RIA and Stockpile Stewardship: The LLNL Perspective

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Abstract:

Lawrence Livermore National Laboratory recognizes the Rare Isotope Accelerator (RIA) as an extraordinary opportunity. Not only are LLNL scientists interested in the nuclear science and astrophysics that RIA will enable, but RIA offers the opportunity to conduct stockpile stewardship relevant measurements that can be performed at no other facility. As such, LLNL has been working hard with the nuclear physics community to convince DOE Office of Science to proceed with RIA. LLNL also recognizes the need to invest its own resources at the early stages in RIA to enable many of the stewardship measurements. Harvesting isotopes, processing the collected material into targets at radiochemistry facilities, and transporting those targets to a neutron source for irradiation are key capabilities needed at RIA for stockpile stewardship and some astrophysics measurements. Additionally, LLNL realizes the benefit its engineering resources would be to the project in solving more general RIA R&D challenges. Over the past two years, LLNL has supported efforts exploring fast RF tuning of cavities, nuclear hazard classification issues, and thermodynamic analysis of a potential beam stop for high power uranium beams. These pursuits have led LLNL to identify two important R&D challenges, beam power on ISOL targets and primary beam handling in the fragment separator, whose resolution will greatly impact how LLNL and the stockpile stewardship community participates in experiments at RIA. The issues as well as the stockpile stewardship case at RIA and LLNL's participation in RIA R&D will be discussed.

Introduction

Determining the neutron flux in regions of enormous instantaneous neutron intensities has been a scientific challenge for LLNL scientists for decades. Addressing this challenge is essential in understanding the interior of stars, interpreting nuclear weapon tests, and will be critical in performing experiments at NIF. While LLNL scientists have tried to solve this problem in several ways, the most commonly used technique requires accurate neutron cross-sections on short-lived nuclei for accurate flux determination. Historically, measurements have yielded the most accurate cross-section information, but it has been impossible to perform measurements on most of the relevant short-lived states due to production limitations. The Rare Isotope Accelerator (RIA) changes that.

The promised production rates at RIA will enable many stockpile stewardship relevant measurements. While several techniques are being explored to obtain neutron cross-section information, it is clear being able to perform direct neutron cross-section measurements on radioactive targets at RIA is critical for some astrophysics and stewardship measurements. Enabling these type of measurements requires three capabilities to be included at RIA: 1) harvesting of isotopes, 2) radioactive target preparation, and 3) neutron irradiation. Thus, LLNL has been investing its own resources to address the R&D challenges associated with adding these capabilities and in general how stewardship relevant measurement can be performed at RIA. These efforts have also led LLNL to investigate some more general RIA R&D challenges such as number of ISOL stations and handling the primary beam in the fragmentation line. Different options for resolving these challenges and how these solutions affects stewardship impact on RIA science programs are discussed below.

RIA's Value to LLNL

In order to obtain information about the neutron flux during nuclear weapon tests, certain isotopes were loaded as neutron flux monitors. For example, assume ^{90}Zr is added to the experimental environment prior to the test. During the test a process similar to nucleosynthesis in stars occurs as neutrons react with the ^{90}Zr atoms to make ^{89}Zr and ^{88}Zr via (n,2n) reactions from 14 MeV neutrons. The amount ^{89}Zr and ^{88}Zr is reduced by competing (n,γ) reactions from lower energy neutrons. Figure 1 below shows a simplified reaction network for zirconium. After the underground test, samples of the material from the highly radioactive center of the explosion can be extracted and subjected to precise radiochemical analyses. Gamma ray spectroscopy can be performed to determine the amount of each reasonably long-lived isotope present in the samples and in particular, the ratio of ^{88}Zr to ^{89}Zr. These measurements would then be compared with the predictions of the computer simulations to determine the neutron (and charged particle) fluxes and thereby the performance details of the device. Similar diagnostics will also be used at NIF.

