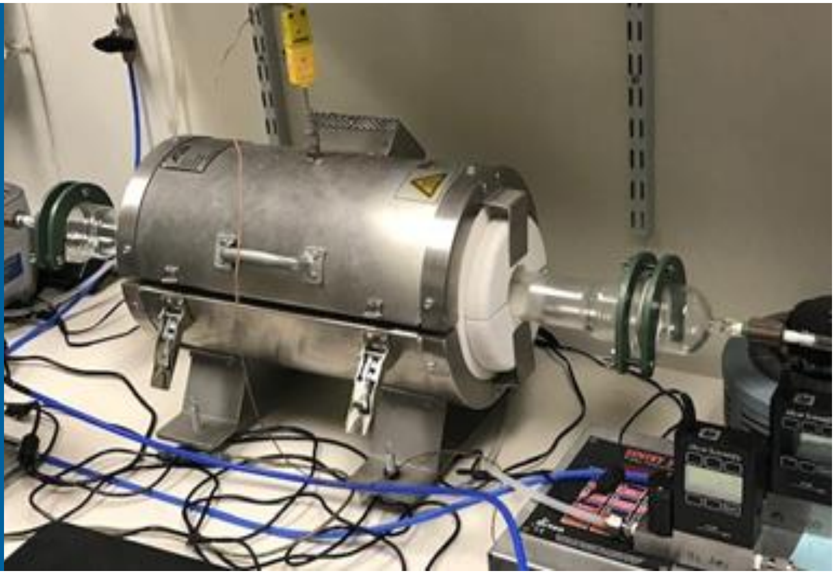
29TH WORLD CONFERENCE OF THE INTERNATIONAL NUCLEAR TARGET DEVELOPMENT SOCIETY

RHENIUM AND IRIDIUM TARGETS PREPARED USING A NOVEL GRAPHENE LOADING TECHNIQUE



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RHENIUM AND IRIDIUM TARGETS PREPARED USING A NOVEL GRAPHENE LOADING TECHNIQUE

ABSTRACT

- For accelerator targets, graphene films are an excellent material choice due to their high thermal conductivity, high temperature tolerance, low outgassing, mechanical integrity, and ease of handling. A variety of targets have been produced using graphene material as a backing or a host matrix. One of the unique advantages of the graphene film fabrication process is the capability to embed target materials, including refractory metals, in the nanoparticle form into a host graphene matrix during target preparation. Targets of ^{nat}Ir and ^{nat}Re have been fabricated as nanoparticle loaded graphene targets for use in nuclear physics research.
- We hope to obtain beam time to evaluate target performance as well as production yields and nuclear decay properties via the ^{nat}Re(α, 2n)^{186}Ir and ^{nat}Ir(α, 3n)^{194}Au reactions, respectively. These rhenium and iridium targets will be irradiated using the ATLAS accelerator and gamma rays measured in-place using the high-precision gamma-ray spectroscopy capabilities of Gammasphere and further analyzed using a multi-parameter detector system. Future plans include the preparation of isotopic targets of these two elements.

OUTLINE OF TALK

- INTRODUCTION AND MOTIVATION
- DESCRIPTION OF THE GRAPHENE LOADING TECHNIQUE
- EXPERIMENTAL RESULTS AND DISCUSSION
- CONCLUSION

INTRODUCTION AND MOTIVATION

Theranostic Radiopharmaceuticals based on ^{186}Ir and ^{194}Au

- A growing field within nuclear medicine are theranostic drugs which allow for low dose imaging and dosimetry estimates followed by a tailored course of therapeutic treatments. The success of this approach is dependent on high specific activity radionuclides. Some recent studies into iridium-based anti-cancer drugs have used photosensitizing complexes to attack cancer cells. Incorporation of the Auger-emitting ^{186}Ir (t1/2 = 16.64 h) could lead to a synergistic killing effect by both improving the overall treatment and by reducing the required dose of chemotherapeutic agent. Additionally, its gamma emissions (γ = 296.9 keV, 62.3%; 137.2 keV, 41.4%) are suitable for SPECT imaging and could have an immense impact on determining dose characteristics. Cytotoxic gold compounds have also been evaluated as anti-cancer drugs. Auger-emitting ^{194}Au (t1/2 = 38.02 h) can provide the same synergistic killing effect and SPECT imaging (γ = 328.5 keV, 60.4%; 293.5 keV, 10.6%) for gold-based anti-cancer drugs.

