The Production and Acceleration of Radioactive Ion Beams at the HRIBF

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The Holifield Radioactive Ion Beam Facility (HRIBF) includes a K=100 cyclotron (ORIC) which provides high-intensity light-ions for producing radioactive atoms, and a 25 MV tandem electrostatic accelerator which is used to accelerate the radioactive-ions for nuclear structure and nuclear astrophysics research. Ion sources and targets suitable for the production of various radioactive ion beams (RIBs) have been developed. Operational experiences, problem areas, and plans for future beam development are discussed.

INTRODUCTION

The HRIBF is a first-generation Isotope-Separator-On-Line (ISOL) type radioactive ion beam (RIB) facility. In an ISOL-type facility, radioactive atoms of the desired species are produced by bombarding a suitable target with high intensity beams from one accelerator, and the radioactive atoms are then ionized, mass separated, and injected into a second accelerator to produce high quality beams for research.

A major part of our operating schedule since beginning operation in late 1996 has been devoted to restoring the light-ion acceleration capability of the cyclotron, improving the performance of the tandem at low terminal voltages and the development of target/ion-source assemblies suitable for the production of radioactive ions. Progress of the various development programs, and our operational experiences to date, are outlined in this paper.

ACCELERATOR DEVELOPMENT

The accelerator used for the production of radioactive atoms at the HRIBF is the Oak Ridge Isochronous Cyclotron (ORIC). The ORIC is a 1.5 m diameter variable energy cyclotron which began operation as a light ion accelerator in 1962. Although the main magnetic field was designed for a maximum rigidity of about 1.45 Tm, corresponding to a proton energy of 100 MeV, the maximum proton beam energy is presently limited to about 42 MeV by the tuning range of the RF system. Several accelerator improvement projects have been carried out, or are in progress, which are designed to improve the performance and reliability of the ORIC. In collaboration with the cyclotron group at Michigan State University, the central region was modified to improve the extraction efficiency and values of 85% are now routinely achieved for both proton and deuteron beams. This can be compared to typical values of 40%-50% prior to modification. Extracted beam currents are presently limited to 50 μA for protons and deuterons, and up to 200 particle-μA for other beams, by our Accelerator Safety Envelope. With the improved extraction efficiencies, these beam currents can be achieved without excessive power dissipation in, and activation of, the extraction system. At the present time, these administrative limits are not the limiting factor for the production of RIBs. The high power density produced by the relatively low beam energies available from the ORIC results in target temperature limitations which, for the RIB production targets presently in use, limit the beam current on target to about 5 μA for protons and deuterons.

Radioactive atoms produced by beams from the ORIC are ionized, mass separated and charge exchanged to produce negative ions which are then injected into the 25URC tandem electrostatic accelerator. The tandem has proven to be very reliable and economical to operate but several additions and modifications were required to meet the needs of a RIB facility. The most significant change was the replacement of the original corona-point voltage grading system with resistors to allow stable operation at lower terminal voltages. This modification now allows stable operation at terminal voltages as low as 1 MV to meet the beam energy requirements of the nuclear astrophysics research program. In addition, since the intensity of the injected radioactive beam is very low, usually below the detection level of standard Faraday cups, particle detectors have been installed at focal points inside the tandem to aid in tuning the beam through the accelerator. Similar detectors have also been installed on the external beam lines.

RIB PRODUCTION PROCESS

Radioactive atoms are produced by fusion-evaporation reactions between light ion beams from the ORIC and the target atoms chosen to produce the radioactive species of interest. Since the energies available from the ORIC are relatively low, the radioactive species accessible are those which can be produced by simple reactions such as (p,xn) or (d,xn) reactions. The cross-sections for such reactions are usually known and the production rate for a specific product atom can be readily calculated. Typical production rates for the radioactive species being studied at the HRIBF are on the order of 10^{-3} atoms per incident beam particle. Thus, for an incident proton or deuteron beam intensity of 5 μA, radioactive atoms would be produced at a rate of about 3×10^{10}/s.