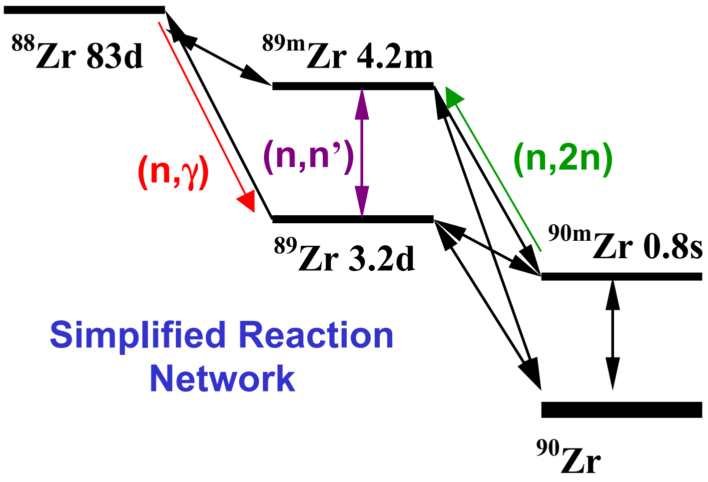

Figure 1: Simplified reaction network for ^{90}Zr.

Even from the simplified network in figure 1, it can be seen that most of the nuclei involved in the reaction network are unstable. In reality, the reactions networks are much more complicated. A more realistic zirconium network has some 60 neutron cross-sections for which accurate neutron cross-section information is required. Only 5 of these have been examined experimentally because it has been impossible to produce the desired isotopes in sufficient quantities. The stockpile stewardship program, however, needs more accurate cross-section information for neutrons from 0.1-20 MeV to accomplish its goals. A facility that dramatically increases the production rates for many short-lived isotopes, thus enabling new opportunities for measurements would be a tremendous resource in improving the nuclear databases used by stockpile stewardship. RIA is just such a facility. Table 1 lists some of the isotope important to stewardship and their expected production rates at RIA. For isotopes with a half-life of a day or longer it is expected that enough of the isotope can be harvested to allow target formation. For isotopes with a half-life much shorter than one day, experiments using radioactive beams could still be used to gain insight into neutron cross sections. Prompt fission fragments, such as ^{95}Sr, fall into this category.

Nuclei	Half-Life	RIA Production Rate	Harvesting Limit
^{87}Y	3.35d	$3\text{x}10^{11}$ pps	18 μg
^{87}Zr	1.71h	$1\text{x}10^{9}$ pps	1.3 ng
^{148}Eu	54.5d	$7\text{x}10^{10}$ pps	117 μg
^{173}Lu	1.27y	$2\text{x}10^{11}$ pps	3.3 mg
^{95}Sr	25.1s	$8\text{x}10^{10}$ pps	0.46 ng

While stewardship science is important to LLNL it is not the only reason LLNL is excited about RIA. LLNL has a long history of involvement in nuclear astrophysics, in part due to the similarities between stars and nuclear weapons. The opportunity to advance our understanding of nucleosynthesis through measurements at RIA fits well into the interest of LLNL scientists. LLNL scientists are also interested in participating in general nuclear science experiments at RIA, which will explore the nuclear force in systems far from stability. Involvement at RIA will also address the recruitment challenge for LLNL since it will be the training ground for future generations of nuclear physicists and radiochemists.

LLNL's Support For RIA

Because of the reasons outlined above, LLNL had worked hard to generate support with in NNSA and have NNSA urge the Office of Science to proceed with the RIA project. The most visible result of this effort has been a letter from Everet Beckner, Deputy Administrator for Defense Programs to Ray Orbach, Director of Office of Science expressing NNSA's interest in conducting research at RIA and the willing of scientist at NNSA's laboratories willingness to help when the Office of Science proceeds with the RIA project. Additionally, Ray Orbach requested a classified briefing to understand in detail the connection between RIA and stockpile stewardship, which was attended by Bill Goldstein, Associate Director for Physics and Advanced Technologies and Ed Hartouni, the division leader for Nuclear, Particle and Accelerator Physics. Mike Kreisler was also present at this meeting and he also participated with RIA Steering committee in presentations to the OMB and the OSTP.

