LLNL-TR-645595

LAWRENCE LIVERMORE NATIONAL LABORATORY

Isotopes and Applications Working Group: Report update October 2013

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November 5, 2013

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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(a) Any changes to the scientific program—Scientific Overview

No significant changes are proposed for the scientific program. The standard mode of operation at FRIB will be to produce a rare isotope beam for a primary user, for example ^{60}Ca from a ^{82}Se beam. At the same time, the fragmentation or fission of the production beam will produce up to 1000 other isotopes that could be collected (harvested) and used for other experiments or applications. The potential applications of these harvested isotopes range from the determination of neutron cross sections for homeland security to kinetic studies of radionuclide uptake in biological processes. Longer-lived samples of the unused isotopes could be collected and used in an ion source for accelerated beam experiments at ReA3, ReA12 or other accelerator facilities outside FRIB.

Given these possibilities, the harvesting working group addresses two general areas:

- The potential uses of rare isotopes at FRIB that fall outside of basic research in nuclear physics, astrophysics, and particle physics
- The collection of selected isotopes that could be used to prepare radioactive targets or samples for experiments and allow a limited multi-user capability at FRIB.

A high-level overview of possible applications for isotopes produced at FRIB is given in the report *Scientific Opportunities with a Rare-Isotope Facility in the United States* written by the Rare Isotope Science Assessment Committee, National Research Council (National Academies Press, 2007) and in the RIA (now FRIB) Applications Workshop – see http://www.lanl.gov/orgs/t/workshop/homepage.htm. More details can be obtained from "Isotopes and Applications Working Group: Report update Feb. 2011".

The general areas of interest fall into 7 broad categories:

- Nuclear power (nuclear data is needed to optimize reactor design, safeguards applications, and for studies related to reprocessing or disposal of nuclear waste)
- Homeland security (nuclear data is needed for modeling of nuclear reactions, detection of nuclear material and other threats, and development and calibration of threat detection technologies)
- Stockpile stewardship (nuclear data is needed for modeling of nuclear reaction networks, similar to astrophysics studies, such as (n,2n), (n,γ), (n,p), and (n,f))
- Medical diagnostics (development of new imaging and treatment technologies, kinetic studies of material uptake in the body, and the possible production of biomedical radioisotopes for diagnostics and therapy)
- Nanoprobes for materials science using radioisotopes (for example the use of polarized ^{8}Li)
- Fundamental symmetries, specifically the measurement of enhanced electric dipole moments of isotopes with significant octupole collectivity. The production and harvesting of ^{225}Ra and/or $^{221/223}$Rn from the beam-dump water will produce

significantly higher rates for ^{232}Th or ^{238}U than are anticipated at TRIUMF/ISAC (for Rn) and from laboratory sources (for ^{225}Ra).

- Industrial and environmental tracers (for example ^{7}Be, ^{210}Pb, ^{137}Cs, etc.)

(b) Description of proposed experiment(s)

Harvesting of isotopes could take place at various stages in the fragment separator, shown below.

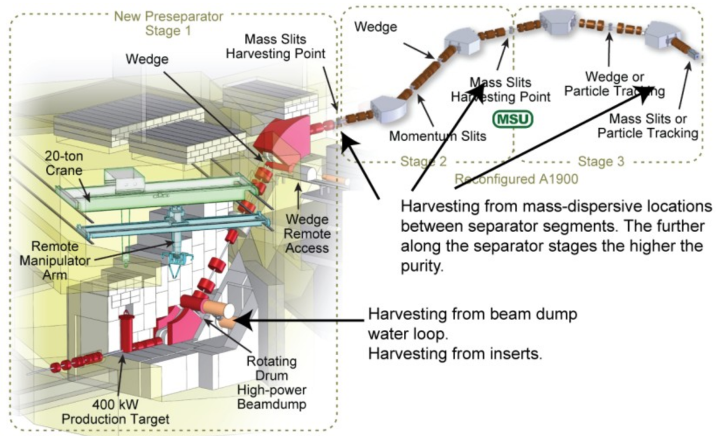

Figure 1: Layout of the FRIB fragment separator area illustrating possible locations of isotope harvesting.

While a large number of isotopes may be available in the cooling water near the production target, the ions of interest may be overwhelmed by additional species. In addition, the cooling water from the beamline magnets has been initially designed to be on the same loop as the beam dump, further complicating separation. The Isotopes Harvesting Working Group has recommended that these lines be separate.

