# CRITICAL TECHNOLOGIES AND FUTURE DIRECTIONS IN HIGH INTENSITY ISOL RIB PRODUCTION*

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*Abstract*

The future frontier of the ISOL technique is to increase the intensity of the Rare Isotope Beams, RIB. The most expedient method is to increase the incident beam on target. Increasing the overall release efficiency and ionization efficiency are the other two dirrect ways to increase the overall RIB intensity. Now with the TRIUMF/Isotope Separator and Accelerator, ISAC facility the ISOL RIB can operate routinely up to 50 kW on target. The factors limiting the driver beam intensity on target currently are (a) radiation damage issues and (b) ion source ionization efficiency and longevity due to the effects of target out-gassing. The other technological challenge for the ISOL technique is the target material itself. The main concern is the capability of the target material to sustain the high power density deposited by the driver beam. Refractory metals foil target are suitable but nevertheless limit in the isotope species that can be produced from these target materials. Composite targets, either from carbide and oxide target materials were developed at ISAC that can sustain operation under high power density. A review of technological challenges and future direction for the production of intense RIB with high reliability is presented in this paper.

## INTRODUCTION

In the past fifteen years the atomic nucleus has been the laboratory for studying to understand the basic forces that bound the nuclear matter. For example three body forces not only explain the light nucleus like $^3$H and $^3$He but also are needed to explain the stability of $^{11}$Li. Beyond the stable nuclei, a wide variety of unstable nuclei exist. We have now discovered more than 3000 of those particle bound combination of proton and neutrons. Some of them have been investigated in detail, but most of them are not very well known. The decay scheme, nuclear spectroscopy, mass, and half-life are among the characteristics that have to be measured. Those nuclei are produced in several places in the universe, stars, novae and supernovas, for example. They splay a crucial role in stellar evolution. Determining their properties provides key input parameters for nuclear astrophysics models.

In the last three decades the emphasis was placed on the production of a wider variety of rare isotope beam for fundamental science, halo-nuclei, nuclear astrophysics and material science. This required state of the art technologies to deal with the various issues of the on-line production of these beams. There are several challenges in the production of intense rare isotope beams on-line, radiation resistant target/ion source and auxiliary component, target material capable of sustaining high power density, beam transport and efficient charge state breeding and mass separation. Furthermore, as the experiments become more complex they are requesting higher intensities over longer periods of time. The requirement for higher target/ion source reliability is of prime importance.

Section one reviews the production of the rare isotope on-line, section two focuses on the various challenges specific to the RIB production using the ISOL method and section three gives some examples of future directions towards high intensity ISOL beam production.

## RIB PRODUCTION

Beams of rare isotopes are challenging to produce, especially the short-lived ones, they do not occur naturally. 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 the nuclei in the 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 accelerated to form an ion beam that can be mass analyzed and be delivered to experiments. The requirements for producing high intensity RIB are:

1) A high energy driver, such as the TRIUMF H⁻ 500 MeV cyclotron,
2) A target material inserted into a oven made of refractory material, connected to an ion source,
3) An ion source at high voltage to produce an ion beam,
4) 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. This target material must also be able to sustain the power deposited from the driver beam. 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 effects 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 remaining proton beam emerging from the target.

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

1) 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.

2) Fragmentation 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.

3) Induced fission occurs when the incoming projectile deposits sufficient energy in the target nucleus to induce a breakup into two roughly equivalent mass products.

The yield of a specific isotope can be expressed using the following equation (1):

$$Y_Z^A = \sigma_Z^A \Phi_p N_t \varepsilon_D \varepsilon_E \varepsilon_I \varepsilon_T, \qquad (1)$$

where: $\sigma_Z^A$ is the nuclear reaction cross section leading to this specific isotope, $\phi_p$ is the primary beam flux, $N_t$ is the number of target nuclei per square cm, and the $\varepsilon$ represents the effusion, diffusion, ionization and transport efficiencies, respectively.

