Thermo-Mechanically Stable Tungsten Powders as Solid Catchers for the Fast Release of Stopped Rare Isotopes

Nuclear Physics SBIR/STTR Exchange Meeting
August 9-10, 2016

Sponsor: Office of Nuclear Physics, DOE
Program Officer: Dr. Manouchehr Farkhondeh
Phase II Contract Number: DE-SC0011346

Small Business
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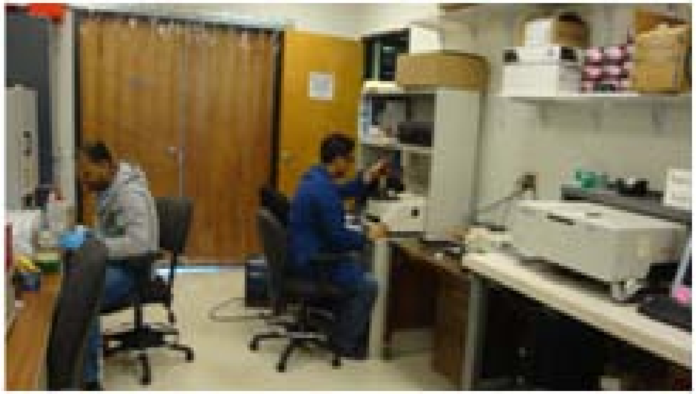
Argonne NATIONAL LABORATORY

Presentation Overview

- About InnoSense LLC
- Commercialization Status
- Motivation
- Relevance to Nuclear Physics Programs
- Year 1 Accomplishments
  - Fabrication of Refractory Solid Catchers
  - Off-Line Extended Heating Evaluations
  - In Beam Evaluations
- Summary
- Acknowledgments

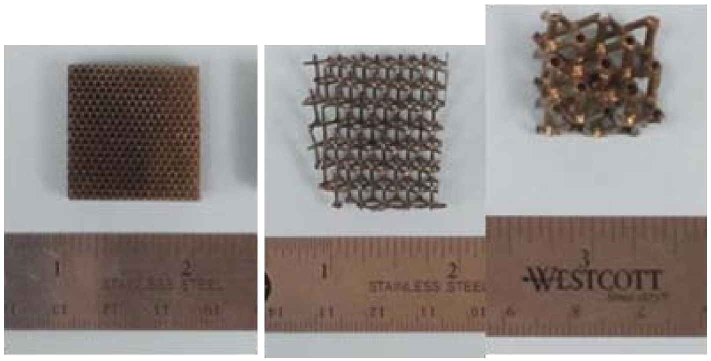
About InnoSense LLC

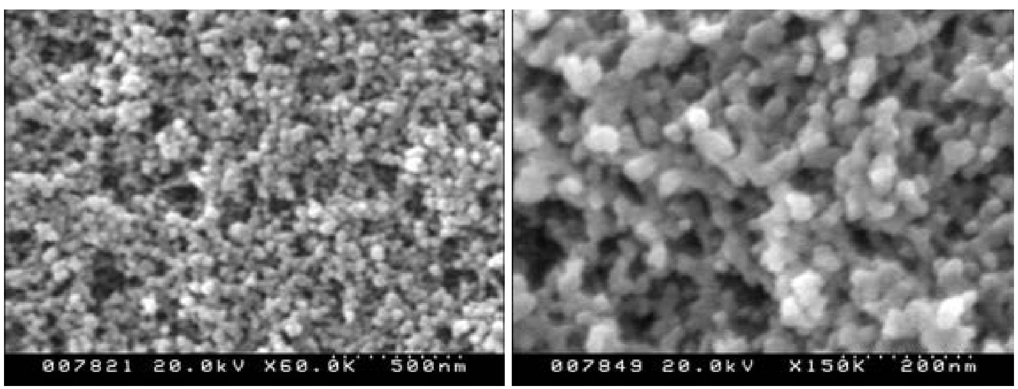
- Established in 2002 by private investment, R&D operations in 2004, housed in a recently expanded 9,000 square feet laboratory facility located in Torrance, California.
- Key laboratories include five "wet" chemical facilities equipped with fume hoods, a clean room, a spectroscopy facility, optics and testing laboratory, and two machine shops.
- 18 employees, including 4 PhD, 5 MS and 2 MBA degree holders.



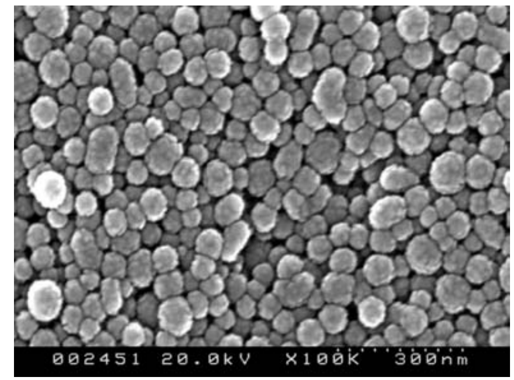
Commercialization – Building from ONP Funding

