# "Super Heavy Elements and Nuclei"

Krzysztof P. Rykaczewski Physics Division, Oak Ridge National Laboratory

This presentation is mostly about the heaviest elements and nuclei at the Island of (enhanced) Stability

#### Russia-US

JINR Dubna - ORNL Oak Ridge - LLNL Livermore UT Knoxville - RIAR Dmitrovgrad - Vanderbilt Nashville

Germany and the rest of the World

Darmstadt-Mainz-Lund-Oak Ridge-Berkeley et al

US

LBNL Berkeley-UC-LLNL-GSI-Mainz-Lund-Oregon

#### Japan

RIKEN- Uni. Kyushu and other Japanese labs



EBSS 2016, July 2016, MSU



Rose Boll and Shelley Van Cleve purifying <sup>249</sup>Bk (~40 Ci) at ORNL



Yuri Oganessian and V. Utyonkov correcting Z=117 Letter at Dubna



MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY



# Mantra for today's talk, EBSS and our research

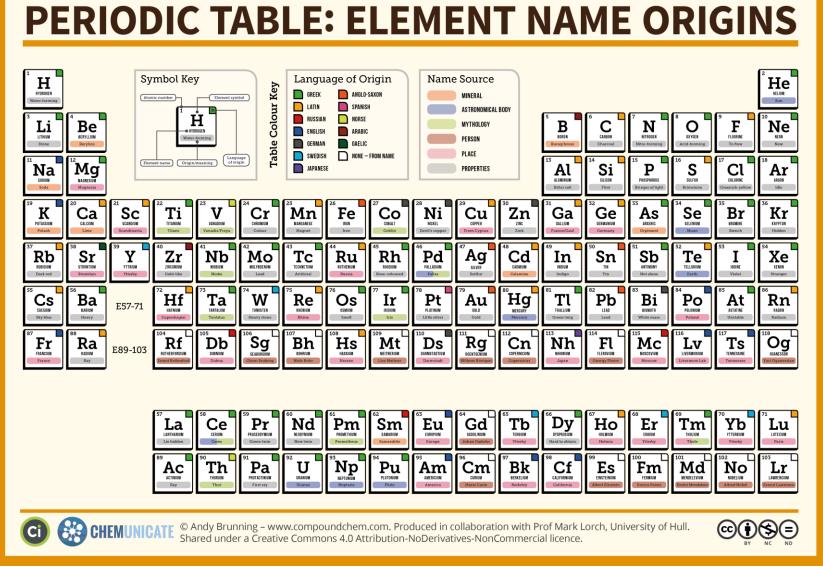


# Why we are studying (decays of) exotic nuclei far from beta-stability path?

effects not present or weak in stable nuclei are often appearing or getting enhanced in nuclei far from  $\beta$ -stability, in nuclei with "unusual" proton-to-neutron content

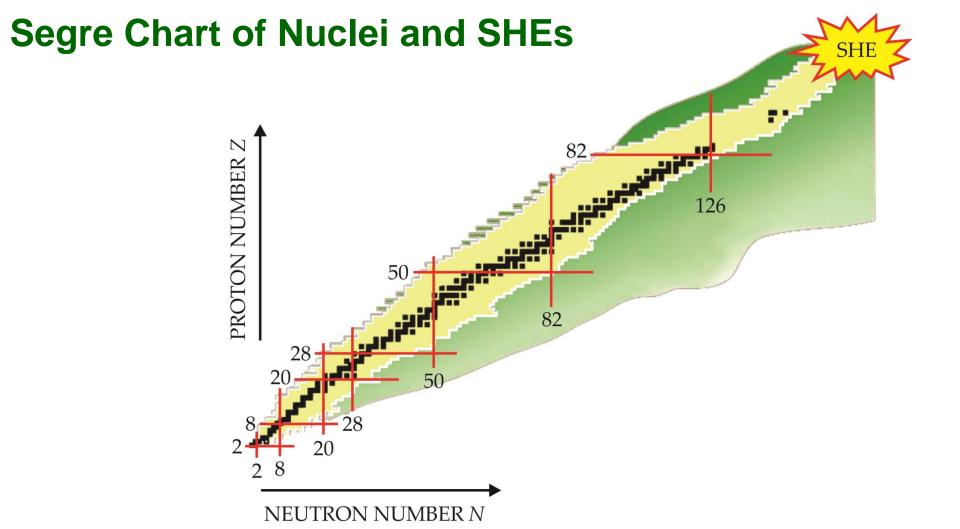
".. decay data offer last verification points for theory before extrapolating into unknown ..."

... and our goal is to understand the origin of structure and processes involving atomic nuclei



Four provisional names accepted/announced by IUPAC/IUPAP, with symbols Nh for 113, Mc for 115, Ts for 117 and Og for 118, are included

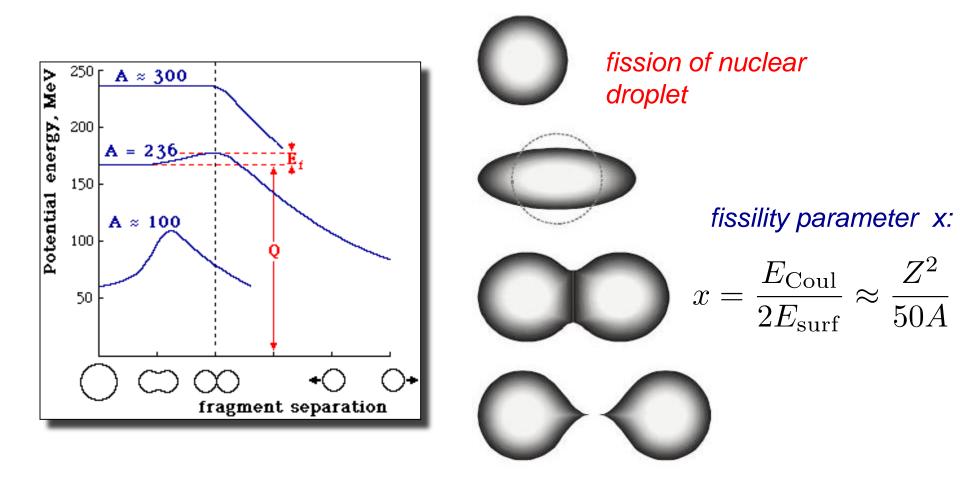
National Laboratory



The definitions of SHE vary, from giving atomic number limit like Z>100, to the "true SHEs", namely atoms of nuclei stabilized against fission by nuclear structure effects like a shell correction to the binding energy.



