PROGRESS TOWARDS NEW RIB AND HIGHER INTENSITIES AT TRIUMF*

Pierre Bricault, Marik Dombsky, Peter Kunz, Jens Lassen, Friedhelm Ames and Victoire Hanemaayer, TRIUMF, Vancouver, Canada

Abstract

Over the past five years TRIUMF has operated routinely the ISAC facility at proton beam intensity up to 100 μA. A major departure from other ISOL facilities, ISAC utilizes a modular assembly for the target station. This is mainly to provide enough radiation shielding for operation at high proton beam intensity. So far ISAC was licensed to operate target materials with Z < 82. Two actinide target (UO2) tests have been performed during the past two years to assess the ISAC systems (vacuum, nuclear ventilation, personnel safety) for actinide operation. The uranium oxide target is limited to 2 μA only because of the material low operating temperature. We are now developing a uranium carbide target using similar techniques as for our other carbide targets (SiC, TiC, ZrC) operating up to 75 μA. These developments are essential for the ARIEL (Advanced Rare IsotopE Laboratory) project for which TRIUMF just received funding. This funding includes the construction of a 500 kW, 50 MeV electron superconducting LINAC and a new target hall building capable of housing two new target stations. One of these target stations is rated at 100 kW beam power from both, electrons and protons. The proton beam line utilizes a modified extraction port on the H- 520 MeV cyclotron. The other target station will be solely dedicated to the 500 kW electron beam for photo fission.

INTRODUCTION

The TRIUMF-ISAC facility was designed for rare isotope beam production using the protons from the TRIUMF H- cyclotron as the driver beam. Short-lived radioisotopes are produced in a thick target from various nuclear reactions mechanisms. The reaction products are stopped in the bulk of the target material. We operate those targets at very high temperature to enhance the diffusion of the products inside the target matrix to the surface of the grain and then the effusion process to bring the isotopes of interest to the ion source where they are ionized and extracted to form an ion beam.

In this paper we will present the resent progress in target fabrication that can withstand high power for the production of intense RI beam. Finally, we will present the AdvancE Rare Isotope Laboratory that just received funding for installation of a new superconducting electron LINAC and the building housing the tunnel, target stations and remote handling equipment.

ISOL METHOD

Beams of rare isotopes are a challenge to produce. Especially the short-lived ones, they do not exist on earth. They have to be produced artificially in the laboratory. The isotopic separation on-line or ISOL method can be described as a process where the isotope of interest is fabricated artificially by bombarding a target material nucleus with fast projectiles. In a thick target the reaction products are stopped in the bulk of the material. The target container is attached directly or indirectly to an ion source, allowing the reaction products to be quickly ionized and turned into an ion beam that can be mass analyzed and be delivered to experiments. The requirements for producing high intensity RIB are:

- A high energy driver, such as the TRIUMF H^- 500 MeV cyclotron,
- A target material inserted into a refractory oven connected to an ion source,
- An ion source at high voltage to produce an ion beam,
- A high-resolution mass separator.

To solve the problem of producing intense rare isotope beams we need to find the best target material that favors the production of the desired RIB. One more thing to consider is contamination of the ion beam by isobars; isotopes having the same mass number, A, but different atomic number, Z.

Another consideration must be the power deposited inside the target material. If the deposited power density is too high, the temperature of the target material will increase above safe operation level and then the target material will begin to evaporate. This can have disastrous effect on the ion source efficiency, especially for plasma ion sources.

To avoid excessive power deposition by the incoming beam we do not stop the primary beam in the target. This is accomplished by choosing the target length such that the energy degradation of the proton beam is only 200 to 300 MeV. A dedicated water-cooled beam dump is located just behind the target to capture the entire proton beam emerging from the target.

There are three main nuclear reactions accessible to produce rare isotope beams at our energy range. They are:

- Spallation, a breakup or fragmentation of the target material nuclei, in which the product distribution peaks a few mass units lighter than the target nucleus. Because neutron emission is energetically easier than proton emission, (due to the Coulomb barrier that the proton experiences) the production of neutron deficient nuclei is favored. A good example is the high production of Rb isotopes from Nb or Zr target.
- Fragmentation, it is the counterpart of the spallation reaction, where the product is one of the light fragments. The fragmentation method is advantageous when producing light, neutron rich products from heavier target nuclei with high

neutron to proton ratios. Because of the neutron excess in the heavy target nuclei, the fragments tend to retain a statistical memory of the neutron to proton ratio and as a result, have higher neutron to proton ratios themselves. A good example of such a reaction is the production of ^{11}Li from Ta, or the production of ^{32}Na from U targets, respectively.