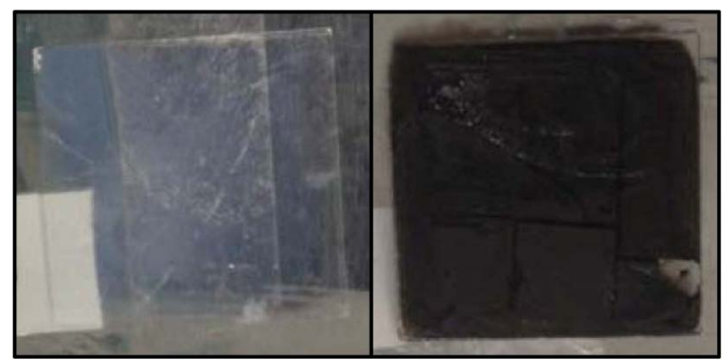
PO for silica aerogel coatings on metal lattices - Invoiced July 2015





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

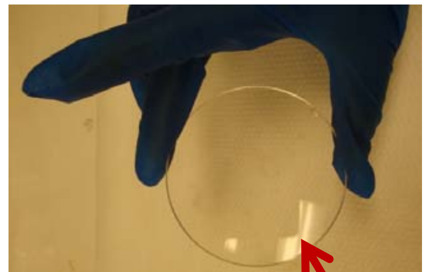
Army: W15QKN-09-C-0153

Passive Temperature Dosimeter

- **Phase III Funding**
- **Correlation Testing completed at Yuma Proving grounds November 2015**
- **Production anticipated in 2017 for 120 mm ammunition**

Army: W911NF-11-C-0056

Permanent anti-fog coating



PC lens and PU visor

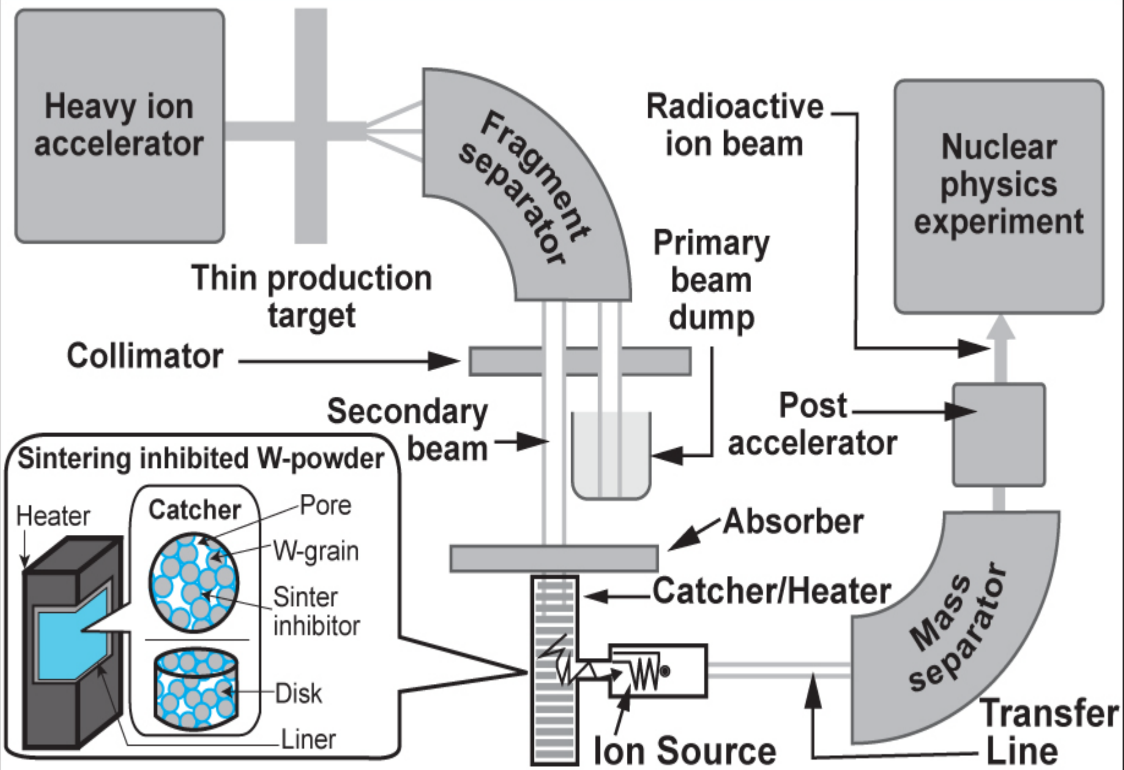
- **DOD DTRA RIF award – 2015**
- **Nanomaterials in coating**
- **Licensing deal underway with safety products manufacturer**
- **Larger commercial markets**

MDA: HQ0147-14-C-7012

Hypergolic Leak Detector for THAAD

- **Second Phase II - 2016**
- **Nanoscale sensing device**
- **Drop-in replacement for leak detector used by MDA**
- **Targeting biomedical diagnostics**

