# Isotope Harvesting at Heavy Ion Fragmentation Facilities

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## Introduction

The National Superconducting Cyclotron Laboratory (NSCL) is a national nuclear physics facility in which heavy ion beams are fragmented to produce exotic nuclei. In this process of fragmentation many nuclei are created, however, only one isotope is selected for experimentation. The remaining isotopes that are created go unused. The future upgrade of the NSCL to the Facility for Rare Isotope Beams (FRIB) will increase the incident energy of these heavy ion beams and amplify the current by three orders of magnitude. An aqueous beam dump will be created to collect the unused isotopes created in the process of fragmentation. Several of these isotopes are of interest for many applications including nuclear security, medical imaging, and therapy and are not currently available or are only available in very limited supply. Harvesting these isotopes from the aqueous beam dump could provide a consistent supply of these important isotopes as an ancillary service to the existing experimental program.

## Material and Methods

A liquid water target system was designed and tested to serve as a mock beam dump for experiments at the NSCL[1]. A 25 pnA 130 MeV/u $^{76}$Ge beam was fragmented using a 493 mg/cm$^2$ thick beryllium production target. After fragmentation the beam was separated using the A1900 fragment separator[2] set up for maximum $^{67}$Cu production using a 240 mg/cm$^2$ aluminum wedge and a 2% momentum acceptance. The secondary beam was collected for four hours in the liquid water target system before being transferred to a collection vessel. Four additional four hour collections were made before finally shipping the five collections to Washington University and Hope College for chemical separation.

Four of the five samples were separated using a two part separation scheme. First they were passed through and 3M Empore iminodiacetic acid functionalized chelation disk in a 1.25M ammonium acetate solution at pH 5. The flow through was collected and analyzed using an HPGe detector. Then 10mL of 6M HCl acid was passed through the chelation disk to remove the $2^+$ transition metals. The 10mL of 6M HCl acid was collected after passing through the disk and added to an anion-exchange column with 2.5 g AG1-X8 resin. The eluate was collected and then an additional 10mL of 6M HCl was passed through the column to remove the nickel. The $^{67}$Cu was then collected by passing 10mL of 0.5M HCl and the eluate was collected in 1mL fractions each analyzed by HPGe for $^{67}$Cu concentration and purity. The two highest $^{67}$Cu fractions were heated to dryness and reconstituted in 50 µL 0.1M ammonium acetate pH 5.5.

2 µL of 7.9 mg/mL NOTA-Bz-Trastuzumab was added to 45 µL of $^{67}$Cu and 3 µL 0.1M ammonium acetate pH 5.5. This solution was placed in a shaking incubator at 37 °C for twenty minutes and then analyzed by radio-instant thin layer chromatography in order to determine the percent of $^{67}$Cu bound to the antibody.

## Results and Conclusion

$^{67}$Cu was collected into the liquid water target system with an average efficiency of 85 ± 5 %. The secondary beam was 73 % pure with the impurities, half-lives greater than 1 minute, listed in TABLE 1.

| *Nuclide* | *Half-life* | *pps* |
|---|---|---|
| $^{66}$Ni | 2.28 d | 4.39E3 |
| $^{65}$Ni | 2.52 h | 1.82E6 |
| $^{70}$Ga | 21.14 min | 7.60E3 |
| $^{66}$Cu | 5.12 min | 9.59E5 |
| $^{62}$Fe | 68 s | 2.79E3 |
| $^{68}$Zn | stable | 5.12E6 |

TABLE 1. Impurities in the secondary beam with half-lives greater than one minute

Separation of $^{67}$Cu from the impurities resulted in an average recovery of 88 ± 3 % for a total recovery of $^{67}$Cu from the beam and separation of 75 ± 4 %. No detectable radioactive impurities were found in the final samples when analyzed using an HPGe detector. TABLE 2 shows the amount of $^{67}$Cu collected from the beam and the amount recovered decay corrected to end of

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bombardment. Labeling NOTA-Bz-Trastuzumab with $^{67}$Cu resulted in > 95 % radiochemical yield.

| *Run* | *Activity in Beam (MBq)* | *Activity Collected (MBq)* | *Activity Recovered (MBq)* |
|---|---|---|---|
| 1 | 518 (33) | 458 (9) | N/A |
| 2 | 517 (32) | 452 (9) | 382 (8) |
| 3 | 492 (30) | 425 (8) | 371 (7) |
| 4 | 482 (29) | 403 (8) | 339 (7) |
| 5 | 492 (31) | 395 (8) | N/A |

TABLE 2. Activity in the beam, collected in the water, and recovered after separation chemistry (all values decay corrected to end of bombardment).

Collection of the 73 % pure $^{67}$Cu beam in water and the resulting separation proved successful. These results demonstrate that radioisotopes can be collected from fragmented heavy ion beams and isolated in usable quantities and purity for many radiochemical applications. Further experimentation with an unpurified beam to better simulate conditions in the beam dump at the Facility for Rare Isotope Beams will be performed in the near future.

**References**


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