## Fission of charged nuclear droplet



The nuclear droplet stays stable and spherical for x<1. For x>1, it fissions immediately. For  $^{238}$ U, x=0.71, for  $^{294}118$  x=0.95. Fissility x is not an universal measure of stability against fission since nuclei are (usually) not spherical.

from Witek Nazarewicz

#### Fission Barriers of Compound Superheavy Nuclei

J. C. Pei, 1,2,3 W. Nazarewicz, 2,3,4 J. A. Sheikh, 3,3 and A. K. Kerman 2,3,5

# Fission barrier in the excited state!

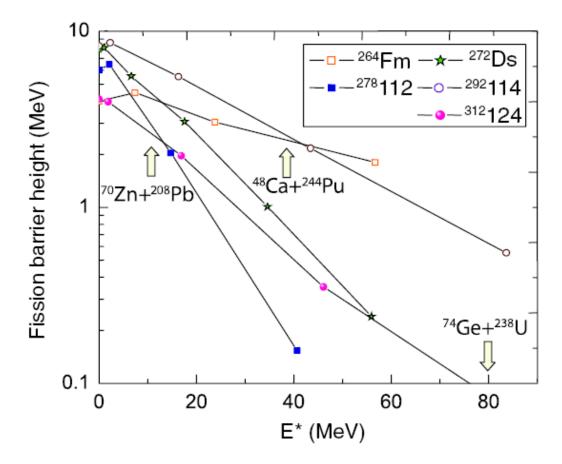
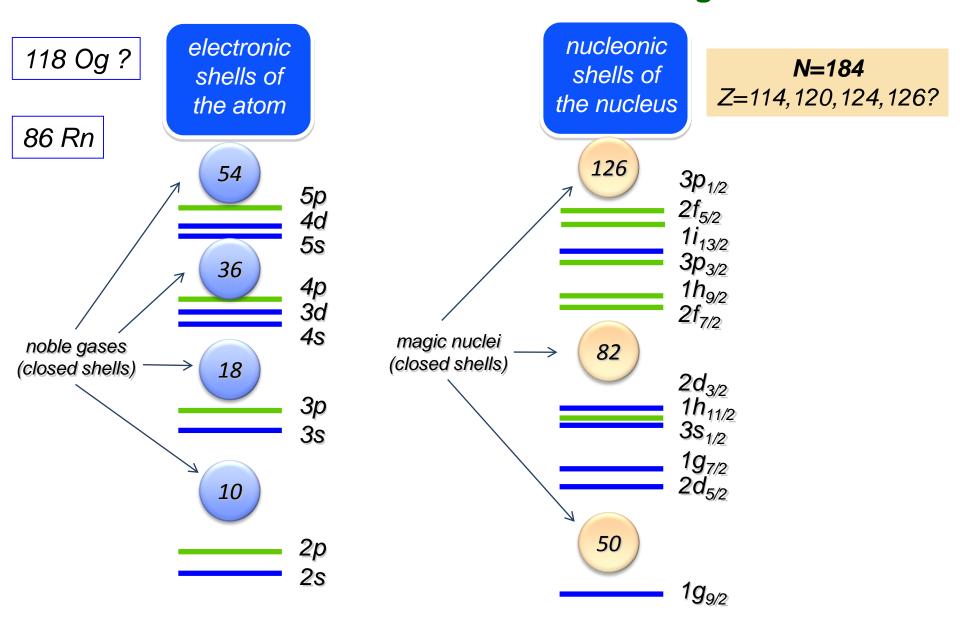


FIG. 4 (color online). The height of the inner fission barrier in  $^{264}$ Fm,  $^{272}$ Ds,  $^{278}$ 112,  $^{292}$ 114, and  $^{312}$ 124 as a function of excitation energy  $E^*$ . The effect of triaxiality on the fission barrier has been included. The experimental values of  $E^*$  corresponding to CN formed in the reactions indicated are marked by arrows.



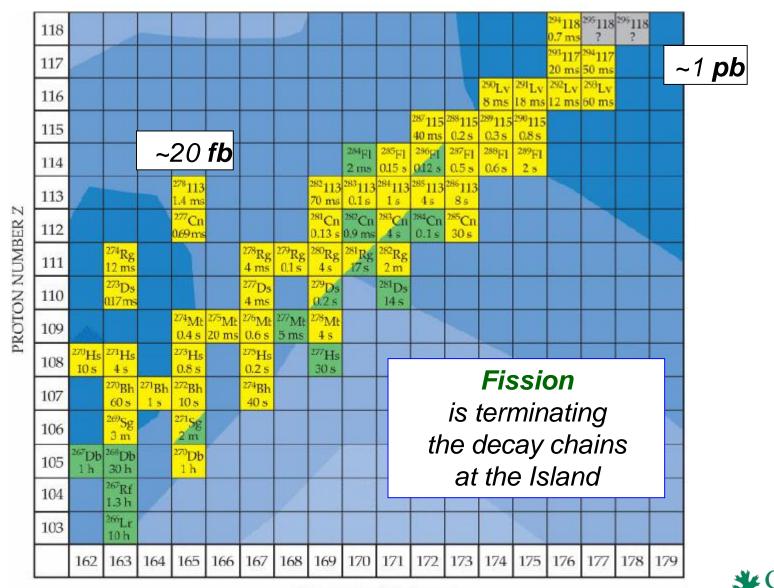
#### Shell effects come to the rescue of nucleus against fission



from Witek Nazarewicz

# The Island of Stability of Super Heavy Nuclei

aka "Hot Fusion Island " of over 50 nuclei produced in <sup>48</sup>Ca+actinides targets reactions



#### Long term goals - science drivers for SHE research

#### New Heaviest Elements and Nuclei

- how many protons and neutrons a nucleus can hold?
- unified description of nuclear properties across varying proton and neutron numbers
- new energy gaps, magic numbers and Island of Stability or rather enhanced stability without shell gaps and magic numbers
- understanding fission process competing with other decay modes ( $\alpha$ , EC)
- structure beyond ground-state properties of super heavy nuclei

#### Understanding production mechanism of the heaviest nuclei

- hot and cold fusion reactions with stable and radioactive nuclei
- multi-nucleon transfer between very heavy nuclei

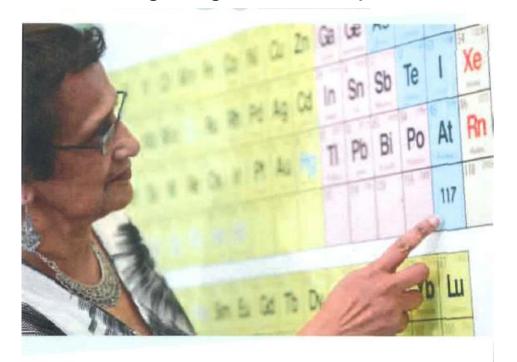
#### Expansion of Periodic Table of Elements

- relativistic effects in chemical properties of atoms (radius, ionization, compounds)
- super heavy atoms in the Universe



