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Design and construction of a water target system for harvesting radioisotopes at the National Superconducting Cyclotron Laboratory

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ABSTRACT

A liquid water target system for harvesting radioisotopes at the National Superconducting Cyclotron Laboratory (NSCL) was designed and constructed as the initial step in proof-of-principle experiments to harvest useful radioisotopes from the Facility for Rare Isotope Beams (FRIB). FRIB will be a new national user facility for nuclear science, to be completed in 2020, at which radioisotopes will be collected synergistically from the water in cooling-loops for the primary beam dump that cycle the water at flow rates in excess of hundreds of gallons per minute. As part of the development of radiochemical expertise required to harvest long-lived radioisotopes of interest in this environment, the water target system described here was constructed and successfully used to collect a test beam of relativistic ^{24}Na ions produced at the NSCL. Future studies will involve collecting interesting transition metal isotopes such as ^{67}Cu from less purified secondary projectile fragment beams.

1. Introduction

The Facility for Rare Isotope Beams (FRIB) is a new national user facility being constructed at Michigan State University (MSU) with funding from the Department of Energy, Office of Sciences (DOE-SC). FRIB is presently scheduled for completion in 2020 and will have the ability to accelerate uranium to 200 MeV/nucleon and light ions to slightly higher energies at intensities up to 400 kW to produce an extremely broad range of rare radioisotopes. In an important advance, FRIB will provide these accelerated beams at intensities several orders or magnitude higher than are presently available, thus producing the most intense beams of these rare isotopes [1]. Since the production process at FRIB is a projectile fragmentation reaction followed by online separation through magnetic analysis, there will be a host of auxiliary radioisotopes produced in the primary target that could be harvested for off-line use in addition to the radioisotope of interest delivered to the primary user. The majority of these auxiliary radioisotopes will be stopped in the cooling water in FRIB's primary beam dump, a spinning titanium container with cooling water circulating at ~300 gal/min to dissipate the large heat load and provide a repository for the storage and disposal of unused radioisotopes. The technology to pass large volumes of cooling water through ion-exchange columns to remove radioisotopes has been well-developed in the nuclear power industry [2]. However, the ability to conduct additional synergistic radiochemical separation and harvesting of certain long-lived radioisotopes from the cooling water of the primary beam dump has also been designed into FRIB [3]. Many of the auxiliary radioisotopes created during routine FRIB operation have useful applications in a wide range of fields, ranging from nuclear medicine to stockpile stewardship [4–11]. Some of these radioisotopes cannot be easily produced by other methods (e.g., nuclear reactors and medical cyclotrons), or in the high quantities that are predicted with FRIB's high beam intensities. Developing aqueous extraction and separation methods for the synergistic harvesting of specific radioisotopes from FRIB's beam dump will be beneficial to many isotope users without interfering with the primary users' experiments.

In order to investigate the feasibility of harvesting useful quantities of isotopes in high purity, it is necessary to demonstrate the isolation of a specific isotope deposited in the water from hundreds of different radioisotopes also contained in the same

Abbreviations: NSCL, National Superconducting Cyclotron Laboratory; FRIB, Facility for Rare Isotope Beams

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aqueous sample. The first step in this demonstration is to use the current projectile fragment separator, the A1900 [12], at the National Superconducting Cyclotron Laboratory (NSCL), to produce analyzed, high-energy beams of long-lived radioisotopes (such as ^{24}Na, ^{67}Cu, ^{48}V, ^{85}Kr, ^{44}Ti, and ^{32}Si) at measured rates, stop them in an aqueous target, and then extract them. The efficiency with which these isotopes can be extracted from water can be measured, optimized and compared to the direct measurement of the production rate in the A1900 separator (see below). Subsequent experiments can be performed in which the secondary beam is not purified and thus contains a mixture of radioisotopes and chemical separations will be critical to providing a pure radioisotope. The A1900 projectile fragment separator has been operated for more than ten years [12] and currently forms the basis of a far ranging nuclear science program using purified beams of projectile fragments [13]. FRIB will use the same techniques with a next-generation accelerator with beam intensities up to 100 times those presently available and a new fragment separator [1,3]. Detailed, tested simulations of the operation of projectile fragment separators are available [14], including systematic predictions of the cross-sections [15], that allow the measured extraction efficiencies from the preliminary work to be scaled to the projected secondary beam intensities from FRIB and predict the yields of radioisotopes that could be harvested at the new facility.

Radioisotope harvesting of ^{13}N has been conducted in the past from the interaction of a fast ^{14}N primary beam with water [16], but harvesting experiments with heavier beams now available at the NSCL have yet to be conducted. In the previous case only a few radioactive products could be produced in the water and ^{13}N was found to be the predominant activity. A key difference in the present work is that the unreacted primary beam is dumped in the A1900 separator and only the secondary beam is stopped in the water. Additionally, while other long-lived isotopes have been extracted from activated materials or beam dumps at high-energy accelerators [17–19], the speed with which water extraction can be performed during normal FRIB operation suggests that many additional isotopes are potential harvesting candidates. Thus, the development of a water-based target system for harvesting radioisotopes at the NSCL is the initial step for the proof-of-principle experiments to harvest useful radioisotopes from FRIB and is reported here.