Once the Rare Isotope has been produced during the interaction of the driver beam with a target nucleus it is stopped in the bulk of the target material. In order to form a useful beam the radioactive atoms has to: 1) diffuse out of the target material grain, 2) undergo surface desorption, 3) effuse from place to place until it reaches the exit hole in the target container to the ion source, 4) get ionized and 5) mass analyzed and being delivered to the experimental facility.

## TECHNICAL CHALLENGES

### ISOL Target Container for High Intensity RIB

Challenging experiments forced us to increase the incident driver beam on target with the goal of producing higher RIB intensity. To do so the ISOL target oven has to be capable of dissipating the power deposited by the incident beam very efficiently.

The ISAC high power target[1] (IHPT) was developed to accommodate the TRIUMF 50 kW proton beam. The conventional target at ISOL facilities such as ISOLDE, SPIRAL and HRIBF can only accommodate for less than 1 kW dissipated power inside the target. There were several attempts in developing the ISOL target for higher power dissipation, by Ravn[2], Talbert[3], Nitschke[4] and RIST collaboration[5]. The most promising being the RIST because of the simple cooling design. But, the target fabrication is quite limiting because it required diffusion bounding and can only be applied to a few target materials. The ISAC High Power Target, IHPT, utilizes the thermal radiation cooling and is made of a 20 cm long and 2 cm in diameter tantalum tube onto which radial fins

are installed. The fins are diffusion bounded to the target container by heating the tube in vacuum at 1500 ºC for a period of approximately 20 hours. The overall emissivity measured is 0.92 which allow operating the target at nominal 2200 ºC up to 20 kW of deposited power by the proton beam.

The challenge with increasing proton beam on target comes in several forms: 1) the radiation damage of the tantalum tube, 2) the thermal shock when the proton beam goes off and 3) chemical reactions between the tantalum container and the target material or the radiological impurities created.

All these processes create cracks in the tantalum container allowing the rare isotope atoms of interest to escape the container reducing the output yield. Fig. 1 shows a photograph of a Ta container that housed a SiC target. The picture was obtained with a USB ProScope HR® installed in the hot-cell for target post-irradiation diagnosis of the target. The image is enlarged by a factor 100 with a lens mounted on the scope.
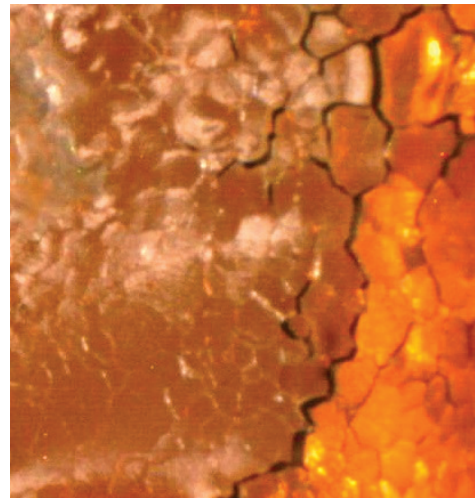


Figure 1: Photograph of the Ta container at 100 X magnification. Damage to the container walls is clearly visible, especially the cracks and crystal growth.

### ISOL Target Material for High Intensity RIB

The challenge for the target material with increasing driver beam intensity is that the target material has to be capable of dissipating the high power deposition in the target. This means that the target material has to have a much larger overall thermal conductivity than routinely used at ISOL facilities.

We have developed a technique to increase the overall target thermal conductivity. This technique can be used for carbide and oxide target material. The heat deposited in the target material is driven to the target container and radiated away to the heat shield[6].

In the case of carbide target material we have developed a casting technique where the carbide target solution, which includes plasticizer and organic solvent is poured over an exfoliated graphite foil. The exfoliated graphite foil has a larger radial thermal conductivity than the

carbide layer. Adding graphite powder to the carbide mixture allows us to even increase the power dissipation from the target material.

For the oxide target the technique is similar. The oxide target material powder is mixed with plasticizers and solvents and using a ball mill we homogenize the suspension and reduce the average grain size at the same time. The resulting slurry is poured over a metallic foil, either Ni, Nb, Mo or Ta, depending on the final product we want to extract from the target.