#### The element name will stay forever in the Periodic Table

Oak Ridge High School, April 2010



**International team discovers element 117** 

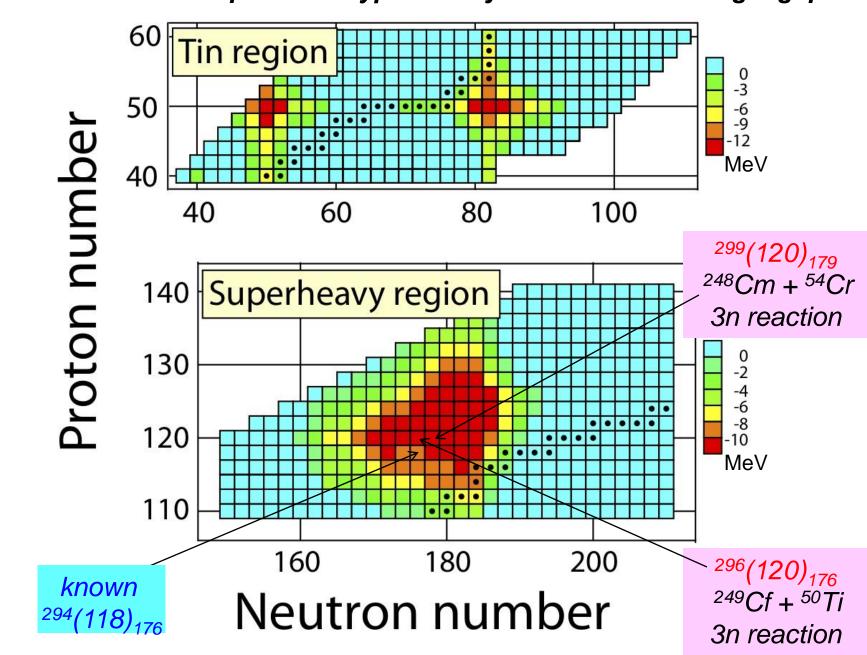
Now tennessine Ts

"Element 117" ice cream
Razzleberry Ice Cream Lab
Oak Ridge



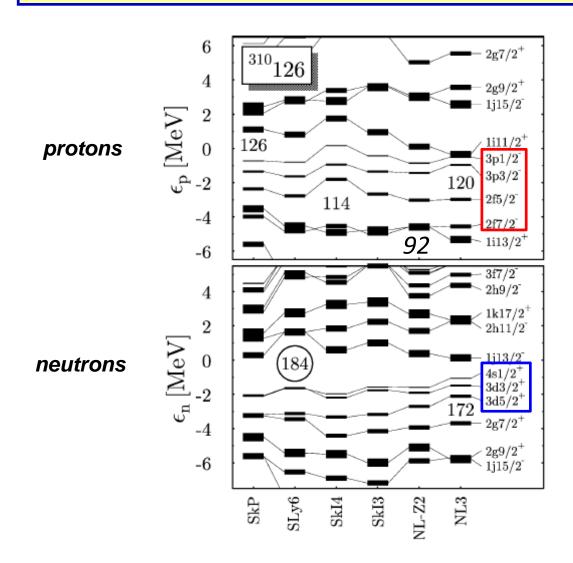
IUPAC/IUPAP press release on 8<sup>th</sup> June 2016, where element 117 was provisionally named tennessine (Ts), might trigger a development of a new brand of Jack Daniels Whisky "No.117"

M. Bender, W. Nazarewicz, P.-G. Reinhard, Phys. Lett. B 515, 42, 2001 "Shell stabilization of super- and hyper-heavy nuclei without magic gaps"



## Single-particle levels in the region of super heavy nuclei

M. Bender, W. Nazarewicz, P.- G. Reinhard, PL B 515, 42, 2001

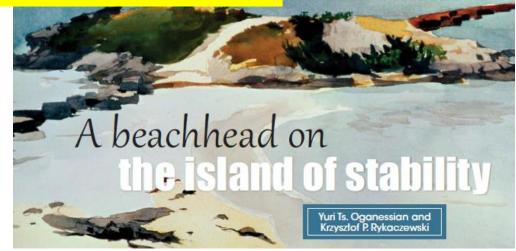


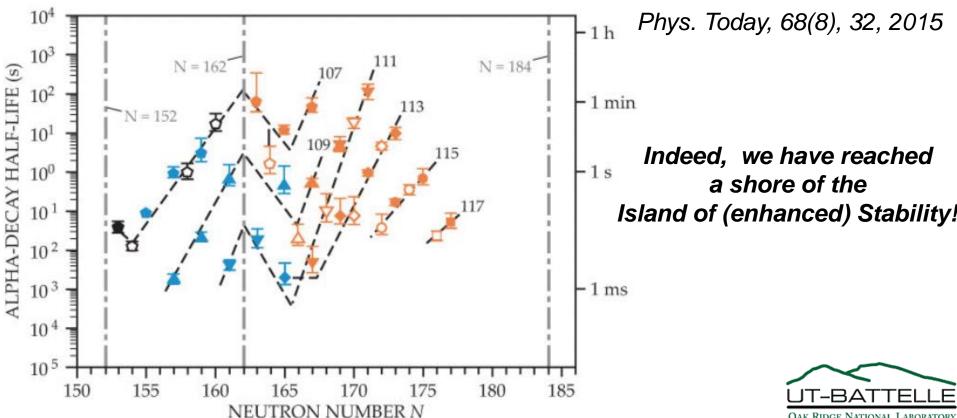


## 2016 - 50 Years of the Island of Stability

The concept of an island of stability has dominated the physics of superheavy nuclei since middle sixties, see, e.g.

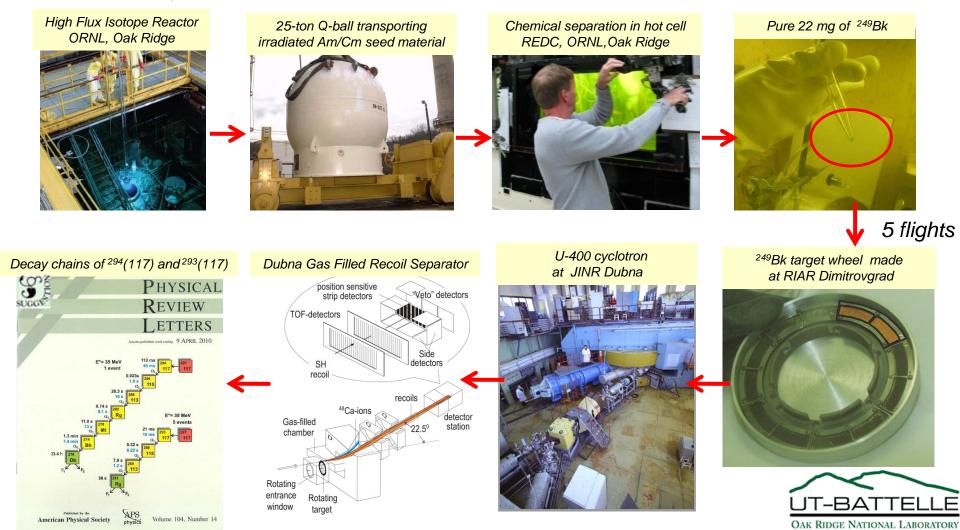
Myers-Świątecki 1966 Sobiczewski-Gareev-Kalinkin 1966 Viola-Seaborg 1966





# 2009-2010: Synthesis of a New Element with Atomic Number Z=117 (Ts) Dubna-Oak Ridge-Nashville-Livermore-Dmitrovgrad-Las Vegas

The identification of a new element Z=117 among the products of the <sup>249</sup>Bk+<sup>48</sup>Ca reaction was enabled by the close collaboration and unique capabilities of the US and Russia laboratories, neither country could achieve it alone. The 327 days half-life of radioactive <sup>249</sup>Bk required a coordination of two years neutron irradiation and chemical separation at Oak Ridge followed by a target production at Dimitrovgrad and six months experiment with an intense <sup>48</sup>Ca beam at Dubna.



