# Refractory Pressed Powders as Porous Solid Catchers of Stopped Rare Isotopes
# Porous Thin Film Targets for Medical Isotopes

**Nuclear Physics SBIR/STTR Exchange Meeting**
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**Sponsor**: Office of Nuclear Physics, DOE
**Program Officer:** Dr. Manouchehr Farkhondeh
**Phase II Grant Number:** DE-SC0011346
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**Small Business**
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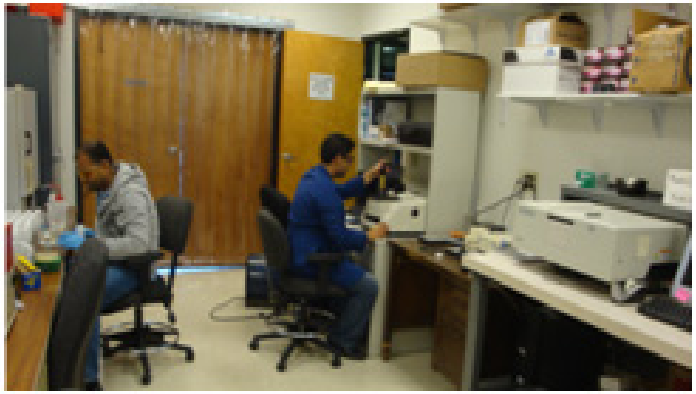
# Presentation Overview

- About InnoSense LLC
- Commercialization Status
- Motivation
- Relevance to Nuclear Physics Programs
- Summary
- Acknowledgments

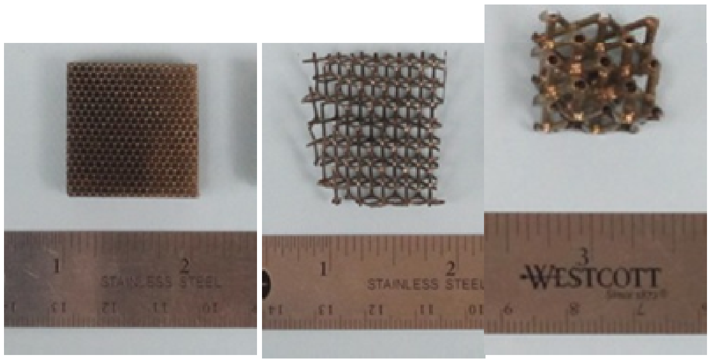
# About InnoSense LLC

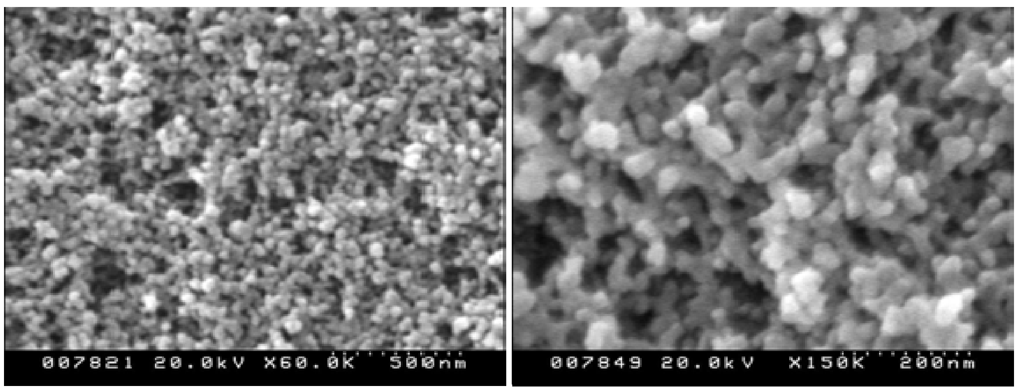
- Established in 2002 by private investment, R&D operations in 2004, housed in a recently expanded 9,000 square feet laboratory facility located in Torrance, California.
- Key laboratories include five “wet” chemical facilities equipped with fume hoods, a clean room, a spectroscopy facility, optics and testing laboratory, and two machine shops.
- 22 employees, including 5 PhD, 3 MS and 2 MBA degree holders.

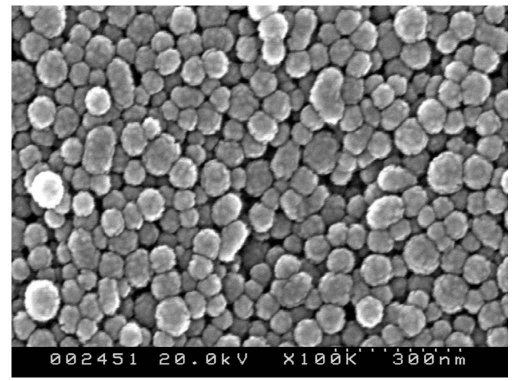

# Commercialization – Building from ONP Funding

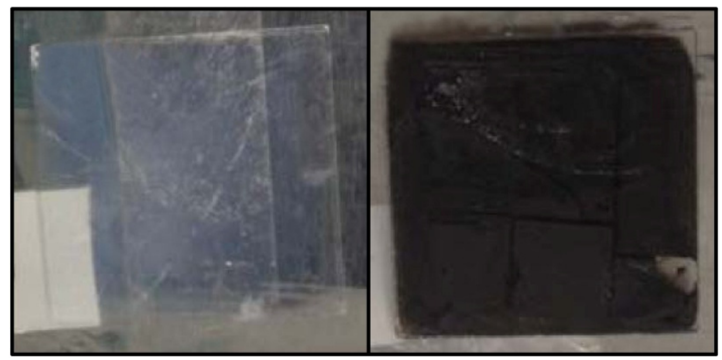
**PO for silica aerogel coatings on metal lattices – Invoiced July 2015**




**Porous Scaffolds for Refractory Solar Selective Coatings – SuNLaMP $**



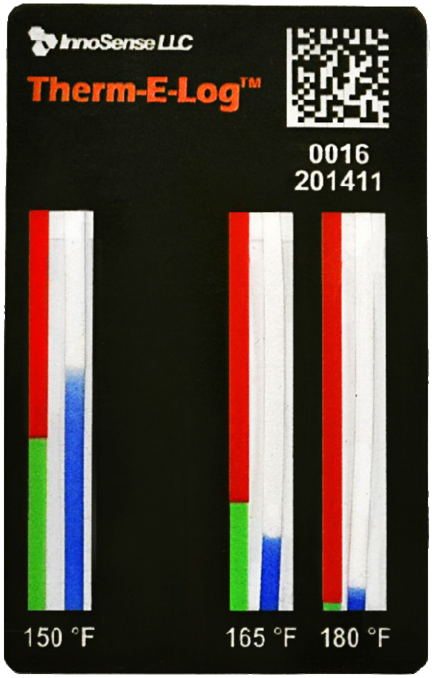



**Prior DOE ONP funding enabled us to develop the technology for porous monoliths and expand the application base for these materials**