Induced fission occurs when the incoming projectile deposits sufficient energy in the target nucleus to induce a breakup into two roughly equivalent mass products. Unlike neutron-induced fission, this reaction mechanism is opened to both fissile target nuclei and to heavy nuclei such as: tantalum, lead, etc. Again because of the higher neutron to proton ratio in heavy nuclei, neutron-rich products in the medium mass region can be effectively produced by this reaction mechanism.

The yield of a specific isotope can be expressed using the following equation:

$$Y = \Phi\sigma(NA/A\cdot\tau)\varepsilon_D\varepsilon_E\varepsilon_I \quad (1)$$

where: is the nuclear reaction cross section leading to this specific isotope, ϕ_p is the primary beam flux, N_t is the number of target nuclei per square cm, and the ε represents the effusion, diffusion, ionization and transport efficiencies, respectively.

Release Processes

After the isotope has been produced by one or more of the three production reactions mentioned above, it is stopped in the target material matrix because its energy is in general not sufficient to escape the target material. In order to produce a RIB the product must diffuse from the inside of the grain or foil to the surface and then effuse out of the target container to the ion source. To avoid decay losses the release and ionization must proceed as quickly as possible. To do so we generally heat the target material to the highest operational temperature. In practice if the desired isotope has a very short half-live it is preferable to have a very short target length to limit the volume from which the atoms have to escape.

The diffusion speed or efficiency is determined by the interaction of the specific diffusing isotopes moving through the target material lattice. The higher the temperature faster is the diffusion to the surface. When using metal foils we tend to limit their thickness as much as possible to speed up the diffusion process. On the other hand we cannot decrease the thickness too much, as to conserve the heat flow toward the container tube. For carbide targets we tend to make them as porous as possible and to limit the grain sizes. To reach the optimum we are mixing three different grain sizes, 1, 7 and 20 μm in equal proportion.

In the effusion process, the product atoms are in a gaseous state and randomly bounce around the target container and the target material surface until they enter the ion source. During each collision with a surface, they stick for a time before continuing in a random direction. As with the diffusion process, a high temperature is required to put the product into the gaseous state and to minimize the sticking time in order to make the product moves as quickly as possible to the exit and then to the ion source. The sticking time is a complex function of the lattice structure, temperature and desorption enthalpy between the product atom and the surface material atoms.

Ionization Process

The ionization is required to convert the desired isotope atom into a charged particle beam. Such a beam can then be guided, mass analyzed and transported to the experimental station or be further accelerated to an energy relevant e.g. for nuclear astrophysics or nuclear structure studies.

The key of the ISOL method is to achieve highest beam intensity and the highest beam purity possible. This is accomplished by passing the extracted beam through a high-resolution magnet dipole. The separation between two isotopes of the same mass number (isobars) at the image focal plane of the dipole varies with the mass difference. The larger the mass difference, the greater is the spatial separation achieved. Using a set of slits one can select the desired isotope that will be transmitted to the experiment. Unfortunately, the mass difference for heavier elements is often not large enough to ensure complete separation at the focal plane, resulting in a mixture of isotopes delivered to the experiments. The contamination level can be quite large depending to the species involved, to a point where the level of contamination makes the experiment impossible.

To limit the contamination there are some tricks that can be applied to the system to improve the selectivity. Here are some of the methods we used at ISAC to purify the rare isotope beams:

Selectively enhance the effusion of a particular species by adding a chemical product in the target container. For example, the addition of fluorine to enhance the release of Al in its molecular species Al-F. The Al-F effusion efficiency is greater than that of Al itself. The ionized Al-F$^+$ is well separated from the otherwise dominating Na contaminant because the Na-F molecule is not stable at the operating temperature.

Selectively reduce the effusion of some contaminant by cooling the transfer tube connecting the target container and the ion source. The best example of this application is for noble gasses. We have demonstrated that by cooling the transfer tube completely removed the ^{18}F contamination from the desired ^{18}Ne beam.