# Actinide production by irradiation of Am/Cm targets in HFIR

Targets specially designed for reactor conditions

Irradiation in the HFIR flux trap

- Composition controls fission and gamma heating (and decay heat)
- Irradiated targets typically remain in the reactor for 3 to 6 24-day cycles (up to 7 cycles/year)
  - Fuel change-out at HFIR

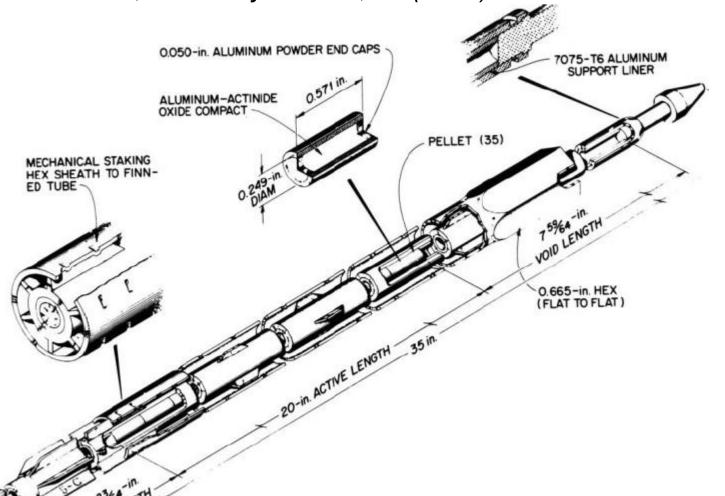
- Intense thermal neutron flux (2.5 × 10<sup>15</sup> neutrons/cm<sup>2</sup>·s)
- 31 target positions (6-8 targets typical for Cf production)
- Typical campaign now: up to 100 mg <sup>252</sup>Cf, 10 mg <sup>249</sup>Bk, ~1 µg <sup>254</sup>Es

Target positions in the flux trap of HFIR fuel assembly



J. Roberto, SHE Symposium 2015

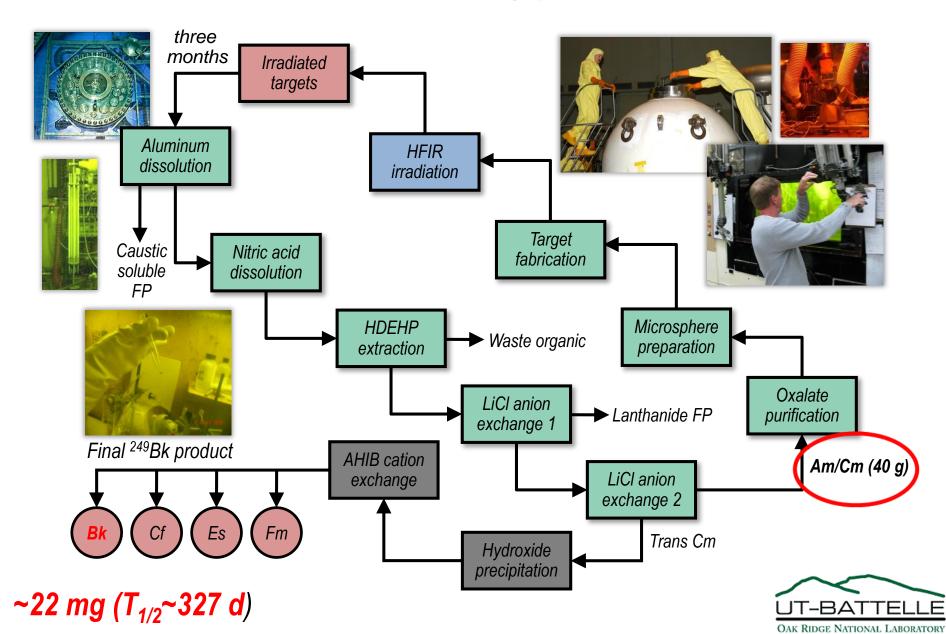
#### J.B. Roberto et al., Nucl. Phys. A 944, 99 (2015)



9 super-heavy elements (Z=104-106, 113-118) synthesized using actinide targets materials mostly from ORNL Oak Ridge and RIAR Dmitrovgrad



# Z=97 <sup>249</sup>Bk production/separation cycle at ORNL 24 months <sup>252</sup>Cf/<sup>249</sup>Bk



# The "voyage" of <sup>249</sup>Bk material started June 6<sup>th</sup>, 2009 at Oak Ridge Oak Ridge - Knoxville - Jamaica (!) - JFK NY - SVO Moscow - JFK NY - SVO Moscow JFK NY - SVO Moscow (frequent flyer miles for Bk + my grey hair)



The barrels with <sup>249</sup>Bk were kept for a week at Sheremetyevo airport custom clearance station. Since no radiation was detected outside the package, the crate was open and containers got unpacked till the Geiger's counters went REALLY HIGH!

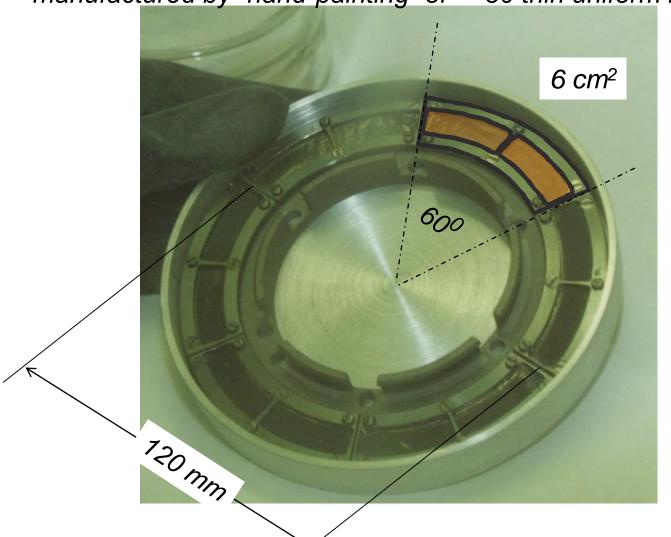
Around 1<sup>st</sup> July 2009 <sup>249</sup>Bk reached target makers at Dmitrovgrad (400 miles from Moscow/Dubna)

It was the fastest transfer of actinide target material from US to Dubna!!



# <sup>249</sup>Bk target wheel manufactured at RIAR Dimitrovgrad (M. A. Ryabinin et al.)

The originally planned electro-deposition of  $^{249}$ Bk didn't work very well. Finally, the target sectors with 0.31mg/cm<sup>2</sup>  $^{249}$ Bk on 0.74 mg/cm<sup>2</sup> Ti foils were manufactured by "hand-painting" of  $\sim$  30 thin uniform layers

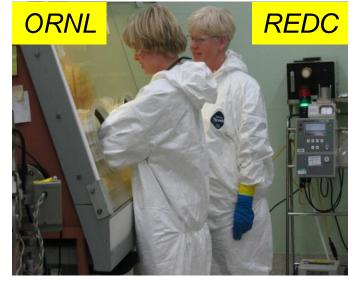


 $\omega = 1700 \text{ rpm}$ 

## Cf isotopic distribution following production and after several decades decay

Isotope	Atomic % (as produced)	Half-life (y)
<sup>249</sup> Cf	3.41	351
<sup>250</sup> Cf	8.7	13.08
<sup>251</sup> Cf	2.60	898
<sup>252</sup> Cf	85.27	2.645
<sup>253</sup> Cf	0.004	0.049
<sup>254</sup> Cf	0.010	0.166





- After several decades, <sup>252</sup>Cf substantially decays (15 half-lives  $\rightarrow$  factor 30,000 down)
- Remaining Cf a mixture of <sup>249, 250, 251</sup>Cf, ~35% <sup>251</sup>Cf (heaviest SHE target material)
- Mixed target must be handled in a shielded glove box due primarily to <sup>250</sup>Cf content
- Possibility of producing two new heaviest isotopes of element 118, A=295 and 296

