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# Isotope Production R&D and Re-establishing Domestic Stable Isotope Production

**Isotope Development for Production and Research DOE Office of Nuclear Physics** 

September 19, 2013



### Nuclear Sciences Advisory Committee on Isotopes

- Charge 2: Develop a long range plan outlining the Nation's future isotope needs
- Recommendations
  - 1. Devise process for enhanced communication and outreach
  - 2. Coordinate production capabilities and research among existing facilities
  - 3. Support research program in base budget to enhance IP capabilities
  - 4. Encourage isotope use in research with reliable supply and affordability
  - 5. Increase robustness and agility of transportation of isotopes
  - 6. Invest in workforce development
  - 7. Construct and operate isotope separator facility
  - 8. Construct and operate a dedicated multiparticle accelerator production facility

#### FINAL REPORT

Second of Two 2008 NSAC Charges on the Isotope Development and Production for Research and Applications Program

# Isotopes for the Nation's Future Along range plan

NSAC Isotopes Subcommittee



#### Nuclear Sciences Advisory Committee on Isotopes

- Charge 1: Prioritize near term compelling opportunities for Isotope Research
- Recommendations
  - 1. New approaches for production of therapeutic alpha-emitting isotopes
  - 2. Coordinate production capabilities and research among existing facilities
  - 3. Create plan and make investments in isotope production to meet needs in heavy element research
  - 4. R&D to address new or increased production of He-3
  - 5. R&D to re-establish a domestic source of mass separated stable and radioactive research isotopes
  - 6. Robust investment into education and training in isotope production

FINAL REPORT One of Two 2008 NSAC Charges on the National Isotopes Production and Application Program

Compelling

pportunities

Research

**NSAC Isotopes Subcommittee** 

using lsotopes



#### Solicitations: \$28.3M total Invested since 2009

- FOA 09-14 (R&D on Alternative Isotope Production Techniques)
  - FY 2009 FY 2010 56 Program Funds/ARRA: ~\$16.4M
    - 56 proposals, \$50.4M Requested
    - Awards: 13 Laboratory, 9 University, 1 Industrial
- FOA 11-448 (Research, Development, and Training in Isotope Production)
  - FY 2011 FY 2012 Program Funds: \$5.8M
    - 35 Proposals, \$40.5M Requested
    - Awards: 3 Laboratory, 4 University,1 Industrial
- FOA 13-743 (Research, Development, and Training in Isotope Production)
  - FY 2013 FY 2014 Program Funds: ~\$6.1M
    - 46 Proposals, \$42 M Requested
    - Peer review completed February 21-22, 2013
    - Awards: 7 Laboratory, 2 University
- Core R&D Support: ~\$1.5M annually

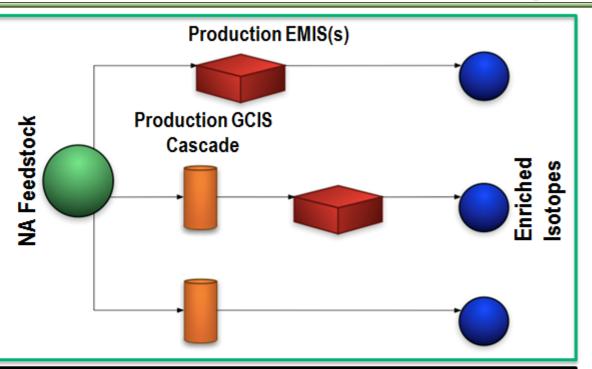


- Therapeutic alpha emitters (At-211, Ac-225, Th-229, Ac-227/Ra-223)
- New radioisotope extraction/separations technologies
- Accelerator and reactor isotope production targetry
- Isotopes for positron emission tomography (Se-72/As-72, Cu-62, Cu-64, Y-86, Zr-89)
- Heavy elements (Cf-252, Bk-249, Am-243, Cf-251, Optimization of the use of Cm feed-stocks in heavy element production)
- Nuclear Forensics, Environmental Research (U-233/Th-229, Si-32, Np-236/Pu-236)
- Therapeutic beta-emitters (Cu-67, As-77, Re-186, Rh-105, Pr-143)
- Workforce development (Most grants and core R&D)
- Stable isotope enrichment (EMIS/ESIPF, Puerto Rico Project, Li-7)
- Isotope harvesting at rare ion beam facility