# Commercialization Status

**Army: W15QKN-09-C-0153**

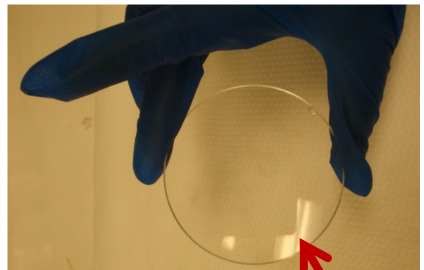
**Passive Temperature Dosimeter**




- **Phase III Funding**
- **Correlation Testing planned at Yuma Proving grounds October 2017**
- **Production anticipated in 2018 for 120 mm ammunition**

**Army: W911NF-11-C-0056**

**Permanent anti-fog coating**



PC lens and PU visor

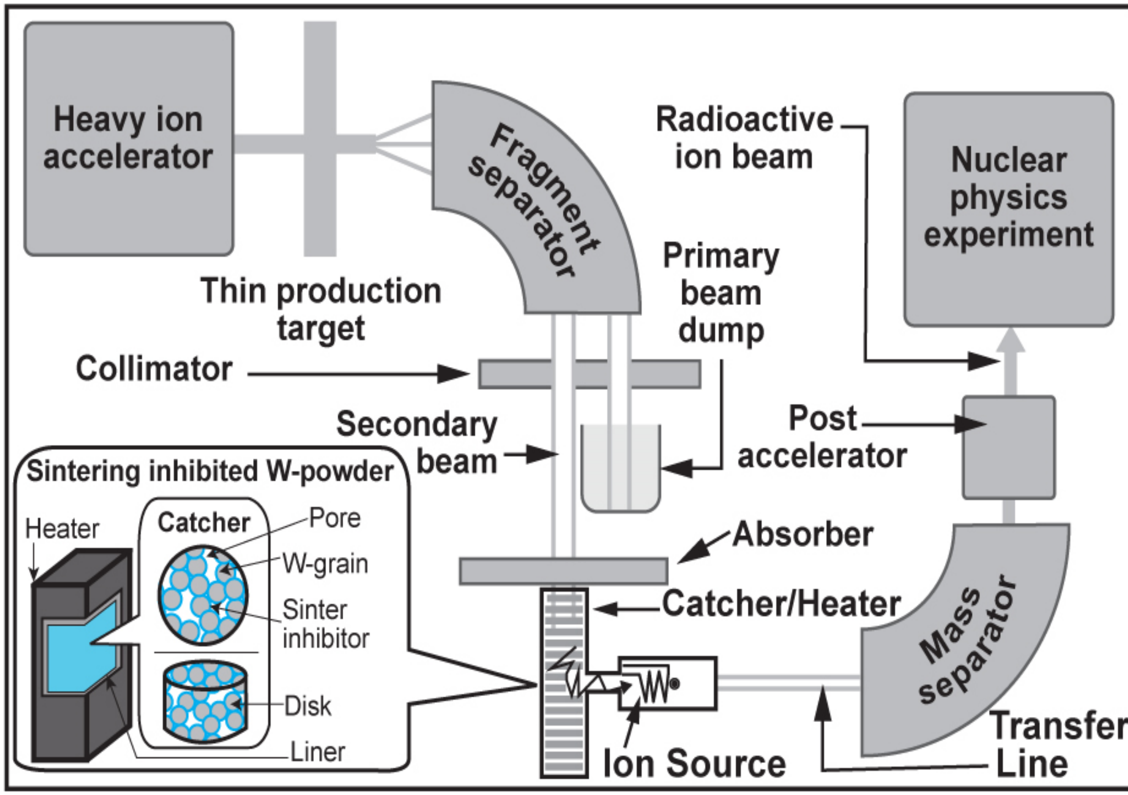
- **DOD DTRA RIF award – 2015**
- **Nanomaterials in coating**
- **Potential to contract coating application for DTRA and other commercial markets**

**MDA: HQ0147-14-C-7012**

**Hypergolic Leak Detector for THAAD**

- **Ongoing Second Phase II**
- **Drop-in Replacement Leak Detector for MDA THAAD missiles**
- **Production Prototype Order in 2017**
- **Targeting biomedical diagnostics**

# Refractory Hot Catchers for Rare Isotopes (no primary beam power)




**Features**

- Sintering inhibited W-powders
  - Maintains porosity at high temperature for long time
- Stable in energetic radioactive beams
- Supplementary to the gas catcher

**Advantages**

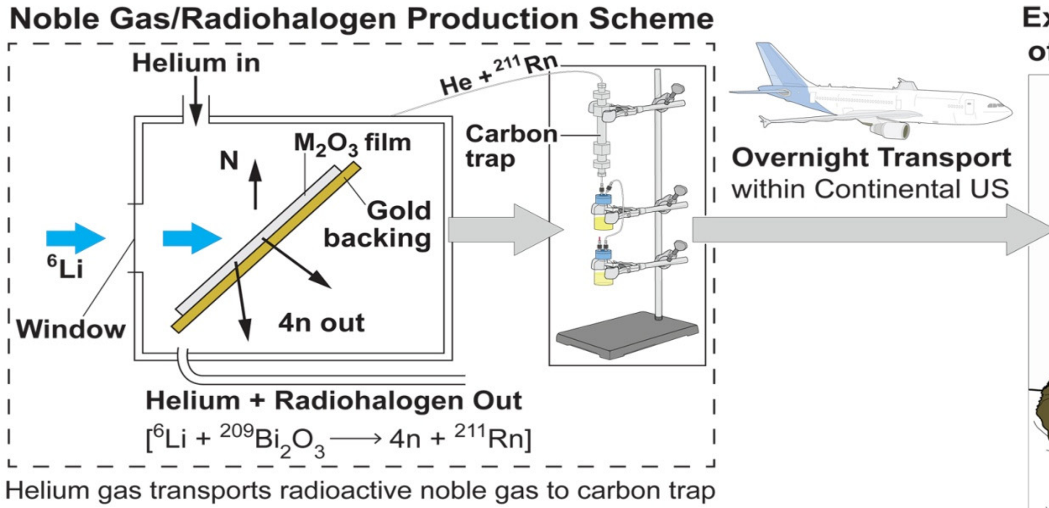
- Higher stopping power (~100 to 1000 times) over gas catcher
- Allows diffusion/effusion of short-lived isotopes as atoms or molecules
- Versatile and cost-effective processing
- Does not have the intensity limits of gas catchers for rare isotope beams

