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Mass Separation of Radioisotopes

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U.S. DEPARTMENT
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New mass-separation technology is needed to expand the impact of accelerator and reactor produced radioisotopes

Challenges Limiting Current Radioisotope Use

- Co-production of chemically inseparable contaminants prevents use of many promising isotopes
- Short half-lives high radioactivity, extreme material value, and extremely small quantities create a unique challenge for mass separation

Mass Separation Requirements

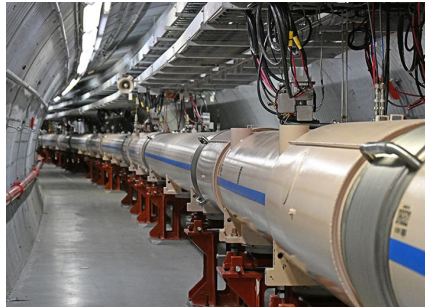
- Beyond state-of-the-art field deployable hot cell-based mass separation capability needed with:
 - 90%+ Collection Efficiency
 - <100 μg (~ 10 Ci) scale with liquid or dried film introduction
 - <1 day process time
 - serviceable in a hot cell

No current mass separation technology meets these requirements

Isotope enrichment has a long history in Oak Ridge; recent major DOE-IRP investments have re-established the capability



Shutdown of calutrons and loss of U.S stable enrichment capability



Routine enrichment of ^{176}Yb for commercial production of cancer-fighting radio-pharmaceutical

1945

1998

2010

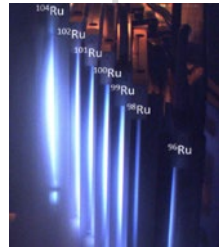
2018

2021

2025



1st enrichment of ^{63}Cu & ^{65}Cu by the Manhattan Project Calutrons



DOE-Isotope R&D and Production Program (IRP) reestablish stable enrichment technology – prototype EMIS device

500 mg of Ru-96 enriched for a quark-gluon plasma physics experiment at BNL's Relativistic Heavy Ion Collider



\$300M investment and full-scale deployment of stable isotope enrichment to reestablish U.S stable isotope program with SIPRC

Basics of plasma-based isotopic enrichment

Atomize

Introduce atoms of the element into a vacuum

Typical techniques: Direct evaporation of solid feed, ion sputter, gaseous feed with electron dissociation, in situ formation of volatile compound

Ionize

Create an atomic ion with +1 charge

Dominant mechanisms are electron impact ionization and possibly penning ionization

Separate

Separate by mass using Lorentz force

Ions of different mass and/or energy have a different bending radius in a uniform magnetic field

$$\vec{F} = m\vec{a} = q(\vec{E} + \vec{v} \times \vec{B})$$

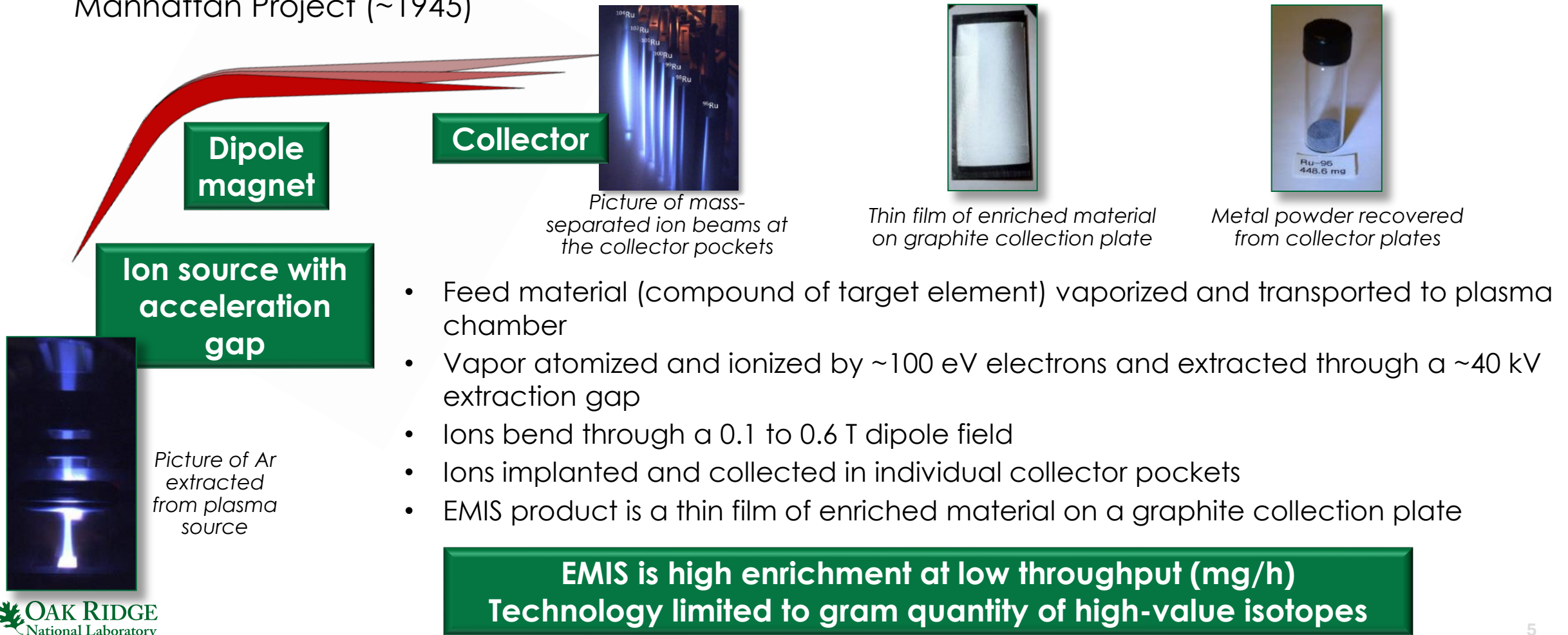
Collect

Selectively collect the separated ions with minimal cross contamination

Typically, ions are deposited as thin metal coatings on graphite collection plates