The separation of the cooling lines may allow collection of longer-lived species if the appropriate infrastructure is in place. For example, additional extraction columns may be placed on a secondary loop streaming a subset of the cooling water volume. These columns may be configured such that they may be replaced with relative ease; the removed columns may be transferred to a hot lab to collect the trapped ions. This collection scheme may work with longer-lived species, such as ^{32}Si ($T_{1/2}$ = 160 y) and ^{44}Ti ($T_{1/2}$ = 11 y).

The collection of isotopes may also take place at the mass slits along the FRIB separator. While these collection sites have the advantage of providing much cleaner separation, the production will be dependent upon the scientific program and the specific experiment being conducted. The ions may be embedded onto catcher foils, comprised of a “sandwich” of materials, similar to how isotopes are collected at the LANL isotope production facility. To limit contaminants, the beam tuning may be conducted on standard mass slits. These slits would then be exchanged for the collection slits for the production run. The exchange of mass slits would have to occur on a short time scale to minimize interference with the science program. Appropriate engineering of the operation, exchange and handling of the mass slits will have to be explored. Isotopes with intermediate half-lives (e.g. ^{182}Ta ($T_{1/2}$ = 114 d) may be collected along the mass slits.

Isotopes may also be collected at the focus of the mass separator. These ions will have the least contaminants, and may be collected directly on a foil and used in a secondary experiment without further processing. Isotope collection in this manner is not parasitic, and will require the user to have the collection time bid for in a competitive process with experiments from the scientific program. For short-lived isotopes that are produced with high yields at FRIB, this may be the preferred collection scheme.

Finally one last idea surfaced during the group discussions at the second workshop for a potential radioisotope production site: namely using the secondary neutrons from the primary target and/or beam dump. FRIB will produce a significant flux (~10^{10} /cm^2 sec) of high energy (~200 MeV) neutrons and one could consider the possibility of using these neutrons for isotope production and/or materials testing. This will require the ability to insert and retrieve targets materials (with and without a moderator) into the neutron beam near the carbon target and/or the beam dump. It was noted that something similar to this capability is already included in the planned facility. Post irradiation the targets/test materials will require transport in shielded casks to hot cells for processing and/or analysis. These may conducted in the same facilities planned for isotope harvesting from the beam dump.

Regardless of the collection scheme, a hot cell facility must be available for isotope harvesting. A modular facility has been discussed with FRIB staff, where users will provide a portable hot cell for use during a collection campaign. Upon conclusion of the collection period, the user will remove the hot cell and assorted items. The infrastructure to support the temporary hot cells, (e.g. crane, ventilation, rails) will be provided by FRIB. Initially, two such temporary hot cells may be supported with expansion plans for up to six cells.

(c) Progress made since last report – current status

There was a second isotope harvesting/applications workshop held at MSU July 23-24, 2012. More details can be obtained from "Proceedings: Second Workshop on Harvesting Radioisotopes from FRIB", 2012.

Water cell harvesting end station was designed and constructed by Hope College and Washington Univ. St. Louis and tested at MSU.

Fig. 2: The isotope harvesting end station designed to collect isotopes from the water dump currently used at MSU.

Several successful test experiments were performed to collect ^{24}Na and ^{67}Cu from the water cell using the apparatus shown in Fig. 2. The collection efficiency of ^{24}Na from water samples sent to Hope College for quantification via gamma-ray counting was determined to be ~50-55% in preliminary runs. In Aug. 2013, tests were conducted to harvest ^{67}Cu. Some contaminations from reactions on water (^{11}C, ^{13}N, ^{7}Be) and the beam (^{68}Zn) were observed. However, separation chemistry successfully produced purified ^{67}Cu suitable for additional radioisotope test labeling chemical studies. Additional analysis is underway.

Priority isotopes were identified in the past and are shown in Table 1. A third workshop was scheduled for Oct. 14-16, 2013 at Washington Univ. St. Louis, but was postponed due to the government shutdown at the start of FY14 to a later as yet specified date. One item of discussion at the workshop is an update of the priority isotopes for harvesting. LLNL would like to add ^{88}Zr to the list to leverage developments for harvesting the medically important isotope ^{89}Zr. Measurement of the $^{88}Zr(n,\gamma)^{89}Zr$ cross-section is important to SSP. Fundamental Symmetries would like to add $^{221/223}Rn$ to the table. There may be other cases where the list needs modification.

Table 1. Priority isotopes for harvesting at FRIB. These isotopes were identified at the Working Group meeting in Santa Fe, NM September 30 – October 1, 2010.