Selective ionization can be accomplished by laser resonant ionization. We are using routinely two or three laser frequencies to resonantly excite the electron into a level from which ionization proceeds. Resonant laser excitation is element selective. The only drawback is that the target and transfer tube are operated at very high temperature to facilitate fast release. This is sometimes enough to obtain surface ionization isobaric contaminants

– usually alkali metals. To resolve this issue we have developed a laser ion source combined with a short, segmented radio frequency quadrupole that replaces the hot transfer tube. Hence surface ionization is inhibited. Subsequently resonant laser ionization is the only remaining ionization mechanism and then the laser only ionizes the desired element. Such created ions are captured and guided by the RFQ to the ion extraction system to form a beam.

ON-LINE HIGH POWER TARGET

As we can see from equation 1 the RI yield is directly proportional to the primary beam intensity. To achieve high rare isotope beam intensity we can work on the release efficiency to a certain extend. But, rapidly we come to very strong limitation because further improvement of the release efficiency would come at the cost of destroying the target or the ion source. The only viable option is to increase the primary beam intensity. But an increase of the incoming flux requires high power targets capable of withstanding and dissipating the deposited beam power.

The ISAC on-line high power target can be described as consisting of two parts, the target material itself and the target container. Both have to sustain the high radiation dose from the primary beam. The target container has to remove the power deposition in such a way that the operating temperature of the target material can be maintained below the evaporation threshold, which in our case is the temperature corresponding to 10-6 mbar vapor pressure.

The ISAC high power target (HPT) has been described in [1]. The tantalum target container was modified to increase the effective emissivity by diffusion bonding radial Ta fins onto the standard target container. The radial fins are installed over the tantalum tube and are diffusion bonded to the target container by heating the overall system in vacuum at 1500_ C for a period of 20 hours. Off-line tests using electron beam heating show that the effective emissivity is 0.92 and the target is capable of dissipating 25 kW of power when operating at 2200_ C. Using resistive heating and beam power we can balance the temperature from 45 to 100 μA proton beam on most high power targets for both metal foil and composite carbide targets. Since we cannot rotate the proton beam on our target we must ensure that the proton beam profile is large enough to avoid puncturing the target entrance window. For a proton beam size of 7 mm FWHM we can reach routinely 75 μA and with a beam profile of 8 mm we can reach 100 μA on Ta and Nb foil targets.

Target Material

Since we operate the target material at its temperature and vapor pressure limits is important to select a target material that has the following properties:

High thermal conductivity,

Low vapor pressure.

Refractory tantalum and niobium foil metal foils and carbides are among the best candidates.

Refractory carbides are ceramic materials that have been used in commercial applications for many years, such as SiC, TiC and WC. They are mainly utilized for their hardness and abrasive properties and as high strength tooling materials. For the ISOL method targets the high operating temperature required to enhance the release processes is essential, especially, when using plasma ion sources, which are sensitive to high pressure and gas loads. At TRIUMF we have developed a production technique for target materials that combines high material porosity and easy handling [2]. In the ceramic industry this technique is known as slip casting. The ceramic powder suspended in a solvent, which contains dissolved polymers that favor the powder dispersion. This mixture in suspension is poured into a mold or onto a backing foil and then allowed to dry. The dried slip cast, which contains the ceramic powder particulates and the polymers, is easily cut into the desired shape. These carbide ceramics were used up to 40 μA. However, due to their relatively low thermal conductivity compared to metal foils, it is necessary to increase the effective thermal conductivity of the carbide ceramics in order to operate targets at higher beam intensity, above 20 μA, in our case.

We have developed a technique allowing us to pour the ceramic powders and polymers onto an exfoliated graphite foil. These composite carbide targets are capable of dissipating very high power. The ceramic layer is typically 0.25 mm thick, while the graphite layer is around 0.13 mm thick.

The same technique can be used for other target materials. For example we have tested Nb_5Si_3 backed onto a Nb foil to dissipate the heat. Normally the Nb_5Si_3 target material can only be operated at the 1 μA level. By using the Nb foil backing we were able to increase the primary beam current on this target material to 15 μA.

Another example for the use of this technique is for oxide target materials. Oxides have a very low thermal conductivity. On average, it is at least one order of magnitude lower than for carbides. The EURISOL target study group was mandated to look at the possibility of using an oxide targets for the release of a high intensity ^{18}Ne beam. A test conducted in March 2009 in collaboration between CERN and TRIUMF was meant to validate the oxide target up to 20 μA under realistic beam conditions.