Refractory Hot Catchers for Rare Isotopes



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

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Argonne National Laboratory

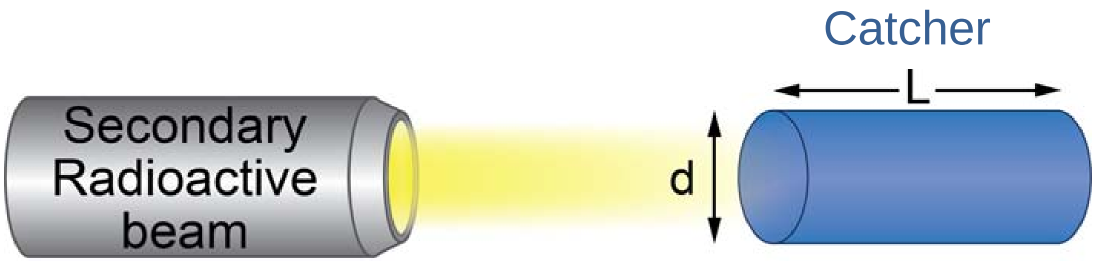
- Porous solid catchers with thicknesses in the range of ~20 g/cm^2 will complement gas catchers which are the FRIB base-line concept for stopping energetic rare isotopes and delivering them for stopped beam research or for reacceleration.
- Tungsten catcher to stop and release ^{11}Li and $^{6,8}He$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

Catchers Under Investigation at ISL and ANL

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

Background on ISOL Target Materials

Isotope Separation On-Line (ISOL) used to generate radionuclides

- Targets are used with high power beams
- Isotopes are produced by reactions of the beam with target material
- Target must be dense enough to stop energetic beam, yet porous enough to allow rapid diffusion of radionuclides to the accelerator source
- Must be thermally conductive to withstand beam power
- Targets are heated to >2000 °C to increase diffusion rates of radioactive nuclides

Benefits When Used in Catcher Mode

- Catchers used to stop high energy radioactive isotopes created in a separate production target up stream
- In the catcher mode, thermal conductivity is less relevant since the beam power is deposited in the thermally separated production target irradiated with heavy ion beams
- No radiation damage when used in catcher mode since only secondary radioisotope beams impinge on it
- Selection of materials is open to new approaches that cannot work with ISOL targets, e.g. aerogels with low thermal conductivity
- The porous refractory materials will theoretically offer more stopping power and fast-release for the generation of intense rare isotopes
- The refractory nature potentially allows them to be used as:
  - Compact isotope catcher/ion source placed in the first focal plane of the fragment separator with the capability of selective harvesting for isotopes for different applications

Technical Objectives and Success Criteria

Objective 1. Refine processing of candidate W powders from Phase I.

Criteria 1: *Minimal grain growth observed via SEM. open porosity retained when vacuum heat aged @ 1200 and 1400 °C.* ☑

Objective 2. Develop non sintering forms of tungsten carbide (WC) and W-coated with atomic layer deposited (ALD) hafnia (HfO_2).

Criteria 2: *Powders remain loosely compacted upon vacuum heat aging; minimal grain growth for WC and W-ALD-HfO_2.*

Objective 3. Evaluate candidate catchers on-line at FSU using stable ^{4}He with RGA

Criteria 3: *On-line tests at the FSU FN Tandem demonstrate the fast release of noble gases (helium) from selected solid catchers after long-term heating to temperatures at 1500 °C or higher. [Screening for on-line testing at NSCL]*

Objective 4: Evaluate candidate catchers on-line at NSCL (Oxygen isotopes stopping in WC or carbon isotopes in oxides to form molecular sidebands).

Criteria 4: *Development of WC and W-ALD HfO_2 powder forms as predicted to be useful for the formation of specific molecular sidebands.*

Objective 5: *Compare on-line test results with HSC predictions to determine the validity of such simulations.*

Criteria 5: *Demonstration of the predictive power of molecular thermochemical simulations for non-equilibrium release of volatile species such as CO.*

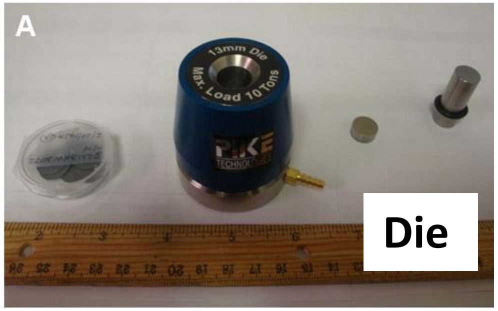
Project Schedule

Tasks	Task	Year 1: 1	2	3	4	Year 2: 5	6	7	8
T1	Refine Hf- and BN-doped W powders from Phase I								
T2	Evaluate powders to confirm sintering inhibition								
T3	Develop sinter-inhibited WC powders as catchers								
T4	Develop ALD process for W-HfO_2								
T5	Evaluate powder characteristics from T3 and T4 after extended vacuum heat aging								
T6	Conduct on-line tests of W catchers at FSU								
T7	Conduct on-line tests of catchers at NSCL								
T8	Validate predictions with on-line release tests								
T9	Evaluate Phase II commercialization								
T10	Submit Deliverables								
Original Milestone Timeline→			❶	❷		❸		❹	❺ ❻
Revised Milestone Timeline→				❶			❷ ❸	❹	❺ ❻

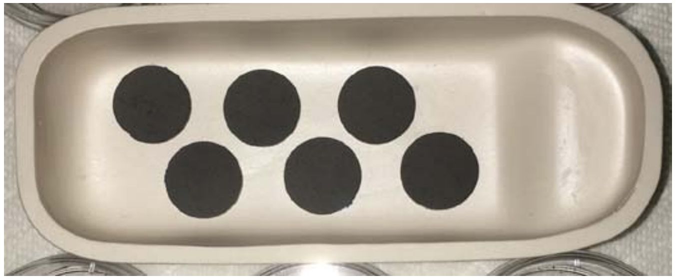
	Revised Task Timeline		Submitted Reports		Milestone Met