**Collaborator**

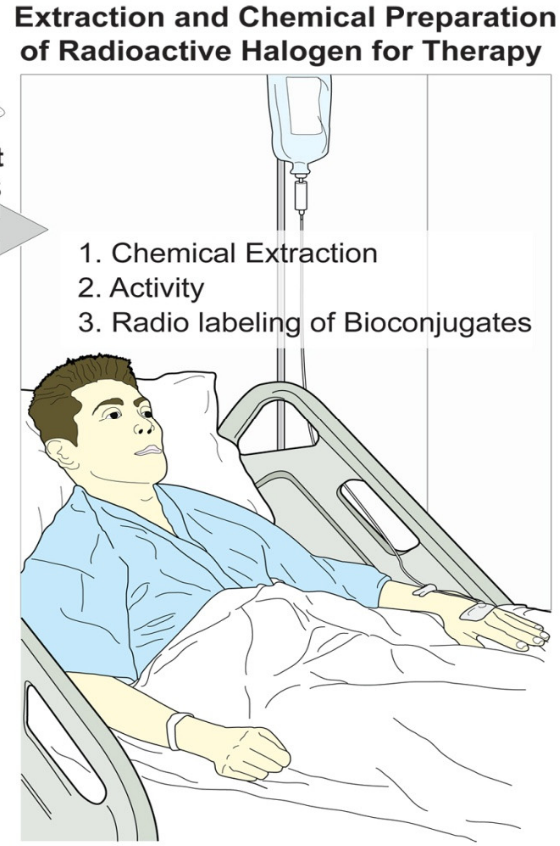
Dr. Jerry Nolen
Argonne National Laboratory

- Porous solid catchers with thicknesses in the range of ~20 $g/cm^2$ will complement gas catchers which are the FRIB base-line concept for stopping energetic rare isotopes and delivering them for stopped beam research or for reacceleration.
- Tungsten catcher to stop and release $^{11}Li$ and $^{6,8}He$ isotopes

# Medical Isotope Production Target Development (must be stable with beam power)




Helium gas transports radioactive noble gas to carbon trap




$M_2O_{3(s)} + {}^{6}Li \longrightarrow N_{(g)} + 4n$

where, $M_{(s)} = {}^{209}Bi$ , ${}^{75}As$ or ${}^{121}Sb$

and $N_{(g)} = {}^{211}Rn$ , ${}^{77}Kr$ or ${}^{123}Xe$ (Noble gases)

that decay to Radiohalogens

${}^{211}At$ ($t_{1/2}$ = 7.2 h), ${}^{77}Br$ ($t_{1/2}$ = 2.78 d), ${}^{123}I$ ($t_{1/2}$ = 13.4 h)

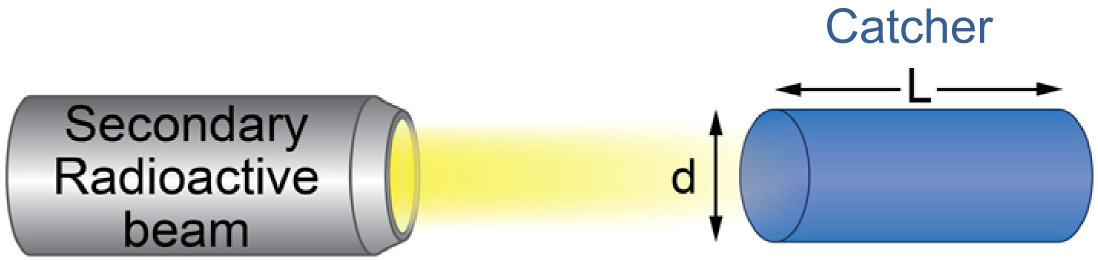
- Efficient production and release of radioactive noble gas precursors at low and room temperature – **Higher production rates of ${}^{211}At$, ${}^{77}Br$ and ${}^{123}I$**
- ${}^{6}Li$ induced reaction for parent/daughter production system, concept for a dedicated linac or cyclotron for radio-halogen production – **Overnight delivery to users from single national facility.**

# Catchers/Targets Being Studied at ISL and ANL

| Refractory Catcher/Target | Production beam | Collected Isotopes |
|---|---|---|
| Tungsten-coated $SiO_2$ Aerogel | $^{18}O$ (typical) | $^{8\text{-}11}Li$<br>$^{6,8}He$ |
| Carbon Aerogel | $^{16}O$, $^{48}Ca$, etc. | $^{12}C^{14}O$–$^{12}C^{24}O$<br>$^{12}C^{14}O_2$–$^{12}C^{24}O_2$ |
| Yttria-Stabilized Zirconia (YSZ) and Hafnia ($HfO_2$) Porous Monolith | $^{12}C$, $^{48}Ca$, etc. | $^{9}C^{16}O$–$^{22}C^{16}O$<br>$^{9}C^{16}O_2$–$^{22}C^{16}O_2$ |
| Sintering-inhibited Disks of Tungsten, Tungsten + ALD-Hafnia and Tungsten Carbide | $^{18}O$, $^{48}Ca$, etc. | "All of the above" |
| Nanoporous CaO Monolith | $^{40}Ca$ | $^{31\text{-}35}Ar$ |
| Nanoporous Metal Oxide ($M_2O_3$) Thin Films* (M = $^{209}Bi$, $^{75}As$, $^{121}Ab$) | $^{4}He$, $^{6,7}Li$ | $^{211}Rn/^{211}At$, $^{77}Kr/^{77}Br$, $^{123}Xe/^{123}I$<br>$t_{½}$ [14 h/7.4 h]; [1.24 h/2.78 d]; [2.08 h/13.4 h] |

* Thin film targetry for medical isotope production

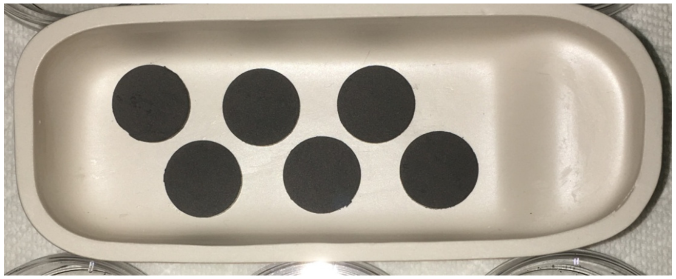
# Catcher Thickness Considerations



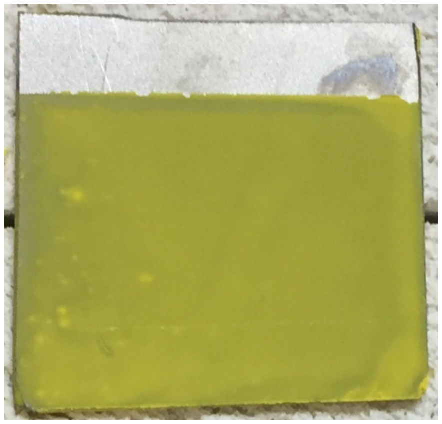

- Desired areal density (η) or thickness for efficient isotope capture can range from 3–20 g/cm$^2$ depending on the material used.
- Areal density can be related to the apparent volumetric density as:
  - $\eta = \rho L$
- This value is used to screen catcher disks after the 1000–1500 °C vacuum heat treatment