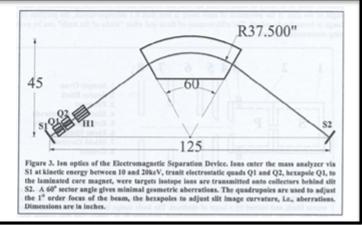


#### ORNL Isotope Enrichment Project

ORNL's Concept Enriched isotopes can be produced in 10's of g quantities by Electromagnetic Isotope Separators (EMIS), Gas Centrifuge Isotope Separators (GCIS) and using a combined method where GCIS is used to pre-enrich feedstock for final enrichment on EMIS



ORNL submitted proposal "Integration of Centrifuge and Electromagnetic Separation for the Preparation of Stable Isotopes", in response to SC Program Announcement LAB 09-14; May 2009





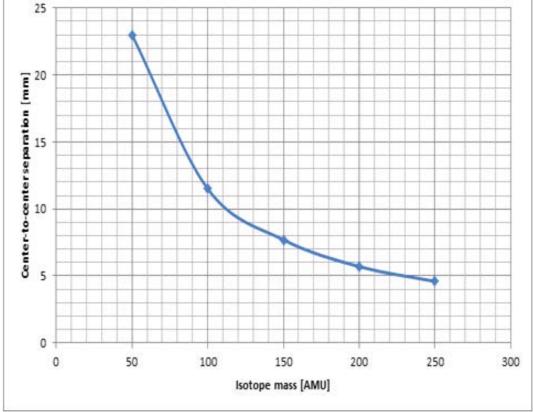
Design time and fabrication risk reduced through use of advanced 3D computer aided design and simulations tools

#### First enriched samples collected Feb 2012 Greater than 98% enriched molybdenum and nickel



10 mA construction completed December 2011 100 mA upgrade scheduled to be complete FY15



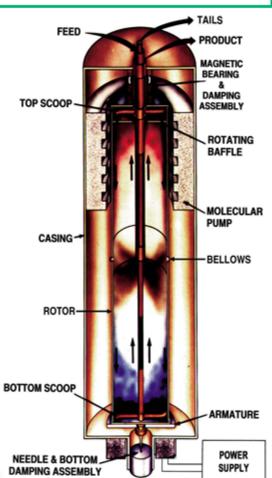


- Magnet designed for 20 amu < A < 208 amu</li>
- Optimized for a A = 100 amu as mid-point
- Magnet capable of bending the path for A up to 450 amu
- Slight amount of distortion in the magnetic field for masses above 250
- Can make adjustments in the flight path to increase the separation for heavier isotopes



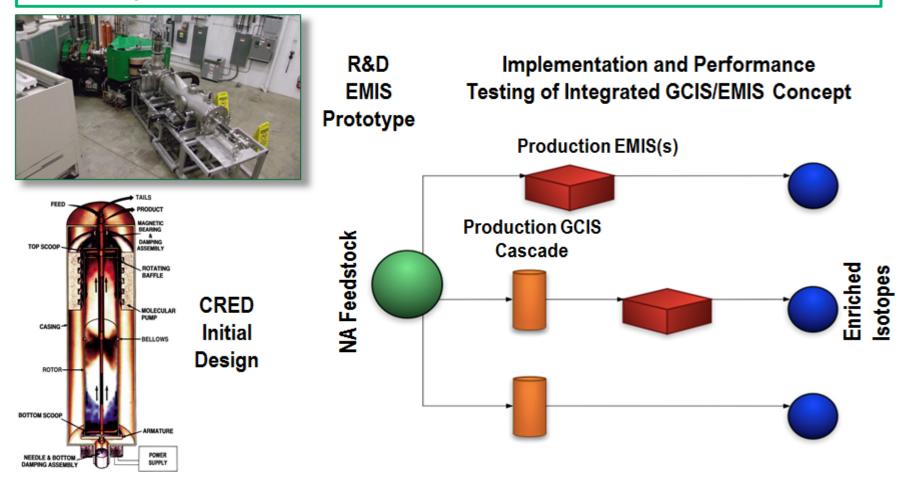
The goal of this study was to examine a number of different isotope systems and centrifuge designs to determine the feasibility of using a GCIS cascade to improve stable isotope production rates using EMIS.