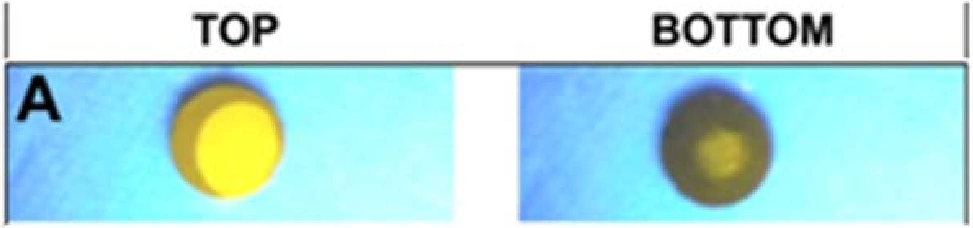
Milestone 1: Grain growth inhibited W powders are prescreened.
Milestone 2: Non-sintering forms of WC and W-ALD HfO_2 powders are prescreened.
Milestone 3: Release efficiency of candidate powders characterized at FSU.
Milestone 4: On-line release measurements with radioactive beams completed at NSCL.
Milestone 5: Predicted release profiles are validated experimentally.
Milestone 6: Phase II objectives successfully demonstrated.

Refine processing of candidate W powders



Hydraulic Press



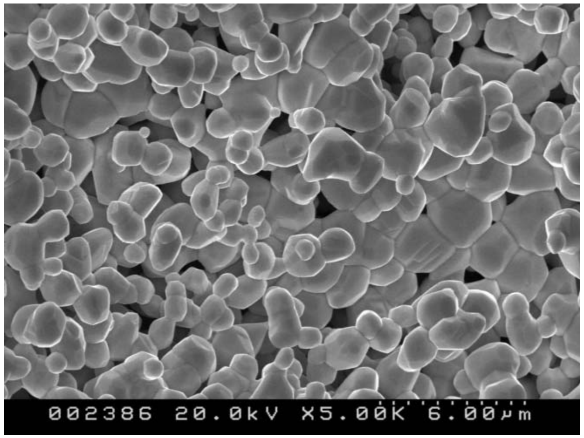


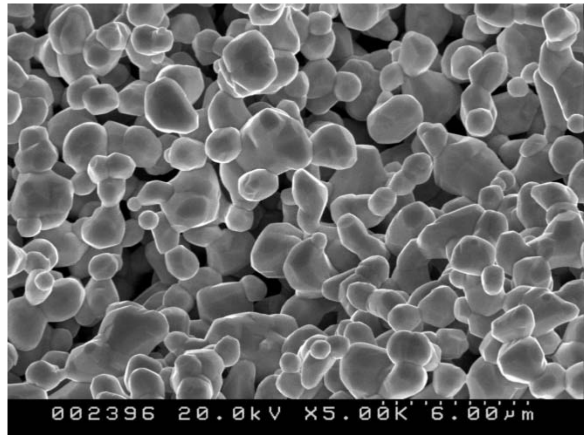
Dye Penetrant Test

Open Porosity Retained After Vacuum Heat Aging @1400 °C for 12 h