Must be thick to stop high energy radioactive beams at FRIB

# Refine processing of candidate W powders and Bismuth Oxide thin films

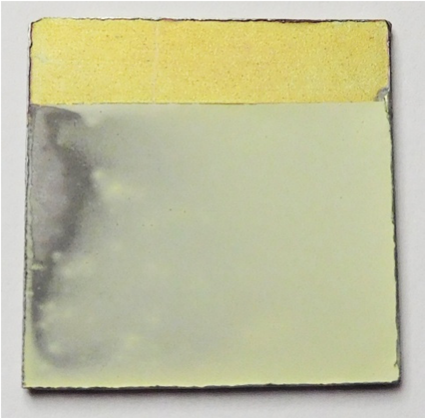


As-coated

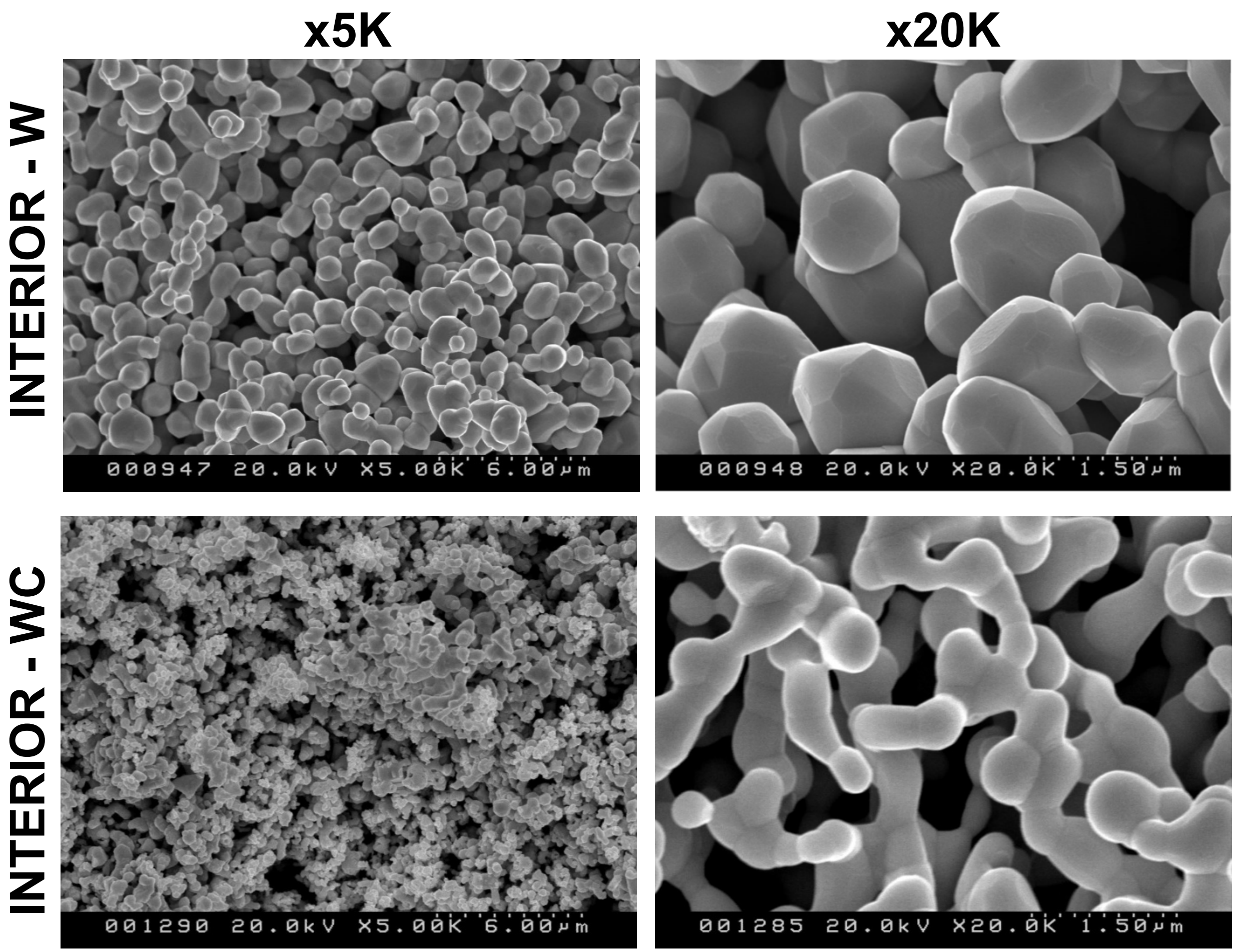


Fired@600°C

3X cycles@600°C



# Open Porosity Retained in W and WC After Heat Aging @1200 °C for 2 h




**Tungsten (<1 μm)**
**App. density ~7 g/cm$^3$ (n =5)**

**Tungsten Carbide (150–300 nm)**
**App. density ~4 g/cm$^3$ (n=6)**

Minimal grain growth and sintering-inhibition achieved with W and WC powders.

Open porosity retained at surface and interior of both W and WC disks.