This technique allow us to run our carbide target up to 75 µA proton beam current and the oxide target up to 20 to 35 µA, representing an increase by a factor 35 and 20, respectively, over conventional ISOL target.

Recently we have operated our composite uranium carbide target at 2 and 10 µA, fig. 2 shows a comparison of the Rb production.
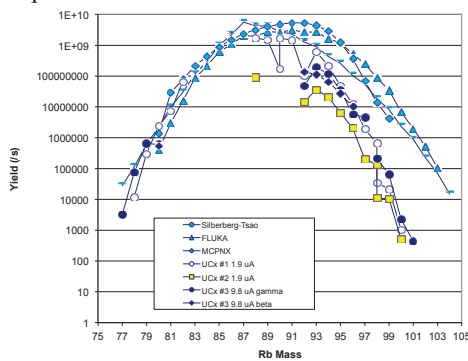


Figure 2: Comparison of the Rb isotopes yields for three UCx targets. The first two were operating at 2 µA while the third on was operating at 10 µA p+ beam intensity. The calculated in-target production using FLUKA, MCPNX and Silberberg-Tsao is also plotted as a function of Rb mass number.

*ISOL Ion Source for High Intensity RIB*

The increase of driver beam power on the target has the effects of increasing the vapor pressure, which in turn can affect the ion source performance.

Another type of ion source that is less affected by pressure increase in the target container is the resonant ionization laser ion source, in this case the ions are generated by sequential photon absorption until the electron gain enough energy to escape the atom.

The simplest ion source, which is the hot Surface Ion Source, SIS, can be affected by the release of either carbon or oxygen from the carbide or oxide target, respectively. The oxide or carbide layer that build-up on the hot surface affects the work function of the surface, which in turn reduce the ionization efficiency. To mitigate this effect we use a rhenium metal tube inserted into the transfer tube. Rhenium does not make stable oxide or carbide at high temperature. This help to keep the surface clean and produce stable ionization efficiency over the entire run.

The other type of ion source that is less affected by pressure increase in the target container is the resonant ionization laser ion source (RILIS). In this case the ions are generated by sequential photon absorption until the electron gain enough energy to escape the atom. The laser ion source utilizes the same structure as the SIS into which high power pulsed laser beams are introduced.

Plasma ion sources on the other hand are sensitive to pressure increase inside the target container. The electron impact driven ion source is less sensitive to pressure increase compared to the electron cyclotron resonance ion source. They are affected by recombination of the positive ion colliding on neutral atoms. To mitigate this effect we can cool the transfer line connecting the target to the ion source volume. It allows volatile atoms or molecules to reach the ion source plasma volume while condensable elements are being trapped in the cold transfer line.

*ISOL Target/Ion Source Reliability*

It is challenging to operate the ISOL target/ion source with high reliability in the hostile radiation and high temperature environment. It is also difficult to assess the problem when a failure occurs because we do not have access to the component due to the high level of radiation.

To improve the ISOL operation reliability we have initiated a failure mode and effects analysis (FMEA) of the whole target/ion source assembly. FMEA is used in product development in manufacturing industries for example, where it helps to identify potential failure modes based on experience. We have applied this analysis method to the design and the processes. Fig. 3 shows an example of template used for the analysis. In this exercise it is very important that the function of the part is identified. Often too many functional aspects are taken for granted and keys function of the component can be ignored. The second column identifies the potential failure modes of the component and then the third column describes the effect of the failure. The next step is to identify the cause(s) of the failure mode. It is important at this point to look at all possibilities and to analyze them. In a complex system, and especially the one like our where it is difficult to have access to the product after failure, sometime the cause may be a succession of event or failures.

To help focusing on the critical failure mode(s) it is important to come up with some sort of rating of the risk. The higher the risk rating number is the more resources and priority should be used to solve the failure mode. Each failure mode is given a severity number (S) from 1 to 10, 1 being benign and 10 severe or critical.

The occurrence rating (O) expresses the failure mode frequency. It can be obtained from the track record during operation of the system. Here we have to be careful because there may be a discrepancy between first observations and the real cause of the failure. This is why post diagnostic of the failed equipment is so valuable. Again, here a low occurrence will be given a lower value for the variable O and a higher failure rate will get a larger value.

