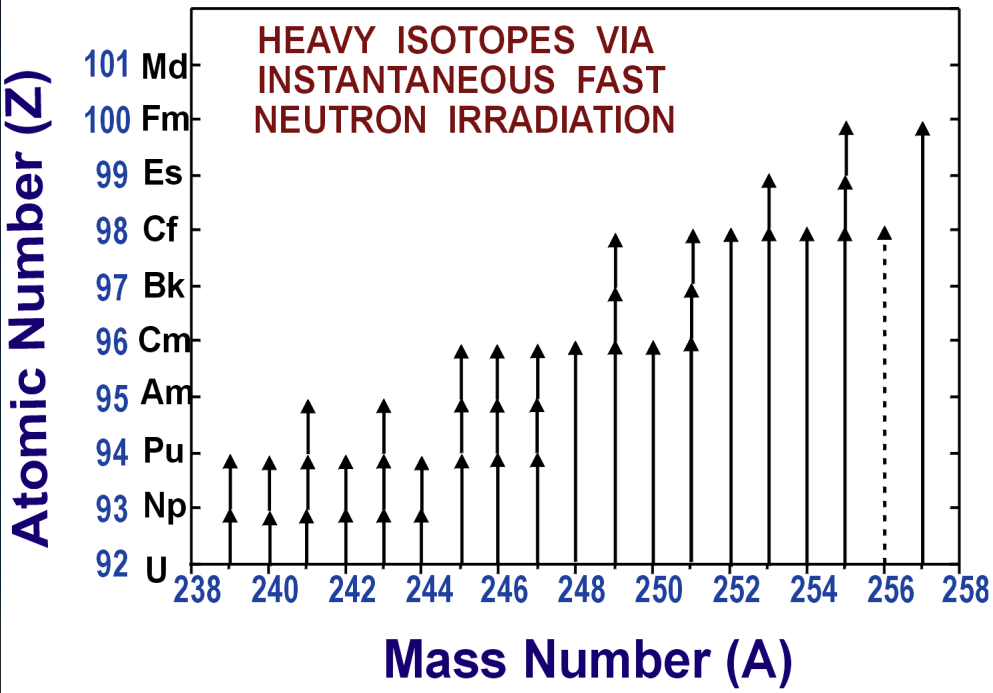
Chemistry professor James F. Haw of Texas A&M University, College Station, a critic of cold fusion, has seen the papers, and he calls them "explosive." One of them "is the bluntest scientific paper I have ever read," he remarks. "This type of direct response in the peer-reviewed literature to the claims of cold fusion is long overdue."

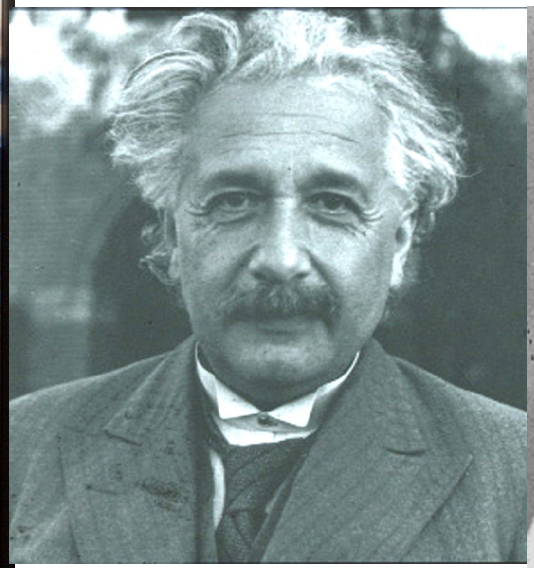
Jones's transformation from cautious cold fusion believer to skeptic came about as a natural consequence of his quest for scientific truth—wherever that quest might lead him. He has also assumed the role of gadfly, pushing other cold fusion researchers to examine their own work with a more skeptical eye.

There had been vitriolic condemnation of John Huizenga's dynamic leadership of our committee and the committee members as well, but John's thorough knowledge of both chemistry and physics was invaluable and enabled him to conduct a careful, fair consideration of all the facts. And his judgement has been ultimately vindicated, even in the case of the very low levels of neutron emission reported by Jones et al!

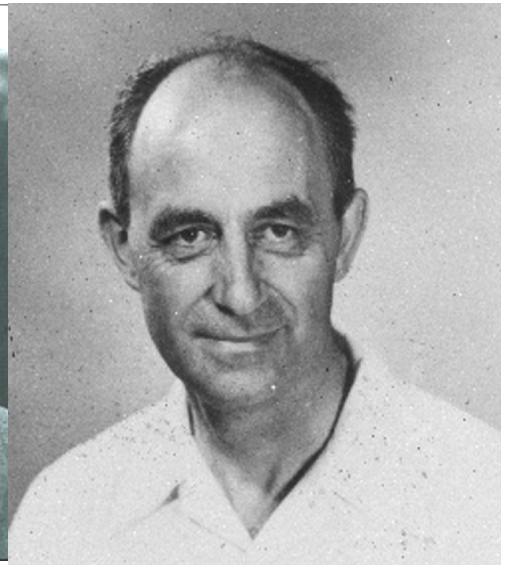


Beta-decay of heavy U isotopes to higher Z elements





Albert Einstein,1932
99-einsteinium (Es)
1879-1955



Enrico Fermi, 1941
100-fermium (Fm)
1901-1954

Seaborg wrote in his journal for June 3, 1955, "I then suggested that Al and I, using the name "einsteinium" for element 99 as preferred by the Los Alamos people, attempt to compose an article for joint publication to see if we could find agreement on it. We then immediately dictated to an Argonne secretary the document, entitled "The New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100," which combined the salient parts of our two documents, UCRL-2981 ("The New Element Losalium, Atomic Number 99" by A. Ghiorso, S.G. Thompson, G.H. Higgins, and G.T. Seaborg) and UCRL-2947 ("The New Element Fermium, Atomic Number 100" by A. Ghiorso, S.G. Thompson, G.H. Higgins, and G.T. Seaborg).

The write-up made it clear that both elements 99 and 100 were discovered at Berkeley first. This was satisfactory, without any changes, to the Argonne group, who also agreed that Ghiorso and the Berkeley group would be senior authors and the Los Alamos investigators would also be included as authors—thus the agreed upon authorship was A. Ghiorso, S. G. Thompson, G. H. Higgins, G. T. Seaborg, M. H. Studier, P. R. Fields, S. M. Fried, H. Diamond, J. F. Mech, G. L. Pyle, J. R. Huizenga, A. Hirsch, W. M. Manning. C. I. Browne, H. L. Smith, and R. W. Spence.

Finally published after declassification:

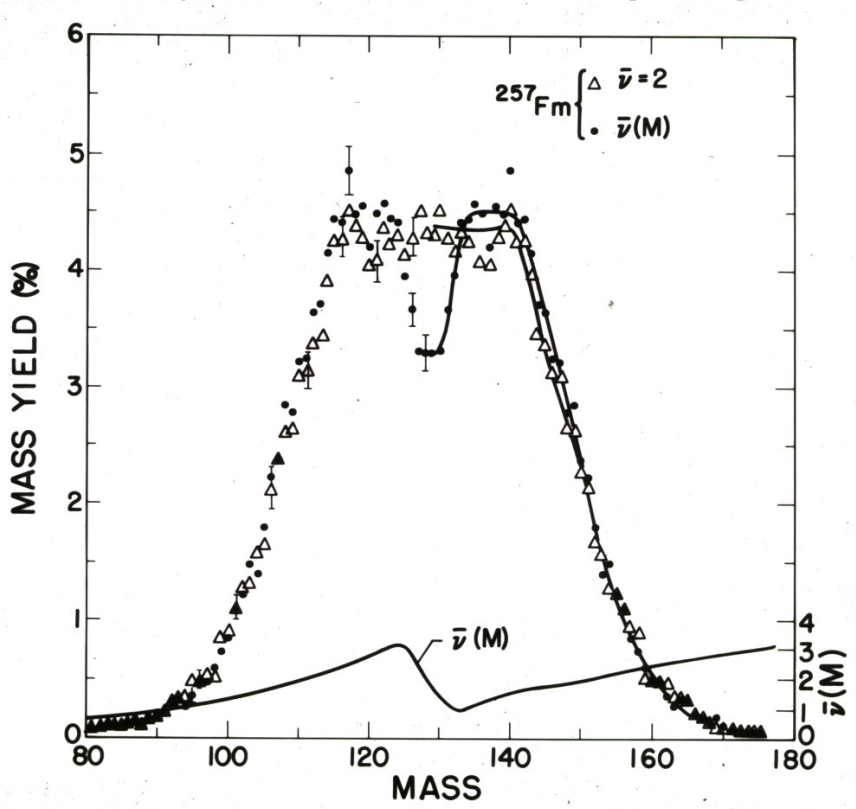
Phys. Rev. 99, 1048 (1955)

APS Meeting,
U. of Chicago
Nov. 1960
Bob & Sue Vandenbosch,
John Huizenga

Symposium on Plutonium Chemistry, ANL, Feb. 18, 1963
From left: E.P. Steinberg, J.J. Katz, S. Fried, J.C. Hindman, F. S.Tompkins, M.S. Freedman, J.R. Huizenga, J.A. Marinsky.

John Huizenga receives E. O. Lawrence Award from AEC Chairman Glenn Seaborg, April 27, 1966 (Vanstrum, Anderson, Agnew, Huizenga)

Symmetric Fission in ^{257}Fm
Phys Rev. Lett. 26, 145 (1971).



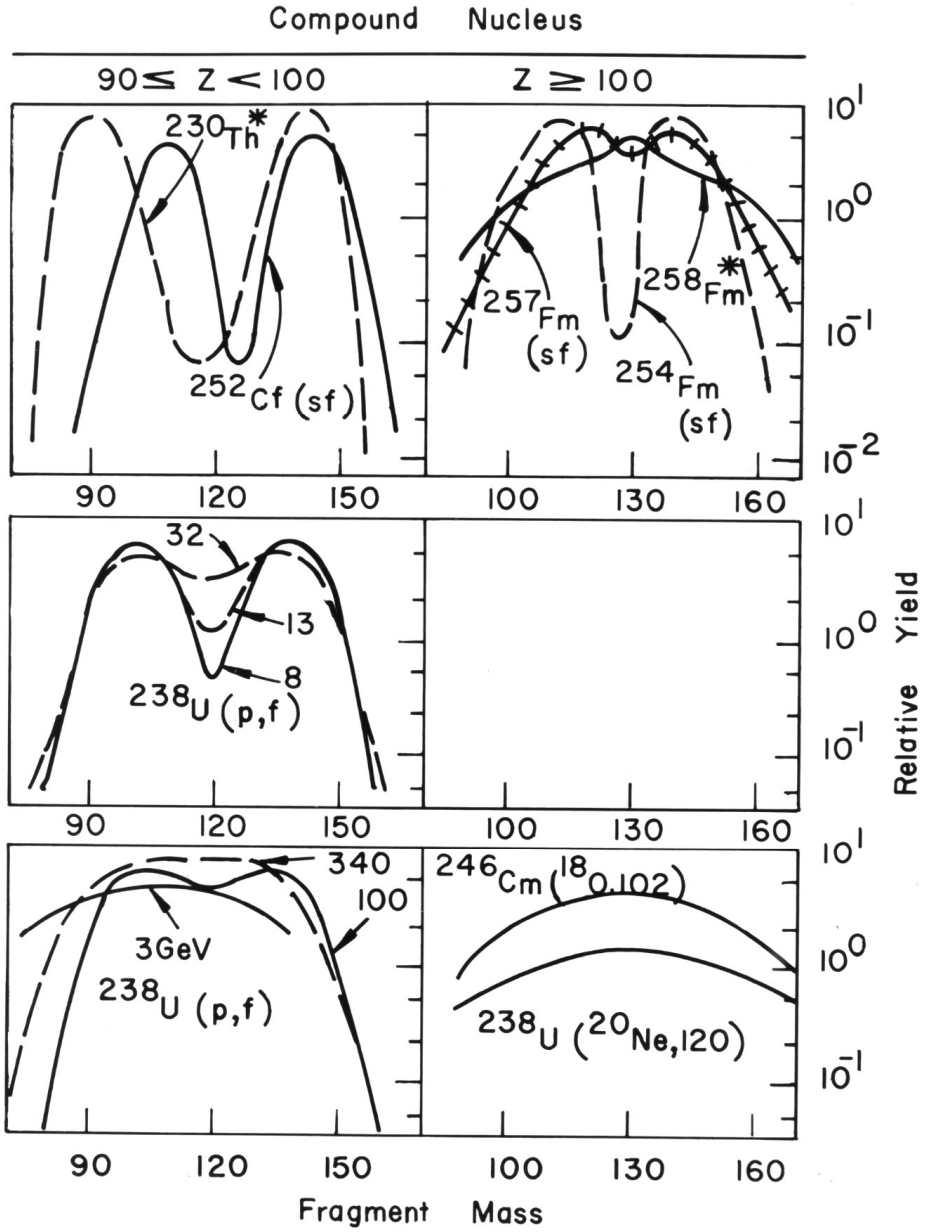
Book on Nuclear Fission (1973)

R. Vandenbosch, J. R. Huizenga

Written at level to introduce students to exciting field of physics & chemistry of fission & also recent developments..for researchers"

Wilkinson Quote: "Fission is a process of deadly fascination…"

Post-Fission Phenomena,
D.C. Hoffman. M. M. Hoffman
Ann. Rev. Nucl. Sci. 1974.



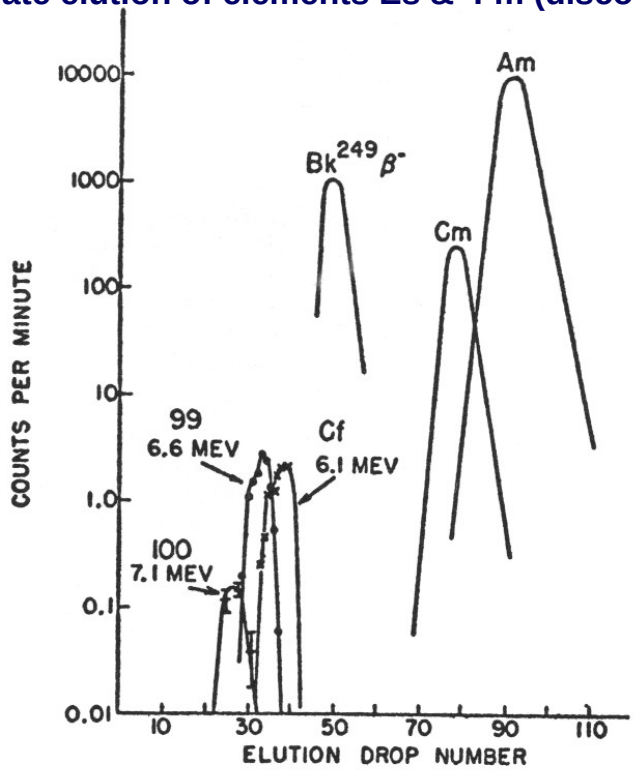
Co-discoverers of elements 99 and 100 at Symposium at Lawrence Berkeley Laboratory, Jan. 23, 1978 commemorating the 25th anniversary of the discovery

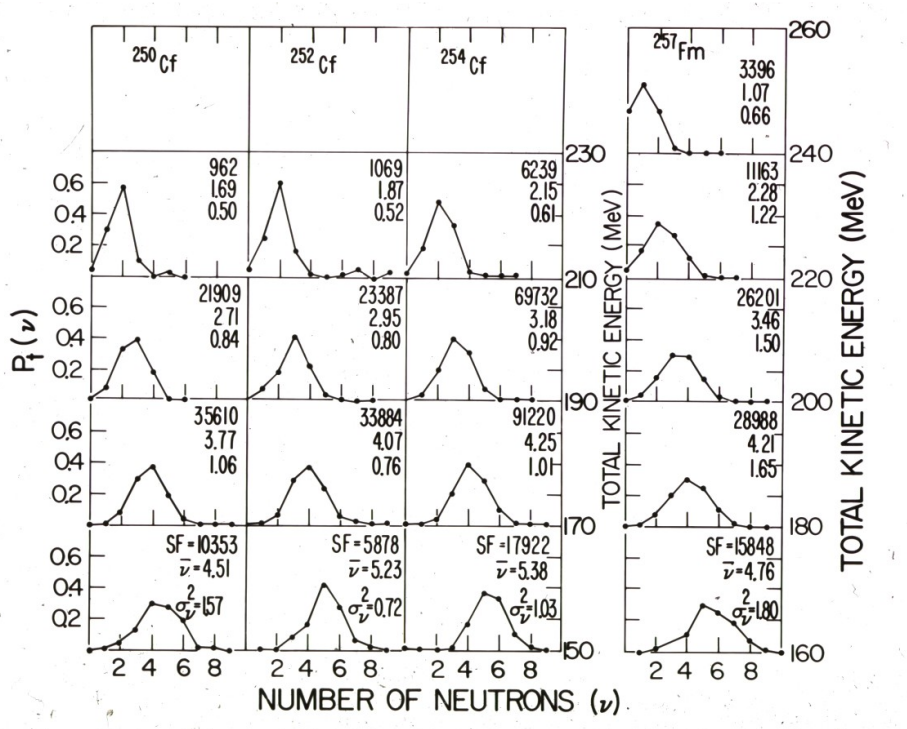
(8 of original 16 co-discoverers)

Back row:
Albert Ghiorso
Rod Spence
Glenn Seaborg
Paul Fields
John Huizenga

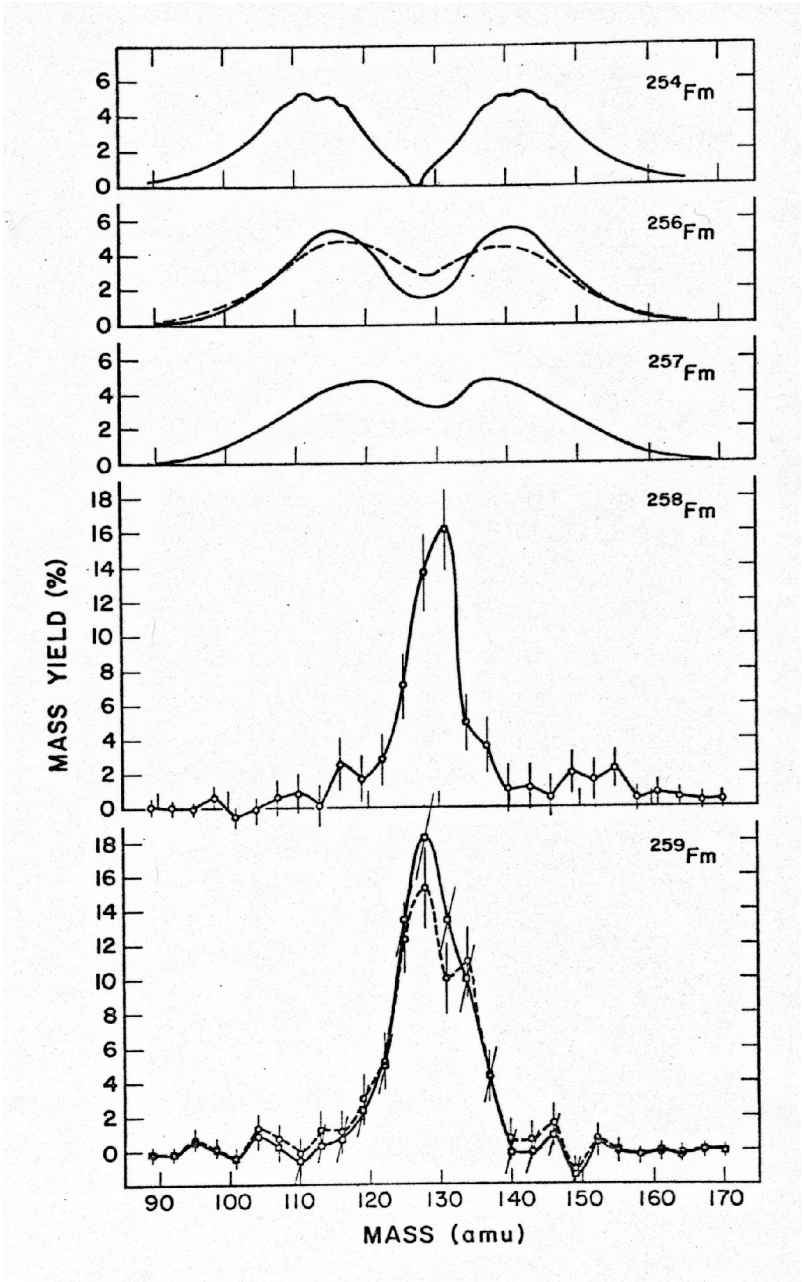
Front row:
Louise Smith
Sherman Fried
Gary Higgins

Citrate elution of elements Es & Fm (discovery paper).