Diameter = 12.1 mm
Thickness = 1.6 mm

**Hg-intrusion porosity**

**W** ~60%

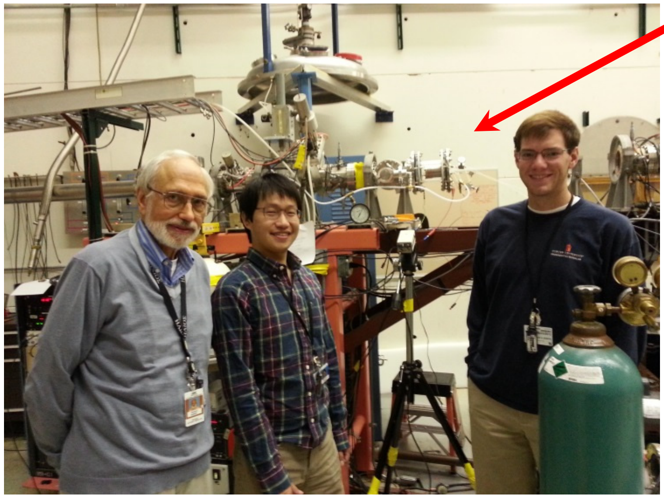
**WC** ~64%

# Nanoporous Bismuth Oxide Thin Films

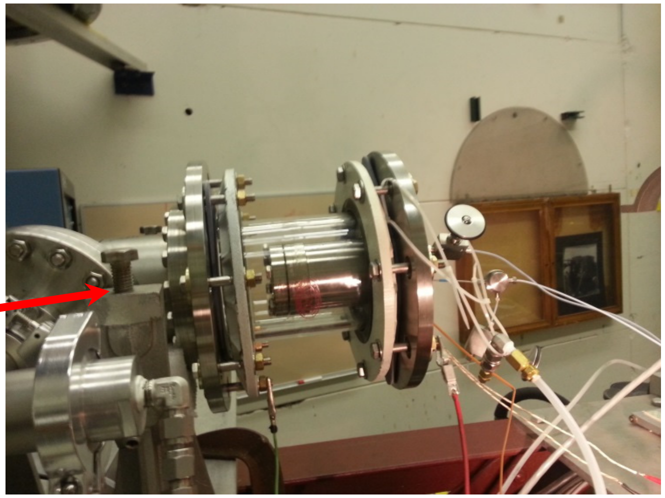
**x5K** | **x20K** | **Imaged @ 44.6 degree**


- **Contiguous nanoporous bismuth oxide films formed on Titanium coupons**
- **Thickness tunable from 2 to 80 μm – targeting 2–20 μm.**
- **Films remain adhered after 3x in vacuum heating to 600 °C.**
- **Improve uniformity of coated film across film surface**
- **Next steps:**
  - **Test at FSU with low energy carbon beams for stability evaluation**
  - **Test at ATLAS with energetic $^{6}$Li or $^{7}$Li beams for formation and release of radiohalogen Radon-211.**

# Initial Tests with a Bismuth Metal Target

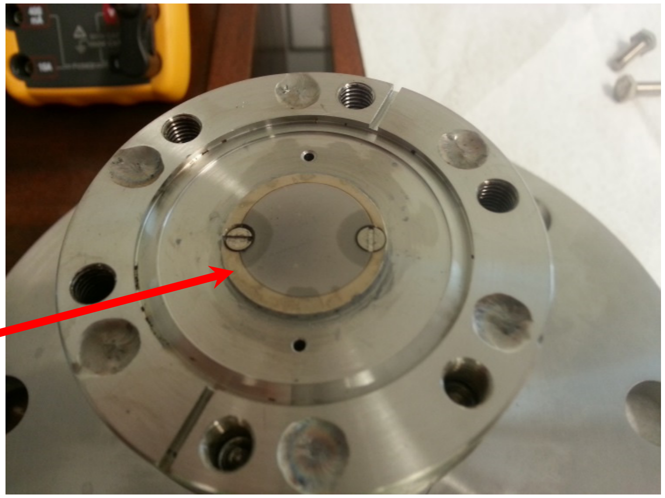


Health physicist,
Post-doc,
Undergraduate



Target/ helium plumbing/ heater assembly

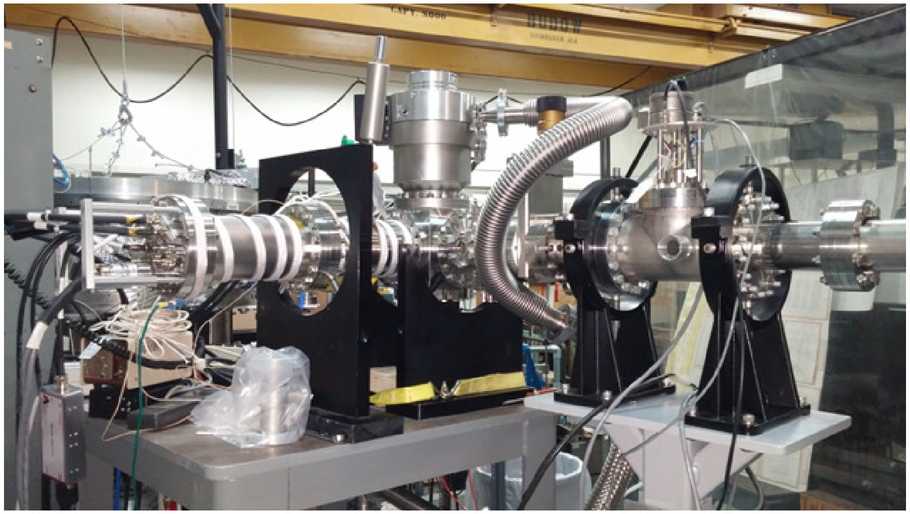

Havar window



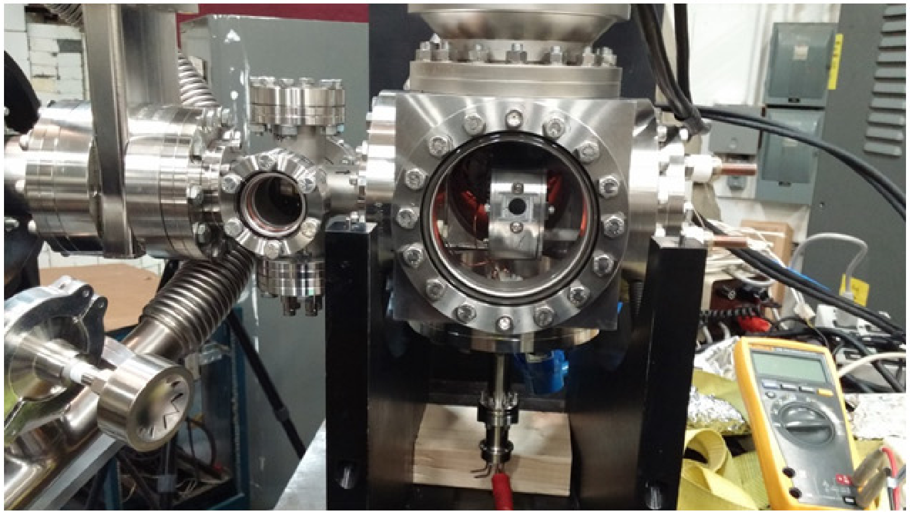
32 $mg/cm^2$ Bi on Ni

# RGA Installed in the FSU Tandem Accelerator

Beam comes from the right, the RGA is on the left, and the sample chamber is in the center below the turbo pump.