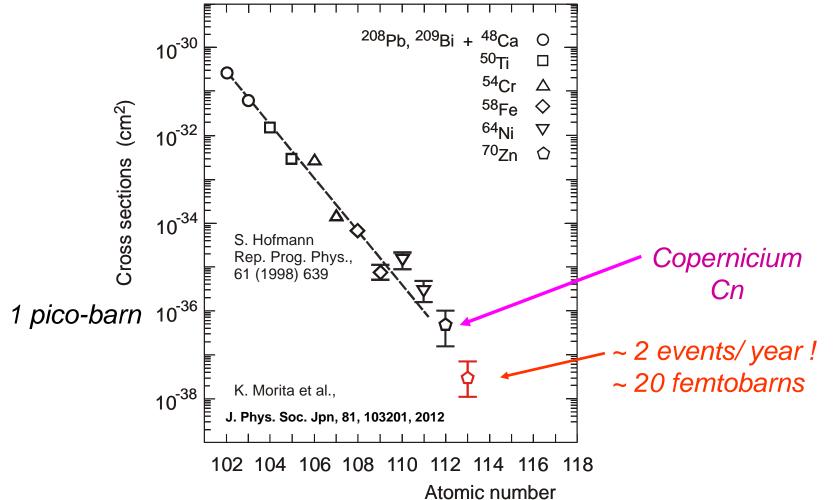
#### Cf recovered from decayed sources

Cf-249 (mg)	Cf- 250 (mg)	Cf- 251 (mg)	Cf- 252 (mg)	Total Cf mg
7.6	2.5	5.7	0.003	15.8
48.1%	15.6%	36.3%	0.02%	3*10 <sup>7</sup> n/s



#### Cross section for formation of SHE nuclei in cold fusion

**Cold Fusion** with magic  $^{208}$ Pb, $^{209}$ Bi targets and n-rich stable beams up to  $^{70}$ Zn  $\rightarrow$  small E\* ~ 10 - 14 MeV barely allowing for ~ 1n emission

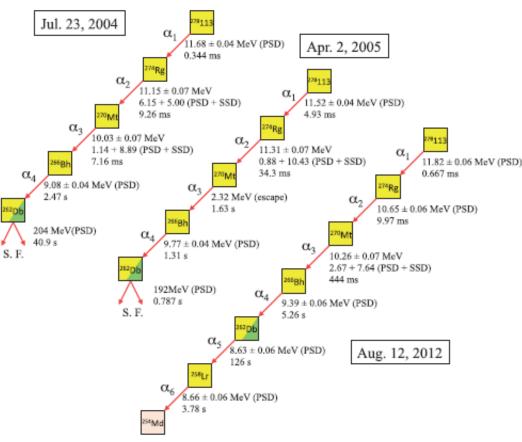




#### Kosuke Morita and his RIKEN team

# $^{70}Zn + ^{209}Bi \rightarrow ^{279}113* \stackrel{1n}{\rightarrow} ^{278}113$





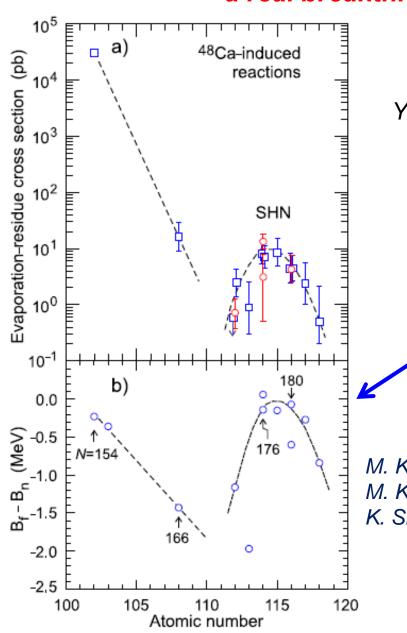
Nearly 600 days of irradiations → 3 events, 22 (+20,-13) fb cross section

## and Z=113 Nihonium Nh



#### Cross sections in hot fusion experiments with actinide targets

"a real breakthrough "Ben Mottelson, June 2016



Yu.Ts. Oganessian and V. K. Utyonkov Nucl. Phys. A 944, 62, 2015

CN survival  $\sim \exp(B_f - B_n)$   $B_f - B_n \sim 0 \text{ MeV}$  $\rightarrow \text{ at the maximum !}$ 

#### **B**<sub>f</sub> and **B**<sub>n</sub> values from

- M. Kowal et al., Phys. Rev. C 82,2010,014303.
- M. Kowal et al. arXiv:1203.5013, 2012.
- K. Siwek-Wilczyńska et al., Phys. Rev. C 86,2012,014611.



# **Separator and detectors**

# Dubna Gas-Filled Recoil Separator DGFRS

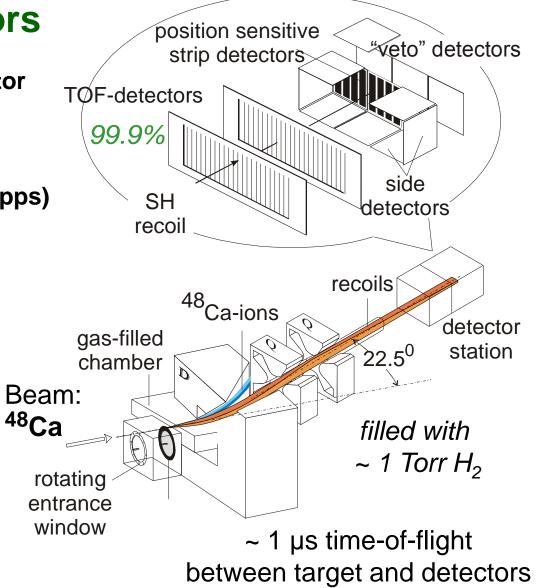
Z=117 experiment with over 1 part×μA <sup>48</sup>Ca beam (7×10<sup>12</sup> pps) for 150 days

#### Transmission for:

evaporation residues 35 % target-like products 10<sup>-4</sup>-10<sup>-7</sup> projectile-like 10<sup>-15</sup>-10<sup>-17</sup>

#### **Detection efficiency:**

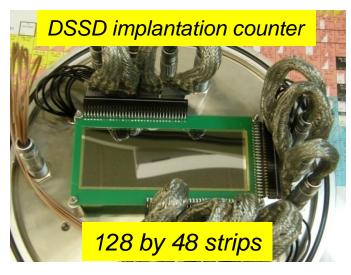
for α-particles 87% for SF: one fragment ~ 100% two fragments ≈ 40%