Multiplicity distributions, Hoffman et al. 1977



Compilation of Mass distributions for SF of Fm isotopes, Hoffman, 1978 Symposium

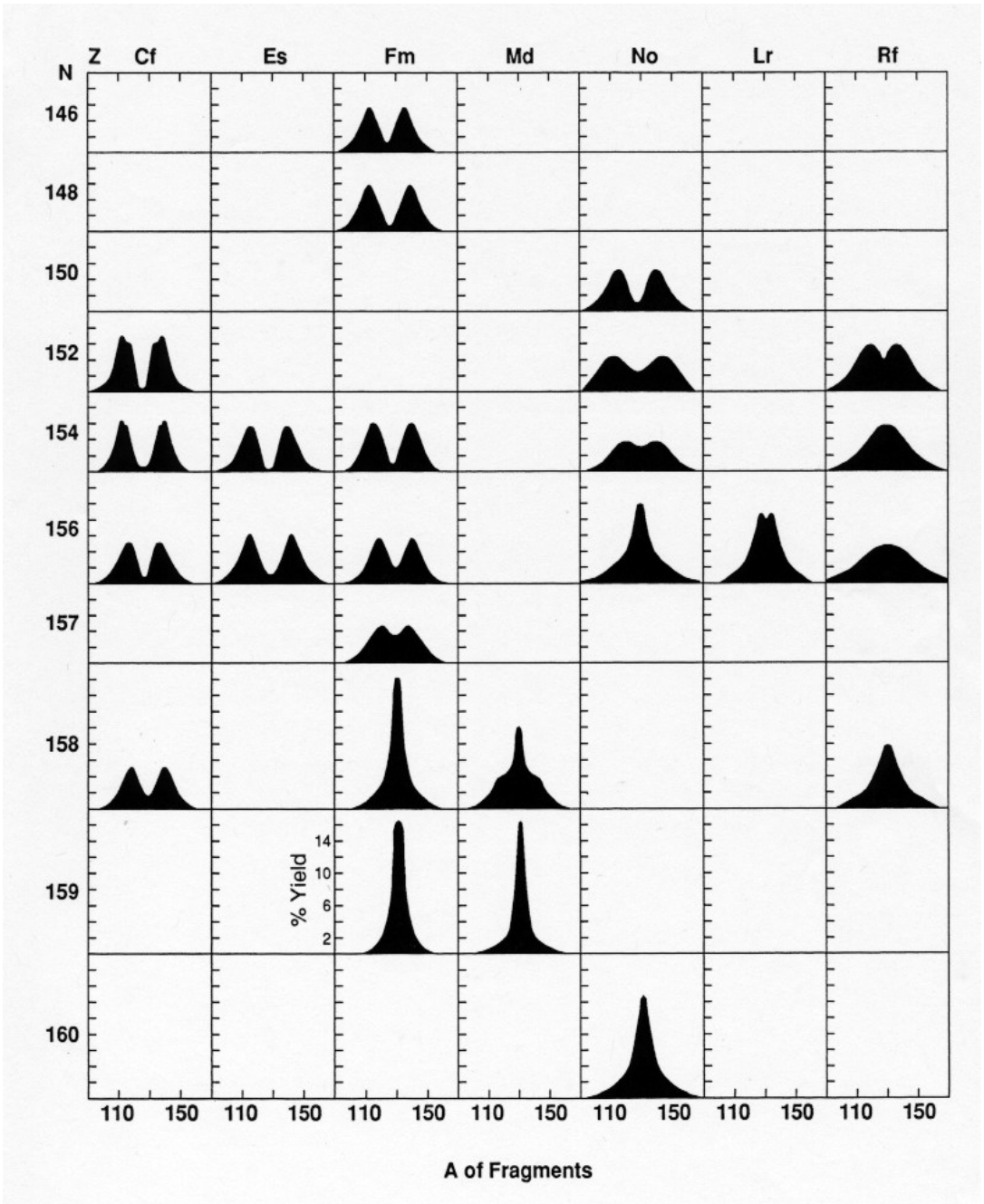
SF SUMMARY-1982

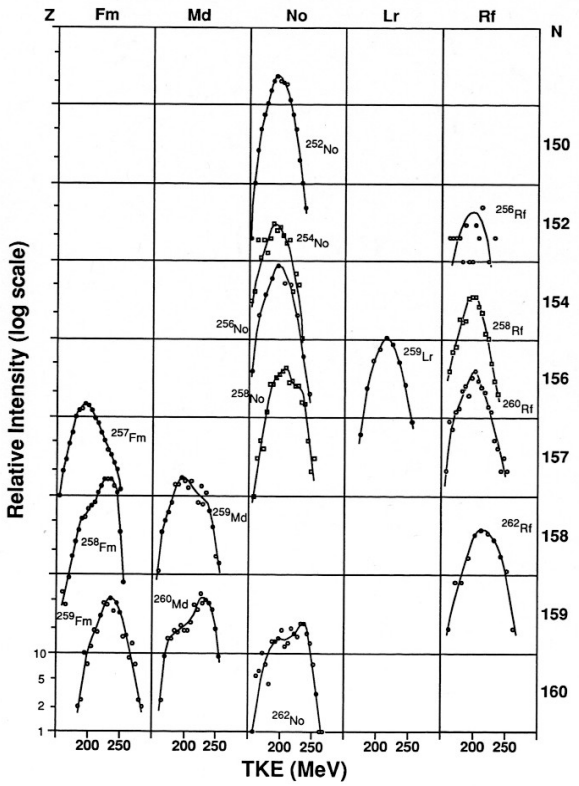
1. Fm ISOTOPES UNIQUE
   "MICROCOSM OF FISSION
   DOMINANT SHELL EFFECTS—
   M.Y., TKE, NEUTRON EMISSION, σ^2
   Explained by Fragment Shells $\rightarrow {}^{132}Sn$
   (spherical, doubly magic), TKE $\rightarrow$Q

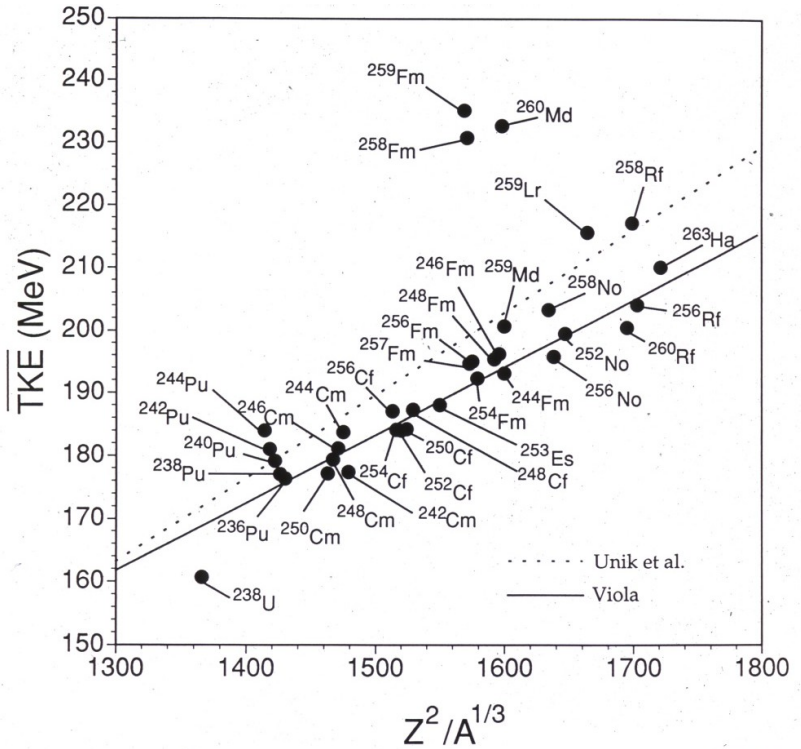
THEORY: Static Scission Point & 2-Center Shell Model

2. PROTON EFFECT? Check fragment properties of elements above and below fermium (Z=100) at higher neutron numbers

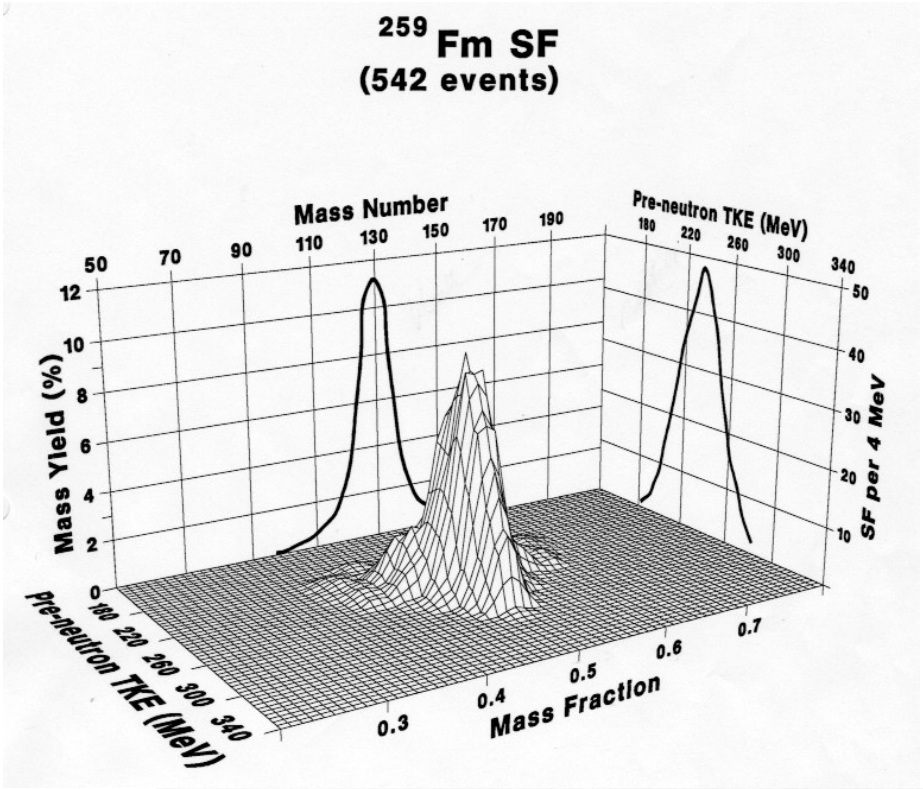
LASL- Sept. 1982

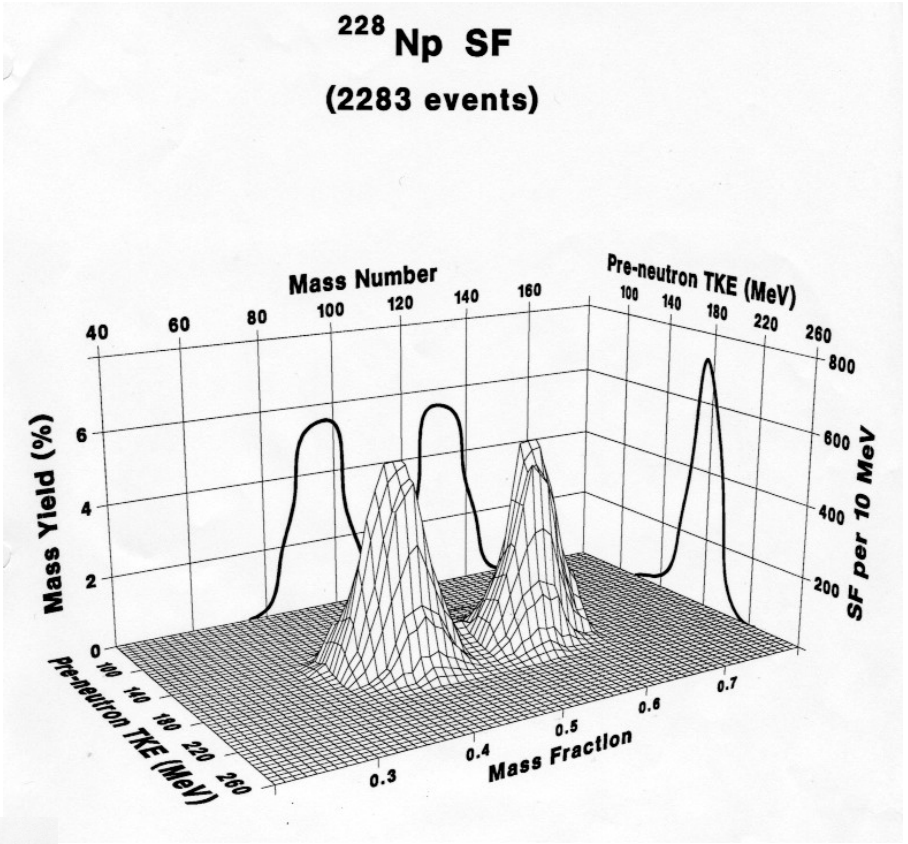

Z
Cf
Es
Fm
Md
No
Lr
Rf
N
146
148
150
152
154
156
157
158
159
160
% Yield
14
10
6
2
110
150
A of Fragments

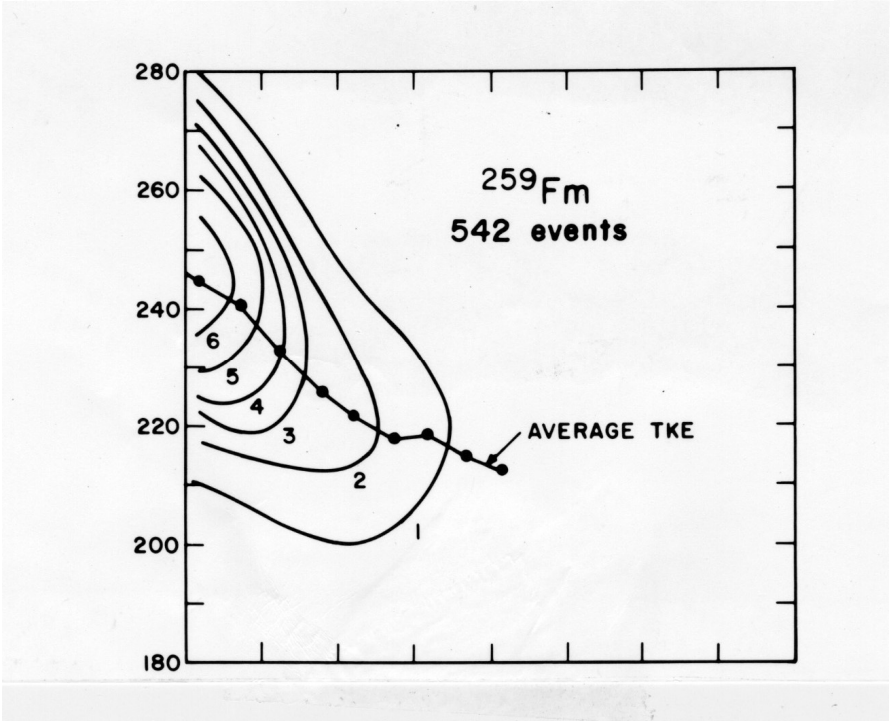

Z
Fm
Md
No
Lr
Rf
N
150
152
154
156
157
158
159
160
^{252}No
^{256}Rf
^{254}No
^{256}No
^{258}Rf
^{259}Lr
^{258}No
^{260}Rf
^{257}Fm
^{259}Md
^{262}Rf
^{258}Fm
^{260}Md
^{259}Fm
^{262}No
Relative Intensity (log scale)
10
5
2
1
200
250
TKE (MeV)