The next step is to determine the ease of detection (D) of the potential failure mode. This can be accomplished by proper inspection methods or test procedures. The

current control of the component or assembly, that prevent failure mode from occurring or detect the failure mode before the assembly reaches the production on-line. The ease of detection of the potential failure mode is a crucial point, sometime the failure mode hard to detect or require long testing time. We may want to change the design to ease the detection. Again the rating goes from 1 for an easy detection to 10 for a difficult detection.

The risk priority number, RPN, is the key of the whole analysis process. It is the product of the S, O and D values. The RPN rating determines the areas of greatest concern for the reliability of the whole assembly. This help to focus the design or process on the critical failure. The next step is to recommend action with deadline. The action is tracked with an Engineering Change Order, ECO, that are kept and recorded. When the ECO is closed the action is completed and the information is kept as a log. This way we can keep track of the progress and design or process changes.

| Item/ function | Potential Failure Mode | Potential Effects of Failure | Potential Cause(s) | Severity (S) | Occurrence (O) | Current Control | Ease of Detection (D) | Risk Priority Number (RPN) | Critical Character Y/N | Recommended Actions, ECO number | Responsibility and Target Date for Completion | Action Taken |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | | | | |

Figure 3: Example of a FMEA template used for the target/ ion source assembly.

## *Example of FMEA*

We have started the FMEA analysis to improve the ISAC target/ion source reliability. We had several issues with water leaks and due to the fact that we did not have access to the assembly after irradiation it was difficult to point out the failure mode. So each component of the assembly has been scrutinized and each potential failure modes ranked according to the occurrence, severity and ease of detection. We discovered that the brazing of the cooling line to the metal gasket joint (VCR) was showing a very high RPN. The detection of a failure in a brazed joint is very difficult, especially the way the brazing of the copper tube to the stainless steel VCR was made. Only destructive testing allows to see if the brazing has penetrated far enough to obtain a good joint. We since changed the design to allow inspection of detection of the brazing and it solved the water leak problem.

## FUTURE DIRECTION FOR THE ISOL TECHNIQUE

The future directions for the ISOL RIB are; increase neutron rich RIB intensity, diversify the RIB species available and increase the charge state breeding efficiency to allow for the delivery of highly charged RIB.

To increase the RIB intensity we can increase the driver beam on the ISOL target to a certain point. With ISAC we have shown that ISOL target can routinely operate with proton driver capable of delivering 50 kW. The ISAC high power target and composite target material can operate up to 100 kW. At higher powers we have to take a different approach. By using neutrons or gammas we can in principle achieve comparable RIB yields with lower power deposition inside the target material. Examples of this approach are the KoRIA, CARIF, SPIRAL-II[7],

EURISOL and the ARIEL[8] projects. The aim is to produce induced fission at a very high rate, $10^{14}$ to $10^{15}$ f/s.

The EURIROL, CARIF and KoRIA projects goal is to extract easy RIB from an ISOL target and than post-accelerated these beam to energy where the in flight fragmentation is more efficient in producing extremely neutron rich nuclei. Such ISOL RIB are e.g. $^{132}$Sn, $^{91}$Kr, $^{142}$Xe, $^{91}$Rb and $^{142}$Cs for example. These isotopes are released quite efficiently from the U targets. The ionization efficiency for the Rb and Cs is close to 100% using a hot surface ion source, 80% for the Xe, 60% for Kr using an electron cyclotron resonant ion source and RILIS efficiency of 10% for Sn is achievable.

Once the ion beam is extracted from the ion source it is mass analyzed and then using a charge breeder one can boost the charge state for a more effective post-acceleration.

Even though the secondary particles are not charged the deposited power from the fission or gamma e-e+ pair conversion is quite large. The heat can only be dissipated to the target container if the target material has a significant thermal conductivity. This is why work on the composite target materials is key to the success of the future facilities. Without the thermal conductivity enhancement these projects will not be feasible.