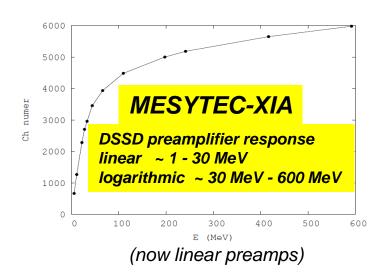


## Position-Time-Energy correlations !!!

# ORNL-UTK 8-detector and digital data acquisition system operating at the Dubna Gas Filled Recoil Separator (DGFRS)

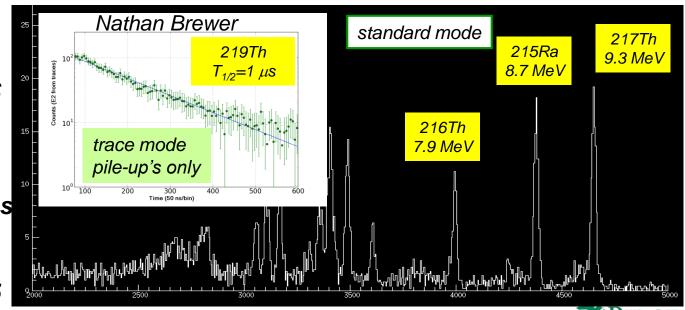






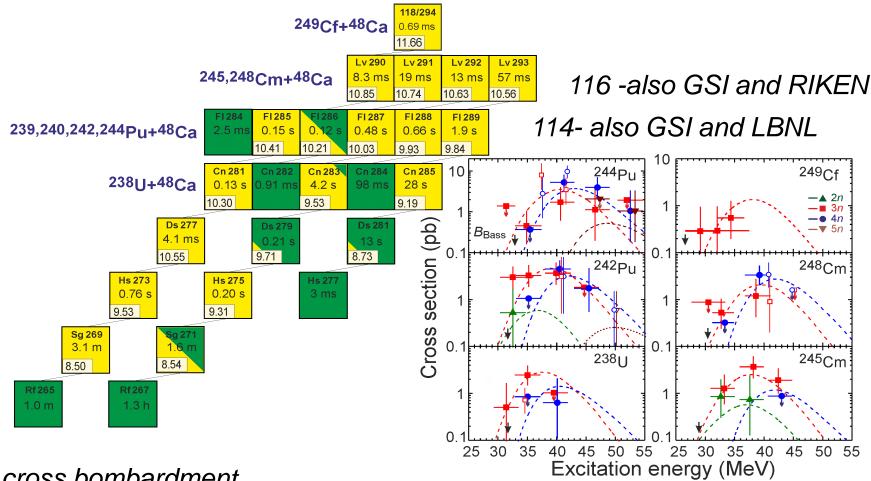
 $\alpha$ -emitters including 1  $\mu$ s activity of  $^{219}$ Th studied at the DGFRS during  $^{48}$ Ca+ $^{nat}$ Yb run Nov.-Dec. 2013.

Search for new isotopes of element Z=114 (FI) with <sup>239, 240</sup>Pu targets Dec. 2013 – June 2015



energy of  $\alpha$ -particles

# RESULTS (Russia-US): even-Z nuclei

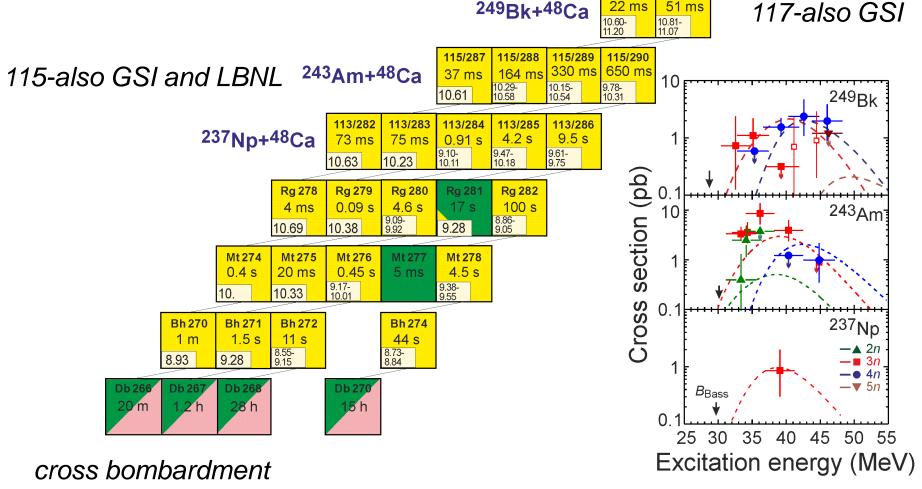


reproducibility, and decay patterns

similarity of excitation functions



# RESULTS (Russia-US): odd-Z nuclei



similarity of excitation function 2n,3n,4n evaporation

22 ms

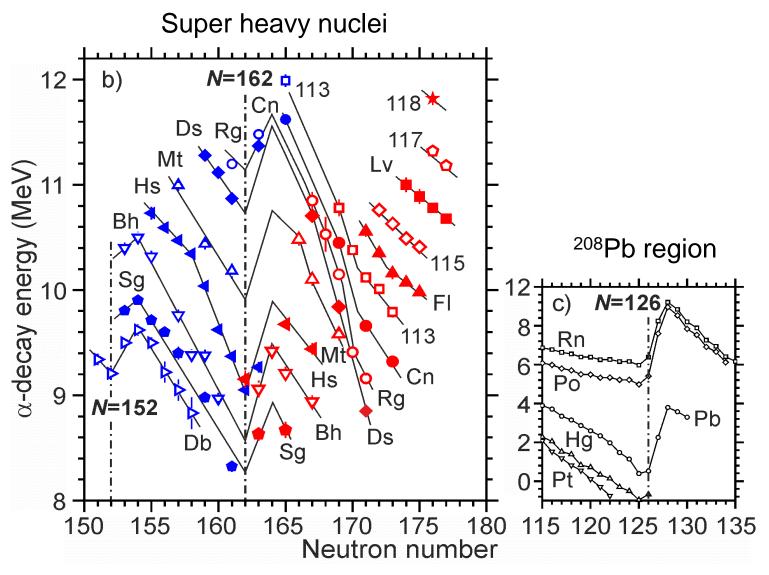
51 ms



reproducibility

and decay patterns

## Shell effects in alpha-energy pattern



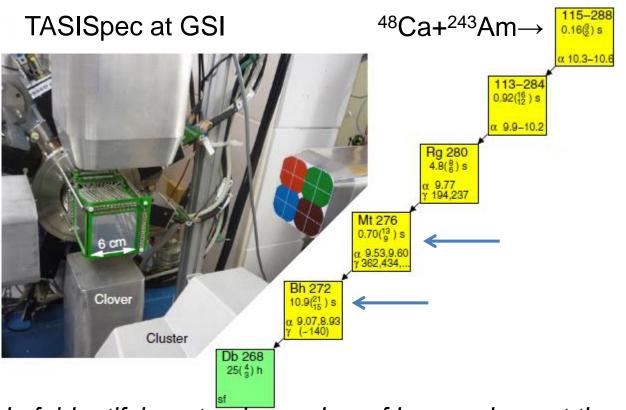




## Beyond ground-states properties of SHE

D. Rudolph et al., Phys. Rev. Lett. 111, 112502, 2013.

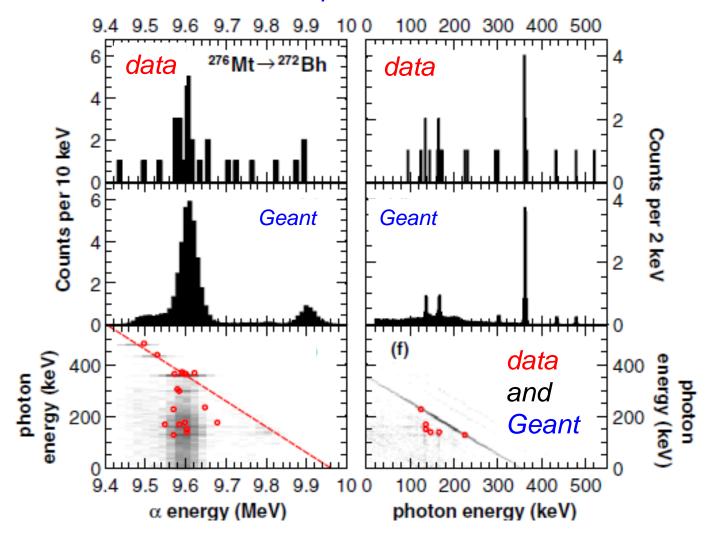
Goal: direct determination of atomic number Z for the element at the Island of Stability



Method of identifying atomic number of heavy element through alpha- Xray observation pioneered by Bemis et al. at Oak Ridge, PRL 31, 647, 1973 (!!)  $^{249}\text{Cf}+^{12}\text{C} \rightarrow ^{261}\text{Rf}^* \rightarrow 4n +^{257}\text{Rf} \rightarrow ^{253}\text{No}^* \rightarrow ^{253}\text{No} + \gamma$  and characteristic X-rays