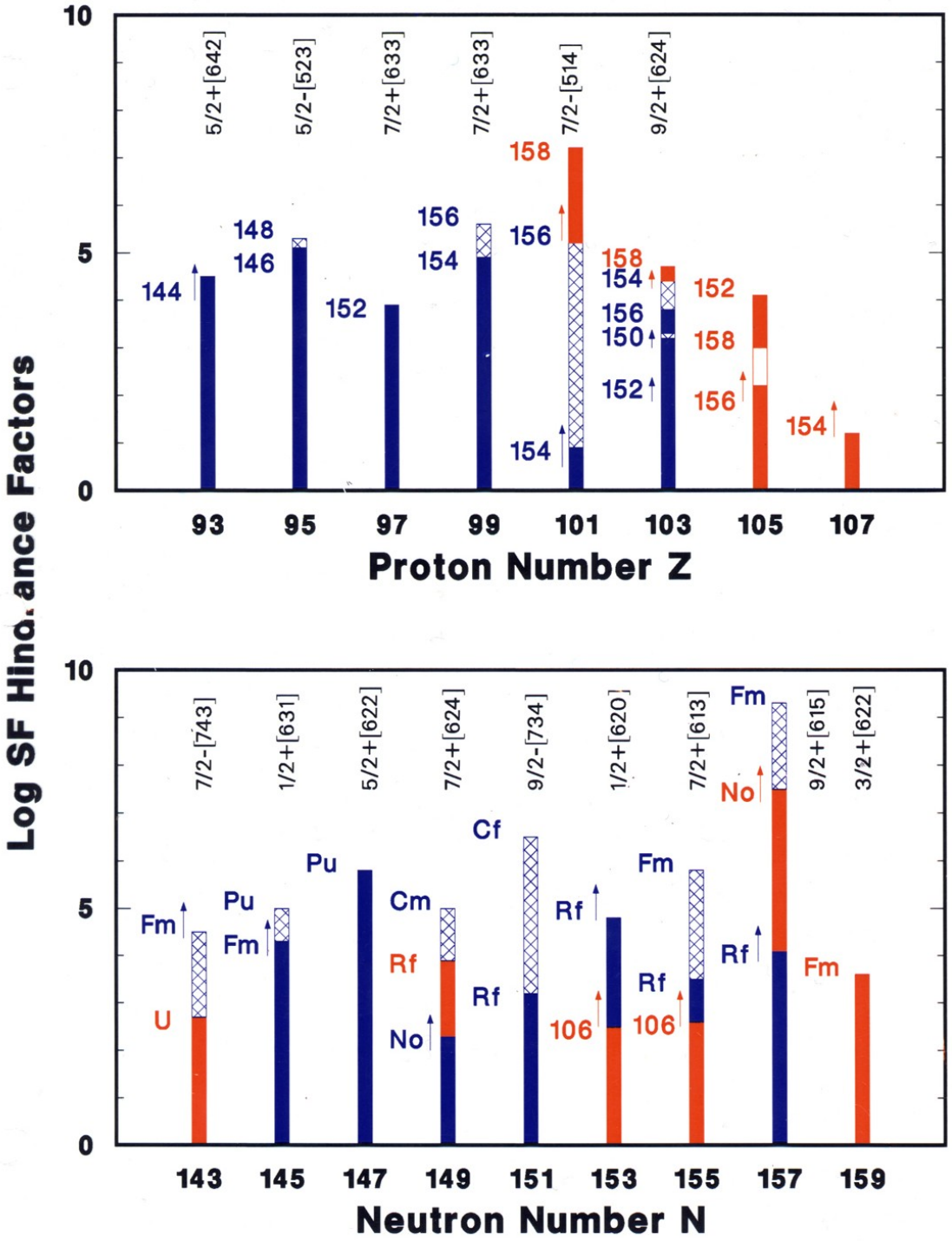

$\overline{TKE}$ (MeV)
250
240
230
220
210
200
190
180
170
160
150
1300
1400
1500
1600
1700
1800
$Z^2/A^{1/3}$
^{259}Fm
^{260}Md
^{258}Fm
^{259}Lr
^{258}Rf
^{263}Ha
^{246}Fm
^{259}Md
^{258}No
^{248}Fm
^{256}Rf
^{256}Fm
^{257}Fm
^{260}Rf
^{252}No
^{256}Cf
^{244}Pu
^{244}Cm
^{244}Fm
^{256}No
^{242}Pu
^{246}Cm
^{254}Fm
^{240}Pu
^{253}Es
^{250}Cf
^{238}Pu
^{254}Cf
^{248}Cf
^{252}Cf
^{248}Cm
^{242}Cm
^{236}Pu
^{250}Cm
^{238}U
Unik et al.
Viola

XBL 927-1625

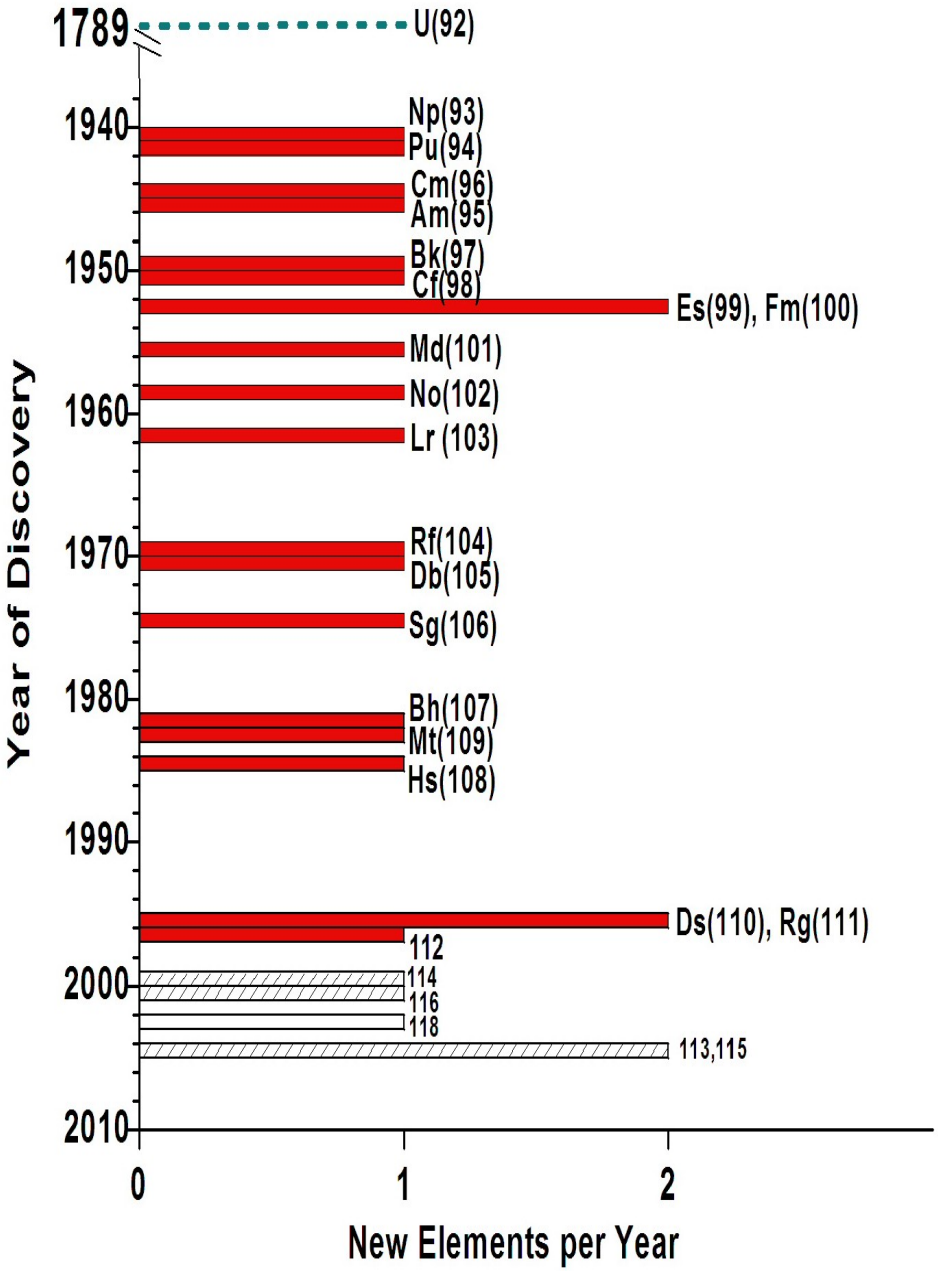

259 Fm SF
(542 events)
Mass Number
Pre-neutron TKE (MeV)
Mass Yield (%)
SF per 4 MeV
Pre-neutron TKE (MeV)
Mass Fraction


228 Np SF
(2283 events)
Mass Number
Pre-neutron TKE (MeV)
Mass Yield (%)
SF per 10 MeV
Pre-neutron TKE (MeV)
Mass Fraction

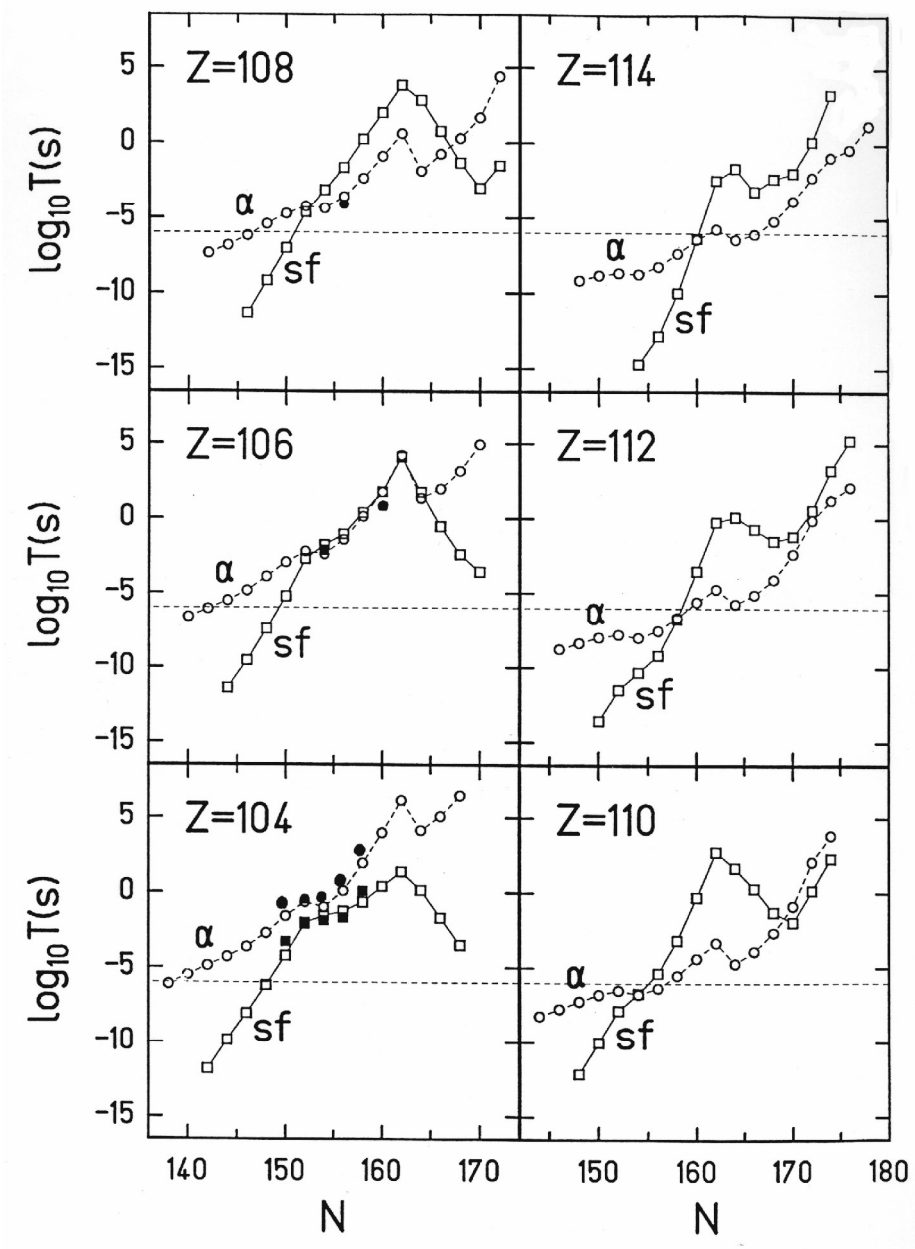

259Fm
542 events
AVERAGE TKE
1
2
3
4
5
6


Log SF Hinderance Factors
10
5
0
5/2+[642]
5/2-[523]
7/2+[633]
7/2+[633]
7/2-[514]
9/2+[624]
144
148
146
152
156
154
158
156
154
158
154
156
150
152
152
158
156
154
93
95
97
99
101
103
105
107
Proton Number Z
7/2-[743]
1/2+[631]
5/2+[622]
7/2+[624]
9/2-[734]
1/2+[620]
7/2+[613]
9/2+[615]
3/2+[622]
Fm
U
Pu
Fm
Pu
Cm
Rf
No
Cf
Rf
Rf
106
Fm
Rf
106
Fm
No
Rf
Fm
143
145
147
149
151
153
155
157
159
Neutron Number N

Time line of discovery of elements



Calculated fission & alpha half-lives vs. neutron number, (1994)



Atom-at-a-Time Chemistry of TANs

Current status of periodic table

- **TANs Names**
- **“Chemical periodic table”**

Importance of Research on TANs

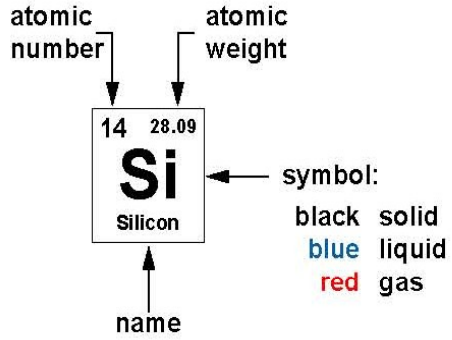
- **Challenges**
- **Relevance**

Status of atom-at-time chemical studies

Future TAN Chemistry?

- **New isotopes & elements**
- **New instrumentation--Collaborations**

PERIODIC TABLE 2006



- alkali metals
- alkaline earth metals
- transitional metals
- other metals
- non metals
- noble gases

1 1.01 H Hydrogen																	2 4.003 He Helium
3 6.94 Li Lithium	4 9.01 Be Beryllium											5 10.81 B Boron	6 12.01 C Carbon	7 14.01 N Nitrogen	8 15.999 O Oxygen	9 18.998 F Fluorine	10 20.18 Ne Neon
11 22.99 Na Sodium	12 24.31 Mg Magnesium											13 26.98 Al Aluminum	14 28.09 Si Silicon	15 30.97 P Phosphorus	16 32.06 S Sulfur	17 35.45 Cl Chlorine	18 39.95 Ar Argon
19 39.10 K Potassium	20 40.08 Ca Calcium	21 44.96 Sc Scandium	22 47.90 Ti Titanium	23 50.94 V Vanadium	24 51.996 Cr Chromium	25 54.94 Mn Manganese	26 55.85 Fe Iron	27 58.93 Co Cobalt	28 58.70 Ni Nickel	29 63.55 Cu Copper	30 65.37 Zn Zinc	31 69.72 Ga Gallium	32 72.59 Ge Germanium	33 74.92 As Arsenic	34 78.96 Se Selenium	35 79.90 Br Bromine	36 83.80 Kr Krypton
37 85.47 Rb Rubidium	38 87.62 Sr Strontium	39 88.91 Y Yttrium	40 91.22 Zr Zirconium	41 92.91 Nb Niobium	42 95.94 Mo Molybdenum	43 (98) Tc Technetium	44 101.07 Ru Ruthenium	45 102.91 Rh Rhodium	46 106.40 Pd Palladium	47 107.87 Ag Silver	48 112.41 Cd Cadmium	49 114.82 In Indium	50 118.69 Sn Tin	51 121.75 Sb Antimony	52 127.60 Te Tellurium	53 126.90 I Iodine	54 131.30 Xe Xenon
55 132.91 Cs Cesium	56 137.33 Ba Barium	57 138.91 La Lanthanum	72 178.49 Hf Hafnium	73 180.95 Ta Tantalum	74 183.85 W Tungsten	75 186.21 Re Rhenium	76 190.20 Os Osmium	77 192.22 Ir Iridium	78 195.09 Pt Platinum	79 196.97 Au Gold	80 200.59 Hg Mercury	81 204.37 Tl Thallium	82 207.19 Pb Lead	83 208.98 Bi Bismuth	84 (209) Po Polonium	85 (210) At Astatine	86 (222) Rn Radon
87 (223) Fr Francium	88 226.03 Ra Radium	89 227.03 Ac Actinium	104 (261) Rf Rutherfordium	105 (262) Db Dubnium	106 (266) Sg Seaborgium	107 (262) Bh Bohrium	108 (265) Hs Hassium	109 (266) Mt Meitnerium	110 (271) Ds Darmstadtium	111 (272) Rg Roentgenium	(277) *112*	(284) *113*	(288) *114*	(288) *115*	(292) *116*	(117)	(118)
(119)	(120)	(121)	(154)														

Lanthanides	58 140.12 Ce Cerium	59 140.91 Pr Praseodymium	60 144.24 Nd Neodymium	61 (145) Pm Promethium	62 150.40 Sm Samarium	63 151.96 Eu Europium	64 157.25 Gd Gadolinium	65 158.93 Tb Terbium	66 162.50 Dy Dysprosium	67 164.93 Ho Holmium	68 167.26 Er Erbium	69 168.93 Tm Thulium	70 173.04 Yb Ytterbium	71 174.97 Lu Lutetium
Actinides	90 232.04 Th Thorium	91 231.04 Pa Protactinium	92 238.03 U Uranium	93 237.05 Np Neptunium	94 (244) Pu Plutonium	95 (243) Am Americium	96 (247) Cm Curium	97 (247) Bk Berkelium	98 (251) Cf Californium	99 (252) Es Einsteinium	100 (257) Fm Fermium	101 (260) Md Mendelevium	102 (259) No Nobelium	103 (262) Lr Lawrencium

Superactinides (122-153)

IUPAC APPROVED HEAVY ELEMENT NAMES

August 30, 1997, Geneva, Switzerland

Element	Name	Symbol
101	Mendelevium	Md
102	Nobelium	No
103	Lawrencium	Lr
Transactinides (TANs)		
104	Rutherfordium	Rf
105	Dubnium (Hahnium)[#]	Db (Ha)[#]
106	Seaborgium	Sg
107	Bohrium	Bh
108	Hassium	Hs
109	Meitnerium	Mt
110 *	Darmstadtium	Ds
111	Roentgenium	Rg

[#]Many publications of chemical studies before 1997 use hahnium (Ha) for 105

*Approved by IUPAC, August 2003; **Approved by IUPAC, November 2004.