At TRIUMF the ARIEL project is based on an electron LINAC; 50 MeV and 10 mA, as a photo-fission driver. FLUKA simulations were performed to determine the yield of some key nuclei. In target production rate for several neutron rich nuclei are given in table 1 for 500 kW electron beam on a Pb converter and 20 $g/cm^2$ UCx target.

The second direction in ISOL RIB is to bridge the gaps between available elements. When we look at the ISOL yield for each element we can see clearly gap. This is due to the fact that some species are released poorly from an

ISOL target. Those elements form alloys or compounds that are highly refractory and consequently reduce the release efficiency dramatically.

Table 1- In target production for a 5 kW proton and 500 kW electron, beam on UCx target.

| Nuclei | 5 kW proton | 500 kW electron |
|--------|-------------|-----------------|
| Ni-72 | 3.8E+08 | 2E+08 |
| Zn-78 | 1.4E+09 | 3.4E+09 |
| Kr-91 | 5.3E+10 | 2.3E+11 |
| Kr-94 | 1.3E+10 | 1.3E+11 |
| Rb-97 | 7.4E+09 | 1.1E+11 |
| Sn-132 | 1.1E+10 | 2.5E+10 |
| Sn-134 | 1.0E+09 | 2.4E+09 |
| Xe-142 | 1.1E+10 | 5.2E+10 |
| Xe-144 | 1.0E+09 | 7.9E+09 |
| Cs-144 | 6.8E+09 | 6.0E+10 |
| Cs-146 | 5.0E+07 | 9.2E+08 |

There are several techniques to improve the release of those refractory metals. They are; He jet with aerosols, Ion guide, chemical reactions inside the target container.

While the first two techniques require He handling to be efficient they are only applicable to thin target. These techniques will not be capable of reaching the same intensity as for the other species using the ISOL method. This means that the application will be limited to experiments that can make use of quite low yields, such as mass measurements and nuclear decay studies, for example.

The third option is using chemicals that react with the desired species to produce more volatile. They can reach the ion source without reacting on the wall of the target container or the ion source itself. One problem with this type of process is the fact that resonant ionization using laser ion source cannot be applied because molecules have a much higher ionization potentials generally.

This technique has been in use for several years at ISOLDE, HRIBF for example. At TRIUMF we did only few tests with injecting $CF_4$, we have observed for the first time short-lived [24,25]Al isotopes from a SiC target at ISAC this spring.

The next direction is to obtain high efficiency charge state breeding. There is two main methods used to produce high charge states, the ECRIS and the EBIS breeder. So far the charge breeding efficiency and beam purity are still a challenge, especially, for the ECRIS. In the past few years there were several improvements. The ANL[9] reported breeding efficiencies approaching the result obtained at ISOLDE/CERN with an EBIS breeder. The main reason for such an improvement with the ECRIS breeder seems to be related to the pressure inside the plasma chamber. The other aspect of the charge breeder is the contamination from stable isotope coming from the breeder itself. At ISAC with the ECRIS breeder we observed large contamination from the stable isotopes coming from the material inside the plasma chamber. We observed a large contamination mainly from Fe, Ni, Cr, Cu and Zn mainly. We took advantage of our annual

maintenance period starting last January to replace all the components inside vacuum with pure Al with the goal of reducing the contamination in the Q/A region of interest for the ISAC RIB program. We machined all the injection and extraction electrodes from Al and we coated the vacuum chamber and the iron yoke with pure Al. We just start up the CSB and can see that the Cr, Fe and Ni stable beams have disappeared, see fig. 4.
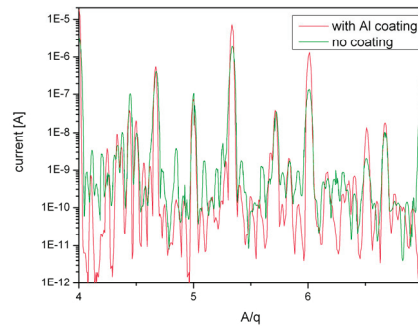


Figure 4: Comparison of the ECRIS charge state breeder before Al coating in green and after modification.

## ACKNOWLEDGMENT

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