2. Materials and methods

2.1. End station design

In order to collect and isolate specific radioisotopes of interest from a cocktail of other isotopes produced at the NSCL, a complete, remote-controlled, liquid water target system was designed, constructed and tested with a radioactive beam. The following features were included in the final system:

The system had to be completely automated, remote-controlled and operated from outside the experimental vault during beam irradiation for radiation safety reasons.

This system had to be capable of performing several remote water collection/storage cycles before being recharged in order to conduct systematic studies with various beam conditions (six in the present case). For example, the purity or intensity of the analyzed beam could be changed and deposited into separate water volumes. Thus, a water manifold system to move volumes of water to/from the target cell into separate collection vessels was necessary.

During the previous light-ion extraction experiments [16], radiolytic production of radioactive gases was observed, thus the target water cell was designed to include a gas manifold system to capture off-gases and to purge the target water with helium to remove dissolved gases before irradiation. In addition the target system was designed to be physically isolated from the accelerator vacuum system by passing the high-energy beam through an air gap before reaching the water target.

These features were realized in a stand-alone unit with a main enclosure housing the hardware and plumbing and a second enclosure housing the electronics, all controlled remotely over ethernet. There are three main components to the stand-alone unit: first, a water-holding reservoir in which aliquots of purified water were held in individual commercially-available high density polyethylene (HDPE) containers. There are six 175 mL conical HDPE bottles (Nalgene, 3143-0175) in the water reservoir system, each filled with 100 mL of distilled water (Sigma-Aldrich, 7732-18-5) prior to beam collection. The water in each reservoir was sparged with helium for ~30 min prior to irradiation. A flow sensor (Omron Electronics Inc. – EMC Div, D6F-P001A1) was used to ensure that the helium, regulated by an electronic valve (SMC, ITV1010-21N1BS4), was properly flowing during sparging and when pushing the water though the manifold system to/from the target cell. A pressure sensor (Honeywell Sensing and Control, SSCSANN001BGAA5) was used to monitor the irradiation cell for gases produced during beam collection. Second, a custom irradiation cell was built, into which water was transferred from the holding reservoir, for stopping and collection of the beam particles. The target cell had a thin entrance window (8 μm Kapton) and was constructed from polytetrafluoroethylene (PTFE). The internal volume of the irradiation cell took the shape of a truncated cone for containing any divergence of the stopping secondary beam, see Fig. 1. Third, a movable carrousel of commercially available PTFE bottles was included for the transfer and temporary storage of the irradiated water samples. After irradiation, the water was transferred to a collection bottle via a tube supported on a pneumatic linear actuator (SMC, CDJ2KB16-45-B) that extended into the bottle to capture any spraying during liquid transfer operations. The actuator was retracted before movement of the carrousel to the next collection bottle. Two magnetic sensors (Cherry, MP201802) on the linear actuator were used to confirm that the actuator rod was in the up position before rotating the carrousel or that it was the down position and the tubing was in the bottle, ready for dispensing water from the target cell. There were ten PTFE collection/storage bottles (Cole-Palmer, 015.150.2) in the 35.6 cm diameter carrousel for post-irradiation retrieval of the water. A stepper motor (Lin Engineering, 85BYGH450A-08), controlled by a stepper motor drive (Lin Engineering, CW250), was used to rotate the carrousel and position the collection bottles as necessary. Four photo sensors (Sharp, GP1A57HRJ00F) viewed notches cut into the carrousel plate to identify each of the ten bottle positions. A fifth photo sensor was used to center each bottle prior to reading the position identification and a capacitive sensor (Carlo Gavazzi, CD50CNF06NO) was used to determine if the collection bottle contained water thus ensuring that a new sample would not be delivered to a filled bottle. All components of the system that could come into contact with water were metal-free (or Teflon-coated) to reduce metal contamination and the loss of aqueous radioisotopes by plating onto metal surfaces. The end station incorporated metal-free, air-operated, normally closed, two-way valves (SMC, LVQ20-Z07N) controlled by low-voltage (5 V) solenoids (SMC, SY3A00-5U1, manifold: SS5Y3-10F2-12BS-N7D0). Fluoroethylpropylene (FEP) fittings (SMC, LQ3 Series) and Teflon tubing (SMC, TIHA07N-33) were used if they came in contact with water. A Measurement Computing I/O device (Measurement Computing, USB-1208LS, USB-SSR24, SSR-ODC-05) was used to communicate with the sensors and to control the stepper motor and electronic valve regulators for both the helium used for