Periodic Table before World War II

No known transuranium elements—Th(90), Pa(91), U(92) placed in body of table as part of *6*d transition series where we now place the transactinides.

Element 93 predicted to be homolog of rhenium (75).

Elements 43(Tc), 61(Pm), 85(At), 87 (Fr) "missing" from table.

1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
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
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	(43)	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57-71 La-Lu	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)	86 Rn
(87)	88 Ra	89 Ac	90 Th	91 Pa	92 U	(93)	(94)	(95)	(96)	(97)	(98)	(99)	(100)				

57 La	58 Ce	59 Pr	60 Nd	(61)	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu

Chemical Periodic Table of the Elements 2006

1																	18
1 H	2											13	14	15	16	17	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg	3	4	5	6	7	8	9	10	11	12	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
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
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
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
87 Fr	88 Ra	89 Ac	104 Rf	105 Ha (Db)	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112		114	115 113	116		(118)

Rf, Ha, Sg Solution & Gas-phase

Bh, Hs Gas-phase

Lanthanides	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
Actinides	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

Chemistry of the Transactinides (TANs)

Atomic number (proton number, Z) >103 (Lawrencium)

CHALLENGES

Must be produced & studied at suitable accelerators.
Low production rates (few atoms at a time).
Very short-lived (minutes to seconds).
Rates & half-lives decrease as Z (atomic number) increases.
Plethora of unwanted elements produced.

UNIQUE CAPABILITIES REQUIRED

Hi-Intensity beams of heavy projectiles:

Berkeley 88-Inch Cyclotron; Dubna, Russia U-400 Cyclotron;
GSI, Darmstadt, Germany UNILAC; JAERI, Riken, Japan;
Jyvaskala, Finland; Lanzhou, China

Facilities & expertise in:

Preparation, handling, & irradiation of radioactive targets.
Fast transport of products from accelerator to separation facility.
Fast radiochemical separations & detection techniques.

Why Study TANs? Importance

- **Unique opportunity to extend knowledge of chemistry to furthest reaches of periodic table.**
- **Assess extent & magnitude of relativistic effects predicted to be especially strong in these elements due to their high nuclear charges.**
- **Compare chemical properties with lighter homologs & with theoretical predictions & periodic table trends.**

Anomalous trends in oxidation states, ionic radii, complexing?

Verify placement of TANS in new 6d transition series.

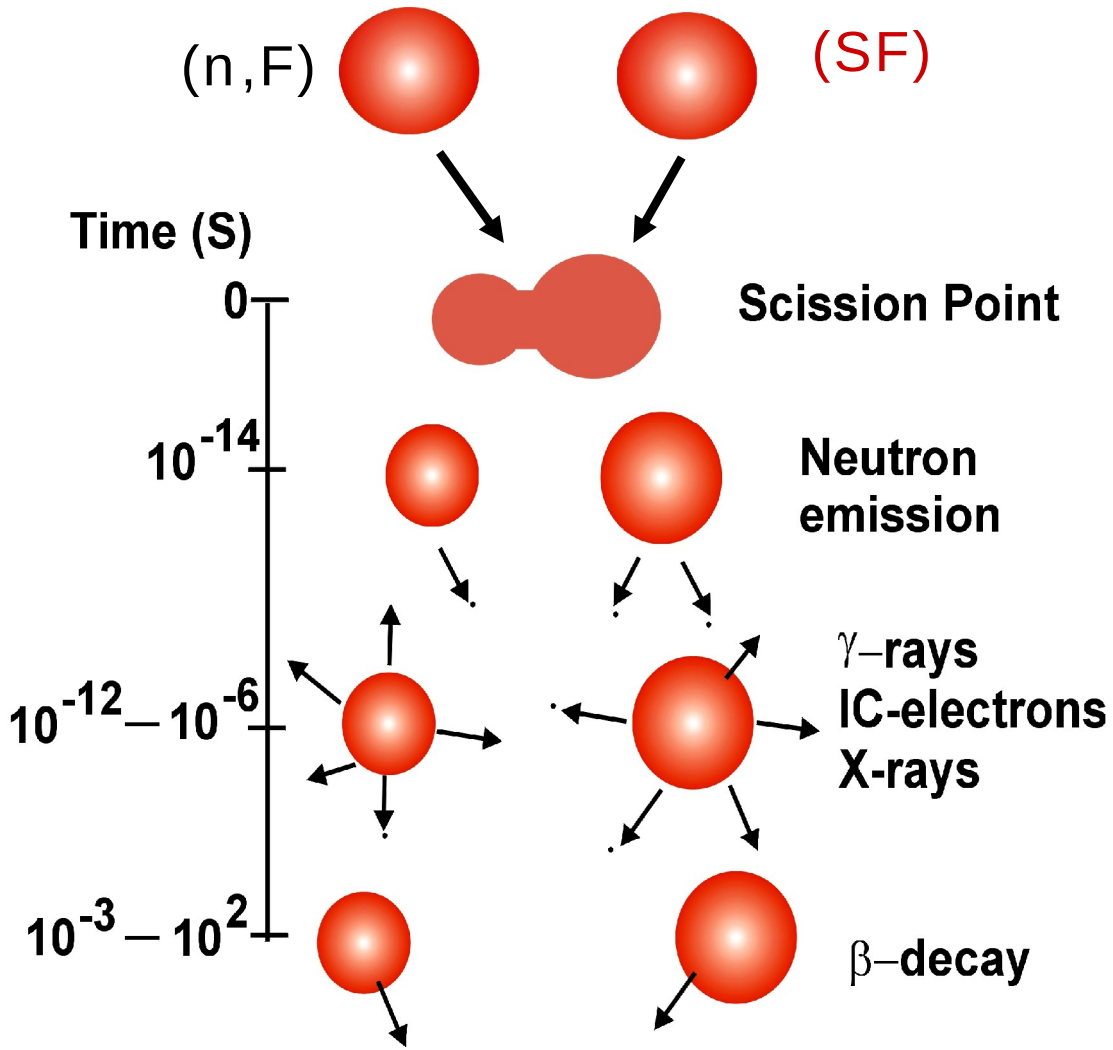
Compare with 5d(Hf, Ta, W, Re, Os) & 4d(Zr, Nb, Mo,Tc) series.

Compare trends within groups 4-8.

Elucidation of the chemical properties of the elements & their placement in the Periodic Table is one of the most fundamental goals of chemistry—sometimes referred to as "Textbook Chemistry"!

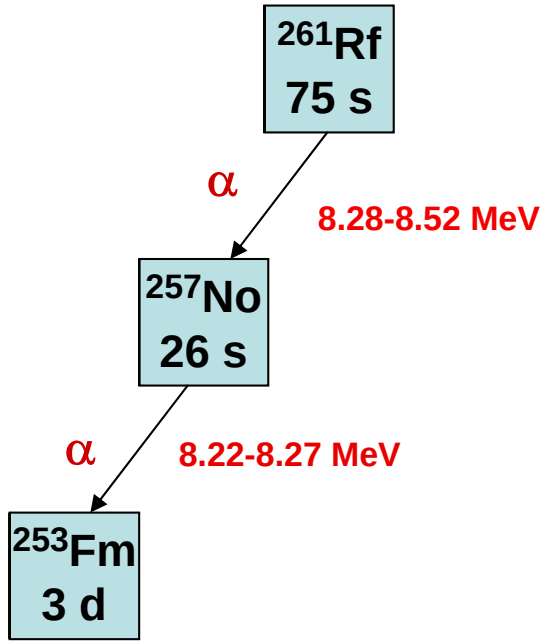
Nuclear Fission

Easy to detect but difficult to know what original fissioning nuclide was—many controversies!

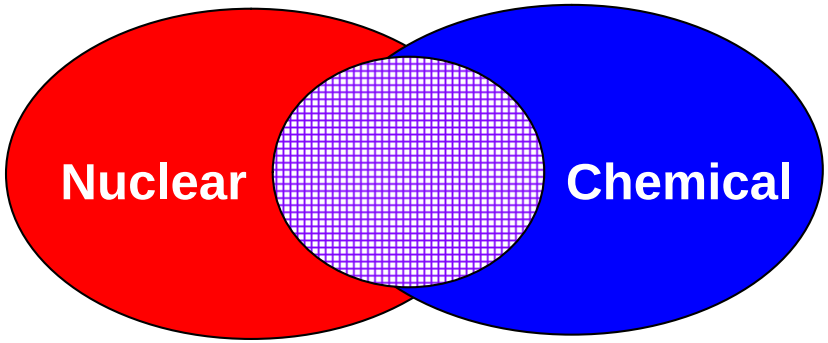


α–α correlation to known daughters

Gives positive i.d. of Z & A, but much more complex instrumentation & analysis needed.



Investigations of chemical and nuclear properties are complementary and should proceed "Hand-in-Hand"

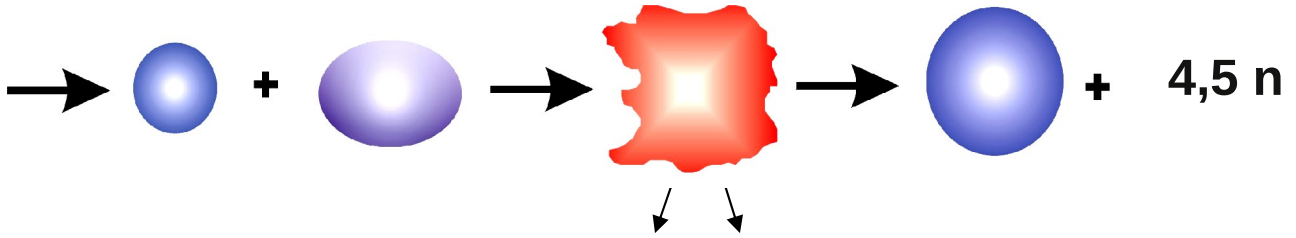


Nuclear properties, production methods, and detection techniques must be known in order to study chemical properties on an *Atom-at-a-Time* basis.

Knowledge of chemical properties permits separation and positive identification of atomic number. Can provide pure samples for study of nuclear properties and discovery of new isotopes.

Production Reactions for TAN Chemistry

Hot Fusion—Elements 104 through 108



Isotopes used in first definitive chemical studies of Rf through Hs

1970	^{18}O + ^{248}Cm	→	^{261}Rf	78s	$\sigma \sim 5$ nb	3/min
1988	^{18}O + ^{249}Bk	→	$^{262}Db(Ha)$	34s	$\sigma \sim 6$ nb	1/min
1997	^{22}Ne + ^{248}Cm	→	$^{266,265}Sg$	21s, 7s	$\sigma \sim 0.03$ nb	1/day
2000	^{22}Ne + ^{249}Bk	→	^{267}Bh	17s	$\sigma \sim .060$ nb	2/wk
2001	^{26}Mg + ^{248}Cm	→	$^{270,269}Hs$	~4s, 14s	$\sigma \sim .005$ nb	1/wk

**For σ ~5 nb, ~2 atoms/min produced. After transport efficiency (50%), chemical yield (80%),detection efficiency(35%), & decay (50%), only detect 0.14/min~200/d.

For Hs, only 0.14/d or ~1-2/week!

CHEMISTRY OF ELEMENTS 104 &105

Much controversy

Short half-lives & Small production cross sections.
Needed development of new detection methods & radioanalytical techniques suitable for atom-at-a-time studies.
Early Russian studies of gas-phase chemistry uncertain due to difficulty in positive id of element.

First solution chemistry of Rf 1970

Silva et al. showed Rf behaved similarly to the +4 elements Zr & Hf & not like+3 actinides in elutions from cation exchange columns. First automated system in 1980 by Hulet et al. again showed Rf similar to Hf, confirming its position in Group 4.

First solution chemistry of Db(Ha) not reported until 1987.

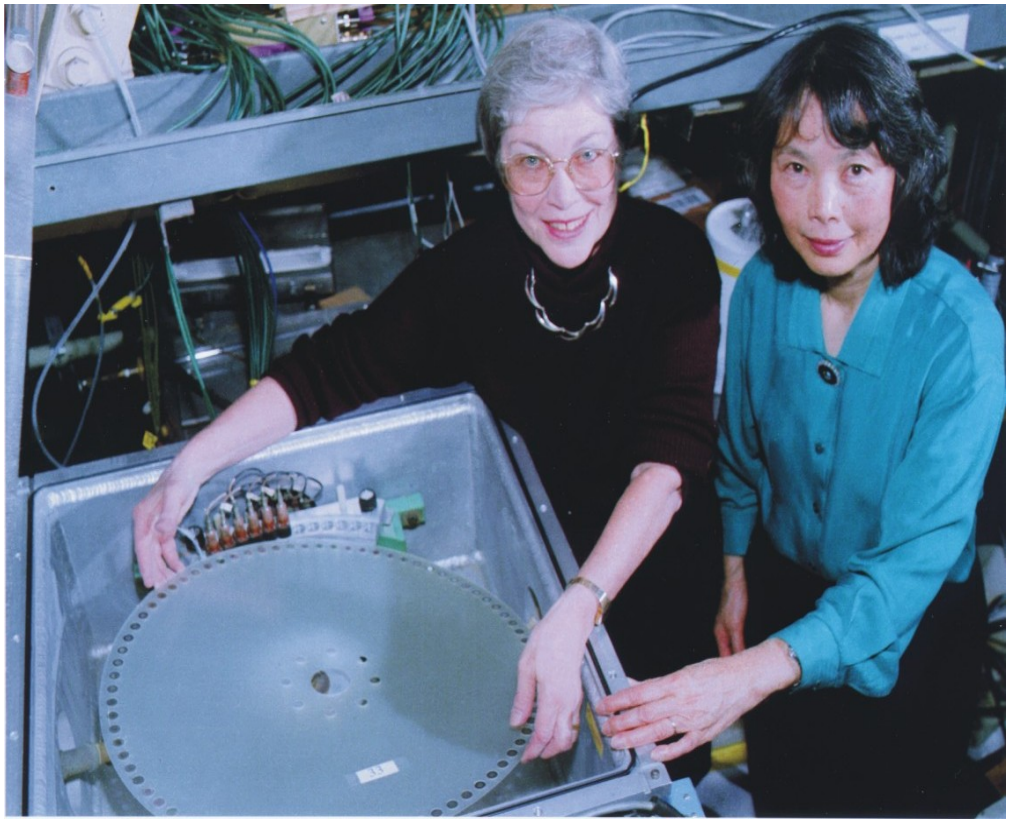
Our group performed some 800 manual experiments showing Ha behaved like Ta & Nb. Sorbed to glass after fuming with nitric acid. But, in extractions into MIBK, Ta extracted while Db remained in solution with Nb. These findings provided impetus for development & use at LBNL of the Automated Rapid Chemistry Apparatus (ARCA) developed by the German group.

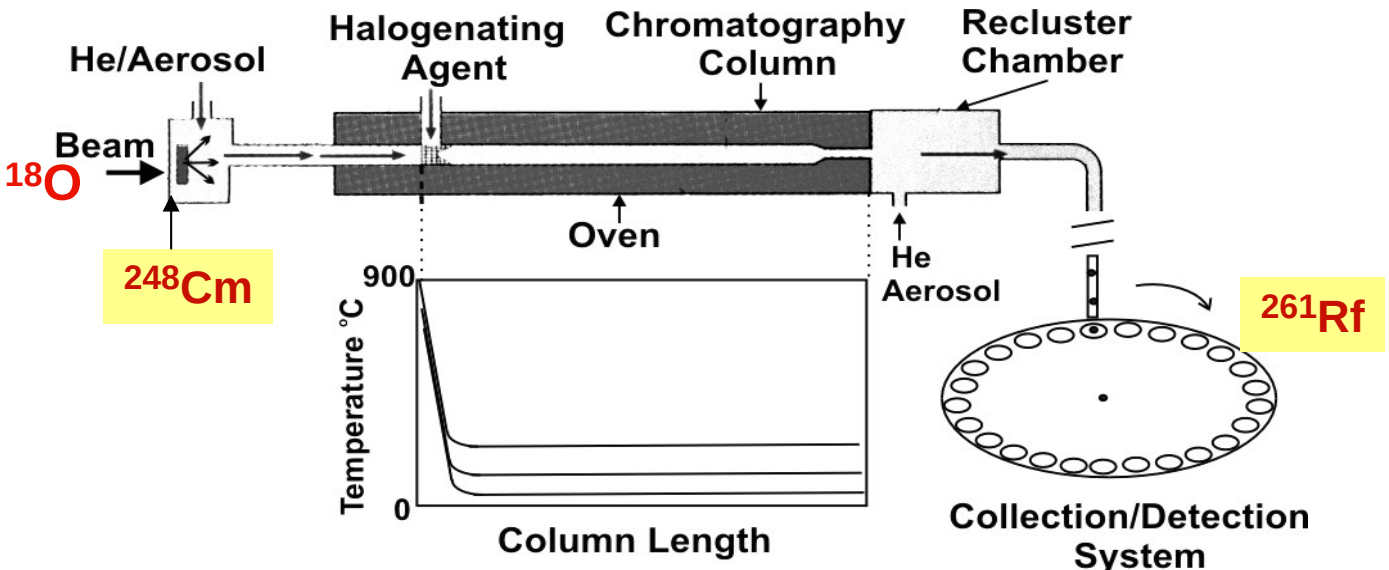
Isothermal Gas-Phase Studies of Element 104 (Rf)