INTRODUCTION AND MOTIVATION

Re and Ir Graphene Targets for Radiopharmaceutical Production

- We propose to produce high specific activity radio-iridium and radio-gold via the ^{185}Re(α, 2n)^{186}Ir [1087 mb; 35 MeV] and ^{193}Ir(α, 3n)^{194}Au [1101 mb; 35 MeV] reactions, respectively, utilizing natural abundance targets for these initial experiments. These isotopes are highly naturally abundant (37.40% for ^{185}Re and 62.7% for ^{193}Ir). Experimental data in the EXFOR database is extremely limited for these reactions, particularly at the alpha energy of interest. Most the data relies on measurements performed in the1990s. Theoretical production yields were calculated based on a 1 mg/cm^2 target and a beam current of 1 pnA. Production rates of 12.9 μCi·nAh-1 and 20.8 μCi·nAh-1 are expected for ^{186}Ir and ^{194}Au, respectively.
- With cancer incidence rates continuing to climb, research into new and advanced, diagnostic and therapeutic radiopharmaceuticals is critical to meeting our future needs.

INTRODUCTION AND MOTIVATION

Rhenium

- Discovered in platinum ores by Noddack, Tacke and Beer in 1925.
  - origin from the Greek word "*Rhenus*" meaning river "*Rhine*"
  - does not occur free in nature or as a compound or mineral
  - widely spread throughout the earth's crust at 0.001 ppm
  - the element is silvery-white with a metallic luster
  - annealed rhenium is very ductile and can be bent coiled or rolled
- Physical Properties
  - Atomic weight – 186.2
  - m.p. 3180 C
  - Density 21.02 g/cc
- Isotopes
  - ^{185}Re 37.40%
  - ^{187}Re 62.60%
  - available as highly enriched metal powder

INTRODUCTION AND MOTIVATION

Iridium

- Discovered in 1803 by Tennant in residue left from Pt dissolved in aqua regia.
  - name comes from its salts which are highly colored
  - occurs uncombined in nature with Pt in alluvial deposits
  - the element is white, similar to Pt but with a slight yellowish cast
  - hard and brittle making it difficult to machine, form or work
  - most corrosion resistant metal known
- Physical Properties
  - Atomic weight – 192.22
  - m.p. 2410 C
  - Density 22.42 g/cc
- Isotopes
  - ^{191}Ir   37.3%
  - ^{193}Ir   62.7%
  - available as highly enriched metal powder

DESCRIPTION OF THE GRAPHENE LOADING TECHNIQUE

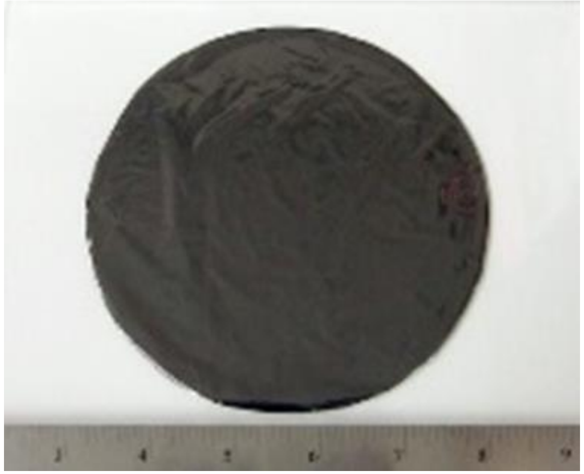
Applied Nanotech, Inc.

- Located in Austin, Texas
- Founded 1988, publicly traded
- In 2014, merged with Nanofilm (Cleveland, OH), both are now subsidiaries of PEN Inc.
- Nanotechnology R&D emphasis:
  1. Graphene foils and films
  2. Printed Electronics
  3. Sensors
  4. Thermal Management
- PEN has staff of 20+ employees, ANI has 6 staff

DESCRIPTION OF THE GRAPHENE LOADING TECHNIQUE

Prior ANI Effort Using Graphene

- Graphene Stripper Foils for FRIB
  - DoE grant DE-SC0000852: The goal of this program was to develop a high thermal conductivity, rigid, large area, uniform graphene foil for charge stripping in accelerators, capable of having long lifetime
  - Fabricated by filtration of reduced GO. Size up to 5”, typical thickness 0.1 to 1.0 mg/cm^2, can be cut per order
  - Longer lifetime (up to 2x) in charge stripping applications compared to conventional carbon foils