## Beyond ground-states properties of SHE

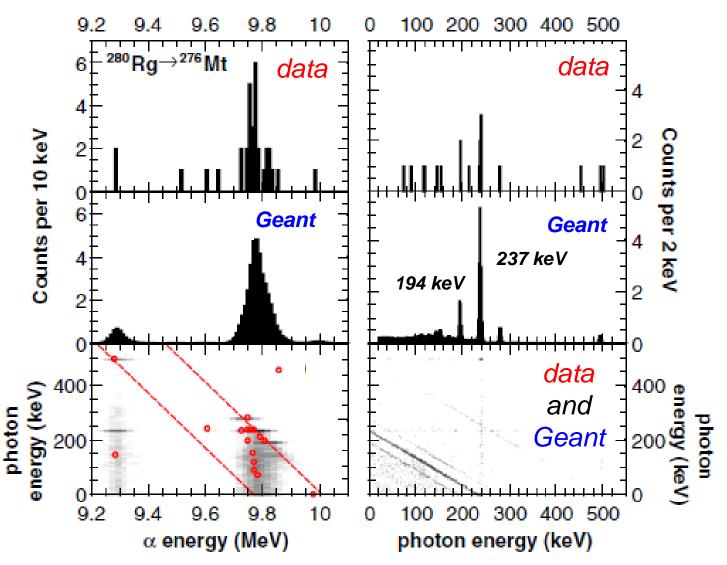
combined TASISpec and BGS+C3 data



J. Gates et al., Phys. Rev. C 92, 021301 (R), 2015
Berkeley Gas-filled Separator (BGS) + corner-cube-clover (C3)



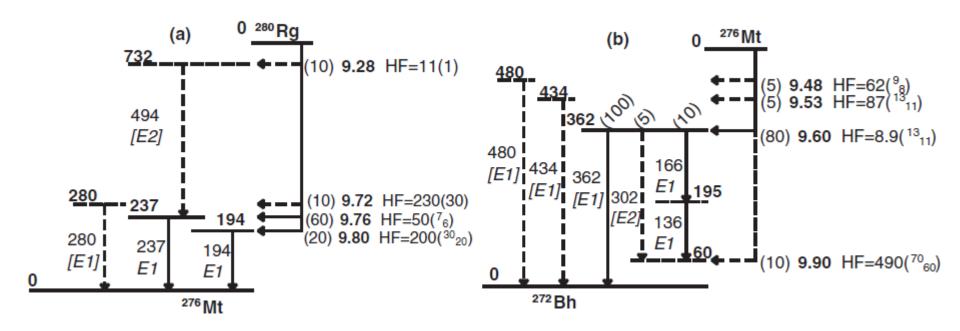
## Beyond ground-states properties of SHE



J. Gates et al., Phys. Rev. C 92, 021301 (R), 2015
Berkeley Gas-filled Separator (BGS) + corner-cube-clover (C3)
combined TASISpec and BGS+C3 results



# First level schemes of superheavy nuclei at the Island of (enhanced) Stability



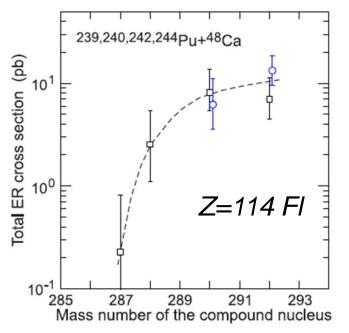
The presence of E1, parity-changing γ-transitions in <sup>276</sup>Mt and <sup>272</sup>Bh already constrains theoretical modeling, see Shi et al., Phys. Rev. C 90, 014308, 2014

Way to go ... to go for much better statistics!



## What we have learned so far about super heavies?

- 1. We reached element 118 (294Og) and neutron number 177 (294Ts and 293Lv) and about 50 other nuclei produced with 48Ca beam and radioactive actinides
- 2. The pattern of alpha decay energies and half-lives indicates that we have reached a beachhead at the Island of (enhanced) Stability. The shell effects make α-decay winning against fission, as was roughly predicted 50 years ago
- 3. For Z=114 Fl we even know where the shore of the Island dives into the sea of fission instability



The total xn cross section drops by a factor ~50 between <sup>244</sup>Pu and <sup>239</sup>Pu targets. It follows the predicted drop of fission barrier in respective Fl compound nuclei. The west shore of the Island of Stability for Z=114 was identified.

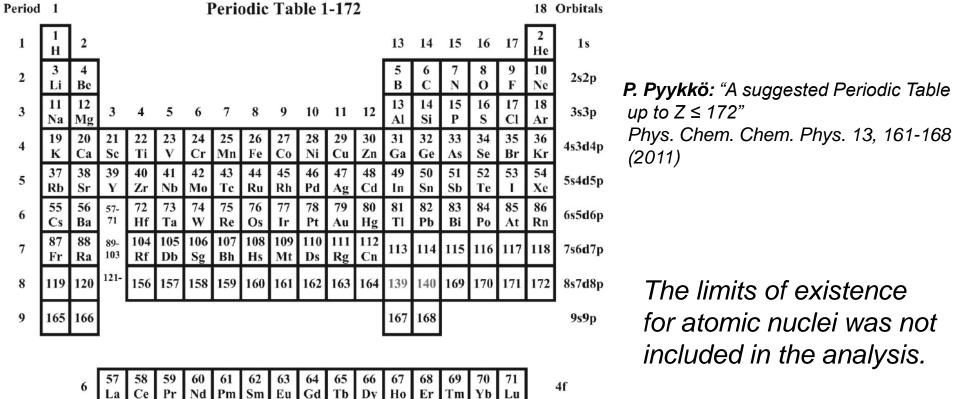
Utyonkov, Brewer et al., PR C92, 034609, 2015.



- We have identified excited states in two odd-odd nuclei at the Island, even with limited statistics we got some guidance about single particle states.
- Fission corridor exists between the Island and Mainland (hard to cross) 5.
- All nuclei at the Island were produced with 48Ca beam. We almost run out 6. of new actinide targets. The end of Nuclear Chart and Periodic Table?

5f

6f



The limits of existence for atomic nuclei was not

122 | 123 | 124 | 125 | 126 | 127 | 128 | 129 | 130 | 131 | 132 | 133 | 134 | 135 | 136 | 137 | 138

98

Cf

150

Pu

Am Cm Bk

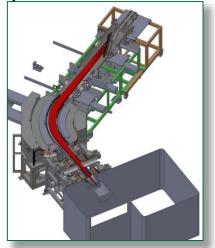
Es Fm

151

### Reestablishing isotope enrichment capability at ORNL

# Electromagnetic Isotope Separator (EMIS)

- Designed and built by ORNL (B. Eagle et al.,)
- Sponsored by DOE Office of Nuclear Physics