Livermore has also recognized the need to invest its own resources in the pre-project phase of RIA. Many of the stockpile stewardship measurements require additions and modification to the RIA layout and working with the rest of the RIA community early on to identify those changes are crucial to minimize their impact. Additionally, LLNL scientific expertise and engineering resources would be valuable in solving many RIA R&D challenges and insuring the best possible facility is built. To this end, LLNL has invested 0.3 FTE's in FY02 and 1.2 FTE's in FY03 in RIA related activities. Those investments include enabling stewardship measurements [1], fast RF tuning [2], nuclear hazard categorization issues [3], and a beam dump for the fragment separator [4]. In FY04, we have proposed to Lab management to use 4.0 FTE's into addressing RIA R&D challenges.

Performing Neutron Cross Section Measurements at RIA

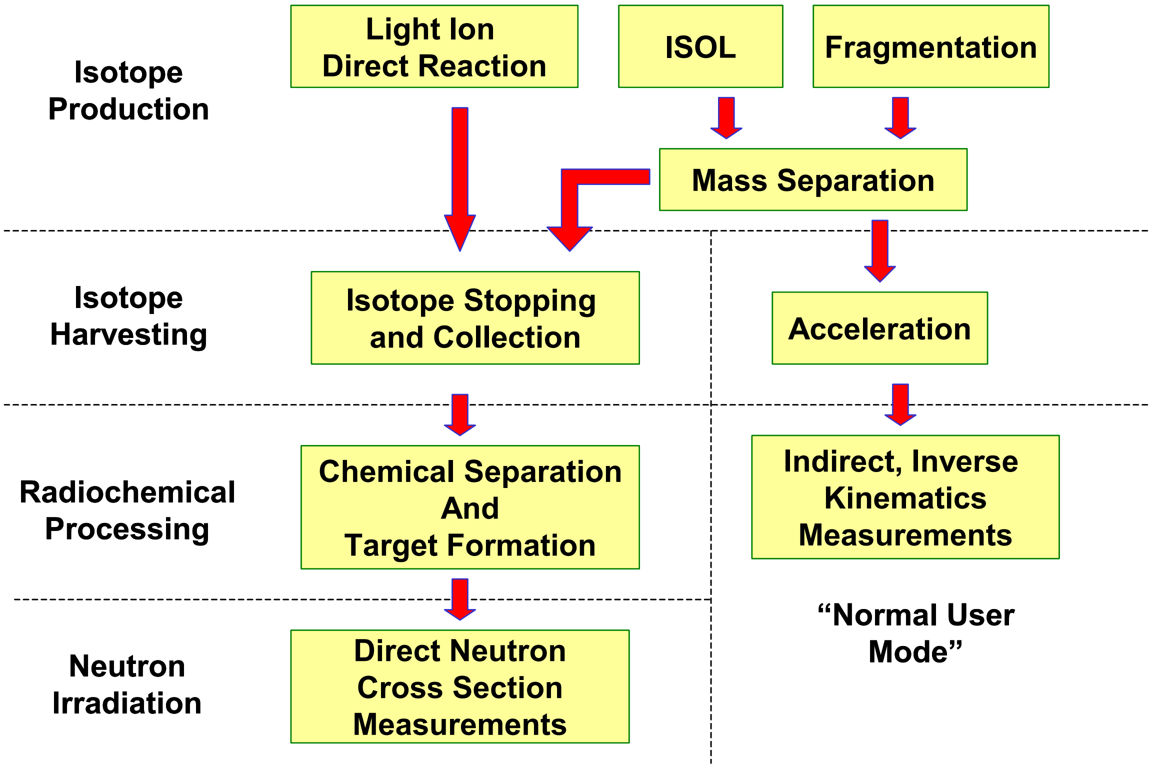

Figure 2: Flow chart describing how neutron cross-section measurements would be performed at RIA.