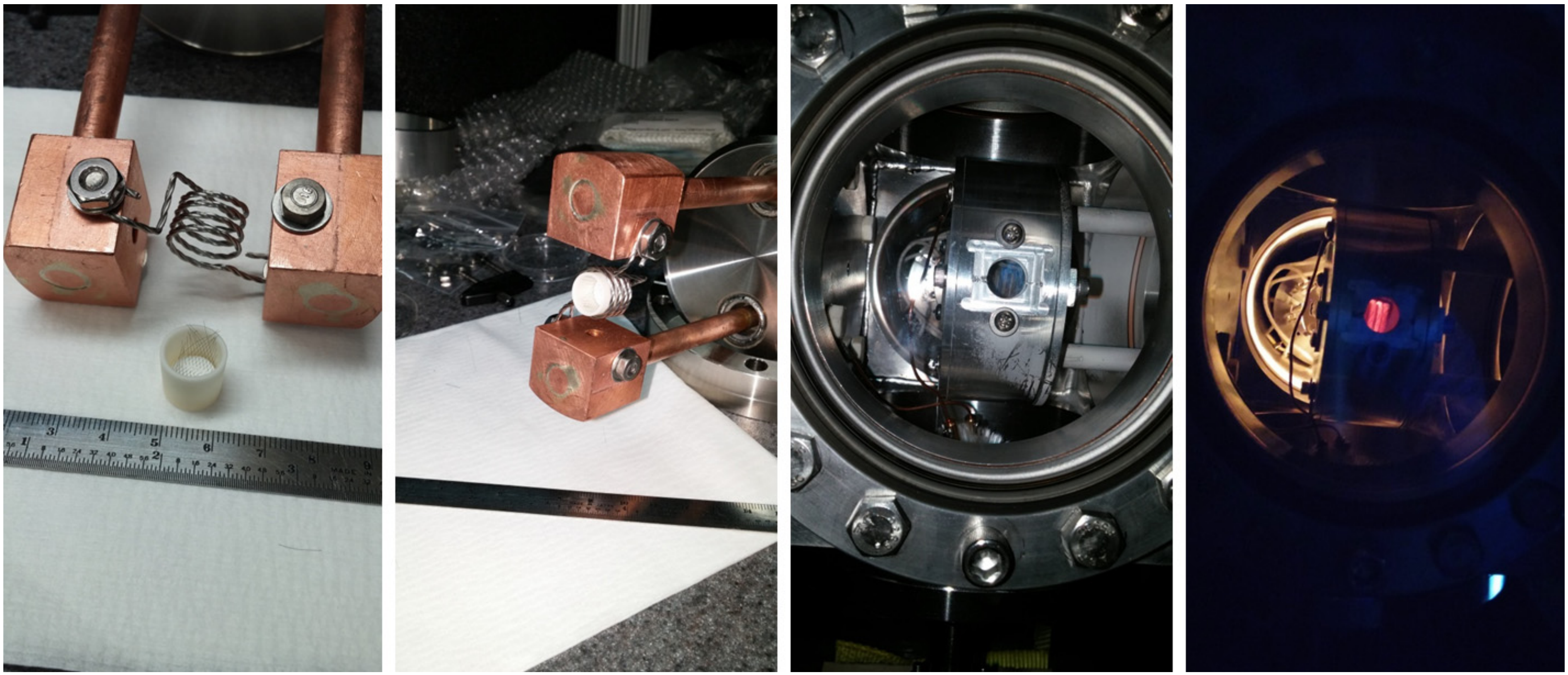


Close-up of the sample chamber with the sample holder visible through the window. Beam enters from the left. A beam diagnostics cross is upstream of the sample chamber.

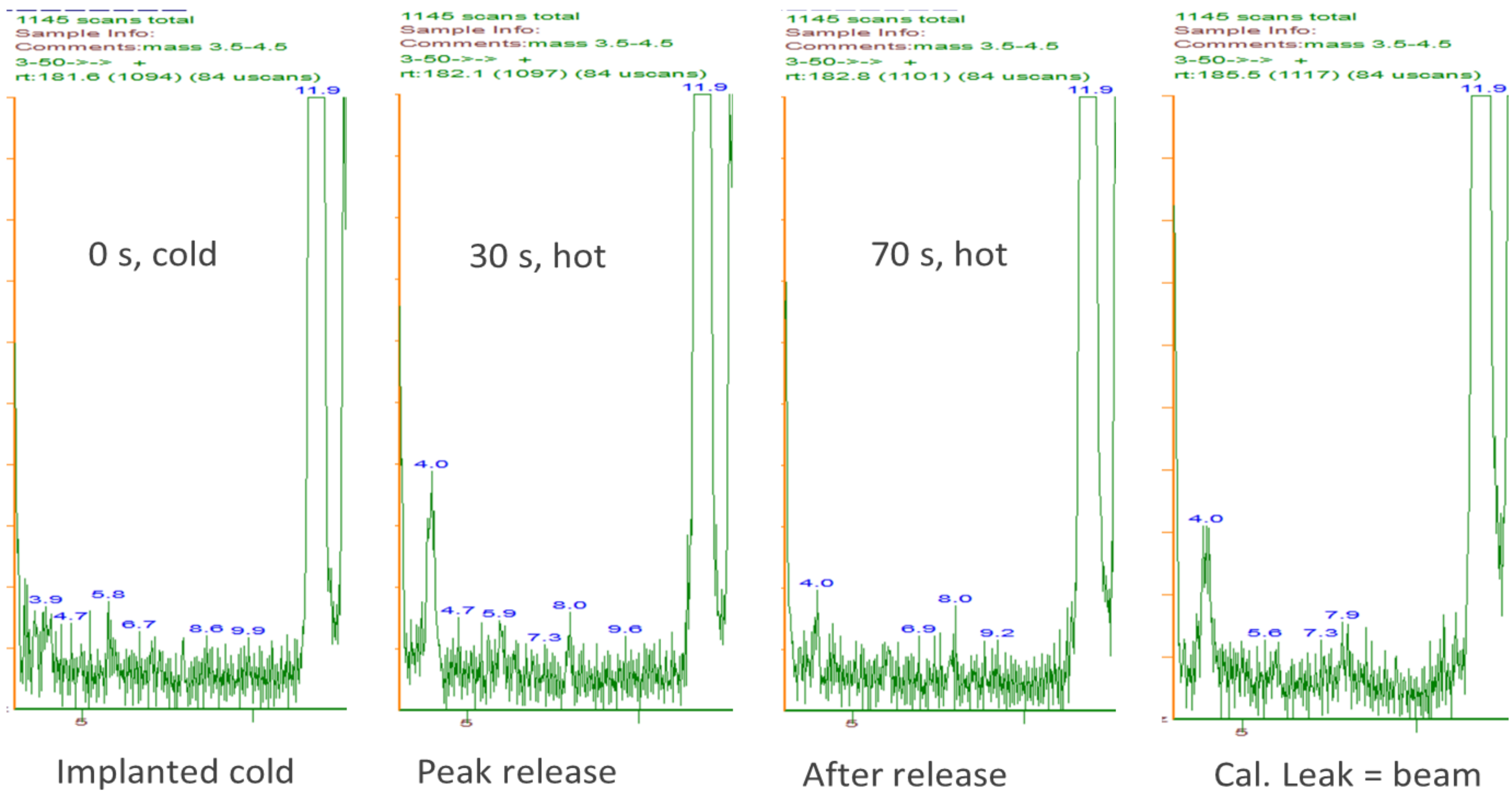


# Sample Holder and Heater in RGA

***Left and left-center:*** Alumina crucible, tungsten heater, and current feed-throughs. ***Right-center:*** View through the chamber window of the sample holder and its heat shield. ***Right:*** View of the sample with the heater on.