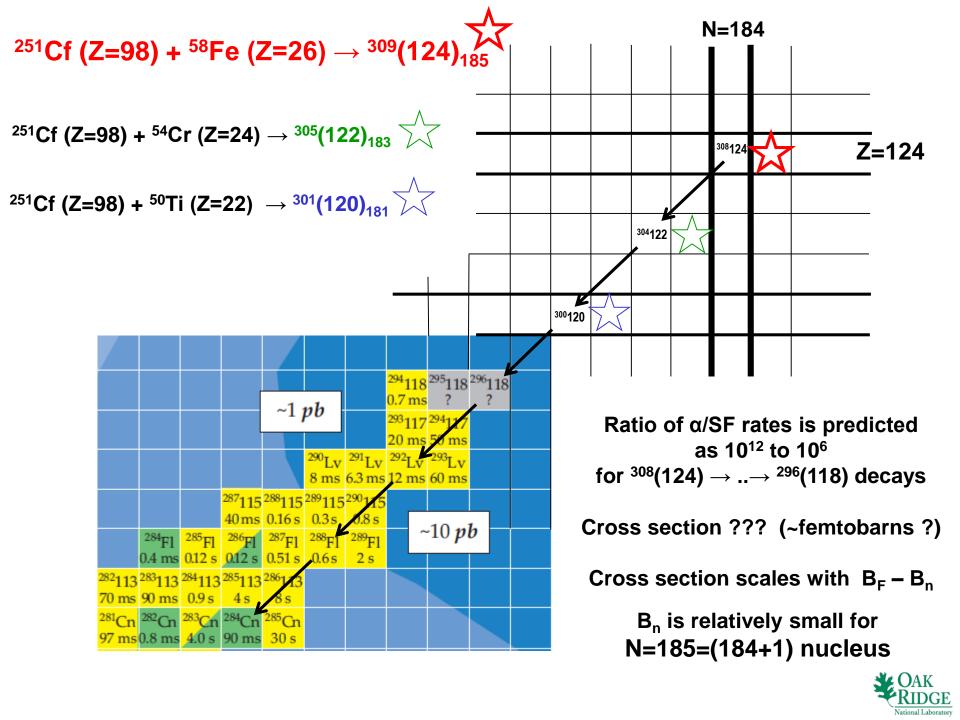




- Currently capable of producing mg to gram quantities of >98% enriched stable isotopes
- Combined with gaseous centrifuge to increase throughput
- Proposed radioactive EMIS for actinides (\$\$\$)







# Fission barriers and probabilities of spontaneous fission for elements with Z≥100

Nucl. Phys. A 944, 442, 2015, "SHE" Issue

A. Baran<sup>1</sup>, M. Kowal<sup>b</sup>, P.-G. Reinhard<sup>d</sup>, L.M. Robledo<sup>c</sup>, A. Staszczak<sup>1</sup>, M. Warda<sup>1</sup>

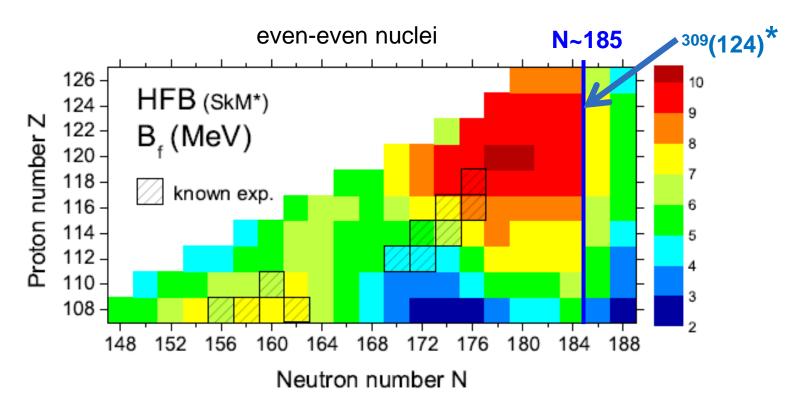


Figure 3: First fission barrier (MeV) of SH nuclei in HFB SkM\* model. Cross-hatched squares represent observed nuclei.

Neutron separation energy values are around ~ 6-7 MeV so for  $^{309}(124)^*$  B<sub>f</sub> - B<sub>n</sub> ~ 0 MeV



"Parameter study" from Krystyna Wilczyńska for a cold/hot fusion of <sup>251</sup>Cf and <sup>58</sup>Fe T. Cap, K. Wilczyńska, M. Kowal, J. Wilczyński, PR C88, 037603, 2013

Mic-Mac Fission Barriers (Kowal et al)  $\rightarrow \sigma$  (xn)  $\sim 0$ 

"a deal" on barrier height:  $2* B_f$  (Mic-Mac 3 MeV) ~  $1/2* B_f$  (HFB 12 MeV) ~ 6 MeV

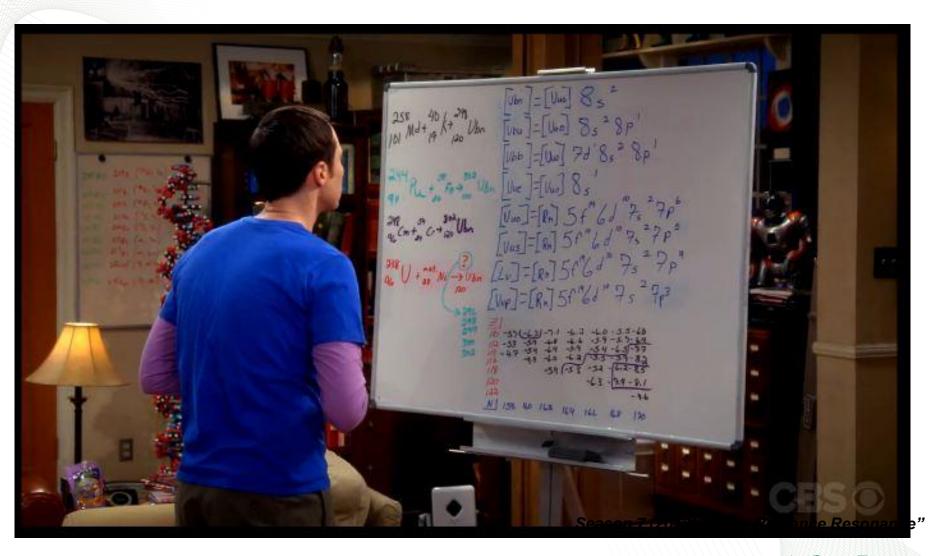
$$B_f \sim 6 \text{ MeV} \rightarrow \sigma (xn) \sim 20 - 40 \text{ picobarn (!!!!)}$$
 (maximum for 2n, 3n channels)

Too good to be true, but we expect measurable cross with fission barrier parameter slightly below 5 MeV

Such study should be doable at  $\sim 10^{20}$  -  $10^{21}$  beam dose and worth to try providing  $^{251}$ Cf target is available from ORNL



# One episode of The Big Bang Theory had Sheldon theoretically discovering a stable SHE



# **Decays of 294117!**



The Big Bang Theory, Season 9 Episode 15: The Valentino Submergence OAK RIDGE National Laboratory

From Mark Stoyer

# Summary – what comes next?

- HFIR/REDC complex at ORNL shall operate for next 25-40 years. New developments in accelerator technology should be complemented by target materials
- New detection techniques including digital signal processing should allow us to reach shorter-lived, heavier nuclei and get data beyond ground-state properties.
- There is no clear theoretical guidance where the Chart of Nuclei and Periodic Table end. Fission barrier and reaction mechanism analysis are critical.
- Experiments with intense beams heavier than <sup>48</sup>Ca, and with <sup>251</sup>Cf target (<sup>248</sup>Cm, <sup>249</sup>Bk, <sup>249</sup>Cf) from ORNL, may help to reach the vicinity of predicted N=184 shell closure and create new elements up to Z=124.

The studies of super heavy elements and nuclei will continue at new or upgraded laboratories with the GFRS/SHELS/Masha at Dubna, GARIS 2 at RIKEN, BGS+C3/FIONA at LBNL, AGFA at ANL, at S3 at GANIL/SPIRAL2 and TASCA/SHIP at new LINAC at GSI.