- Studied various scenarios
  - Four target gases: MoF<sub>6</sub>, WF<sub>6</sub>, GeF<sub>4</sub>, and Ni(PF<sub>3</sub>)<sub>4</sub>
  - Single machines in series
  - Cascades of different designs (number of stages/machines)
- All centrifuges were capable of separating all of these gases
  - Smaller machines better for lower mass flows and product requirements, more flexible for range of isotopes
  - Must consider more units in a cascade or passes through a single machine
  - Machine geometry must be adjusted to make some of the machines effective for a specific gas



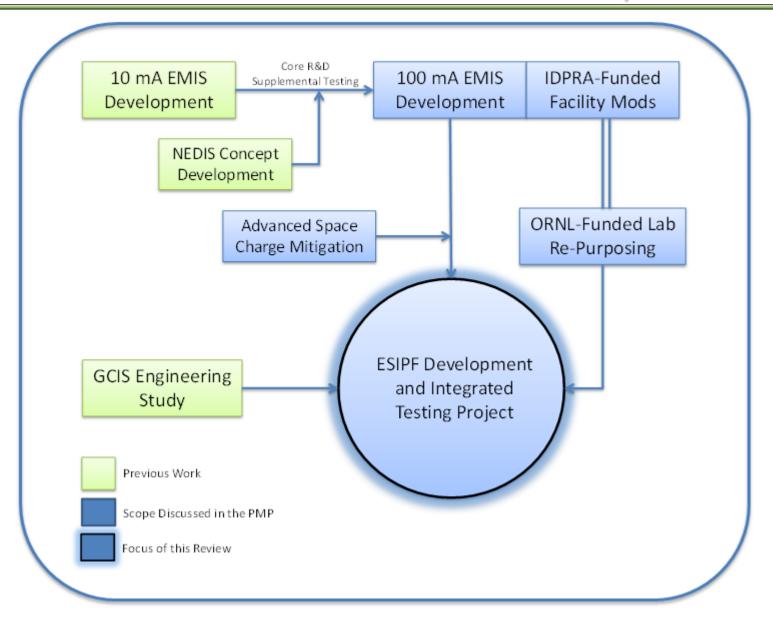


The goal of this project is to complete final reliability and automation tasks to achieve a modern, production-class electromagnetic separator and a 9-unit gas centrifuge cascade (with a 2-unit test stand), capable of producing milligram to tens of grams of enriched stable isotopes.



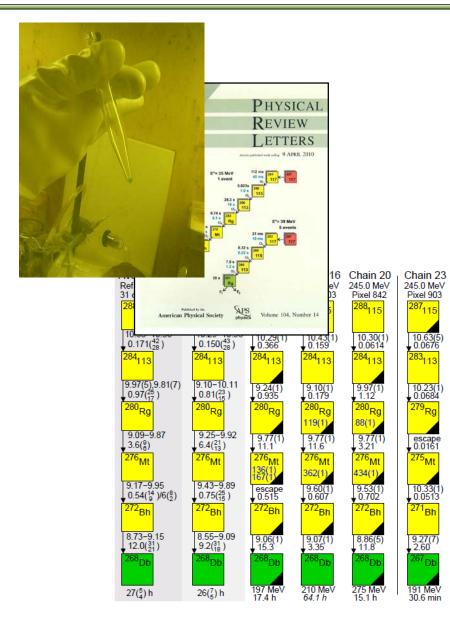


#### ESIPF Technology Development Project





Isotopes for Super Heavy Element Research



## Production of Cf-252

- Optimization of irradiation of curium targets at HFIR
- Cf-252 used primarily by industry (neutron sources for oil exploration, R&D)

## Bk-249, Am-243, Cf-251

- "Hot fusion" to produce isotopes of heavy elements 117, 115
- New Isotopes of element 118
- SHE factory in Dubna