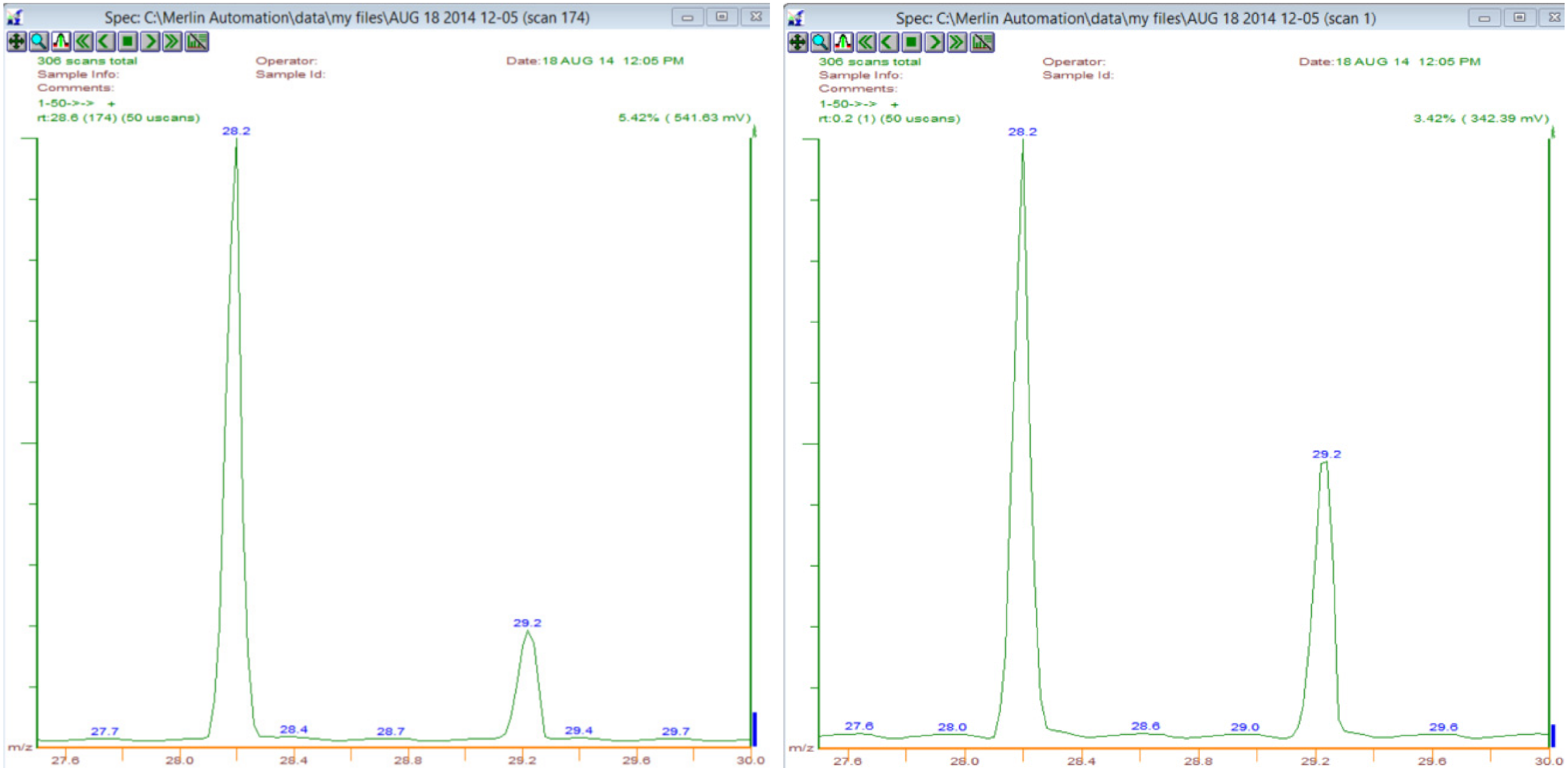


# RGA Spectra of $^{4}He$ from Calibrated Leak and from Alumina Stopper Implanted with $^{4}He$



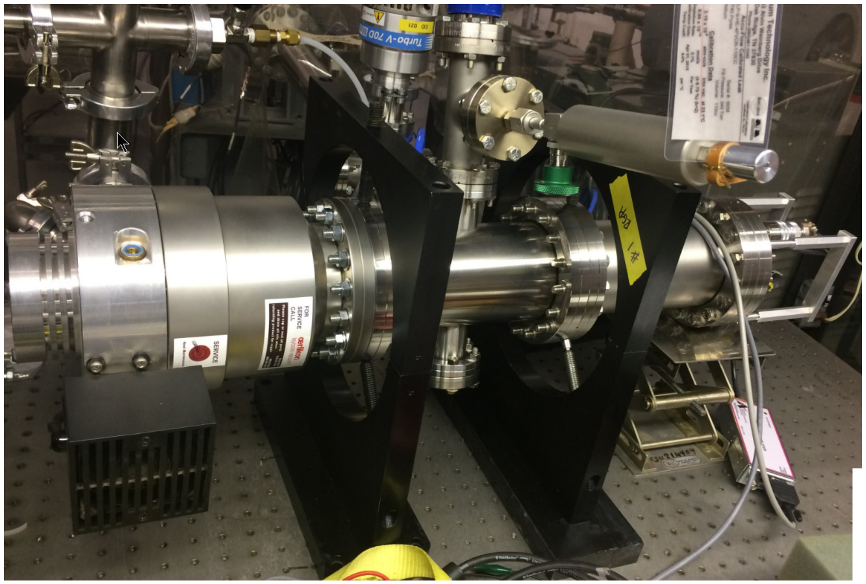

**RGA spectra between masses 3 and 12. *Left:* After He beam implantation; *Left-center:* After heating to ~1000 °C in 30 s; *Right-center*: 40 s later showing depletion of He from sample. *Right*: Same region with sample cold and calibrated He leak open to chamber.**