Composite Al_2O_3/Nb foils were made by reactive brazing of the oxide layer onto Nb disc to enhance the low intrinsic thermal conductivity of the oxide material and to obtain a good thermal contact between the Nb foils and the tantalum tube. The prototype operated in a stable mode at nominal 25 μA proton beam current – with a peak at 30 μA – for 9 days [3].

Table 1 shows a summary of the composite carbide targets used at ISAC over the past five years and gives a comparison with metal foil targets.

Table 1 – Comparison of the primary beam intensity for the composite carbide, metal foils and oxide targets.

Target Material	Proton intensity on target
$SiC/C_{graphite}$	70 μA
$TiC/C_{graphite}$	70 μA
$ZrC/C_{graphite}$	75 μA
Ta	75 μA
Nb	100 μA
Nb_5Si_3/Nb	15 μA
Al_2O_3/Nb	25 μA

Radiation Enhancement and Associated Issues

The most striking beneficial effect we observed with increasing proton beam intensity on ISAC targets has been the observation of Radiation Enhanced Diffusion (RED) [4], [5]. The increase in diffusion or enhancement of atom mobility in an irradiated material is due to two factors: (1) enhancing the defects concentration, and (2) creating new defect species.

This can be viewed as a hidden term proportional to the incident beam flux, in the diffusion efficiency term, m can be equal to _ or 1 depending of the defect annihilation process taking place with the target material.

With intense proton beam flux the yield in equation 1 can result in a proportionality of to rather than . This effect is most remarkable for the short-lived products because most of the decay losses occur during the diffusion process. The long-lived species are less affected or enhanced since they can take longer time to diffuse to the surface having a longer half-life.

We have also observed with an Al beam that the yield increases with time. For example yield measured of ^{26g}Al and ^{26m}Al increases by a factor 10 and 7 respectively in one week, while the ^{22}Na and ^{24}Na yield remains constant. This suggests that there are cumulative irradiation induced changes to the SiC target material that affect the mechanisms of diffusion of Al in SiC. The assumption is that the lattice experiences a phase transition from α-SiC to β-SiC. The fact that Na yields do not change suggests that the Na diffusion mechanism is different from the one for Al. Na diffusion in carbides may be fast enough so the radiation-induced changes are insignificant.

The RED effect can be seen positive as it enhances the RIB intensity, at the same time it also proved to be detrimental by decreasing the target longevity. Under long time irradiation a substantial decrease in yield is observed. For example we have observed target aging effect for an ISAC high power Ta target. The target has received an integrated charge of 42000 μA-hr, corresponding to $9.4*10^{20}$ protons. The yields ratio of measurements taken 17 days apart is plotted as a function of the isotope half-life. We can see the yield has dropped by a factor 10 in some cases. On average the yields have dropped by 80% [Dom2007]. The simplest explanation for the uniform yield decreases is the loss of integrity of the target container itself. With cumulative radiation damage and buildup at the lattice boundaries, cracks open up between the crystal grains of the Ta target container. Eventually, the total area of the cracks becomes significant and allows large quantities of products to escape the target container before they have a chance to reach the ion source.

Such grain boundary cracks are observed during post-irradiation inspections of the target container.

UC_x Target Development

The physics program at ISAC demands a high power UC_x target and it is even more important for the ARIEL project that is presented in the next section. Our simulations show that the power deposition in the UC_x target for the phase one is about 22 kW for the 100 kW phase, while it is 66 kW for the 500 kW second phase [6].

At the moment the UC_x targets are in use at other ISOL facilities like, ISOLDE, HRIBF and ALTO. They are operating in the range of only few kW dissipated beam power. We have started the development of an UC_x target for the ISAC facility based on techniques similar to those used for our other carbide targets.

The difference to other carbide target materials is the fact that we cannot obtain directly the uranium carbide powder from a commercial supplier. We have prepared the uranium carbide production following these steps:

Milling UO_2 and C,
Casting the UO_2/C slurry,
Carbonization under vacuum, ($p< 10^{-4}$ mbar),
Making the target discs,
Milling the UC_x and C,
Casting the UC_x/C slurry,
Cutting the target discs from the green cast,

Conditioning and sintering the target material is done by loading the target discs into the Ta container in the green form and heating it up to 1750 _C under vacuum, ($p< 6.5*10^{-5}$ mbar).