The number of radioactive atoms which can be delivered

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to an experiment is, of course, much smaller than the number produced and can vary over several orders of magnitude depending on the operating conditions in the target/ion source assembly (1). The major loss mechanism for many of the radioactive species studied thus far has been the decay of the radioactive atoms during the time required to diffuse out of the target and drift to the ion source. For chemically active elements, losses will occur due to reactions between the radioactive atoms and the materials used in the target, vapor transport tube, and ionization chamber. Such elements require the formation of nonreactive molecules in order to be extracted from the target and transported to the ion source with any reasonable efficiency.

The target/ion source assembly is the 'heart' of any ISOL-type RIB facility. It must be capable of long term operation in a harsh environment of high temperature and high radiation fields while maintaining high voltage and high vacuum integrity. The design presently in use is the Electron-Beam-Plasma (EBP) positive-ion source (1) which is based on the design of the target/ion source used at ISOLDE (2).

Extensive testing is essential for any ion source prior to its use for the production of radioactive ion beams, and facilities have been built or modified for both off-line and on-line testing. The initial off-line testing of new or modified ion source designs is carried out at the Target Ion Source Test Facility (TISTF) (3). The TISTF allows convenient measurement of ion source performance over a wide range of operating parameters in a controlled, radiation-free, environment.

An ion source test stand has been constructed to assemble ion source components, check dimensional tolerances, ensure the integrity of vacuum seals and cooling water circuits, and measure electrical parameters prior to installation at either the on-line test facility or the RIB Injector. The stand also includes target heating capabilities for outgassing target materials and for efficiency and release time measurements of various radioactive species from irradiated targets (4).

For on-line testing, the UNISOR isotope separator has been modified (5) to allow target/ion source assemblies to be installed and tested with low intensity proton and deuteron beams from the tandem electrostatic accelerator. The UNISOR facility allows measurement of ion source efficiencies for the radioactive species under investigation while minimizing radiological exposure and contamination hazards.

RIB TRANSPORT/INJECTION

The target/ion source assembly is mounted on a high voltage platform, referred to as the RIB Injector, which also supports electrostatic beam optics elements, the first-stage mass-analyzing magnet, and charge-exchange cell. The high voltage platform is designed to operate at up to 300 kV for compatibility with the existing tandem accelerator stable-ion injector. The high voltage platform is presently operated at a voltage which provides 200 keV ions for injection into the tandem. A lower operating voltage was chosen to reduce leakage currents and prevent sparks since studies of the tandem showed that there was no significant difference in beam transport efficiency between 200 and 300 keV. Electronic components are mounted on a separate high-voltage platform which is radiologically shielded from the ORIC beam to prevent radiation damage to solid-state devices. The target/ion source is electrically isolated from the high-voltage platform and can be operated at voltages up to 60 kV although, at the present time, the ion source is normally operated at 40 kV.

Following extraction of the positive ion beam from the EBP ion source, the desired beam species is selected using the first-stage mass analyzing magnet which has a measured mass resolving power, $M/\Delta M$, of approximately 1,000. The beam, now consisting of a single mass but possibly several isobars, is then charge-exchanged to produce the negative ions required for injection into the tandem. At the present time, cesium vapor is used as the charge-exchange medium, but other charge-exchange media are being investigated.

A new beam line was constructed to connect the RIB Injector with the tandem accelerator. All of the focussing elements are electrostatic, and therefore mass independent, allowing initial beam tuning to be done with stable ion beams. Included in the transport line is a high resolution mass-analyzing magnet which was designed to provide a mass resolving power of 20,000 to allow for possible isobar separation. However, to achieve this level of mass resolving power requires very high beam quality. The energy spread introduced by ripple in the RIB Injector high voltage supplies has been measured and found to be less than the design goal of ±10 eV. However, measurements of the energy-loss straggling introduced by the charge-exchange process, for ^{70}Ge beams and typical charge-exchange vapor densities, gave a FWHM of approximately 42 eV. This effectively limits the mass resolution to about 1 part in 5,000 for beams in this mass region.