# Release of $^{13}CO$ Measured at T<1000 °C Matching Theoretical Simulations



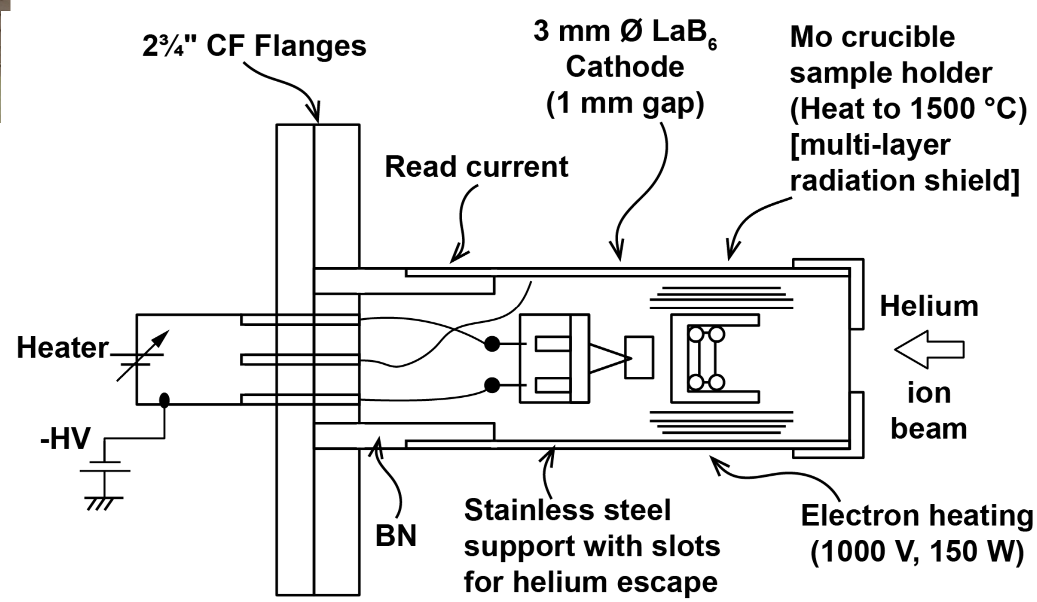

**Left Panel -** Mass 29 ($^{12}COH$ + $^{13}CO$) is larger relative to mass 28 ($^{12}CO$) in the right panel. The **right panel** was recorded near the peak of the $^{13}CO$ release and the left panel 30 minutes later.

# Revised Test Setup for FSU



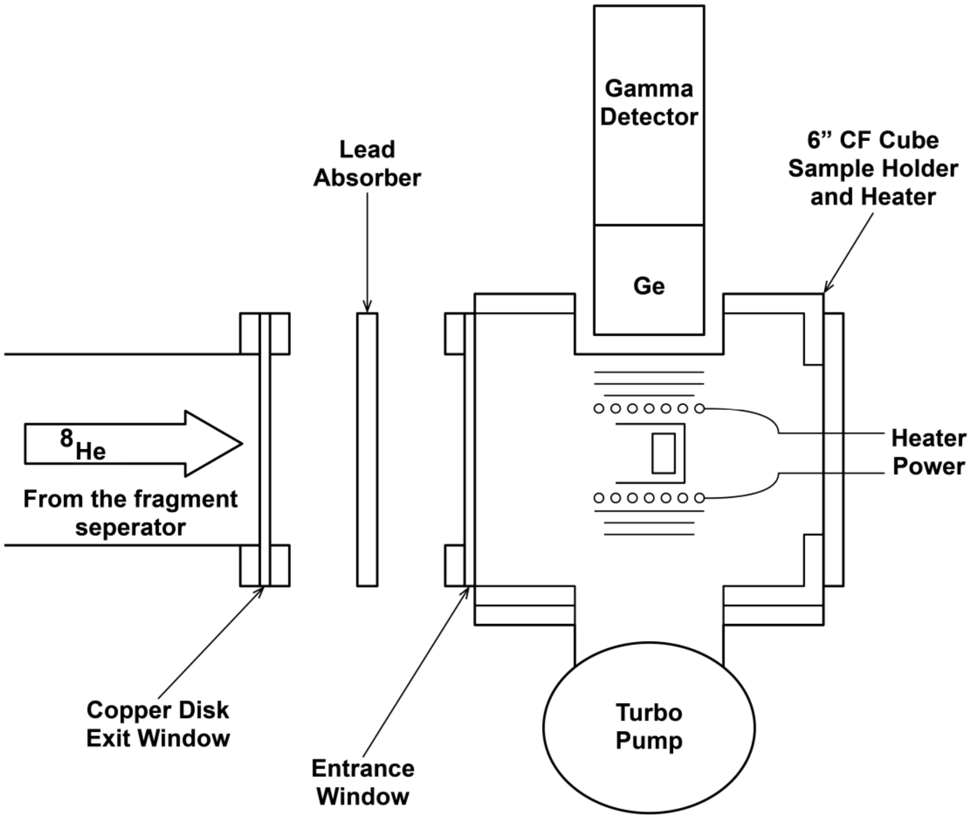
**Upgrade to UHV and much more compact sample chamber and heater. Will add plasma "cleaner" to reduce hydrocarbon background.**

- Turbo Pump connected to RGA body
- Sample in smaller chamber to minimize surface area/complete system baking
- Eliminated porous insulating materials



# Setup for solid stopper tests at NSCL

- **Beam time is approved to do develop on-line solid catcher evaluation at NSCL using short-lived isotopes**
  - Collaboration of Argonne and NSCL scientists



# Summary

- Refractory nanoporous tungsten and tungsten carbide solid catchers developed
  - Apparent density
    - Tungsten ~7 g/cm$^3$
    - Tungsten carbide ~4 g/cm$^3$
  - Open (intrusion) porosity
    - Tungsten ~60%
    - Tungsten carbide ~ 64%
- New RGA method for release characteristics of stable isotopes developed and demonstrated mass 4 ($^4$He release)
- Revised heater and UHV upgrade design completed for in-beam studies at FSU
- Beam time approved and apparatus designed for on-line testing at NSCL
- Extending development of porous solid catchers to porous thin film targets of oxides – to be tested for radiation damage with ion beams at FSU

# Acknowledgments

**DOE and the Office of Nuclear Physics to support these efforts through the following grants DE-SC0011346 and DE-SC0007252**

**Program Officer(s) – Dr. Manouchehr Farkhondeh**
**Dr. Michelle Shin**

**Dr. Georg Bollen for technical discussions and sustained interest to evaluate the catcher materials at FRIB**

**Dr. Ingo Weidenhover at FSU for beam-line studies at the FN Tandem accelerator**