Figure 1 shows a scanning electron microscopy (SEM) image of the sintered UC_x material. We can see that the grain sizes are quite small and the porosity is such that we obtain approximately 45% of the theoretical density. The first test of our UC_x target will take place in December 2010 at ISAC.

One of the outstanding questions regarding the usage of UC_x at ISAC is its chemical stability. This has been a longstanding question for many years. We recently, finalized tests of the UC_x chemical stability when exposed to air at different temperatures up to 400_ C and in water. The sintered UC_x discs exposed in air do not show any sign of chemical reaction and after 5 days in water at room temperature have no visible sign of chemical reaction.

We can conclude from those chemical reactivity tests in air that the UC_x is quite stable. We did not observed rapid oxidation. After 5 days we can only see a small surface discoloration. The tests in air at high temperature (400 ^{o}C) and in water show a slow oxidation.

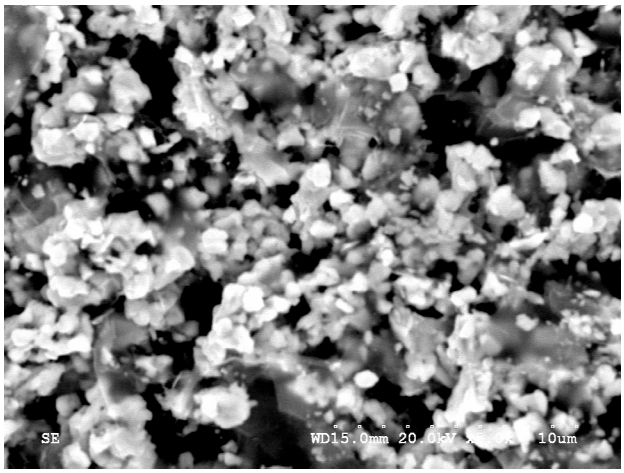

Figure 1: Scanning Electron Microscopy image of the UC_x target developed for the ISAC facility. The magnification is 5000 x.

ION SOURCES COMBINED WITH HIGH POWER TARGETS

In the previous section we saw that we were able to develop a target material and a target container capable of operating at high power. The next step in the chain for successful production of intense rare isotope beams is the ion source.

At ISAC we are using primary three types of ion sources,

Hot surface,
Resonant laser and
FEBIAD

The hot surface ion source and the laser ion source are not so much affected by the target outgas while plasma ion source are.

A Forced Electron Beam Induced Arc Discharge, (FEBIAD) [8] is one of the best plasma ion sources to operate in large range of pressure. We have developed our based on the hollow cathode developed by Sundell [9] at ISOLDE-CERN. Our FEBIAD has a smaller volume mainly constraint by the available envelope on board of our target module.

In order to satisfy the demand for isotopes for the nuclear astrophysics program we must be able to ionize with high efficiency the gaseous elements or molecules of C, N, O, F, and Ne. These elements are only ionized efficiently with an electron cyclotron resonance ion source (ECRIS). A new ECRIS capable to operate on-line with high power targets is being developed. A prototype having the following improvements is being operated at a test stand:

Improved electron confinement,
Increased operating frequency, 6 GHz,
Movable extraction electrode,
Higher throughput, increased pressure tolerance.

The new ECRIS is based on MONOBOB [10], which was developed at GANIL, where four coils made from hollow copper conductor surrounded by a ferromagnetic structure produce the desired magnetic field. The magnetic field has an axial and radial confinement. The extraction is an adjustable, three electrode system [11].

ARIEL PROJECT

The Advanced Rare IsotopE Laboratory (ARIEL) project proposed the construction of a new electron superconducting LINAC capable of delivering up to 500 kW electron beam at 50 MeV, a new proton beam line BL4N, and two new target stations combined with a high mass-resolution separator in a new building. The LINAC and the ARIEL building have been funded by the Canadian Foundation for Innovation from the Government of Canada and the Province of British Columbia provincial, representing a total funding amount of $60.7 M. Figure 2 shows the TRIUMF site layout, in blue the actual layout and the proposed ARIEL and UCN projects in red.

The ARIEL target stations and the associated equipment for servicing the area will be quite similar to the existing ISAC facility. From our experience of operating high power ISOL targets we would like to retain the following aspects:

Two-stage mass separator system, the pre-separator inside a heavily shielded hall. From our experience we have very low contamination spread in the adjacent mass separator room and beam line. The first selection slit is directly accessible by the remote handling equipment for maintenance.