Figure 2 is a flow chart describing how neutron cross-section experiments would be performed at RIA. Isotope production is the first step at either an ISOL station or a fragmentation line, both of which would provide mass separation. Also listed in the chart are light ion direct reactions that could take place at the first ion stripper of the driver linac. More about this production method is discussed later. After isotope production, a radioactive ion beam can be prepared for inverse kinematics experiments designed to indirectly gain insight into the desired neutron cross-sections. This is the only option for very short-lived nuclei. If the half-life of the isotope is long enough, then the desired isotope can be stopped and collected, transported to radiochemical facilities for chemical separation and target formation, and finally delivered to a neutron source for irradiation.

For nuclei with a half-life much shorter than one day, indirect methods are the only option for obtaining experimental information. At the very least this will involve making mass, life-time, and structure measurements to give theory the best input possible in making accurate calculations of the appropriate reaction cross section. Level schemes will play a particularly important role in this avenue of approach. The surrogate reaction technique [5], however, is one experimental method that offers the possibility of obtaining at least some cross section information for short-lived nuclei. The idea behind the surrogate approach is to look at another reaction that generates the same compound system as would be produced by the neutron reaction, such as (d,p) reactions. Much development on this method remains to be done. In either case, the experiments will involve inverse kinematics experiments using radioactive ion beams and LLNL would participate in the same manner as any other user of RIA.

For nuclei with a half-life a day or longer, the possibility exists for target formation and direct neutron irradiation experiments. There are two basic approaches to measuring neutron cross sections via neutron irradiation on radioactive targets. In the delayed method the target is irradiated with neutrons and spectroscopy is performed on the target afterward to determine the amount of the reaction product produced. Target purity is one of the most difficult issues to overcome for this technique. Given a neutron flux of 10^{11} neutrons/second on target and an irradiation time of 1-2 days, then only 1 in 10^9 target atoms will undergo the desired nuclear reaction of interest. Thus, the target must be free of the reaction product at that level, otherwise the experimental signal will be swamped by background. And if the interest is in (n,2n) and (n,γ) reactions as is the case for stewardship, then chemistry can not help purify the target, since the atomic number does not change. Therefore, either the production method must not produce this background or mass separation is required to obtain the appropriate purity.

The other approach to directly measure neutron cross sections is a prompt experiment that measures each nuclear reaction as it occurs. The choice of technique depends on the exact nature of the reaction of interest. In all cases, the prompt approach greatly reduces the requirement on target purity to the level of 10^{-2} or better, but now the detector must be able to handle the radiation coming from the target. Also, the detector must be shielded from the neutron source, usually requiring the radioactive target to be some distance away from the neutron source. This reduces the intensity of neutrons on target. It is important to maximize neutrons on target since this will reduce the amount of target material needed and thus reduce the background seen by the detector.

Harvesting Isotopes

As mentioned above, direct reactions at the first stripper offer an alternative for near stability isotope production to ISOL and fragmentation. Light ion direct reactions such as (p,X) or (α,X) can be used to create isotopes near stability at high production rates. At this point in the linac, the beam will be 100’s of particle mircroamps of 50 MeV protons or 10’s of MeV/A ions. There will be no mass separation possible for the production technique since the beam will stop in the production target and the produced isotopes will not release. Additionally, this method works best for producing low Z, proton rich nuclei, near stability. The production cross-section fall with the Z of the production targets since the coulomb barrier grows and these reactions tend to emit neutrons over protons. The medical isotope community is also interested in this production area, though they are interested in putting heavy ions on production target to strip off a few neutrons. The difference in stopping of the various reaction products would be use to do a rough mass separation.

The Isotope Separation On-Line (ISOL) beam line offers two advantages for isotope harvesting, large production rates, up to 10^{12} pps, and low energy beams, typically 60 keV. The high production rates are important to minimize the half-life limit for making a target. The low energy allows easy collection by implantation in a foil since the nuclei will stop quickly without inducing unwanted nuclear reactions in the foil that would increase the impurity level. The disadvantage of the ISOL method is the chemistry dependence of the production rate. The ISOL method relies on release of the desired product from an amorphous material at high temperatures and then efficient ionization. Elements like noble gases release well, but elements with a high melting temperature like zirconium, do not.