Isotope	Half-life	Application
^{32}Si	160 y	Tracer; geology and botany
^{44}Ti	10.8 y	Medicine, astrophysics, nuclear structure
^{48}V	16 d	Stockpile Stewardship
^{67}Cu	2.6 d	Medicine
^{85}Kr	10.0 y	Astrophysics, stockpile stewardship
Eu*		Stockpile Stewardship
^{211}Rn	14.6 h	Medicine
^{225}Ra	14.9d	Medicine, Electric Dipole Moment
^{225}Ac	10.0 d	Medicine

*A range of Eu isotopes are of interest, A~147 – 154.

(d) Near term activities and required resources and funding

For each of the isotope harvesting scenarios described above, it was recommended that a single dedicated experimental radiochemical facility for harvesting, separation and characterization of the harvested material would be needed as part of FRIB to accommodate both internal as well as external users of the radioisotopes. Even for off-site usage of the harvested radioisotopes, a wet-lab facility to characterize and package the radioisotopes for transport would be needed. Several suggestions made by the FRIB staff indicated additional space adjacent to the primary target facility that might be used, depending on the size of the radiochemistry lab and facilities required. Details of the suggested space and capabilities of this facility can be found in the "Proceedings: Second Workshop on Harvesting Radioisotopes from FRIB", 2012.

Continued development of the harvesting end station is desired. Begin developments of harvesting from slits including simulations of expected production rates of targeted isotopes. Hold third isotope harvesting/applications workshop (most likely in Spring 2014).

(e) Collaboration list and organizational structure (if any):

Note: The current collaboration list is probably some subset of the attached, although there remains significant national and international interest in harvesting of isotopes.

Initial 2010 Collaborators		
Ani	Aprahamian	University of Notre Dame
Daniel	Bazin	MSU
Sebastien	Bianchin	GSI
Georg	Bollen	MSU
Todd	Bredeweg	LANL
Giovanni	Burgada	CAEN Technologies
Xinfeng	Chen	Washington University
Seonho	Choi	Seoul National University
Jolie	Cizewski	Rutgers University
Jason	Clark	ANL
Daniel	Coupland	MSU
Aaron	Couture	LANL
Yves	Dardenne	LLNL
Hugh	Evans	Eckert & Ziegler
Rafeael	Ferrer	MSU
Don	Geesaman	ANL
Amanda	Gehring	MSU
John	Greene	ANL
Marc	Hausmann	MSU
Wen Chien	Hsi	Procure Treatment Centers, Inc.
Filip	Kondev	ANL
Suzanna	Lapi	Washington University
Steve	Libby	LLNL
Sean	Liddick	MSU
Milan	Matos	Louisiana State University
Witold	Nazarewicz	University of Tennessee
Jerry	Nolen	ANL

Jorge	Pereira	MSU
David	Radford	ORNL
Andrew	Ratkiewicz	MSU
Robert	Reba	Georgetown University Hospital
Jennifer Jo	Ressler	LLNL
Lee	Riedinger	University of Tennessee
Mark	Riley	Florida State University
Ryan	Ringle	MSU
Thomas	Ruth	TRIUMF
Brad	Sherrill	MSU
Karl	Smith	MSU
Mark	Stoyer	LLNL
Hiroyuki	Takeda	RIKEN
Dave	Vieira	LANL
Kathy	Walsh	MSU
Ryan	Winkler	MSU
Additional 2011 Collaborators		
Rene	Reifarth	GSI
Roger	Henderson	LLNL
Julie	Gostic	LLNL
Graham	Peaslee	Hope College
Francois	Nortier	LANL
Tom	Ruth	TRIUMF

Workshop held July 23-24, 2012 at the Henry Conference Center, Michigan State University, E. Lansing:

List of participants:

Jill Berryman MSU
Georg Bollen MSU
Aaron Couture LANL
Heather Crawford LBNL
Cathy Cutler MURR
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Joe Devore ORNL
Scott Essenmacher Hope
Doug Gage MSU

Marc Hausman MSU
Alan Ketring MURR
Suzanne Lapi* Wash U.
Alain Lapierre MSU
Tara Mastren Wash U.
Milan Matos ORNL
David Morrissey* MSU
Meiring Nortier LANL
Graham Peaslee* Hope
Aranh Pen Hope
Dennis Phillips DOE
Reg Ronningen MSU
Brad Sherrill MSU
Suzanne Smith BNL
Dan Stracener ORNL
Andrew Wooten Wash U.
Nicholas Wozniak UNLV
Remco Zegers MSU

*Co-organizers