The approach we use at ISAC allows us to operate ISOL targets at unprecedented beam power. We routinely operate our target at proton beam currents between 70 and 100 μA.

All non radiation resistant components are located in a low radiation field thanks to the target module-shielding plug.

On the other hand there are issues that we will address, for example:

For budget reasons when ISAC was build it was decided to build only one mass separator for the two target stations. The optics from each station merge at the pre-separator. The exit module beam optics box does not block the neutron flux back streaming from one target station to the other. This implies that we cannot service one target station while operating the other one. It reduces the flexibility in operation and forces beam off period to attend the target module for the next run.

The actual containment box at the bottom of the target module is not hermetically sealed. It creates difficulties when operating air sensitive target materials such as LaC_2. Furthermore, the target/ion source assembly cannot be pre-conditioned off-line.

All electrical, vacuum and signal connections on the top of the target module are done manually imposing long delays of at least one week and some times two weeks after beam off to allow for cool down before personnel access to the area.

Target/ion source exchanges takes from 3 to 5 weeks. This is forcing us to operate our target for much longer period than we should. We would like to speed up the process in order to reduce the target irradiation time to 2 to 3 weeks. This will reduce the impact of the yield drop due to radiation damage, metal fatigue, target sintering, etc.

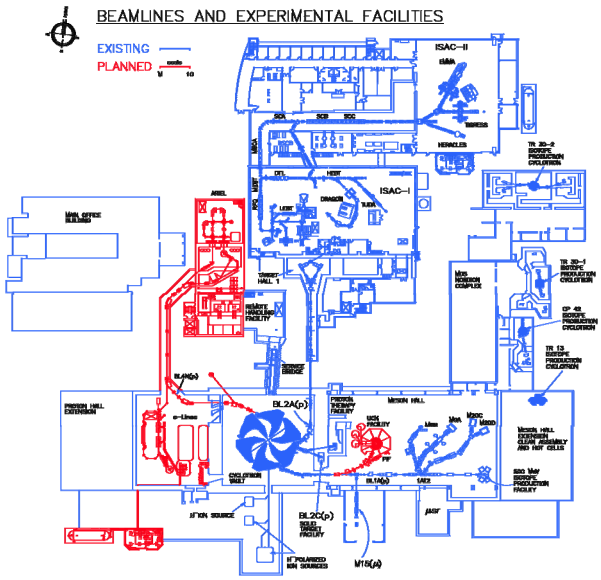

Figure 2: Layout of TRIUMF site (actual in blue), ARIEL project in red with the proposed UCN on the right of the cyclotron.

The new concept for the target module in ARIEL will be quite different than the ISAC target modules. The containment box will be a vacuum box equipped with a radiation hard valve to seal the heavy ion port.

The ARIEL facility will have a conditioning station allowing the preparation and test of the complete target module equipped with the target/ion source before it goes on-line.

The target module services will be remotely connected using a similar design as the one developed for the actual ISAC facility.

The front end beam line which houses the optics and the diagnostics and vacuum pumps is built into 1 m long sections. Each section can be disconnected and transfer to the hot cell for maintenance and repair.

CONCLUSION

We have made progress on the development of a uranium carbide target material. Using technique similar to the one used for the other composite carbide target materials we have developed an UC_x target material, which can operate up to 40 to 50 μA, depending on the thermal conductivity.

We have made tests to demonstrate the chemical stability of the UC_x. It appears that the material is quite stable when exposed to air even at high temperature (400 °C) and in water. These results give us confidence about our technique to deal with spent target in the hot-cell and long-term storage vault.

These progress on the UC_x are essential for the success of the ARIEL project, which is based on using mainly uranium target for the physics program both for the photo-fission and the spallation products like Rn and Fr for the EDM and Fr experiments.

The new ARIEL target stations have several improvements with respect of the first generation of the ISAC target station. Using new radiation resistant all metal seal gate valves and double pillow seals we have significantly simplified the vacuum system. It allows a true confinement box for the target module housing the target/ion source assembly. With the remote services connection we will have a much quicker turn around for target exchange.

Finally, once completed the ARIEL project combined with the ISAC facility will allow three RI beam users simultaneously.

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