Heavy Element Volatility Instrument

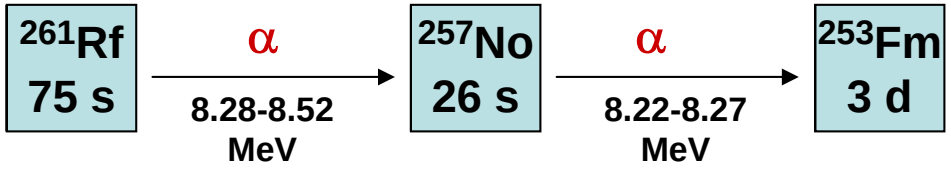
HEVI 1992

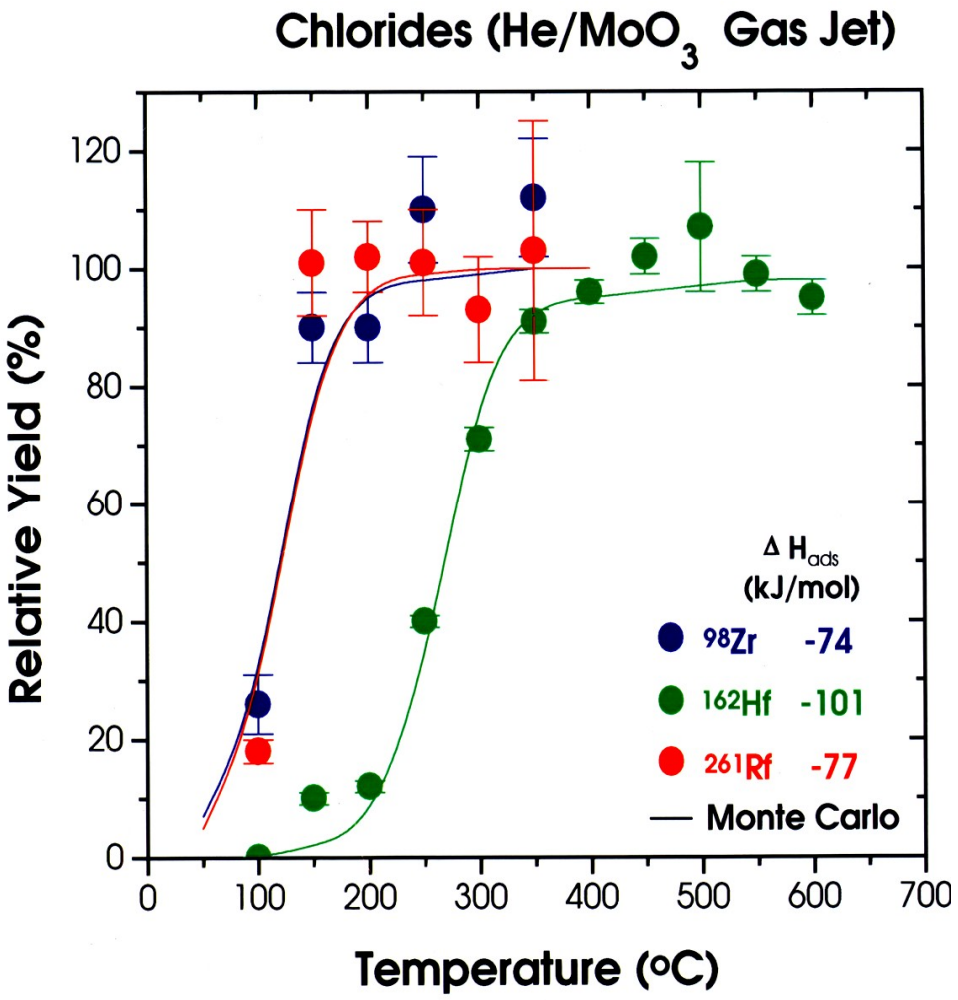
Merry-Go-Around (MG) 1994

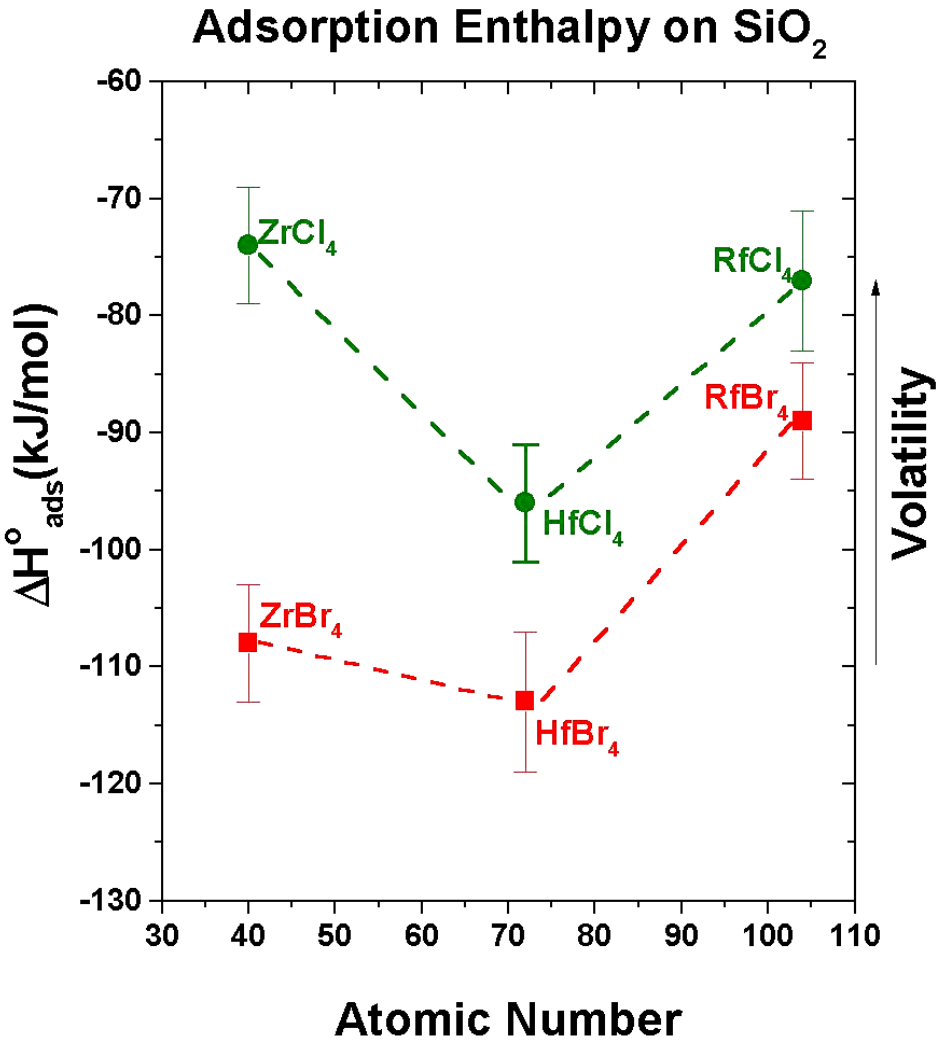




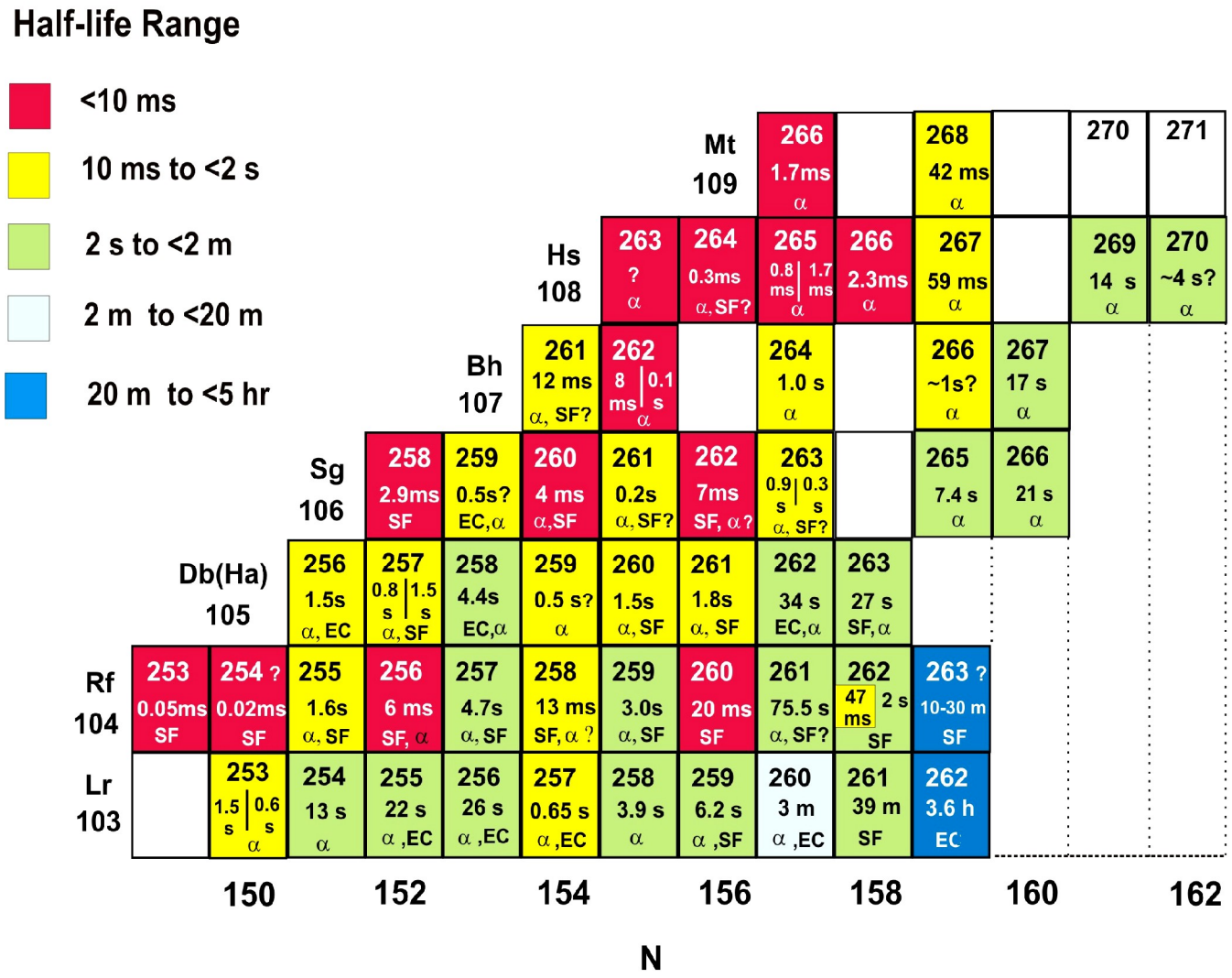
Volatility of Rf, Hf, Zr chlorides & bromides

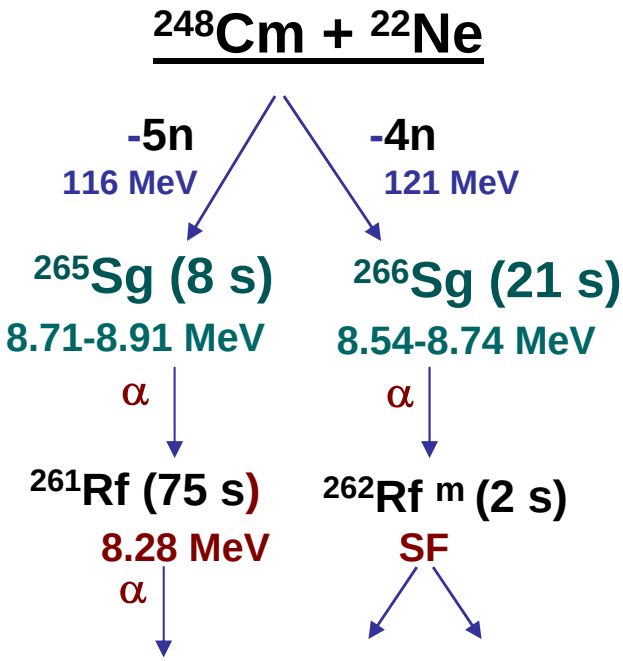






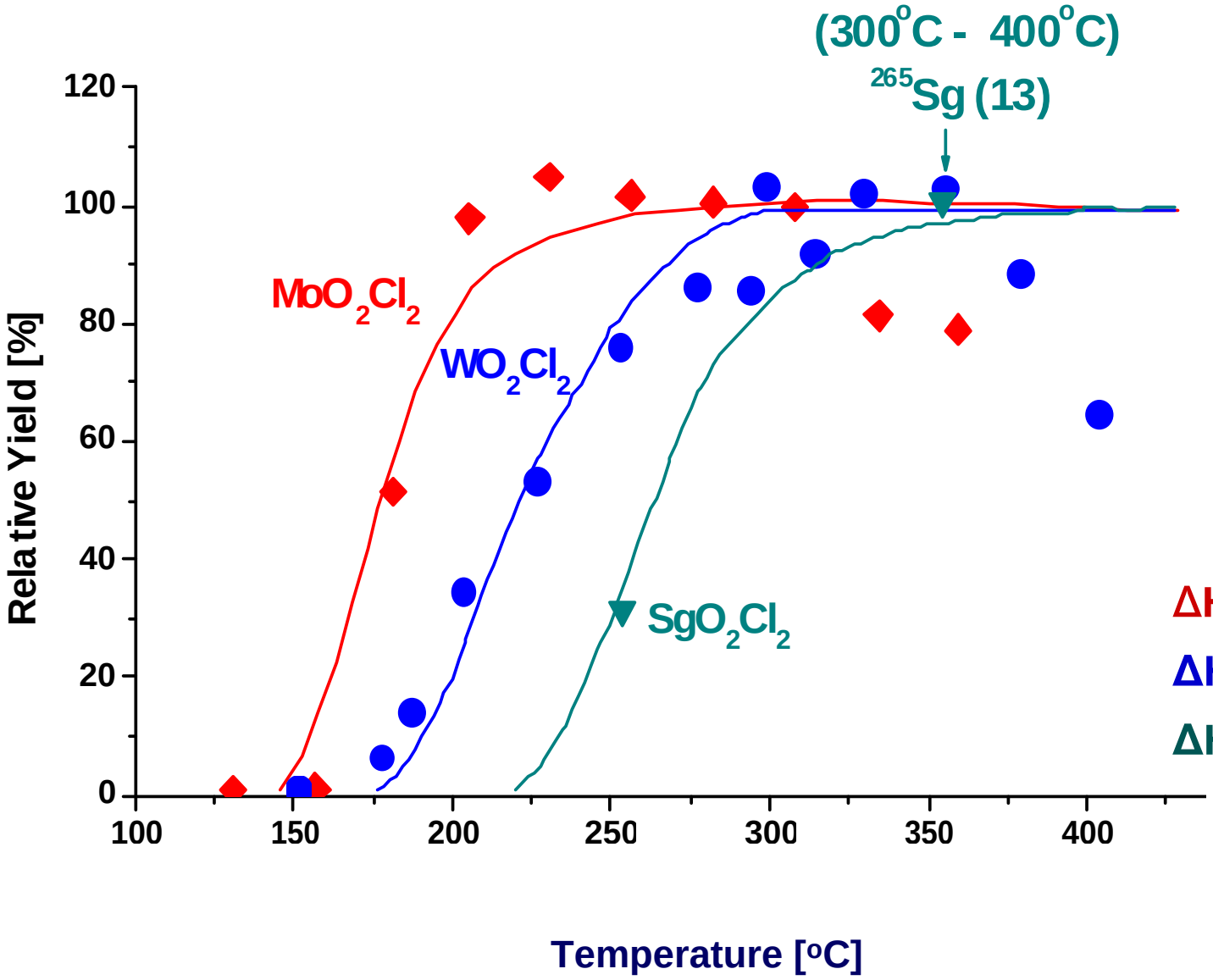
Discovery of longer-lived isotopes of Sg (1996-97), Bh (2000) & Hs (2002) near predicted region of extra nuclear stability ~N=162 made chemical studies possible





Gas-Phase Experiments with Sg

International Collaboration at UNILAC, GSI, Darmstadt, Germany



Volatility: Mo>W>Sg

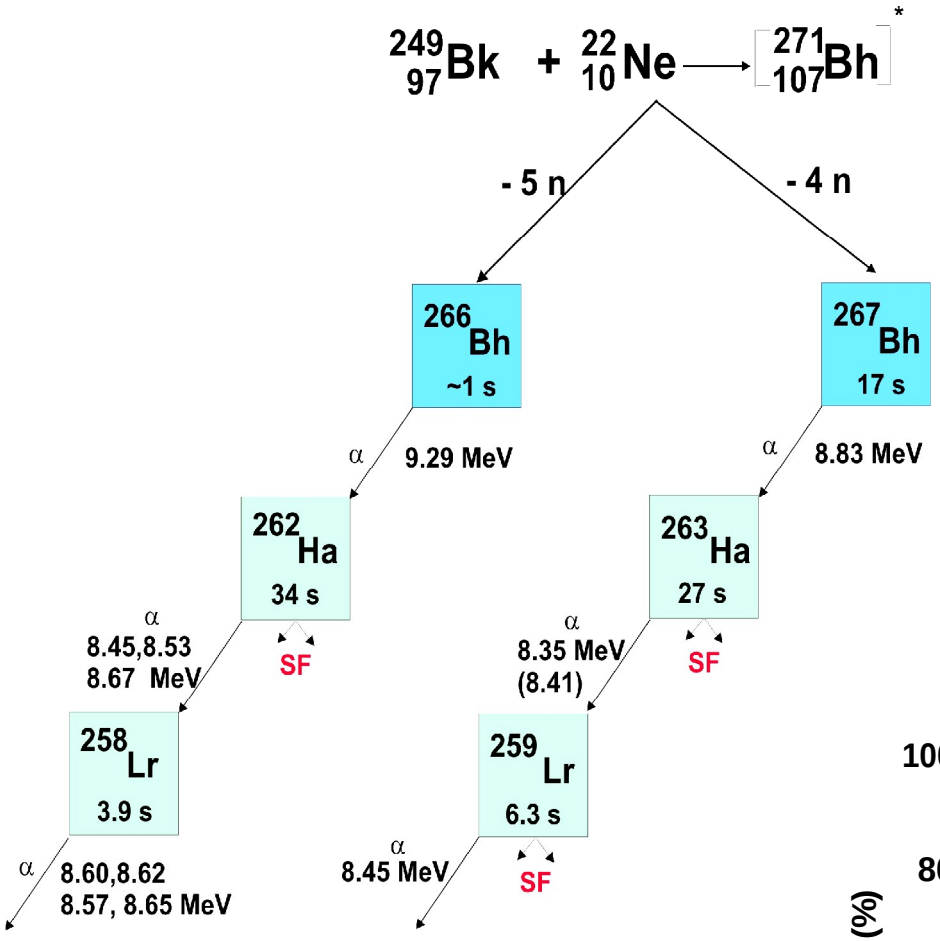
ΔH_{ads} = - 90 kj/mol (59 s)