Ga and As Beams

A liquid germanium target was developed for the production of arsenic and gallium radioactive ion beams. The target consists of enriched ^{70}Ge contained in a graphite target cell having internal dimensions approximately 9 mm diameter by 4 mm thickness. Radioactive ion beams which have been produced by bombardment of the target with 42 MeV proton beams from ORIC are ^{69,70}As from ^{70}Ge(p,xn) reactions and ^{67}Ga from ^{70}Ge(p,α) reactions.

In the first RIB experiment performed with this target, which was carried out in May-June 1997, the ORIC beam current delivered to the RIB Injector was limited to less than 4 μA due to count rate limitations of the experimental apparatus. At a proton beam current of 4 μA, a maximum of 1.4×10^6 ions/s of ^{69}As beam was delivered to the experimental station. This corresponds to a total efficiency, defined as the number of accelerated radioactive ions delivered to the experimental station divided by the number of radioactive atoms produced, of 6×10^{-5}. The total efficiency includes the transmission through the exchange cell of 70%-80%, charge-exchange efficiency of approximately 40% for producing As$^-$ or Ga$^-$ ions, transmission through the tandem, including charge state

fractionation due to stripping in the terminal, of about 20%, and the target/ion source efficiency. The target/ion source efficiency includes losses in the target and transfer tube plus the ionization efficiency of the ion source for producing As^+ or Ga^+ ions. Measurements at UNISOR (6) have given target/ion source efficiencies up to 0.85% for the production of $^{69}As^+$, but in this experiment, the efficiency appeared to be significantly lower, about 0.11%, for reasons which have not been determined.

Initially, the mass 69 beam was dominated by ^{69}As. However, the release time for As from the molten Ge target is long (6) compared to the half-life of ^{69}As ($T_{1/2}$= 15.2 m) resulting in a buildup of ^{69}Ge and ^{69}Ga in the target and after several days of intermittent operation, the mass 69 beam was dominated by ^{69}Ga. Since the mass difference between ^{69}As and ^{69}Ga is only one part in 10^4, no significant isobar separation could be achieved. The beam line was then tuned for mass 67 and approximately 10^5 ions/s of ^{67}Ga beam was delivered to the experimental station.

A second experiment was carried out in September 1997 with the intent of increasing the ORIC beam intensity and yield of the ^{69}As beam. A beam of ^{69}As was successfully transported to the Recoil Mass Separator but the maximum beam intensity which could be achieved was only 10^5 ions/s for proton beam intensities up to about 6 μA. As the ORIC beam current was increased to 10 μA it was discovered that the yield of ^{69}As decreased rapidly. Post-run examination of the target revealed that most of the Ge had been vaporized and deposited in the vapor transport tube connecting the target and ion source apparently due to excessive temperatures induced by localized beam heating. A new target is now being designed which will incorporate baffles intended to condense and recirculate the Ge and allow operation with higher ORIC beam currents.

Fluorine Beam Development

Aluminum oxide in various physical forms has been investigated at the UNISOR test facility in an effort to develop a target suitable for the production of ^{17}F beams (6). Release measurements on solid alumina targets showed <0.1% release of ^{18}F($T_{1/2}$=1.83 h) but in similar tests of low density fibrous material, composed of 3-μm diameter fibers, >80% release of ^{18}F was observed (7) at a temperature of 1400 °C. Since fluorine is chemically active, it reacts with elements in the target and it was found that 88% of the fluorine extracted from the ion source is in the form of AlF^+ molecules. The best target/ion source efficiency obtained thus far is about 0.2% (8) and efforts are continuing to improve the yield.

In order to be injected into the tandem, the AlF^+ must first be converted to a negative ion. Charge-exchange studies (9) carried out at UNISOR indicated that little or no AlF^- is formed. However, it was determined that the AlF^+ molecule can be dissociated and F^- ions formed with a combined molecular-breakup/charge-exchange efficiency of approximately 10%. Similar charge-exchange efficiencies were observed using both Cs and Mg charge-exchange vapors.