Finished Graphene Foil

H. Chen, M. B. Müller, K. J. Gilmore, G. G. Wallace, D. Li, Adv. Mater. **20**, 3557 (2008)

I. Pavlovsky, R.L. Fink. J. Vac. Sci. Technol. B **30**(3), May/Jun 2012, 03D106-1

DESCRIPTION OF THE GRAPHENE LOADING TECHNIQUE

Prior ANI Effort Using Graphene

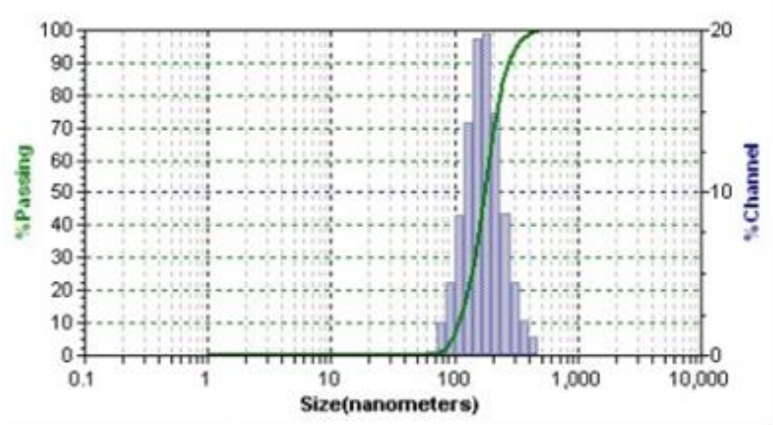
- Isotopic Carbon Graphene Foil Targets
  - DoE grant DE-SC0015140: The goal of this program is to develop ^{13}C and ^{14}C carbon targets for nuclear experiments
  - Fabricated ^{nat}C graphene targets, while commercially available ^{13}C did not graphitize
  - Low risk approach for ^{14}C that has high levels of impurities is to print 3-5 mg/cm^2 films using a binder (2% wt.)
  - Test run complete at ANL in June 2018

Removal of Al Backing

"Isotopic Carbon Targets" & "Isotopic Targets with Graphene Backing"
Presented at CAARI2018, August 12-17, 2018 – submitted for publication

DESCRIPTION OF THE GRAPHENE LOADING TECHNIQUE

Iridium Loaded Graphene Foils

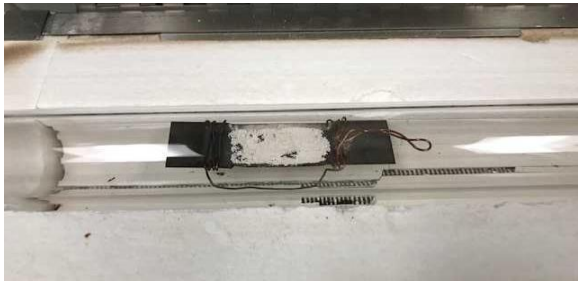


- We fabricated iridium nanoparticles dispersions in ethylene glycol by reduction of hexachloroiridate ($H_2IrCl_6*H_2O$) at elevated temperatures up to 150 C. The average particle size that we obtained was in the range of 170 nm to 600 nm and the distribution was determined using a Nanotrac particle analyzer. The particles were dispersed in DI water to obtain 10 mg/ml Ir stock dispersion.
- The ^{nat}Ir nanoparticle dispersions were added and mixed with the prepared graphene dispersions before the final filtration. To ensure that the Ir loaded graphene targets kept good mechanical integrity under the beam, a mass loading of Ir of 1:1 over graphene was not exceeded (higher Ir loadings might interfere with van der Waals attraction forces between graphene crystallites). Area densities of the obtained films were in the range of 0.11 to 0.5 mg/cm2. Several 40x40 mm^2 rectangular ^{nat}Ir targets were made using a 125 mm pressure filtration system.