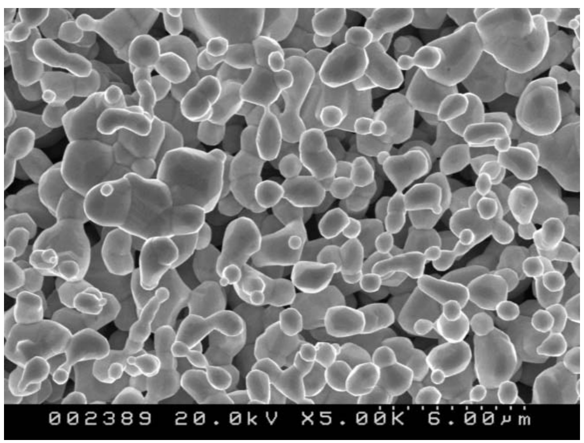
x5K | x5K

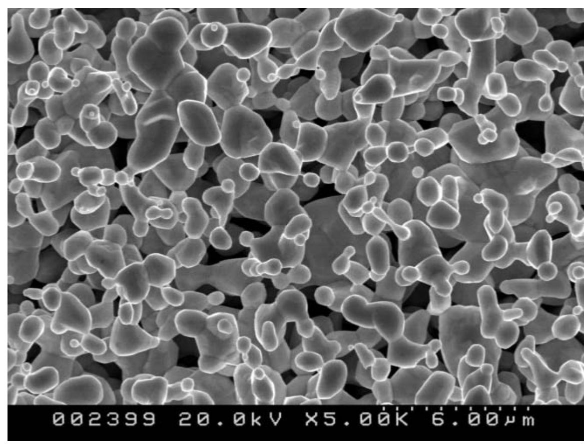
INTERIOR





SURFACE





Tungsten
App. density = 9.20 ± 0.26 g/cm^3
App. Porosity ~ 52%

Sinter-inhibited tungsten
App. density = 8.77 ± 0.11 g/cm^3
App. Porosity ~ 54%

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

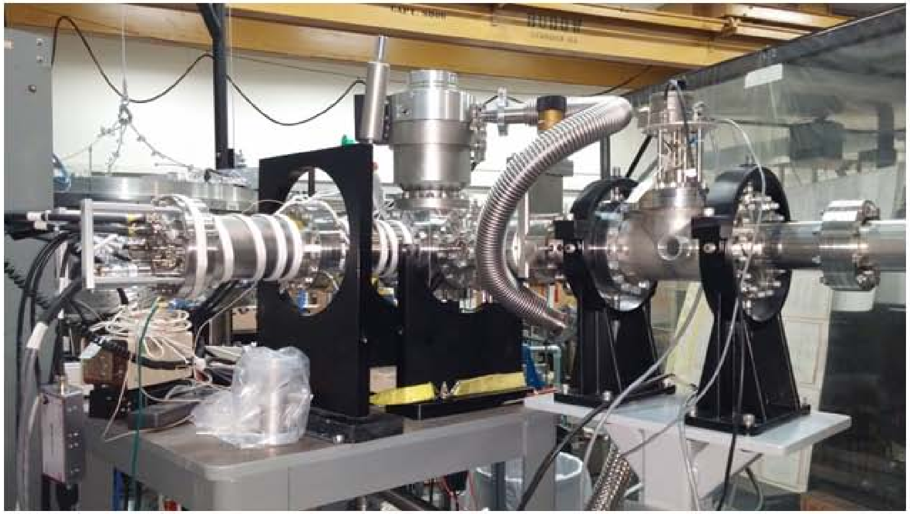
Open porosity retained at surface and interior of W disks.

Diameter = 12.1 mm
Thickness = 1.6 mm

Hg-intrusion porosimetry
Skeletal Density = ??
Intrusion porosity = ??

RGA Installed in the FSU Tandem Accelerator

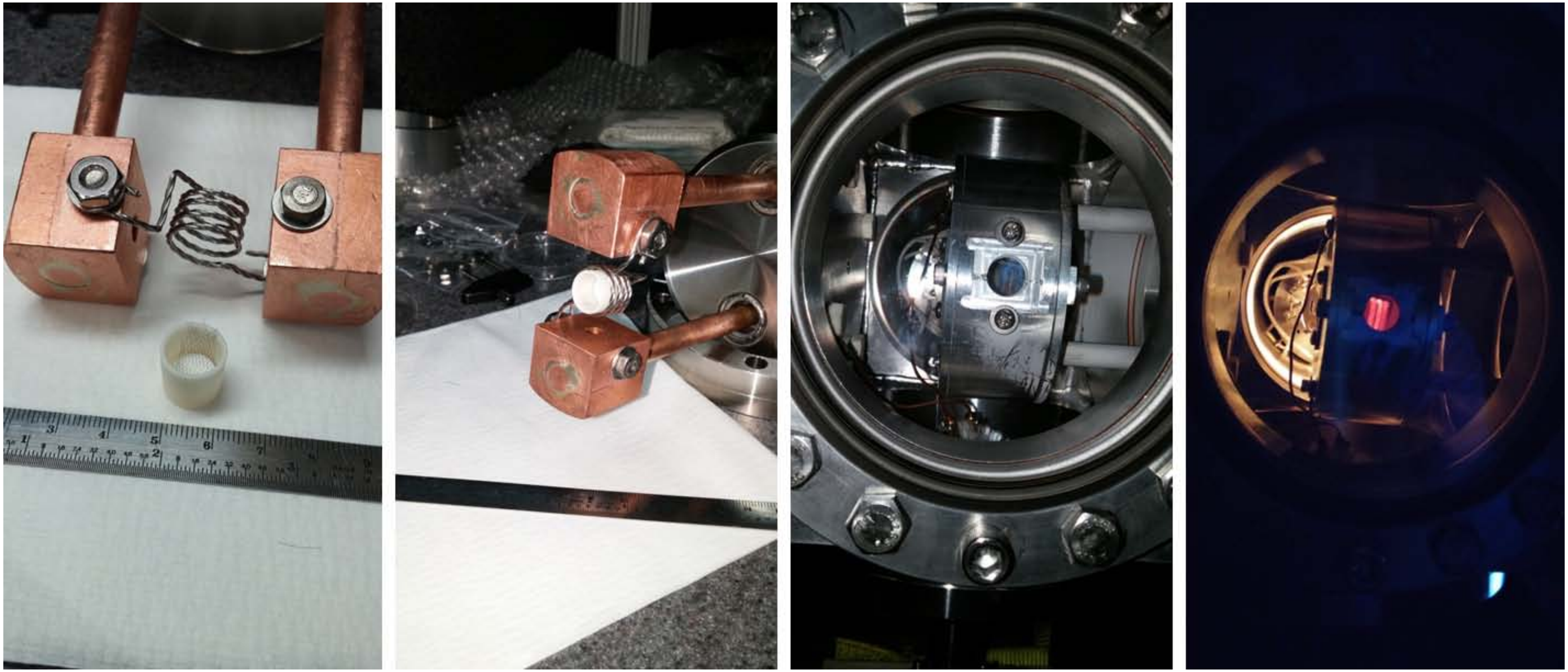
Beam comes from the right, the RGA is on the left, and the sample chamber is in the center below the turbo pump.