Assuming a good release and ionization of the desired product, there are two locations where it might be possible to collect separated isotopes. Depending on the ion source used in the ISOL target, a large number of different isotopes will be extracted from the ISOL system. Mass separation is then applied in two stages to purify the ion beam. The pre-separator, which does a rough separation, is one location where collection can occur. It is unclear whether the purity that can be obtained at this stage would be sufficient. For prompt measurements, it probably would be sufficient, but for delayed measurements, it probably would not be. After this low-resolution mass separator, a high-resolution mass separator is then used. This would give much better target purities. Both locations offer the possibility of collecting isotopes parasitically to other experiments, but to achieve the highest data purity samples, the collection would have to be done as a primary user of one of the ISOL production lines.

The fragmentation line overcomes the chemical limitation of the ISOL line, but at the cost of lower production rates and higher beam energy. The production in the fragmentation target can still be quite high, around 10^{11} pps, for lighter mass beams where the driver linac can deliver on the order of 100's of particle-microamps. But for heavy mass beams (A>100), the beam current is down to 10's of particle-microamps and as little as 1 particle-microamp for a uranium beam. Some of this is due to the limitations of the ion source for the driver linac but beam heating in the production target is also limiting.

The nuclei of interest are produced as beam fragments and thus have a similar energy as the production beam, up to 350 MeV/nucleon. This creates a challenge in stopping the ions for collection. The gas-stopping cell currently being developed uses electric fields to quickly guide ions with a half-life under one second to another ion source for reacceleration. Thus, the beam current in the cell must be kept low so that plasma formation and space charge does not alter the electric fields. This is presently believed to keep the output of the gas cell down to 10^9 pps. This limitation however may be overcome for target production since one is interested in much longer-lived isotopes. Complicating this issue is the nuclear reactions the ions will undergo as they are stopped. This will increase the impurities in the collected sample. Exactly how this might be accomplished will require further development.

Neutron Source With Radiochemistry Facilities

At the heart of creating a target for neutron bombardment is radiochemistry. A 10 μg sample of a 1 day half-life isotope implies about 10 Ci of activity. The minimum amount of material needed will depend on the reaction rate, the measurement method, and the desired measurement accuracy. In most cases 10 μg should be enough to perform measurements, though sometimes less material maybe required. But there is also the possibility that other radioactive species will be collected in the sample. Thus, a radiochemistry lab will need to be designed to handle a hundred Curie sample of gamma radiation. Hot cells capable of handling 1kCi of gamma radiation are not uncommon. These hot cells would allow chemistry to be performed to purify the collected sample and form a target suitable for neutron irradiation.

Transportation to and from the radiochemistry lab is another important issue. Presently, transportation from the collection area to the radiochemistry laboratory will be done above ground via some lead container. Having some sort of underground transport system for this part has been considered, but given the different possible locations of isotope collection some means of above ground transport will be needed. It is planned to have an underground transportation system from the radiochemistry laboratory to the neutron irradiation areas.

Given the desired neutron energy range, 0.1-20 MeV, and the need for high fluxes, up to 10^{10} n/cm^2/s, a “monoenergetic”, tunable energy neutron source has been the pursued option. Several different production reactions are needed to reach the entire range and two different accelerators are used in order to maximize flux. For neutrons, below 200 keV either the ^{7}Li(p,n)^{7}Be or the t(p,n)^{3}He reaction can be used to produce a white source of neutrons. Both reactions have a negative Q value, -1.64 and -0.76 MeV respectively, which makes them ideal for producing low energy neutrons. ^{7}Be has an excited state at 429 keV, which makes it problematic to produce neutrons above several hundred keV. The t(p,n) reaction has no such issues. For neutrons above 3 MeV, the reactions d(d,n)^{3}He, t(d,n)^{4}He and X(d,pn)X can be used to generate “monoenergetic” sources of neutrons. The d(d,n) reaction has a 3.27 Q-value and works best for neutrons up to 10 MeV. Above this energy neutrons the deuteron breakup reaction become comparable. The t(d,n) has a 17.59 MeV Q-value and is used to make 14 MeV neutrons. The deuteron breakup reaction using high Z targets can also be used to produce neutrons and the neutron distribution is much more forward focused at similar neutron energies, especially the t(d,n) reaction.