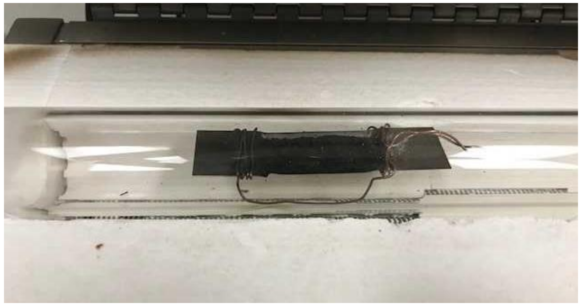
DESCRIPTION OF THE GRAPHENE LOADING TECHNIQUE

Rhenium Loaded Graphene Foils

- To prepare Re nanoparticles, we reduced ammonium perrhenate (APR) powder in a tube furnace. APR was purchased from Sigma Aldrich. Approximately 0.5 g APR powder was spread uniformly over 10 cm^2 tungsten boat. The boat was then moved into the center of the tube furnace, and 100 sccm N_2 flow was used to purge the tube for 20 min. Oven temperature was ramped at 10 °C/min to 320 °C, and then held at this temperature for 1 hr. Dark gray ReO_2 was formed in the boat. Then, 50 sccm H_2 was introduced to further reduce rhenium oxide to metal powder at the same temperature for another 2 hrs. The aqueous powder dispersion was then processed in a SpeedMixer and sonicated.



