Closeout Report

Rare Isotope Generators at the NSCL and FRIB

PI: Gregory Severin

Institution: Michigan State University

Findings:

At the start of the Rare Isotope Generators at the NSCL and FRIB project, the isotope harvesting experimental program had already been very successful at recovering implanted radioisotopes from static-water collectors. Our task was to begin testing isotope harvesting in environments that more closely resembled the conditions at FRIB. This meant moving to higher beam power and transitioning from harvesting the isotopes produced in-flight to those produced in a flowing-water beam dump. To achieve this, students Paige Abel, and Hannah Clause, and I created a flowing-water target that would allow us to harvest radionuclides produced from a ^{40}Ca "primary beam" as it interacted with the water [1].

In the experiment, we observed the production of various radioisotopes and were able to recover them from the water with simple anion and cation exchange columns. Prior to the irradiation, we predicted we would find that the radionuclides would exist in their highest water-stable oxidations states. This assertion was supported in the experiment by the behavior of ^{48}V and ^{48}Cr, acting as high-valent oxo-anions, and accumulating strongly on the anion exchange resin bed. Further, the non-specific accumulation of the isotopes of Sc demonstrated that hydrolysable metals would be difficult to isolate cleanly. Our other important observations were that the radiolytic production of H_2 gas was suppressed from an escape-yield based prediction; fusion-evaporation reactions with ^{16}O at the end of the beam tracks were non-negligible when predicting production yields; [^{11}C]CO_2 will evolve from heavy-ion irradiated water if it is not sufficiently degassed of atmospheric CO_2; and the titanium alloy that is planned for use in the FRIB beam dump does not experience rapid deterioration at the beam-water interface during low-power irradiation.

Following on the ^{40}Ca experiment, we began designing a flowing-water target that could accommodate the highest available beam powers from NSCL. We called this the "harvesting beam blocker", and it was built to mimic the FRIB beam dump in its material properties. The blocker was fabricated via additive manufacturing with grade 5 titanium alloy, which is known for its corrosion resistance. Additive manufacturing allowed the design to incorporate a thin (590 µm) entrance window and similar internal baffle, all in one continuous piece without seals, o-rings, or screws. In order to validate the fabrication we used a preclinical CT scanner, and were able to confirm that the internal geometry of the blocker was correct.

While building the beam blocker, we also created a robust water system including remotely operable controls and data acquisition. The water system could circulate water at 10-20 L/min through the blocker, while simultaneously passing 500 mL/min through cation and anion exchange resins for harvesting. It also included a sealed reservoir with an active purge gas that allowed us to completely degas the water and to send gas-phase radionuclides to a cold trap for harvesting [2].

The harvesting beam blocker was used in several experiments, one of which was to collect and purify ^{47}Ca that was created during ^{48}Ca irradiation. ^{47}Ca is useful as a generator parent for the therapeutic radionuclide ^{47}Sc. This experiment allowed us to try harvesting at higher power than before, providing more information on the radiolysis rates and isotope production rates during bombardment. Importantly, we were able to pull over 95% of the ^{47}Ca produced in the irradiation from the water, and successfully made a radionuclidicly pure ^{47}Sc source from generation. This was a particularly valuable finding because many other scandium isotopes are co-produced in the water; without the generator route to production of ^{47}Sc, it would not have been possible to achieve high radionuclidic purity. Also, as was found in the ^{40}Ca irradiation, hydrolysable metals are difficult to harvest, but in this case, the generator product was easy to obtain, and was viable for radiolabeling DTPA-TOC, a somatostatin receptor targeting peptide that is used in targeted radiotherapy [5].

As an example of application of a harvested isotope, for [6] we employed harvested ^{47}Ca in a nuclear data measurement. Prior to our work, there was a 15% absolute uncertainty in the ground state beta decay branching ratio in ^{47}Ca decay, which, when propagated to the gamma-ray branching ratios, made determining the absolute amount of ^{47}Ca in a sample very imprecise. Since we had isolated pure samples of ^{47}Ca by isotope harvesting, we were able to use the ingrowth of ^{47}Sc (which has a well-known half-life) to quantify ^{47}Ca independently from its gamma-emission spectrum. This allowed us to determine the gamma-ray branching ratios through measurement with a calibrated HPGe spectrometer, and then to use those results to compute the beta-decay branches. We were able to improve the relative precisions to less than 1% for the three highest intensity gamma-ray branches and the highest intensity beta-decay branch and improved the precision of the absolute ground state beta branching ratio to 0.5% [6].

Another application for harvested material appears in [4]. In this study, our group was able to examine the relative uptake of Zn(II) in cress culture with and without the presence of the chelator DTPA by radiotracing with harvested ^{62}Zn. The findings were not groundbreaking but nevertheless illustrated the proof of concept that harvested materials could have an impact on horticultural research. Since this work, we have deepened our interaction with the MSU Plant and Soil Sciences Department and are preparing to support our collaborators with radiotracing studies to examine the role of the soil microbiome in trace nutrient uptake in plants.