To deliver the proton beams for low energy neutron production, a Dynamitron from IBA would be used [6]. The Dynamitron can accelerate high currents, up to 10’s of milliamps and down in energy to 50 keV. As designed, the Dynamitron delivers a DC current, but beam choppers have been with a Dynamitron to deliver a pulsed beam for neutron time-of-flight experiments [7]. The “monoenergetic” high-energy neutrons are produced via deuteron beam that would be generated from a 40 MeV linac. The linac would start with a 2 MeV RFQ and then continues to accelerate with DTL’s up to the desired energy. The DTL’s allow complete energy flexibility between 2 and 40 MeV. The expected beam current would be around 1 milliamp though others have done paper studies for a 4-milliamp machine [8]. Figure 3 is a drawing of the conceptual design for such a facility. In addition to showing the accelerators and radiochemistry facilities it also shows three experimental areas. One of the areas is for low energy neutrons and the other two are for high-energy neutrons with beam stops for the deuteron beam.

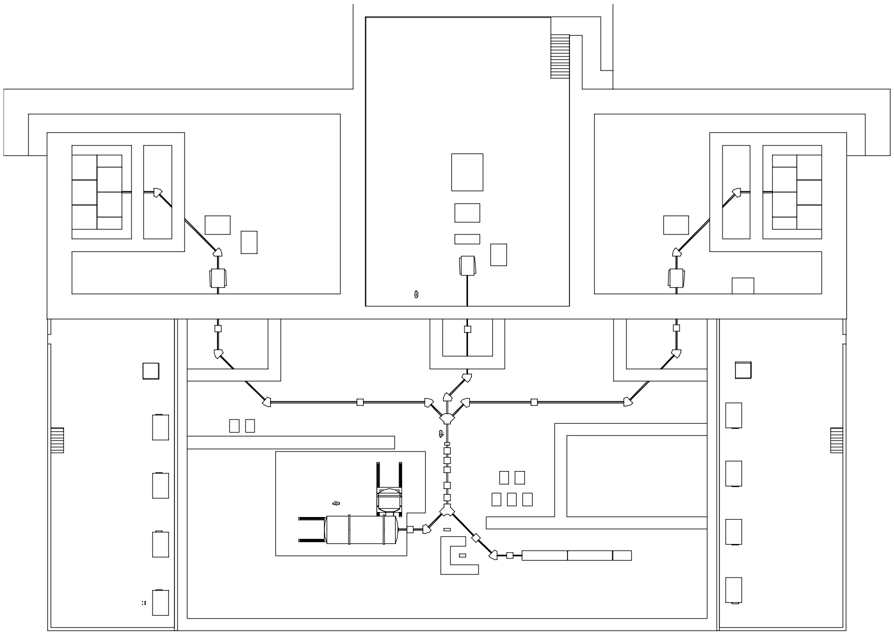

Figure 3: A drawing of a possible design for a neutron production facility at RIA. The experimental areas are up top, with a low energy neutron area in the middle. The two room on either side at the bottom are areas for radiochemistry. The dimensions of the entire facility are approximately 80 x 60m. See text for other details.

RIA R&D Issues

In addressing how stewardship measurements can be performed at RIA LLNL has identified several important RIA R&D issues whose resolution will effect how stewardship measurements will impact with the rest of the RIA community. These are more general RIA R&D challenges, whose solution will also impact how the RIA basic science missions are accomplished, and in particularly the number of simultaneous users at RIA.