Ammonium Perrhenate

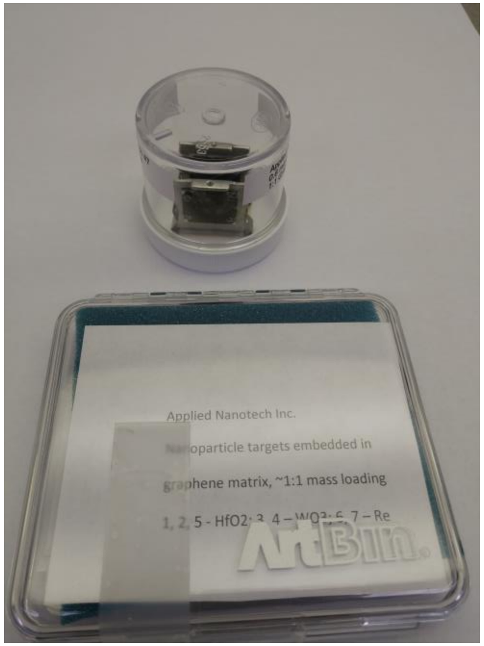


Decomposition to Rhenium Oxide

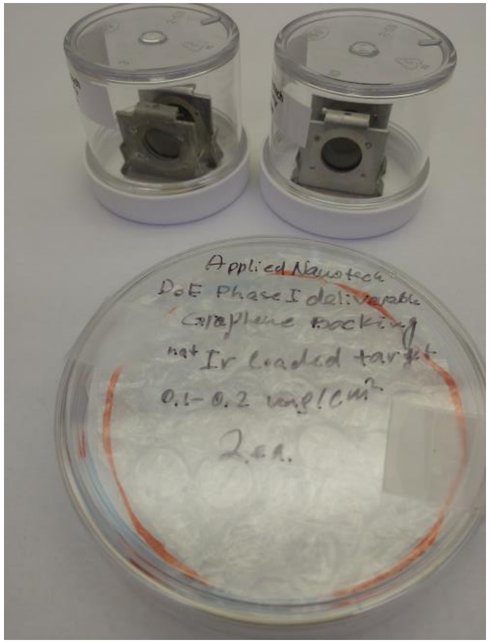
Rhenium Nanoparticles

EXPERIMENTAL RESULTS AND DISCUSSION

Rhenium and Iridium Targets



TARGET	THICKNESS (mg/cm2)	No. of TARGETS
^{nat}Re	0.6	2
^{nat}Ir	0.2	4

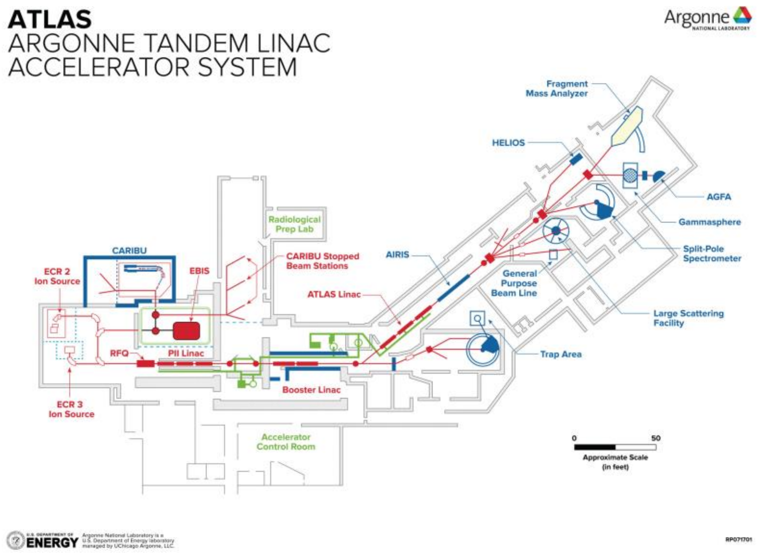


- The Ir targets have loading of 1:1 with respect to the mass of graphene, where the areal mass density of graphene was in the range of 0.1 to 0.2 mg/cm^2. Rectangular samples were cut from the 125 mm disk samples.
- Natural rhenium targets were produced the same way as Ir targets, using a 1 inch diam stainless steel pressure filtration system.

EXPERIMENTAL RESULTS AND DISCUSSION

Proposed ATLAS Experiment

- We propose to irradiate these novel rhenium and iridium targets using the beam time to evaluate ^{186}Ir and ^{194}Au excitation functions and production yields via the $^{nat}Re(\alpha, 2n)^{186}Ir$ and $^{nat}Ir(\alpha, 3n)^{194}Au$ reactions. The method should enable the measurement of the excitation functions of these reactions using single targets of each element. This approach is only achievable at ATLAS combining its unique beam capabilities with Gammasphere.

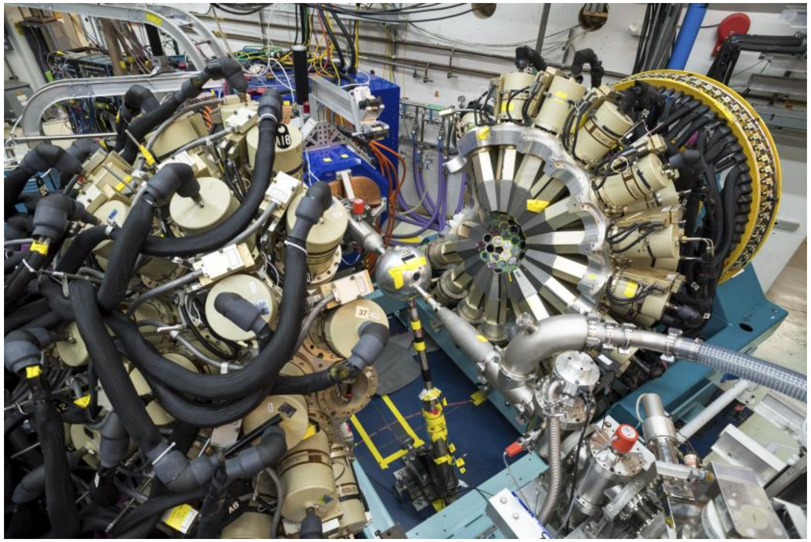


- We will measure at three alpha particle energies (38 MeV, 35 MeV, and 32 MeV). The beam will be tuned and the target irradiated and counted for 1 hr then counted offline for 1 hr while the beam is adjusted to the next energy. Once all 3 measurements have been made, the target will be counted overnight and repeated with the next target put in place. These adjustments to the beam energies can be achieved by scaling down the resonators so that a complete retuning would not be required.

EXPERIMENTAL RESULTS AND DISCUSSION

GAMMASPHERE Measurement

- We propose 2 days of beam time to evaluate ^{186}Ir and ^{194}Au excitation functions and production yields using single targets of each element. Prompt gamma rays will be measured in-beam and decay gamma rays will be measured offline in-place using the high-precision gamma-ray spectroscopy capabilities of Gammasphere.