#### Accelerator Production of Ac-225/Bi-213

U 227	U 228	U2		U 2		U 23		U 23		U 233		U 234			
1,1 m	9,1 m 4 6.68; 6.59	58 4: 4 6.36 6:339:6	297	20,8 + 5,888	5.818 4:	4,2 ( 6. n. 5.458 5.471: 5.4		68,9 4 x 5,320; 5,2 Ne 24;	82	1,592 · 10 n 4,824; 4,78 Ne 25;	3 2	0,005	0'a		
6.74 y247:310;e <sup></sup>	γ (246: 187) σ	123: 8 100 a	B;	230h:e n 25	r-	9 20: 84 1 9 . m - 25	02	y (58; 129 ar 73) m 74		v (42: 97); a: 47; n; 530	• H	9 88, 194, 19 193 1 # 196, 19 # 1800	111.		
Pa 226 1,8 m	Pa 227 38,3 m	Pa 228 22 h		Pa 229		Pa 23		Pa 23	04.0	Pa 23		Pa 23	3		
u 6.88; 6.82 ¢	46,466; 6,416 ¥65; 110	c: x 6.07 5.799: 6 y911: 46 965		c; u 5:08 5.0 0: 5.0 119:40		6:8-0.5 = 5.345 5.3 = 951-944	100	+ 5.012, 4.952 3.034 (2.1-300, 903 + 200, m + 0.0	-	y 969, m			< 0,1		
Th 225 8,72	Th 226	18.7	d	Th 2		Th 21 7880		7.54 10		n 23 25,5 h		Th 23	2		
n 6,48255,445 6,504 - 17 7,321,248 359:306	u 6,336; 6,230 y111: (242: 131) g~	a 6.00 5.75 	0:256	a 6,423; 1 964; (21) O 20	6.340 5 × 0.3	104 201 26 11.110 11.110 11.110 11.110	4,815	= 4,687 γ (68) N = 23 < 0.0005	1.6	β <sup></sup> 0.3: 0.4. γ 26; 84 θ <sup></sup>		405 10 4013 3.950 64			
Ac 224 2.9 h	Ag 225	Ac	h	Ac 2		Ac 2		Ac 22 62.7		Ac 23		Ac 23 7.5 m			
α 6.142 6.060; 6.214 γ 216; 132	= 5.830 5732 y 100 (1	67 0.9; c; a 5.3 y 230, 1 254; 160	1.1	β <sup>-</sup> 0.04. α 4.953. γ (100.0 α 650.01	1	P 1.2.2.	1	11 <sup>-1,1</sup> y 105; 569 262; 146; 1		0 <sup>-2.7</sup> 3 455:508. 1244 6 <sup>-</sup>	P				
Ra 223	Ra 224 3.66 d	Ra 4,	22	Ra 2	226	Ra 2 42.2		Ra 2: 5.75		Ra 22 4.0 m		Ra 23		233	U 234
u 8,7162: 5,6067 v 269: 154: 324	× 5.6854. 5.4486	8-0,3:0	4	o 4,7843; > 186; 0	4,601			β <sup>+</sup> 0.04 γ (14: 16			1	0.8 72:63:20		2 · 105 a 4, 4,783	2,455 . 10
C 14. a 130. a 0.7	¥241 C 14 # 12.0	¥ 40		ci = 13 ci < 0.00	105	β <sup>-1.3</sup> . 7 27: 300	303	e" # 361 m < 1	2	β=1.0 1	4	70		97	ia: 4,775; 4,725;; sl Mg 28; Ne; 5 (23; T WT: x 96; xj 4,8005
		226 3 m	Pa 2 38,3			228 2 h		a 229		a 230		231		a 232	Pa 233
	o 8.86	6.82	a 6,466; 6,416		5.799!	078; 8,105; 6,118, 463; 669;	c: o 1 5.670	5,60 1: 5,8 9: 40; 14	0.3.3	0.5/. 15: 0.326 1919: 455:	+ 5.014 5.028	4.952 No 24, F-23 0:303 _ 141	β <sup>-0</sup> γ96	(3, 1,3,)» 9: 894:	130 A
	9	005	y 65: 11		965	-			U da l	M - m 1500 -	m 200; <i>m</i>	+ = 0.000	100	σ; σι 700	o 20+1 m -
	8,7	225 2 m	Th 2 31	m	18	227 ,72 d	1.	h 228 913 a	2	229	7,54	- 10 <sup>4</sup> a		h 231 25,5 h	Th 132 100
	0.6.483 6.504 7.321 369 30	: 6,445; ; 4 246; (8	4 6,336) 9111: (2 131) 9 <sup></sup>	6,230 4Z:	5.757.	8: 5,978; 50: 256 200	VB4: 0 20	23: 5.340 (216): e : m < 0.3	1434 1104 21	211;00;	¥ 068:	7: 4,621 144 ): e : # 23,4 0005	β~0 725 8	3: 0.4	1,405°101 4013 3.950 964 30 9737 90,000
		224 9 h	Ac 1			226 9 h		c 227		c 228 3.13 h		229 2.7 m	A	122 s	Ac 231 7.5 m
	e 0.6.14	de la	= 5.830 5732	5,793;	870.9 cta 5	1.1.1	B-0		β-1 u.43	2:2.1	871.	1	10-1 3-4	7	6-
	6.060. γ 216;	6.214	y 100. (1 100- 63	50:		158	7(10	0; 84). e <sup>-</sup> l; ay < 0,029	7.91	1,969	y 185	569. 46; 135	124		7 282, 307, 221, 188, 38
		223 43 d	Ra			1 225 4,8 d		a 226 600 a		a 227 2.2 m		228 75 a		ta 229	Ra 230 93 m
	a 5.716	5,5067	» 5.6854 5.4436		87 0,3		0 4,7	843: 4,601 ; C 14			ST 0.				β <sup>+</sup> 0.8 γ 72: 63: 20: 470
	G 14, 0 010,7	130	y 241 # 12.0	0.14	y 40		a = 1 er <	3 0.00005	15-1 7-27	.3 . 300: 303	e # 36;	m < 2	871	.0	470 e