ΔH_{ads} = - 96 kj/mol (51 s)

ΔH_{ads} = - 98 kj/mol (8 s)

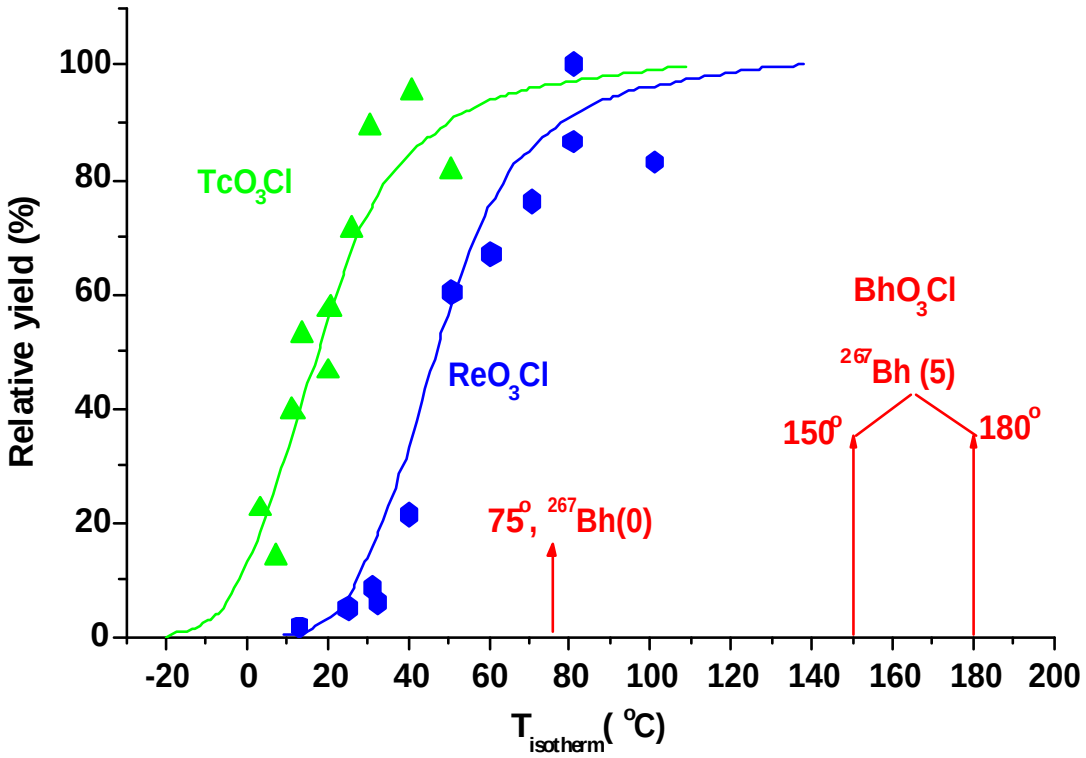
First Chemical Studies of Element 107 (Bh), 2001-2

(International collaboration)



Gas-phase studies at PSI/GSI

Tc>Re>Bh



Discovery of ~17-s ^{267}Bh LBNL, 2000

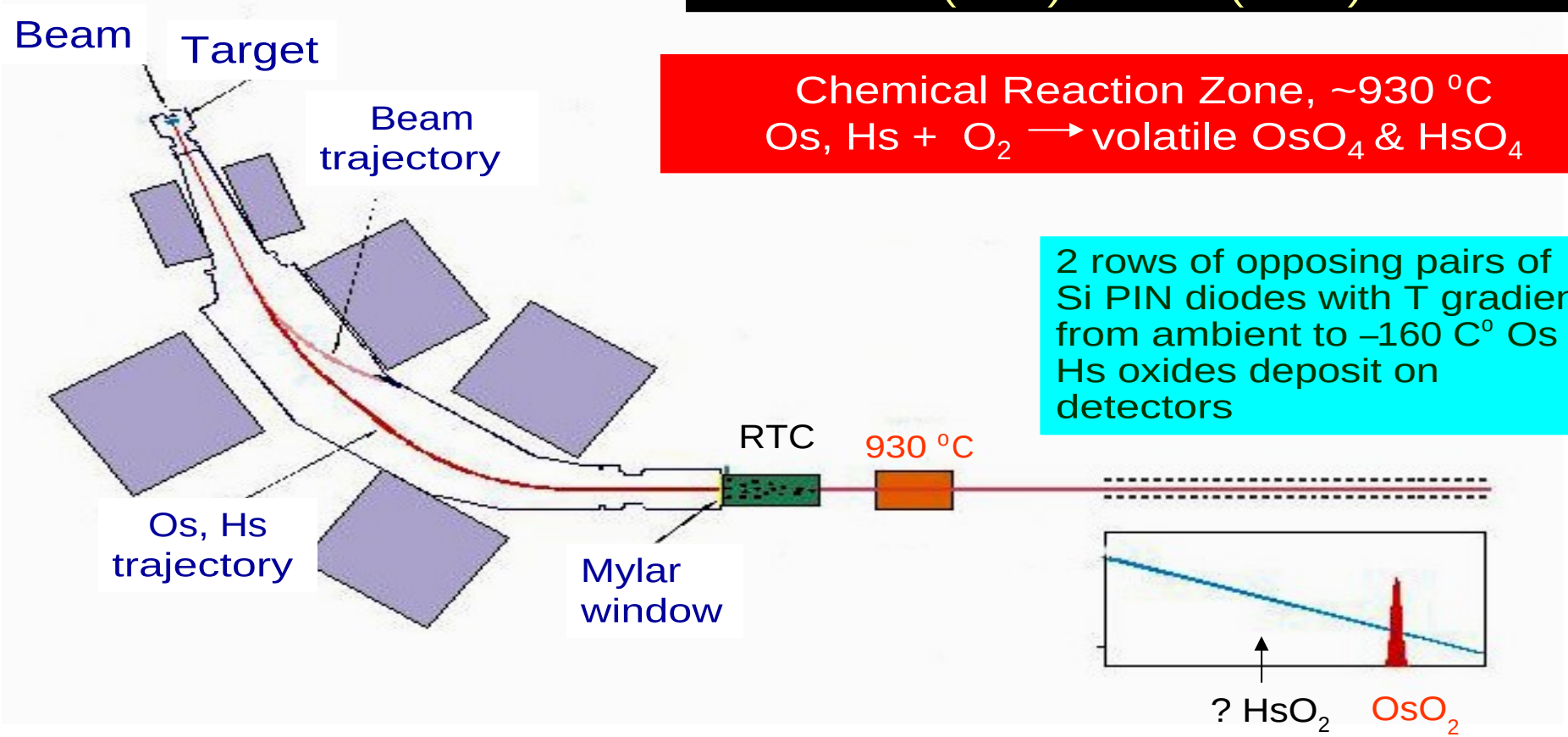
Chemical Studies of Hassium (Element 108)

$^{248}Cm(\ ^{26}Mg, 5n)$ 10 pb—>^{269}Hs (9 s)

Cryo-Thermochromatographic Separator (CTS) coupled to BGS

Recoil Transfer Chamber (RTC)
Transport activity from BGS
(0.2T) to CTS (550T)

Chemical Reaction Zone, ~930 °C
Os, Hs + O_2 → volatile OsO_4 & HsO_4

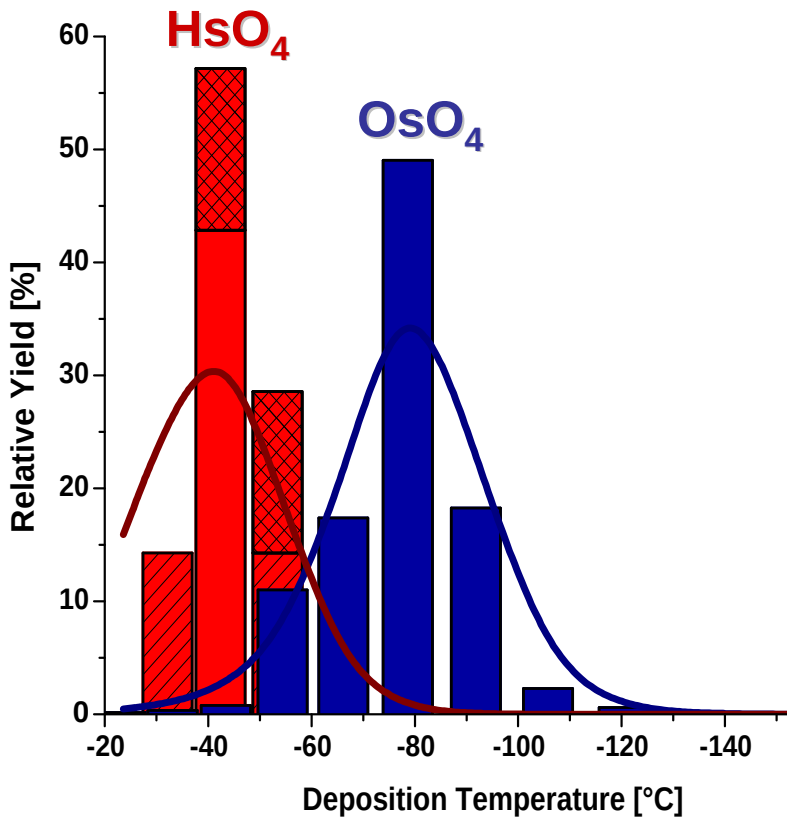


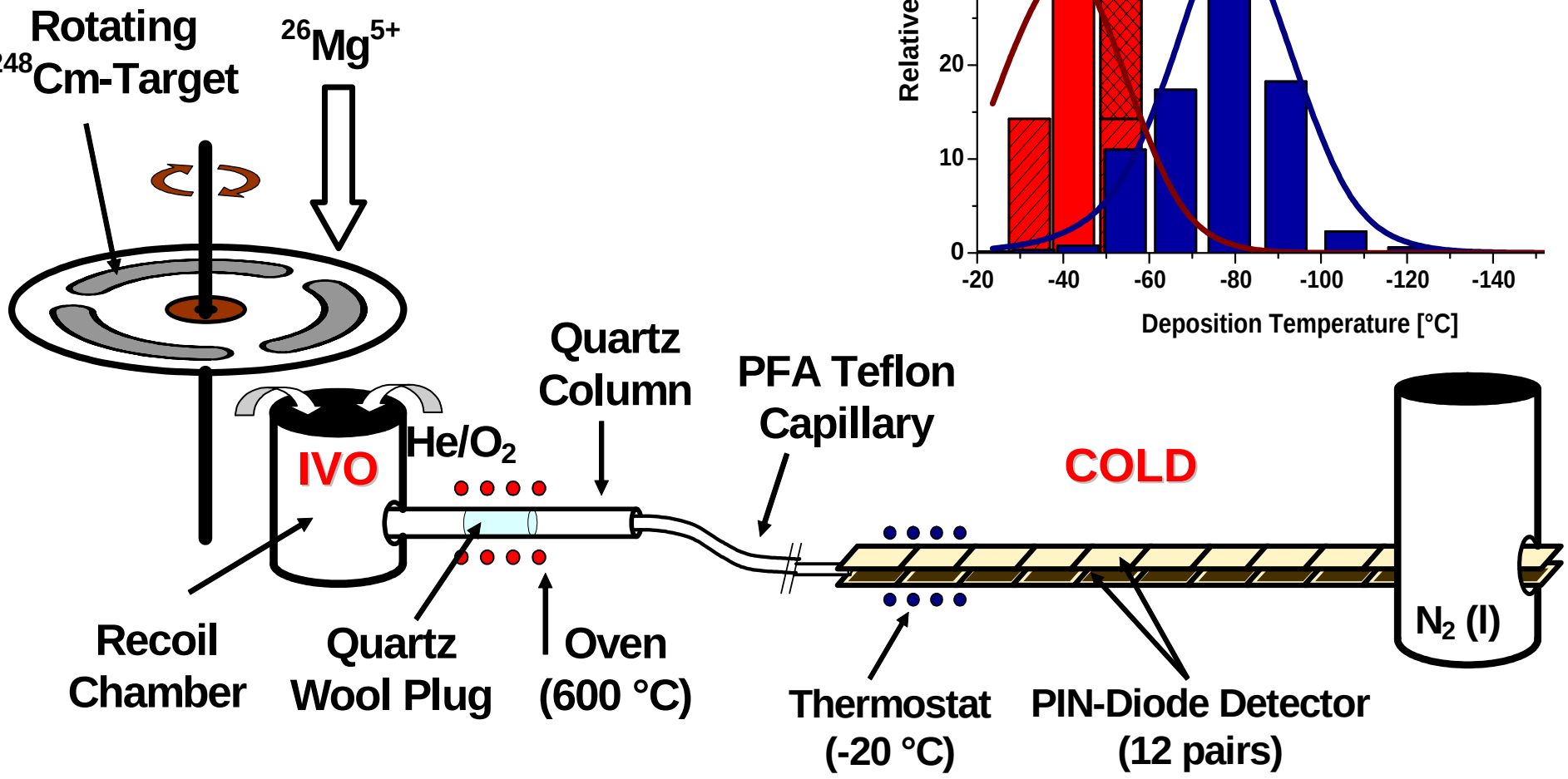
2 rows of opposing pairs of Si PIN diodes with T gradient from ambient to –160 C° Os & Hs oxides deposit on detectors

First Chemical Studies of Hassium

International Collaboration: PSI, GSI, LBNL

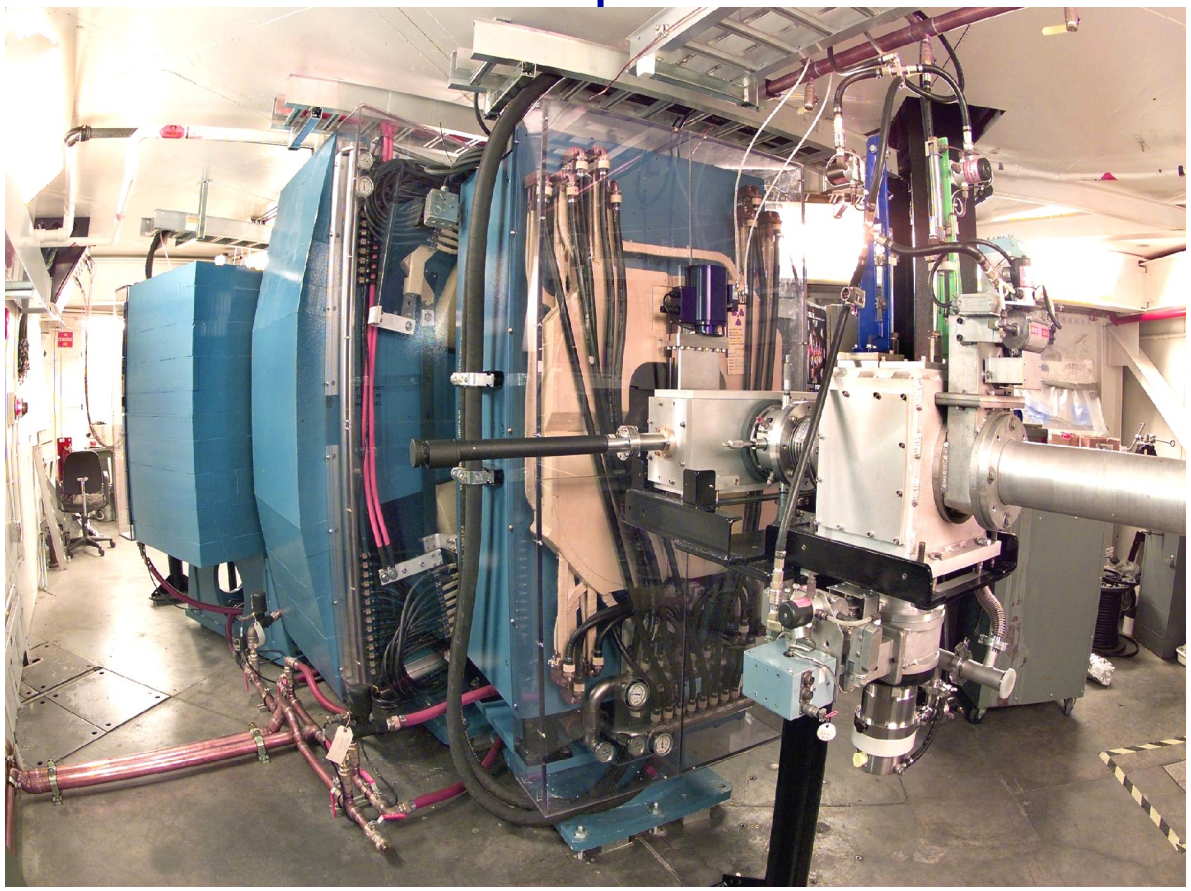
In-situ Volatilization Chamber (IVO) & Cryo On-Line Detector (COLD)