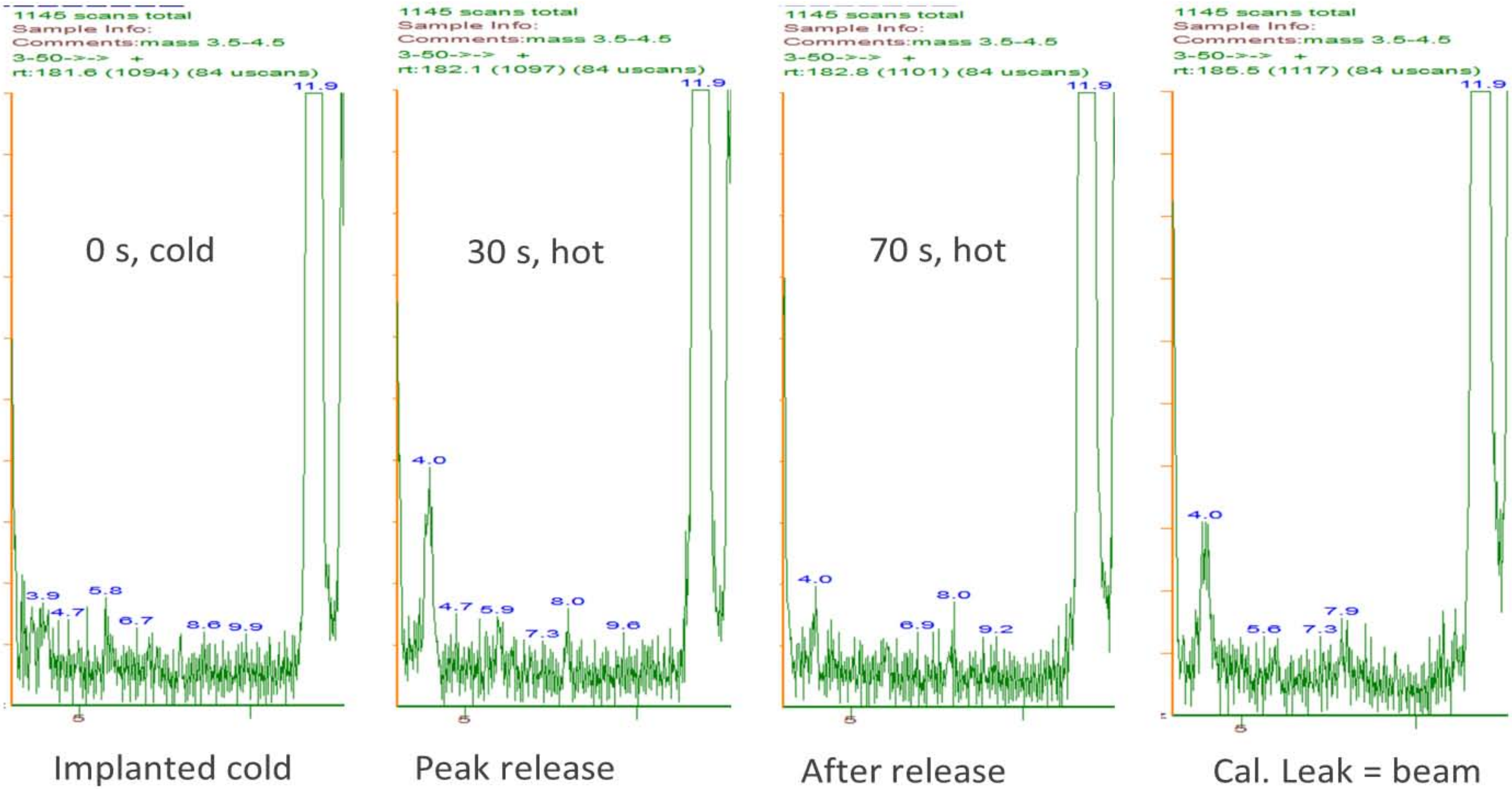
Close-up of the sample chamber with the sample holder visible through the window. Beam enters from the left. A beam diagnostics cross is upstream of the sample chamber.

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.