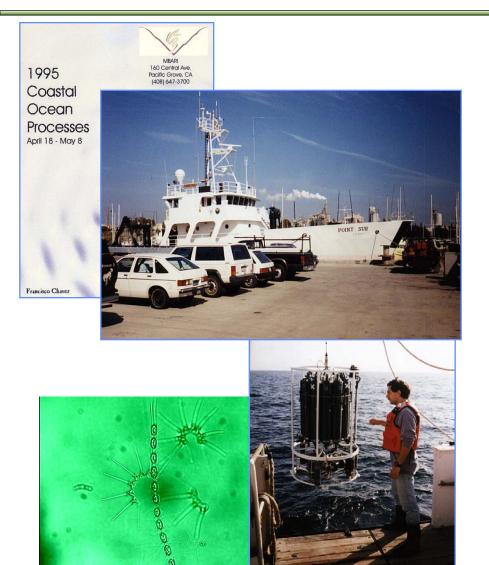
#### Protons on Thorium Targets

- High energy reactions
  - <sup>232</sup>Th(p,x)<sup>225</sup>Ac
  - $^{232}$ Th(p,x) $^{225}$ Th  $\rightarrow ^{225}$ Ac
  - $^{232}$ Th(p,4n) $^{229}$ Pa $\rightarrow ^{225}$ Ac
  - $^{232}$ Th(p,x) $^{225}$ Ra  $\rightarrow ^{225}$ Ac
- Low energy reactions
  - $^{230}$ Th(p,2n) $^{229}$ Pa $\rightarrow ^{229}$ Th
  - $^{232}$ Th(p,4n) $^{229}$ Pa  $\rightarrow ^{229Th}$
  - <sup>232</sup>Th(p,p3n)<sup>229Th</sup>

#### Processing

- Scale up ORNL technology
- Develop new for scale up





- Si-32 is a radioanalytical tracer to measure bloom rates of diatoms
- Rate of bloom of diatoms is a key parameter in carbon cycle
- Can only be effectively produced by spallation reactions on KCI targets
  - KCI(p,x)<sup>32</sup>Si
  - Long irradiation
  - Complicated chemistry



- Recovery of high purity U-233 and Th-229
  - Collaborative effort with NNSA Office of Non-Proliferation and International Security
  - Recovered, separated Th-229, analyzed, re-packaged ~100 g of 99.9875% pure U-233; provided ~20 g of the U-233 to New Brunswick Lab for CRM
- R&D project to investigate feasibility of accelerator production of Np-236g and Pu-236 for IDMS applications in Nuclear Forensics
  - -<sup>238</sup>U(p,3n)  $\rightarrow$ <sup>236m</sup>Np  $\rightarrow$ <sup>236</sup>Pu
  - <sup>235</sup>U(d,n)  $\rightarrow$  <sup>236g</sup>Np
- R&D project to evaluate feasibility of a new process to enrich Li-7
  - Solvent extraction technology
  - Goal to demonstrate enrichment to greater than 99.9%



#### Discussion



Reaction	Advantages	Disadvantages					
<sup>238</sup> U(p,3n) <sup>236m,g</sup> Np LANL-IPF: 1μA of 30 MeV protons	<ul> <li>Target material readily available</li> <li>Larger amounts of <sup>236m</sup>Np produced per irradiation:</li> <li>Estimated 1 mCi of <sup>236m</sup>Np for 1h of proton beam <i>or</i></li> <li>0.4 μCi of <sup>236</sup>Pu after decay</li> </ul>	<ul> <li><sup>237</sup>Np impurity in product would require isotope separation to purify <sup>236g</sup>Np</li> <li><sup>238</sup>Np from (p,n) decays to <sup>238</sup>Pu, potentially contaminating grown-in <sup>236</sup>Pu.</li> </ul>					
<ul> <li><sup>235</sup>U(d,n)<sup>236m,g</sup>Np</li> <li>University of</li> <li>Washington: 1µA of</li> <li>24 MeV deuterons</li> </ul>	<ul> <li>Anticipated higher radioisotopic purity of <sup>236g</sup>Np (no production of <sup>237</sup>Np)</li> <li>0.2 ng of <sup>236g</sup>Np for 1h of deuteron beam</li> </ul>	<ul> <li>Lower total cross section for deuteron-induced reactions compared to proton</li> <li>Available deuteron beam currents are a factor of 5 smaller than available proton currents, reducing yields significantly</li> </ul>					