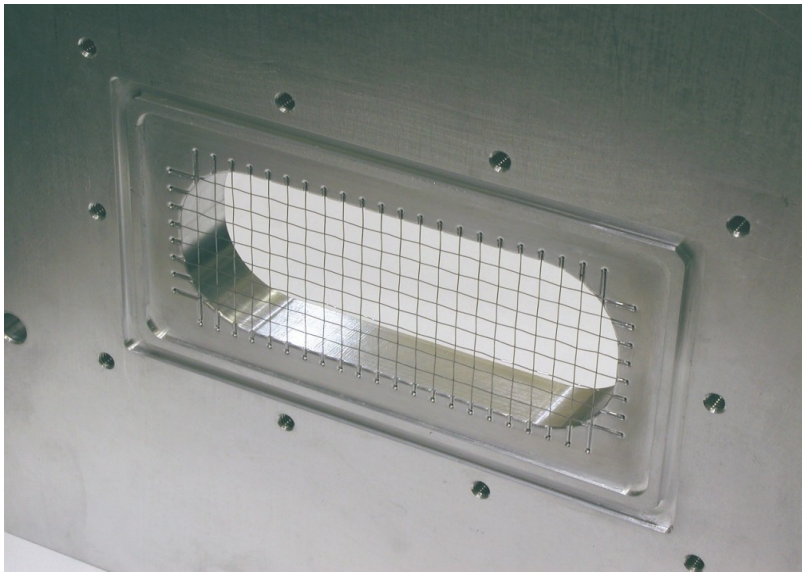




Ch.E. Düllmann et al. Nature 418 (2002) 859

BGS as Pre-Separator for Chemical/Nuclear Studies





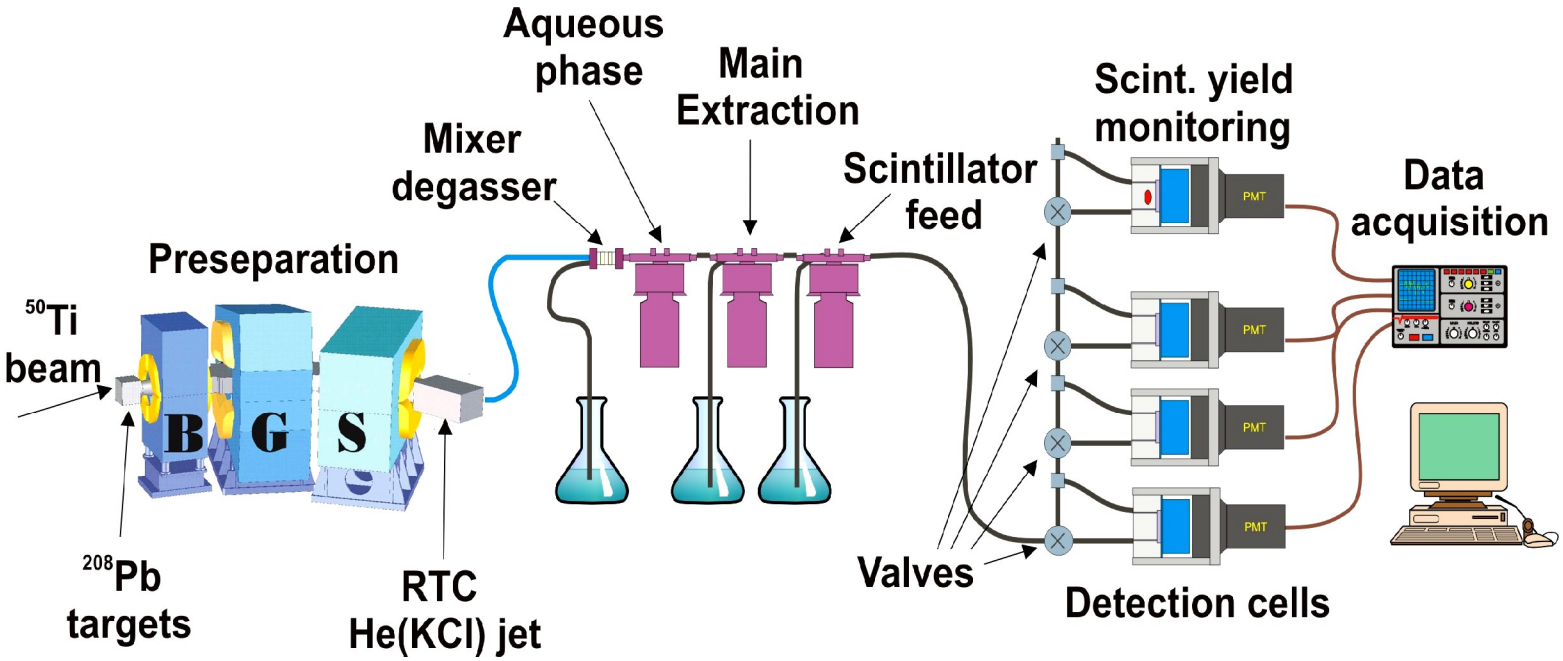
- **Provides decontamination from plethora of unwanted products.**
- **Uniquely suited for use at high beam intensity accelerators.**
- **Demonstrated with Cryogenic Thermo-Chromatographic Separator to study volatilities of Group 8 tetroxides, Os, Hs.**
- **BGS-SISAK studies of Rf show feasibility for studying solution chemistry of other short-lived transactinides.**

Recoil Transfer Chamber

Important for pre-separation prior to other chemical studies.

SISAK (Short-lived Isotopes Studied by the AKufve technique)

Continuous liquid-liquid extractions & detection

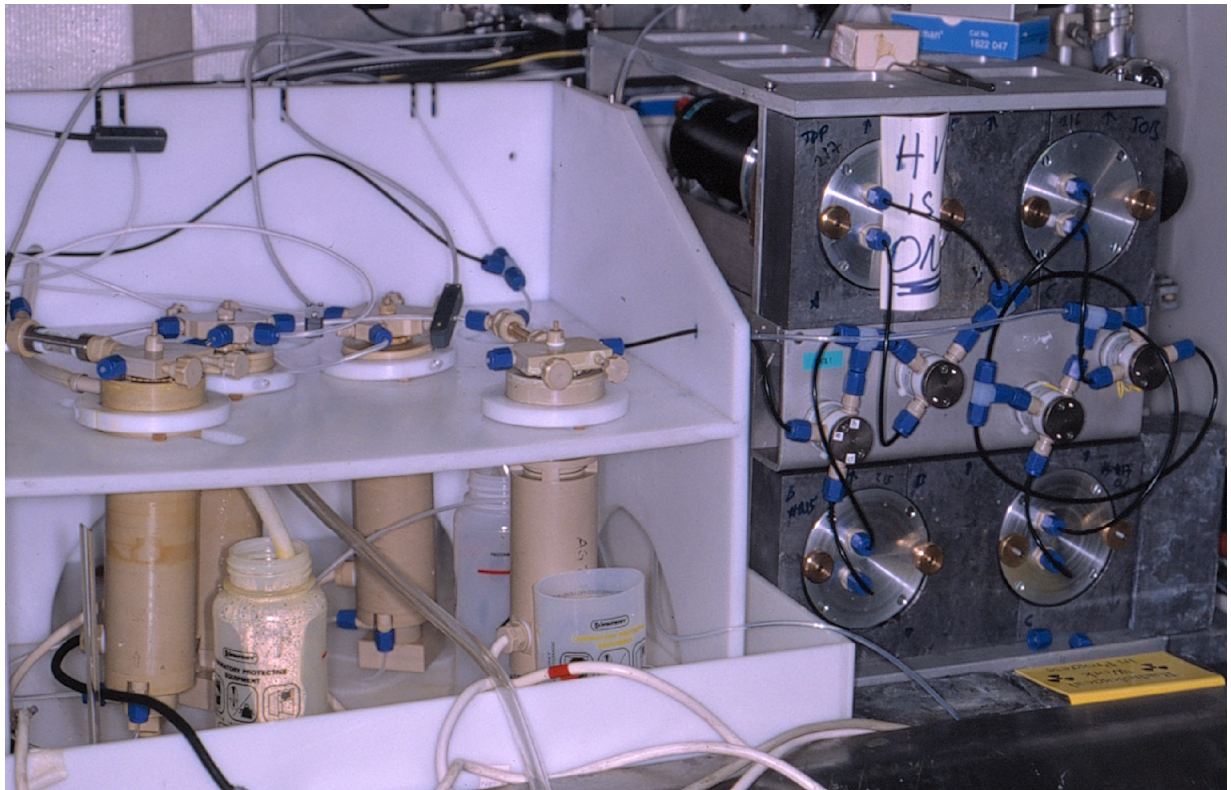


First successful transactinide chemistry experiment with SISAK.

Detected 24 ^{257}Rf (4s half-life) α-decays in 17 hours.

Proved flowing liquid scintillator system can be used for TANs.

Demonstrated advantage of using BGS as a pre-separator.



SISAK Collaboration Group, Norway, Sweden, Germany, USA

Berkeley, November 2000

BERKELEY LAB

Technical Approaches

CHEMICAL SEPARATION

THEN-MANUAL 1985

Repeated *"SRAFP"* collections of recoil products transported via He-jet followed by rapid liquid-liquid extractions or column chromatography.

NOW & FUTURE- AUTOMATED

ARCA & SISAK for Solution Chemistry
HEVI & OLGA for Volatility Studies
BGS as Pre-Separator/Recoil Transfer Chamber (RCT)
In-situ Volatilization On-line (IVO) & Cryo-On-Line Detector (COLD) for rapid cyrogenic gas-phase separations.

DETECTION

Passivated, ion-implanted planar silicon detectors (PIPS)/ Pin Diodes for alpha & SF detection & kinetic-energy measurements.
Multiple detector systems for aqueous chemistry.
Rotating wheel system (MGA) for collection & detection.
Flowing Liquid Scintillation Systems.
Record time, energy, position via computer.

Chemical Periodic Table of the Elements 2005

1																	18
1 H	2											13	14	15	16	17	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg	3	4	5	6	7	8	9	10	11	12	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
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
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
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
87 Fr	88 Ra	89 Ac	104 Rf	105 Ha (Db)	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	*112*	***113***	***114***	***115***	***116***		(118)

Rf, Ha, Sg Solution & Gas-phase

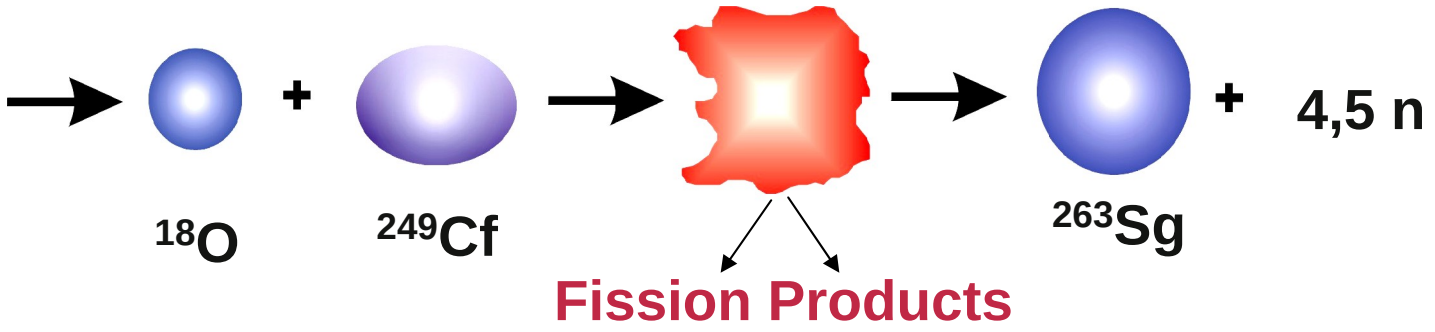
Bh, Hs Gas-phase

Mt, Ds, Rg ? ? ?

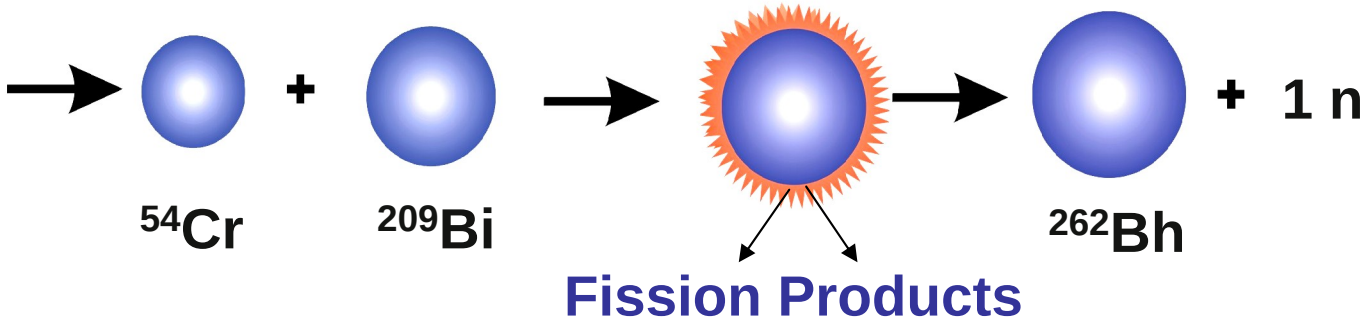
Lanthanides	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
Actinides	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