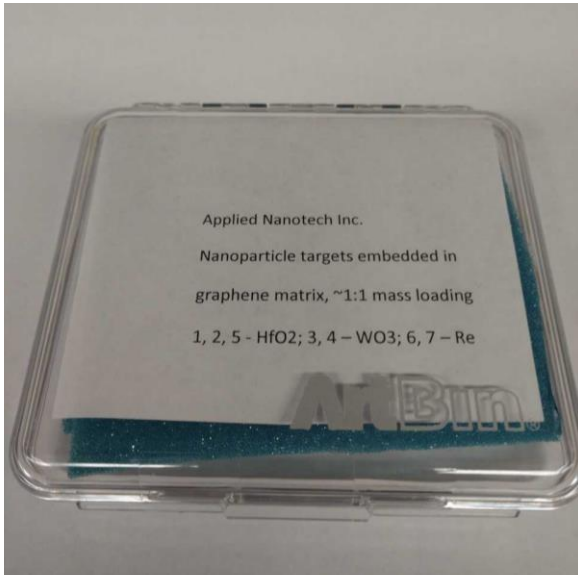


- The reaction products will be stopped inside the target, which can be used for in-beam and off-line activity measurements where the beam current, collected in a Faraday cup, can be continuously monitored. In order to quantify the absolute efficiency for Gammasphere, a calibrated standard source will be placed with the target and measured at the same time. The gamma intensities from Gammasphere can be used to precisely calculate the production yields and determine the reaction cross-sections at each beam energy. Offline decay measurements enable validation of production rates and provide a baseline count for further irradiations.

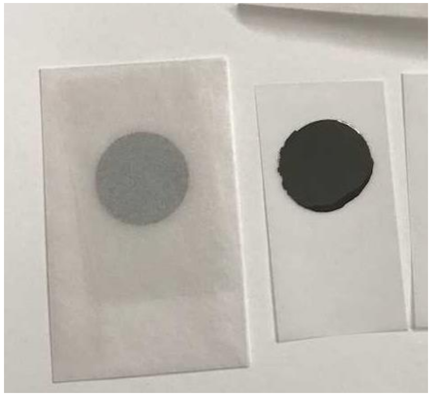
EXPERIMENTAL RESULTS AND DISCUSSION

Further Development

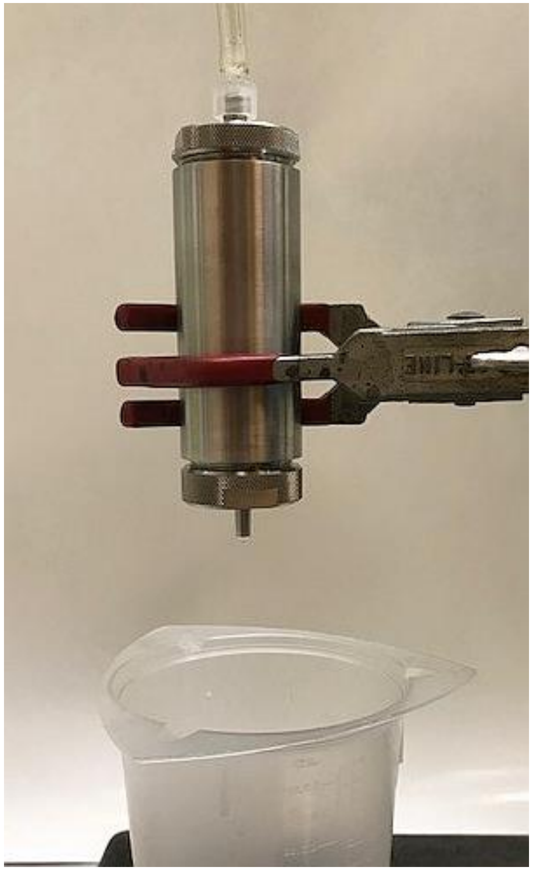
- This process was repeated to form $^{nat}WO_3$, and $^{nat}HfO_2$ nanoparticle-loaded graphene targets for use in nuclear physics research. The $^{nat}WO_3$, and $^{nat}HfO_2$ nanoparticle were obtained from commercial sources.



Hf, W and Re Graphene Foils



Target Foils



Pressure Filtration System

CONCLUSION

- IN CONCLUSION, RHENIUM AND IRIDIUM LOADED GRAPHENE TARGETS HAVE BEEN DEVELOPED AS AN ALTERNATIVE APPROACH TO PREPARING THESE REFRACTORY ELEMENTS.
- TESTING THESE TARGETS IN BEAM WILL COMMENCE SOON.
- IF SUCCESSFUL, THESE TARGETS ARE AVAILABLE FOR MEDICAL ISOTOPE PRODUCTION AND FOR LOW-ENERGY NUCLEAR PHYSICS RESEARCH.
- THE APPLICABILITY OF THIS METHOD TO OTHER REFRACTORY ELEMENTS AND COMPOUNDS IS UNDER INVESTIGATION.

Argonne NATIONAL LABORATORY

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