During the RIA Experimental Equipment Workshop at Oak Ridge in March, there was some discussion as to whether a third ISOL target station would be beneficial. The optimum number of target stations will depend on many factors including, expected maximum beam power on target, need for target development, target change out time, and cost. Given the history of ISOL research and radioactive ion beam facilities in general, there will be significant need for target development beam time, which coupled with the expected change out time between targets argues for more than just two ISOL stations. TRIUMF currently has a three-week change out time, though it is believed this could be shortened to one week. But the most important specification is the expected maximum beam power on an ISOL target. If 100 kilowatts is the maximum, than the driver linac could deliver enough beam power for four ISOL stations. One could be reserved for nuclear structure experiments, another for astrophysics measurements, the third for experiments requiring no acceleration, and the last would be shared between harvesting and target development. This scenario would require the post-accelerators for the nuclear structure and astrophysics experiments to be decoupled, but otherwise the challenge is figuring out the mechanical layout. Even if 200 kilowatts is the expected maximum, a third ISOL station for harvesting and target development may still be desirable in part due to the expected change out time.

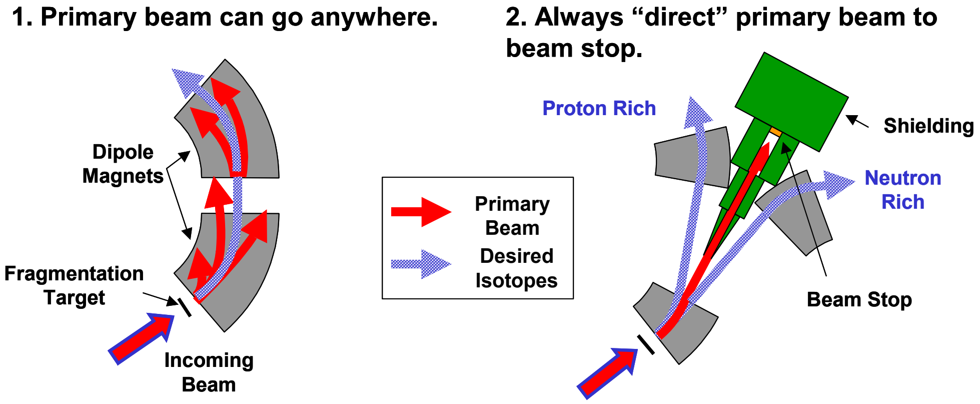

Figure 4: Diagrams of different concepts for handling the primary beam in the fragment separator.

There is a similar issue for the fragmentation line. Handling the primary beam in the fragment separator is difficult challenge that could be addressed in a number of ways. One plan has the primary beam deflected away from the desired isotope early and stopped before it reaches the first quadrupole. This could be problematic for harvesting many of the near stability isotopes, given the similarity in the charge to mass ratio to the primary beam. This could make it impossible to send the isotopes through the fragment separator and simultaneously stop the primary beam before the first quadrupole. Additionally, the primary beam is allowed to be in variety of locations, increasing the challenge of designing the beam dump and dealing with the neutrons generated when the primary beam hits the beam stop. Another alternative would be to always direct the primary beam to a specific location and continue the fragment separator on both sides of the primary beam. Figure 4 is a picture of what such a system might look like. This type of design would ease the design challenge of the beam dump, allow the possibility to

shield many of the critical fragment separator components, and might allow for more than one user from per fragmentation target. Determining which of the two options is the optimum will require a detailed design study involving beam optics, radiation hard engineering, thermal analysis, neutronics issues, mechanical design, and nuclear safety.
LLNL is very interested in the resolution to these two issues for the scientific reasons mentioned above. Resolving these issues will require a cross discipline analysis exploring the impact of many factors. LLNL has expertise in many of the required disciplines and is willing to contribute its resources in solving these important challenges to the RIA community.

Summary

Lawrence Livermore National Laboratory values the Rare Isotope Accelerator because it will enable improved neutron cross-section evaluations on short-live nuclei important stockpile stewardship. As such, LLNL has worked hard to generate support within NNSA and has recognized the need to invest its own resources in the pre-project phases of RIA. In part this is due to the need to enable many of the stewardship measurements, which require isotope harvesting, radiochemical processing, and neutron irradiation capabilities. LLNL is also willing to commit its own resources to solving some more general RIA R&D issues, many of which have been identified by LLNL as important in determining how stewardship measurements will be carried out. We are looking forward to continuing and strengthening our relationship with the rest of the RIA community.

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