HEAVY ELEMENT PRODUCTION REACTIONS

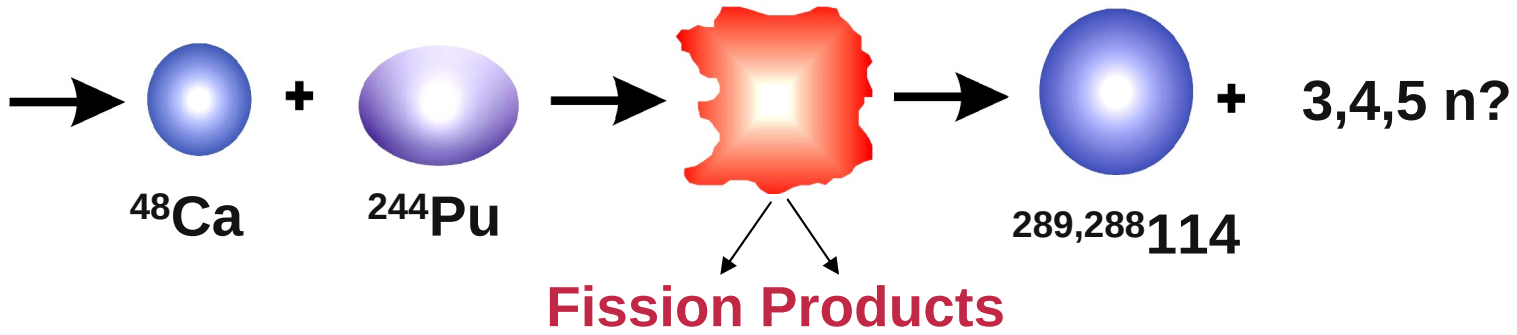
Hot Fusion—Elements 104 through 106



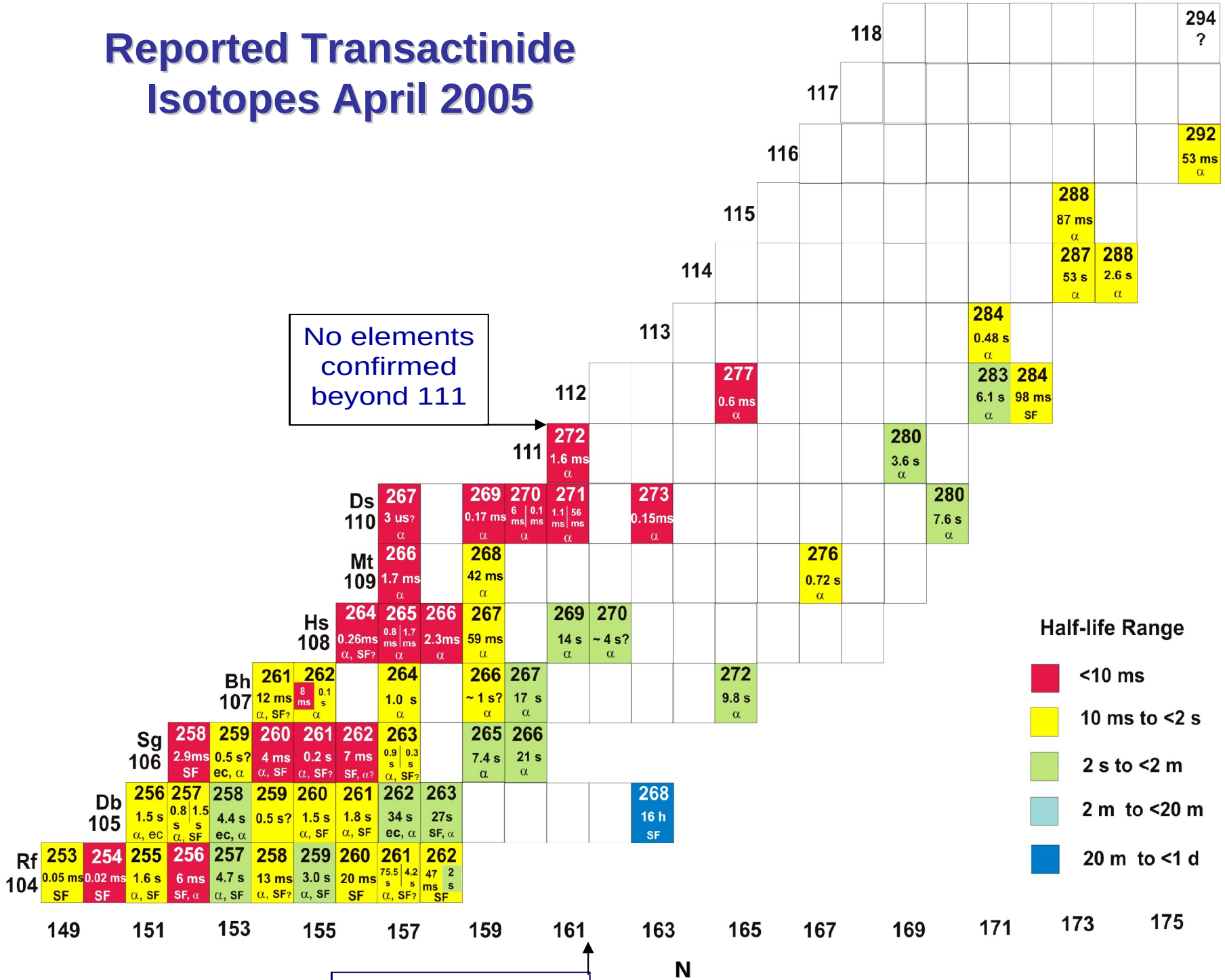
Cold Fusion—Elements 107 through 112



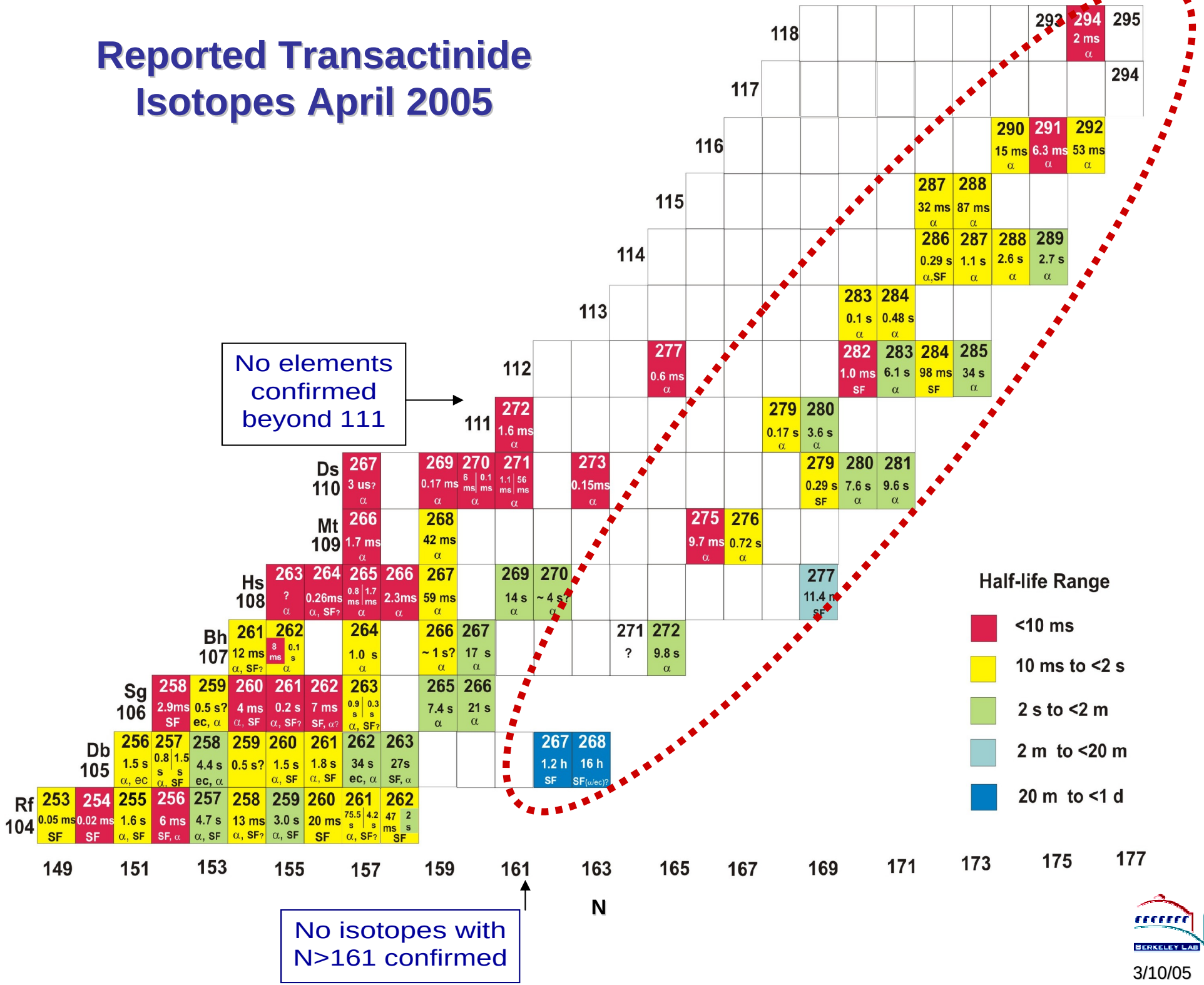
Hot (warm) Fusion- reported elements 114,115 & 116



Reported Transactinide Isotopes April 2005



Reported Transactinide Isotopes April 2005



Advantages of Pre-separation– *Opportunities for Chemistry*

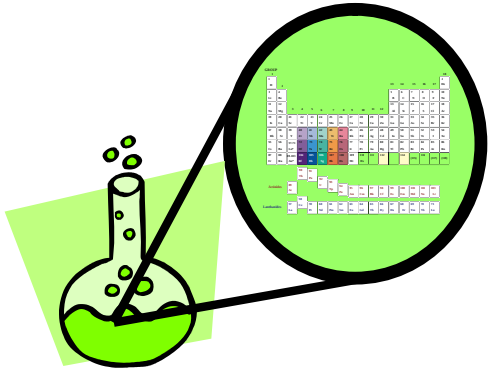
Chemistry without pre-separation

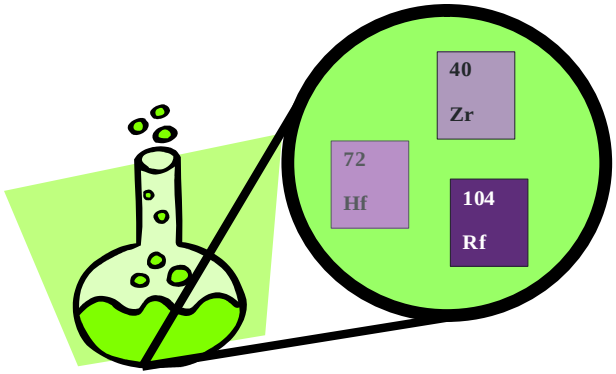
Chemical system needs to separate out all interfering nuclides.

Chemistry with pre-separation

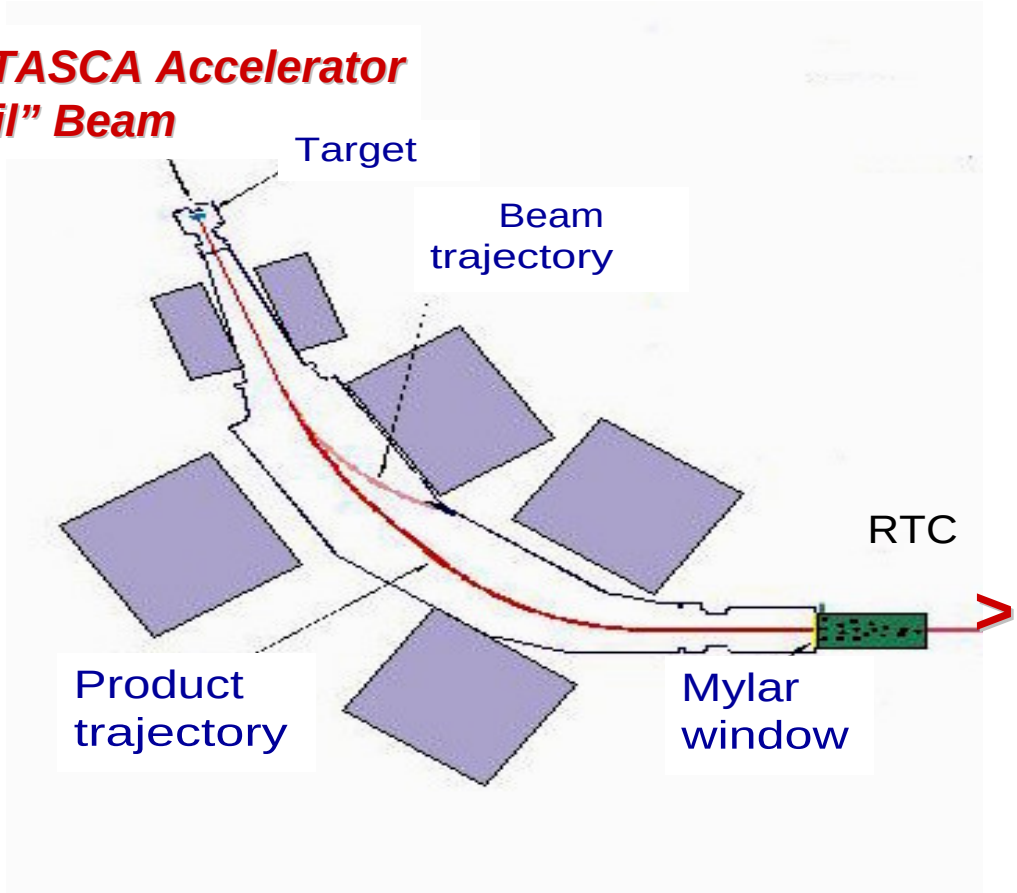
Chemical system can favor selectivity between homologues over removal of interfering nuclides..

Opens way to classes of chemical systems previously deemed unsuitable.



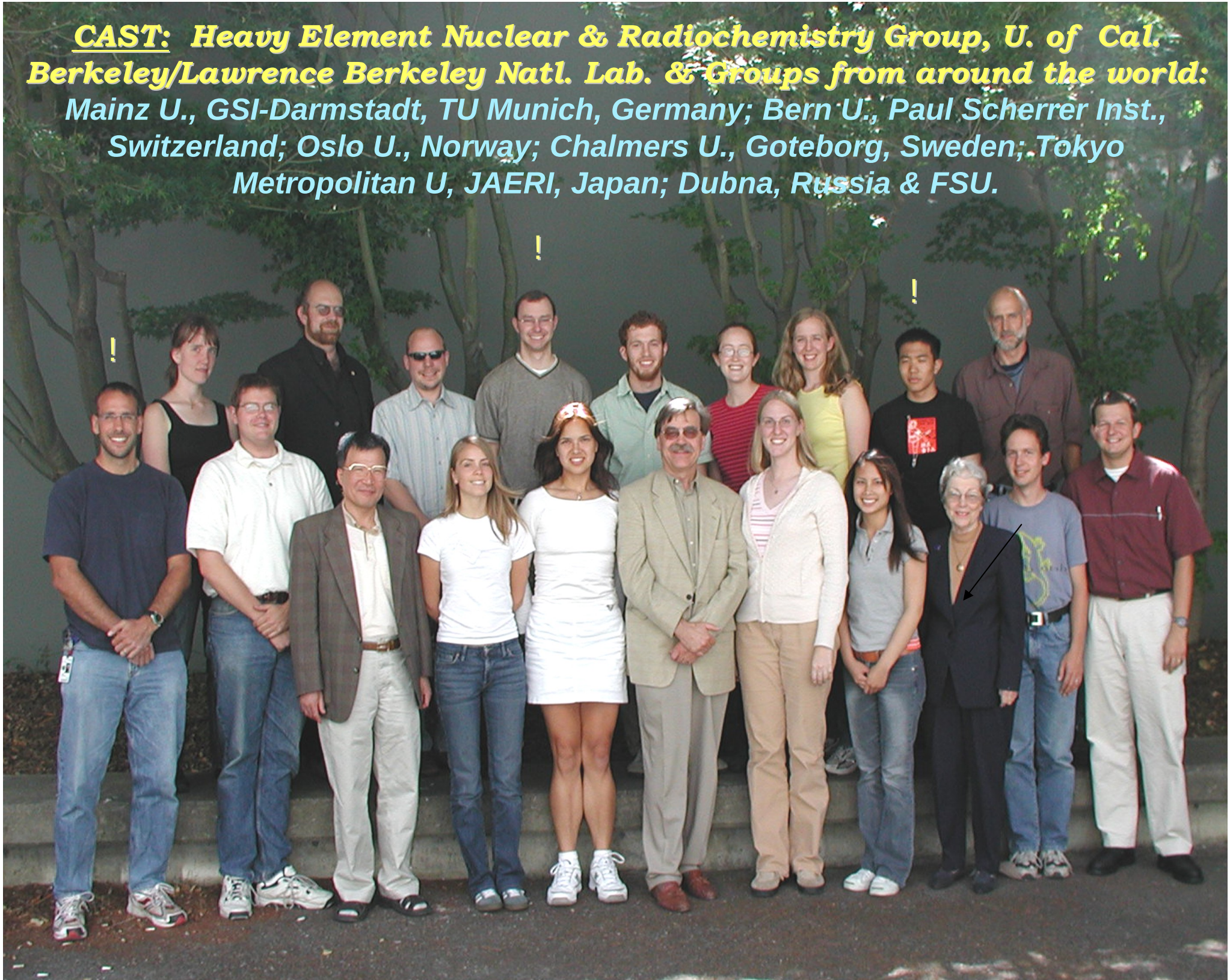


BGS or TASCA Accelerator
"Cocktail" Beam



Separation Sites

- SISAK
- HEVI/OLGA
- IVO-COLD
- CTS
- L/L extractions (crown ethers)


CAST: Heavy Element Nuclear & Radiochemistry Group, U. of Cal. Berkeley/Lawrence Berkeley Natl. Lab. & Groups from around the world:
Mainz U., GSI-Darmstadt, TU Munich, Germany; Bern U., Paul Scherrer Inst., Switzerland; Oslo U., Norway; Chalmers U., Goteborg, Sweden; Tokyo Metropolitan U, JAERI, Japan; Dubna, Russia & FSU.

RELEVANCE

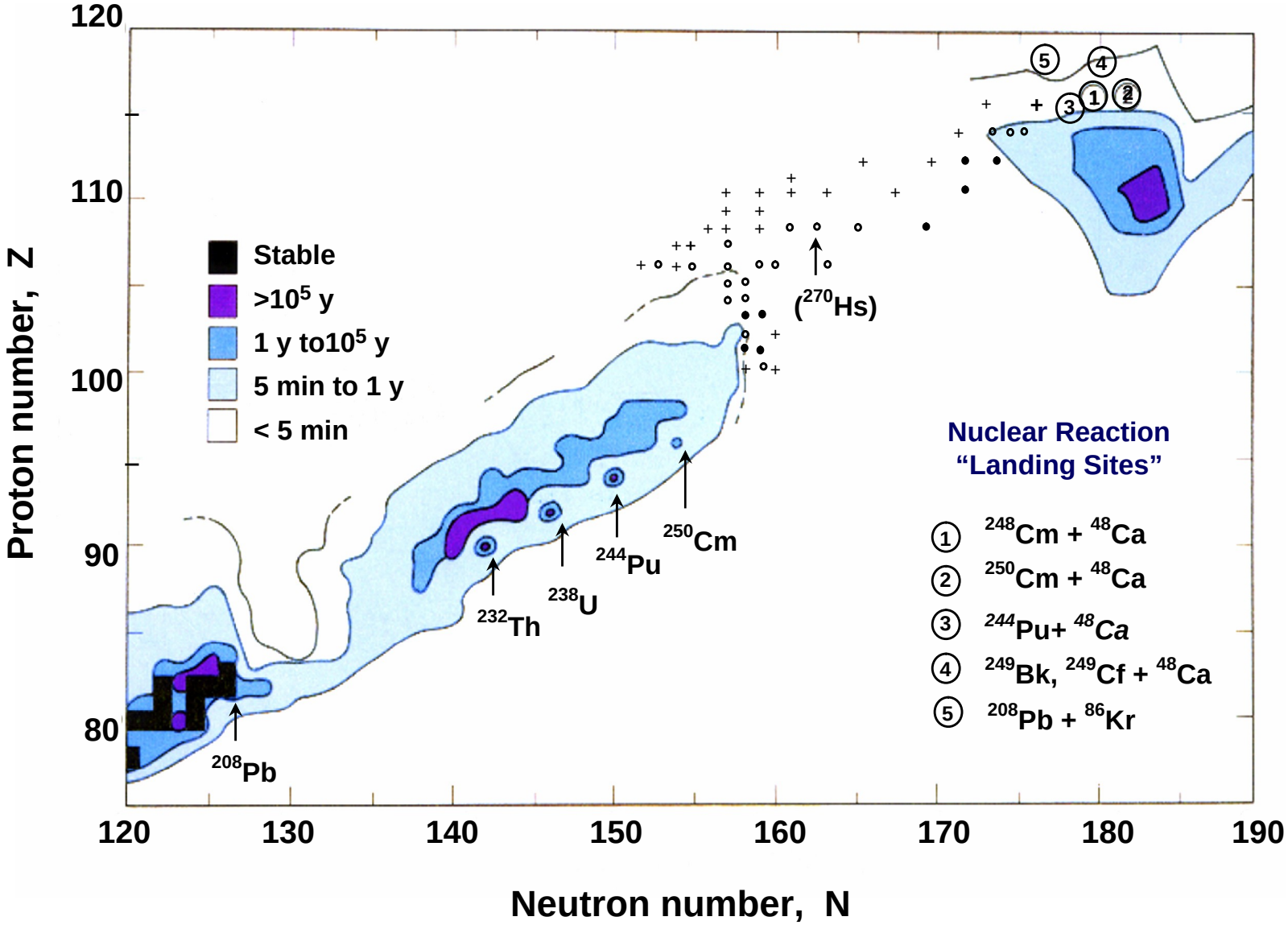
SEVERE CURRENT & FUTURE SHORTAGE OF NUCLEAR SCIENTISTS

Exotic, frontier studies attract many undergraduate and graduate students to nuclear & radiochemistry.

Excellent education & training for future careers & contributions to basic research & teaching as well as a variety of applied areas:

- **Ultrasensitive & radioanalytical analyses.**
- **Surveillance of clandestine nuclear activities.**
- **Automated, computer-controlled remote processing systems.**
- **Nuclear medicine, isotope production, radiopharmaceutical preparation; diagnostics & therapy.**
- **Nuclear power: reactor design & performance.**
- **Treatment, processing, & minimization of nuclear waste.**
- **Nuclear waste isolation & site remediation.**
- **Environmental studies: prediction & monitoring of behavior of actinides & other species in the environment.**
- **Stockpile stewardship & nuclear surveillance.**

Contour Plot 2005



<0.1 s (+) 0.1 s to 0.5 min (o) >0.5 min (●)

FUTURE CHEMISTRY

1. **Use BGS to Identify longer-lived isotopes of Mt,Ds,Rg:**

   Mt: ^{238}U (^{37}Cl, 4,5n)^{271,270}Mt, s?; Ds: ^{238}U(^{40}Ar, 4,5n) ^{274,273}Ds. 0.3 pb,?s;

   Ds: ^{244}Pu(^{48}Ca,3n)^{289}114→^{281}Ds, 9.6 s??

   Rg: ^{243}Am(^{48}Ca,3n) ^{288}115→α,α → ^{280}Rg, 3.6 s

   (Theoreticians predict behavior similar to Pt, Au, Hg.)

2. **Identify 112 α- emitter: ^{238}U(^{48}Ca, 3n) → ^{283}112, 6 s, 3 pb??**

   Is it more like Rn or Hg as others predict?

3. **Use BGS as pre-separator for chemical studies.**

   Prepare to use ^{244}Pu targets in BGS.

   More detailed studies of solution & gas-phase chemistry of Rf through Sg–volatile complexes .

   Study solution chemistry of Bh & Hs.

4. **Continue close interaction with both nuclear & chemical theorists.**

5. **If much longer-lived SHEs identified, devise methods for increasing yields, and "stockpiling" them.**

(TASCA similar to BGS being built at GSI by international cooperation with various working groups, etc.)

Happy 85th Birthday!
Prof. John R. Huizenga
21 April, 2006