Basics of electromagnetic separation (EMIS)

- Ions of different masses with equal energy and charge bend at different radii in a uniform magnetic field
- First modern mass spectrometer developed by A. J. Dempster in 1918; implemented on a massive scale by E. O. Lawrence during the Manhattan Project (~1945)



Ion source efficiency and the properties of the feed material are the major limiting factors of EMIS enrichment

Starting abundance of target isotope of feed material (0.1%, 1%, or 10%)

- The primary driver of final enrichment is the starting abundance – not the resolution of the mass spec
- EMIS throughput is limited by the total ion current of all ion species exiting the plasma source. This includes molecular ions species and other elements. Present technology is limited to approximately 50 mA

Ionization Efficiency is the biggest limitation of EMIS to radioactive materials

- Typical only 5 to 10% of feed material is vaporized, ionized, transported, and collected
- Yb is exceptional at 15% due to an ideal vapor pressure and low ionization potential.
- At 10% efficiency – 90% of the feed material becomes hold up in the machine or exhausted – both are major issues for radioactive operations.
- Heat and halogenated gases are used in situ to increase the mobility of the targeted element and improve ion source performance and efficiency
- It is typical to form thin metal films, which can have pyrophoric properties and additional safety considerations

EMIS is a mature and highly flexible technology with high first pass enrichment in the milligram to 100 g scale

¹⁷⁶Yb case study (actual)

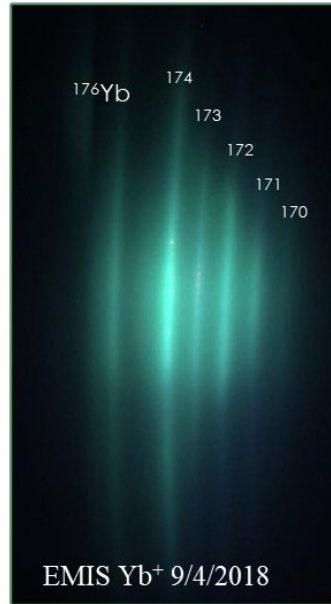
Routine enrichment of ¹⁷⁶Yb and all other Yb isotopes since 2021.

¹⁷⁶Yb: 99.8% enrichment from 12.9% natural abundance

¹⁶⁸Yb: ~65% enrichment from 0.13%

~15% Ionization Efficiency

(mass of ^{Nat}Yb introduced vs collected mass)



Example of ²⁵¹Cf enrichment

Goal of study: enrich 100 mg of ²⁵¹Cf in 1-year using available feed material that has a starting abundance of 16% ²⁵¹Cf

Technology: EMIS style device with less 0.1 mA of current to the collectors

Ionization Efficiency	Estimated working inventory in facility	Facility type
1%	4,500 mg	Haz Cat 3
10%	450 mg	Haz Cat 3
20%	225 mg	Radiological

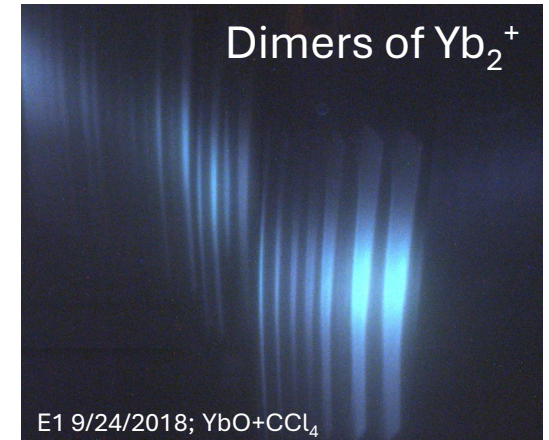
Mass Scale

1,000 g		^{nat} Yb processed by EMIS
150 g	15%	Total of all Yb isotopes collected
19 g	12.7%	¹⁷⁶ Yb collected and recovered

A new, beyond state-of-the-art, ion source and mass separator is needed for high-specific activity applications

**Order of Magnitude Requirements: 90% collection efficiency,
1 day processing of 10 Ci (<100 µg) in a hot cell**

- Isotopic separation requires atomic ion species.
 - Molecular ions, dimers, etc cause mass overlaps.
 - “soft-ionization” methods such as electrospray and liquid based techniques have limited use
- The Argon Torch of typical ICP-MS approaches 100% ionization efficiency; however, the transition from the high-pressure Argon Torch to the mass spec’s vacuum drops the overall collection efficiency to ~1%.
- Thermal surface ionization sources have reported up to 70% efficiency.
 - A typical TIMS analytical unit are ~50%
 - Works only for elements with low first ionization potential such as the Ln and Ac series.
- The current EMIS and the previous calutron ion sources typically achieved between 1% and 20% total efficiency.



**Low efficiency is a concern for both the loss of valuable material
and rad-holdup in the machine, waste and emissions concerns.**

100 μg versus 100+ g scale requires very different equipment

A “rad-EMIS” is usefully at the greater than 100 mg scale and requires glove box-based operations and maintenance– full hot cell maintenance would be extremely challenging

Commercial multi-collector analytical equipment could be used at the 100 μg scale with a new ion source

This mission space likely requires custom developed ion source and paired separation technologies.



New mass-separation technology is needed to expand the impact of accelerator and reactor produced radioisotopes

Mass separation technology needs to have: 90%+ collection efficiency; ~100 μg (~10 Ci) scale with liquid or dried film introduction; 1 day process time; serviceable in a hot cell

Current ion source technology is the primary limiting factor