

Demonstration of the Stopping and Release of Rare Light Isotopes at NSCL-MSU Facility and Medical Isotope Production at ATLAS

Uma Sampathkumaran¹, Raymond Winter¹, John Abolencia¹, Cole Gladfelter¹, Jerry Nolen², Brahim Mustapha², Robin de Kruijff², Jeongseog Song², Ravi Gampa², Shaofei Zhu², Matthew Gott² and John Greene² ¹ InnoSense LLC, 2531 West 237th Street, Ste 127, Torrance, CA 90505; ² Argonne National Laboratory, Physics Division, 9700 S. Cass Avenue, Lemont, IL 60439

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ABSTRACT

InnoSense LLC (ISL) has developed a manufacturing technology to produce refractory Yttria-stabilized zirconia (YSZ) and Hafnia (HfO₂) and Calcium oxide CaO (CaO) monoliths with >50% open interconnected porosity, for use as versatile, fast-release solid catchers for rare isotopes (SCRI). These catchers are targeted to stop rare isotopes with conversion to and release as shown below.

Refractory Catcher	Production Beam	Collected Isotopes
Nanoporous YSZ and HfO ₂	¹² C, ⁴⁸ C, etc.	⁹ C ¹⁶ O— ²² C ¹⁶ O ⁹ C ¹⁶ O ₂ — ²² C ¹⁶ O ₂
Nanoporous Tungsten (W) W + ALD-Hafnia; Tungsten Carbide (WC)	¹⁸ O, ⁴⁸ Ca, etc.	⁸⁻¹¹ Li; ^{6,8} He; ⁹ C ¹⁶ O— ²² C ¹⁶ O ⁹ C ¹⁶ O ₂ — ²² C ¹⁶ O ₂
Nanoporous CaO	⁴⁰ Ca	³¹⁻³⁵ Ar

Solid catchers can play an important role at in-flight fragmentation facilities for stopped and reaccelerated beams, and for isotope harvesting. Such solid catchers can be used in parasitic or primary user modes and are complementary to gas catchers (See Figure below).

ISL demonstrated proof-of-concept for SCRI by developing refractory monoliths of nano- YSZ, HfO₂ and CaO with 52-74% interconnected porosity. The collaborator at ANL developed/demonstrated:

- A novel method to implant and release ⁴He ions in a porous oxide stopper using a residual gas analyzer.
- A catcher/heater apparatus for stopping and release of ⁸He with a 118 msec lifetime at NSCL/MSU.



Schematic depiction of the use of porous oxide catchers in an isotope production setup. [Inset] Cross-section view of the oxide catcher.

INNOVATIONS		
Feature	Advantage	Benefit
Novel refractory formulations	Porous tape cast monoliths	Thin structurally stable disks at high temperature
Fugitive acicular or spherical pore formers	Large interconnected porosity at multiple length scales	Fast release of short-lived isotopes
Sintering inhibitors	Retain open porosity during extended vacuum heating	Extends operational life in beam line
Tape casting method to form thin disks as catchers	Applicable to oxides/carbides/nitrides/ silicides	Tune radioisotope release by catcher or additive composition

Nanoscale powders (YSZ, HfO₂ and CaCO₃) were used as starting materials. The processing steps are shown in the images below.





900 ° C/1 h and 1200 ° C /1 h at 1E5 HPa)

- (mass and geometric dimensions)
- porosimetry)



Scanning Electron Microscope (SEM) images of YSZ, HfO₂ and CaO disks following sequential heat treatments in air, and vacuum - Retained >50% open pores and limited particle growth.

FABRICATING CATCHERS

Oxide Catcher Processing: A. Tape casting, B. Heat lamination of disks, C. Green disks, D. Fired disks, E. Vacuum tube furnace for heat aging studies (Ar capable).

CATCHER CHARACTERISTICS

Post sequential heat processing (1400 °C/2 h in air; • 1-2 mm thick disks robust to mechanical handling • Apparent density - n-YSZ (n=16) = 2.82 ± 0.04 g/cm³

• Open porosity - n-YSZ (n=4) = 52±1% (Hg intrusion

• Apparent density - n-HfO₂ (12)= 3.0 ± 0.09 g/cm³ • Open porosity - $n-HfO_2(n=3) = 63\pm 1\%$ (Hg intrusion) • Apparent density of CaO (n=12)= 0.87±0.06 g/cm³ • Open porosity- CaO (6) = 74±2% (Hg intrusion) • Varying degrees of particle coalescence observed

TESTING SOLID STOPPERS AT NSCL

- Test setup was constructed and tested at ATLAS
- First on-line test of the porous solid catchers (WC) was conducted at NSCL using a very short-lived isotope (⁸He, 119 msec) in May 2018.
- Collaboration between ANL and NSCL scientists.



Test Participants: Jeongseog Song, Ravi Gampa, Jim Specht, John Greene, Matt Gott, and Jerry Nolen from Argonne, and Mauricio Portillo, Antonio Villari, Mathias Steiner, and Tom Ginter from MSU/NSCL

- Test apparatus has a dedicated station at the NSCL beam line from the Fragment Separator.
- Will enable reacceleration of very short-lived light isotopes, and achieve beam intensities that exceed the theoretical limits of gas catchers (~1E9 ions/s).

TARGETRY FOR ²¹¹RADON/²¹¹ASTATINE

Extending **SCRI** technology to medical isotopes, ISL adapted the porous oxide fabrication to develop bismuth oxide films on metal targets (Titanium and 303SS). In-situ production of the noble gas ²¹¹Radon (**Rn**) via the

²⁰⁹Bi(⁶Li,4n)²¹¹Rn>²¹¹Astatine (At) nuclear reaction was investigated at ATLAS.²¹¹At is a critical alpha-emitter.



production scheme for ²¹¹Rn/²¹¹At generator (GentopeTM) for potential delivery to other domestic locations in the continental U.S.





⁸He decreased with heater power up to present limit of 400 W,

 Release estimate ~20% - very good for a noble

TARGET CHARACTERIZATION

Bismuth oxide (Bi_2O_3) target on 303SS or Titanium support

- Processed by spin coating here, newer method agnostic to shape
- Adheres to substrate upon thermal cycling to 600 °C.
- Retains 50–60% porosity
- Film thickness tunable from 2–80 μm.



monoclinic Bi₂O₃.





(Left) Heat-treated Bi_2O_3 coatings on 303SS and (Right) sample mounted for in-beam test at ATLAS. Area density of Bi₂O₃ coatings ranged from 9–15 mg/cm².

CONCLUSIONS

- The high density and large open porosity of the SCRI are ideal for capturing rare isotopes, allowing their diffusion into and release from the catcher.
- The first-generation of a catcher/heater apparatus was built and tested at both NSCL and ATLAS.
- The apparatus successfully heated samples up to ~400 °C while implanting ⁸He fragments at NSCL.
- Very promising first result with an "effective half-life" decreased to ~117 msec (limited by present heater) estimated at ~20% release efficiency.
- Several fabricated porous catchers (oxide, tungsten, tungsten carbide, carbon) await on-line testing at FRIB.
- Heating the ceramic-like Bi_2O_3 target to 500 °C in helium atmosphere did not release ²¹¹Rn significantly. Next test is to heat in vacuum and go to higher T if necessary. (run scheduled for Sept 3-5, 2019.)
- Targetry development continues through an ongoing SBIR Phase I (DE-SC0019587).

ABOUT INNOSENSE LLC

- Founded in 2002
- Located in Torrance, CA
- 30 full-time employees 11,400 sq ft. of lab & office space