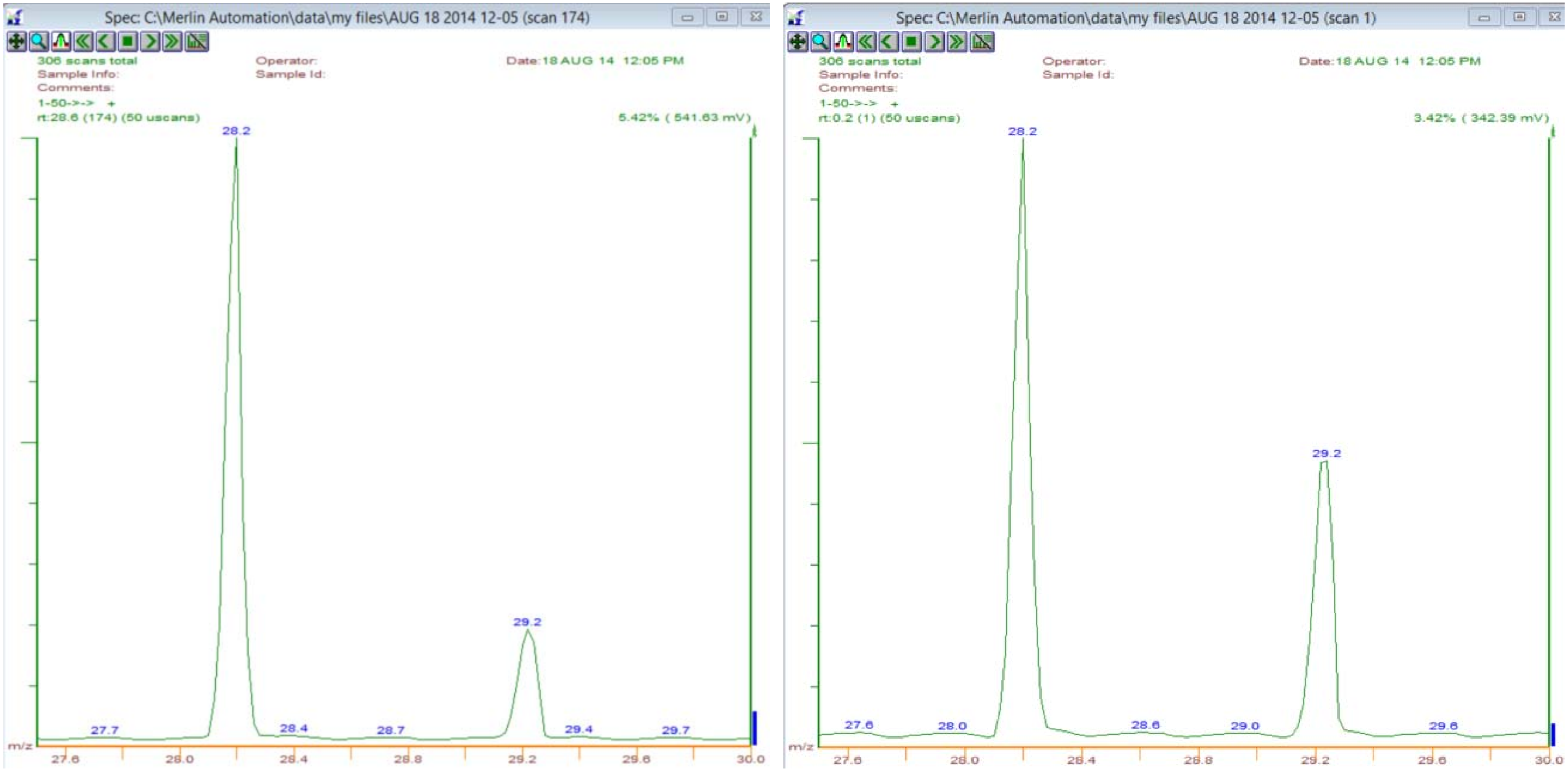


RGA Spectra of ^{4}He from Calibrated Leak and from Alumina Stopper Implanted with ^{4}He



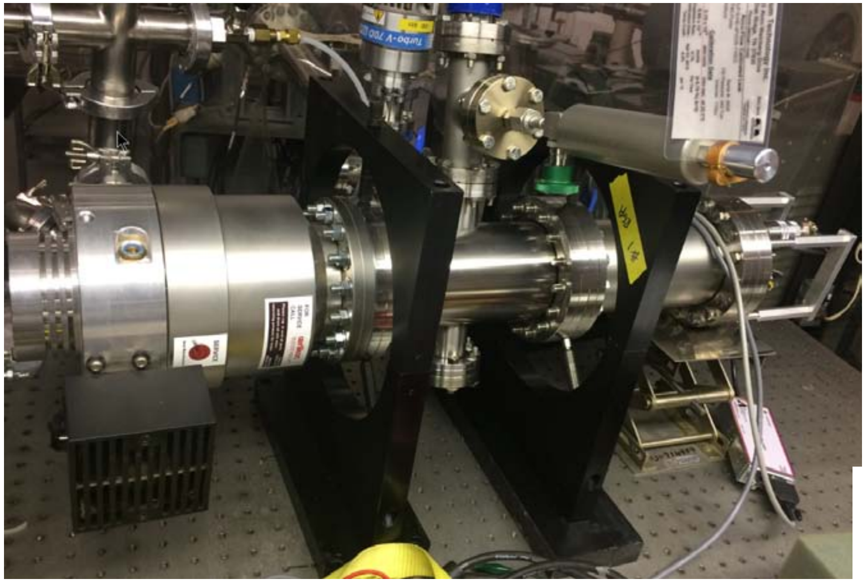
RGA spectra between masses 3 and 12. *Left:* After He beam implantation; *Left-center:* After heating to ~1000 ºC in 30 s; *Right-center*: 40 s later showing depletion of He from sample. *Right*: Same region with sample cold and calibrated He leak open to chamber.