Based on the success of the UNISOR measurements, a first high intensity study of a fibrous alumina target was carried out on the RIB Injector in March-April 1998 using a beam of 28 MeV deuterons from the ORIC. The calculated ^{17}F production rate was $3.7x10^9$/s per μA of deuterons. The maximum ion source efficiency was obtained at a deuteron beam current of about 2 μA. At higher deuteron beam currents, the total yield increased but the source efficiency decreased due, at least in part, to a rapid increase in the Al^+ beam emitted by the ion source at higher deuteron beam currents. The maximum yield of $Al^{17}F^+$ observed was $\approx 3x10^6$ ions/s at a deuteron beam current of 4 μA which corresponds to a source efficiency of about 0.02%. Further increases in the deuteron beam current resulted in reduced yields and, because of the decline in source output, no attempt was made to produce a F^- beam. Post-run examination revealed that the target material had suffered significant shrinkage and deterioration.

A second high intensity test of this target material was made in August 1998. In this test, the deuteron beam current was limited to 2 μA to avoid target degradation. The yield of $Al^{17}F^+$ was $\approx 2x10^6$ ions/s. After breakup of the AlF molecule and charge exchange in the charge-exchange cell, the yield of $^{17}F^-$ was measured to be $\approx 5x10^4$ ions/s. This gives an efficiency for molecular dissociation and charge exchange of ≈2.5%, or a factor of 4 smaller than the value obtained in the low intensity measurements at UNISOR. Attempts to transport this small amount of $^{17}F^-$ beam through the tandem were unsuccessful.

At least part of the problem encountered in transporting the fluorine beam can be attributed to the large energy spread and emittance growth introduced by the molecular breakup process. Measurements in the isobar separator of $^{19}F^-$ beams from the breakup of $Al^{19}F^+$, produced by introducing SF_6 into the ion source, gave energy widths of ≈400 eV. The energy width is also more sensitive to the density of the charge exchange vapor, presumably due to the low energy, 16.5 keV, of the ^{19}F atoms following the molecular breakup process. The emittance growth has not yet been measured.

Studies of other metal-oxides having higher working temperatures are in progress in an effort to develop targets capable of withstanding higher deuteron beam currents. A major part of the development program for these target materials is the addition of a vapor feed system to provide a reliable source of aluminum or other metals to form metal-fluoride molecules.

A negative-ion surface ionization source is also under development (10) which would avoid the problems associated with the molecular breakup and charge-exchange processes. Measurements made on a prototype negative-ion source, using SF_6 to produce $^{19}F^-$ beams, gave an ion source efficiency of about 3.5% at the TISTF without target material present, and about 1.5% at the UNISOR facility with target material present. On-line tests at UNISOR, using low intensity 22 MeV deuteron beams from the tandem, have resulted in target/ion source efficiencies as high as 0.02% for the production of $^{17}F^-$ and 0.5% for the production of $^{18}F^-$. The difference in efficiencies for $^{17,18}F^-$ and $^{19}F^-$ is presumably due to losses associated with the decay of the radioactive species due to the hold-up time in the target. A hold-up time of 16.4 minutes (6) has been

reported for release of fluorine from alumina fiber targets operated at 1470 °C in an EBP ion source, but later measurements suggest that a fast release component with a hold-up time of about 3 minutes is also present.

FUTURE DEVELOPMENT

The accelerator development plans include replacement or upgrading of the most troublesome power supplies, installation of a recirculating gas stripper in the tandem and developing ^{3,4}He beams from the ORIC. The ORIC trim coils, several sections of which are no longer operable due to water leaks, will be replaced if required for accelerating beams needed for RIB production.

In the area of RIB development, the immediate plan is to concentrate on the development of negative-ion sources to eliminate the need for charge exchange, and modification of the liquid germanium target holder to handle higher beam power. A 'batch-mode' target/ion-source is also being fabricated for use with longer lived radioactive species. This source will allow a target to be irradiated with beam from the ORIC, then rotated to a negative-ion cesium-sputter source for production of a RIB beam while a new target is being irradiated by the ORIC beam. The first tests of this source, using Ni targets for the production of a ^{56}Ni beam, are expected to be made late this year.

Low-density uranium carbide targets are being investigated for the production of neutron-rich RIBs through proton induced fission of ^{238}U. Low-intensity studies are being carried out at the UNISOR facility to measure fission fragment release from targets consisting of thin UC_2 coatings on carbon fibers.

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