From the point of view of development of the isotope harvesting project, this experiment showed that we were capable of harvesting from the irradiated water and from its offgas simultaneously (we were also collecting ^{76}Kr and ^{77}Kr. A manuscript describing that work is currently under review for publication). This is a key point because it truly highlights that the isotope harvesting project makes efficient use of dumped beams by enabling multiple experiments at once. In fact, during the same chemical processing, we were also able to isolate pure ^{22}Na and pure ^{7}Be, implying that our process is robust, and does not require sacrifice of one product in favor of another.

Output:

- 2 PhD theses:
    - Abel, Emily Paige, “Isotope Harvesting of Aqueous Phase Ions from Heavy-Ion Fragmentation Facilities for the Production of a 47Ca/47Sc Generator”, Michigan State University, 2020.

    - Clause, Hannah K., “Demonstrating Gas-Phase Harvesting Capabilities at the NSCL through the Production and Collection of ^{76}Kr and ^{77}Kr”, Michigan State University, 2021.
- 6 published manuscripts:
    1. Abel EP, Clause HK, Severin GW. Radiolysis and radionuclide production in a flowing-water target during fast 40Ca20+ irradiation. Appl Radiat Isot 2020;158:109049. https://doi.org/10.1016/j.apradiso.2020.109049
    2. Domnanich KA, Abel EP, Clause HK, Kalman C, Walker W, Severin GW. An isotope harvesting beam blocker for the National Superconducting Cyclotron Laboratory. Nucl Instruments Methods Phys Res Sect A Accel Spectrometers, Detect Assoc Equip 2020;959:163526. https://doi.org/10.1016/j.nima.2020.163526
    3. Abel EP, Domnanich K, Kalman C, Walker W, Engle JW, Barnhart TE, et al. Durability test of a flowing-water target for isotope harvesting. Nucl Instruments Methods Phys Res Sect B Beam Interact with Mater Atoms 2020;478:34–45. https://doi.org/10.1016/j.nimb.2020.05.011
    4. Domnanich KA, Vyas CK, Abel EP, Kalman C, Walker W, Severin GW. Harvesting 62Zn from an aqueous cocktail at the NSCL. New J Chem 2020;44:20861–70. https://doi.org/10.1039/d0nj04411c
    5. Abel EP, Domnanich K, Clause HK, Kalman C, Walker W, Shusterman JA, et al. Production, Collection, and Purification of 47 Ca for the Generation of 47 Sc through Isotope Harvesting at the National Superconducting Cyclotron Laboratory . ACS Omega 2020. https://doi.org/10.1021/acsomega.0c03020
    6. Abel EP, Kleinfeldt C, Kalman M, Severin GW; Branching ratios for the three most intense gamma rays in the decay of ^{47}Ca. Applied Radiation and Isotopes, 179, 109994, 2022. https://doi.org/10.1016/j.apradiso.2021.109994
- 1 manuscript under review
- 1 manuscript in preparation

Milestones

- *MS1: NBB and purification system operating in non-irradiation setting*
    - Forecast- FY18 Q4, completion: FY18 Q4
    - Achieved
- *MS2: first full collection f isotopes under 48Ca beam (goal:^{47}Ca)*
    - Forecast- FY19 Q1, completion: FY19 Q1
    - Achieved
- *MS3: first full collection of isotopes under ^{78}Kr beam (goal: ^{76}Kr)*
    - Forecast: FY19 Q3, Completed FY19 Q3
    - Achieved
- *MS4: first generator ^{47}Sc- demonstrated the radionuclidic purity of ^{47}Sc produced from ^{47}Ca*
    - Forecast: FY19 Q3, completion: FY19 Q1
    - Achieved

- *MS5: first generator ^{76}Br- demonstrated the radionuclidic purity of ^{76}Br produced from ^{76}Kr*
    - Forecast: FY19 Q4, completion: FY19 Q3
    - Achieved
- MS6 radiolabeling with 47Sc- demonstrated the chemical reactivity of generated 47Sc with a generic chelator-protein pair
    - Forecast: FY 20 Q1 Complete: FY 20 Q1
    - Achieved
- MS7 radiolabeling 76Br- demonstrate the chemical reactivity of generated 76Br with a generic protein
    - Forecast: FY 20 Q2 New Forecast: Canceled due to accelerator closure
    - Not Possible Due to COVID 19
- MS8 SOPs written describing the harvesting process for obtaining radionuclidicly pure and radiochemically viable 47Sc & 76Br
    - Forecast: FY 20 Q3 Complete: FY21 Q2 (^{47}Sc and ^{76}Br harvesting procedures are documented and awaiting publication)
    - Achieved