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



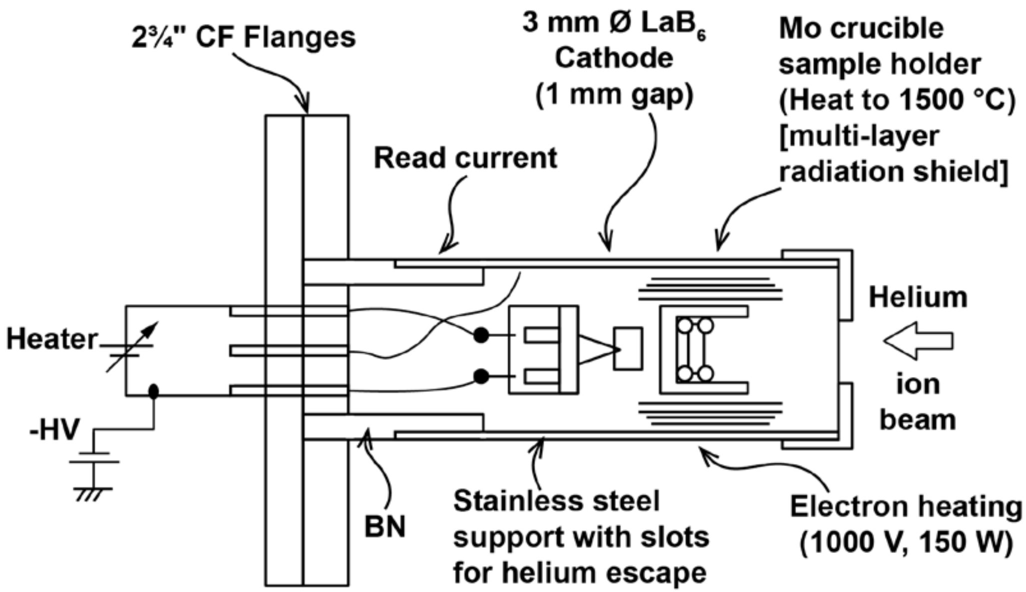
Left Panel - Mass 29 (^{12}COH + ^{13}CO) is larger relative to mass 28 (^{12}CO) in the right panel. The **right panel** was recorded near the peak of the ^{13}CO release and the left panel 30 minutes later.

Revised Test Setup for FSU



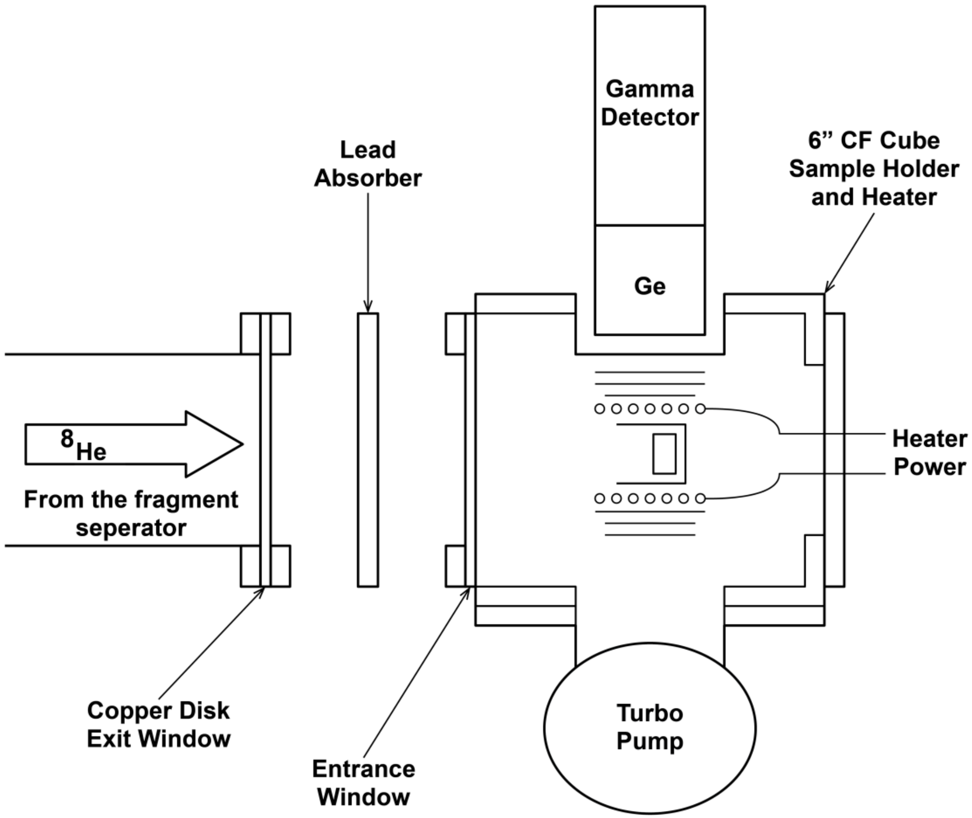
Upgrade to UHV and much more compact sample chamber and heater. Will add plasma "cleaner" to reduce hydrocarbon background.

- 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 tungsten solid catchers 1.6-1.7 mm thick after vacuum heat aging @1400 °C/12 h
  - Apparent density
    - Tungsten ~9.20 ± 0.26 g/cm^3
    - Sinter-inhibited tungsten ~8.77 ± 0.11 g/cm^3
  - Apparent porosity
    - Tungsten ~52.3%
    - Sinter-inhibited tungsten ~ 54.1%
- Beam line tests demonstrated molecular ^{13}CO release from nanoalumina powder with trace C additive and calibrated ^{4}He leak.
- Work on tungsten carbide and W-ALD-HfO_2 to be completed
- 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

Acknowledgments

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

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