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# Liquid-lithium cooling for 100-kW ISOL and fragmentation targets

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## Abstract

Advanced exotic beam facilities that are currently being developed will use powerful driver accelerators for the production of short-lived rare isotopes. Multi-beam drivers capable of producing high power beams from very light to very heavy ions are now technically feasible. A challenge for such facilities is the development of production targets to be used for a variety of reaction mechanisms with beam powers of about 100 kilowatts. This paper presents engineering concepts that have been developed recently for using liquid lithium coolant for two types of targets, one for use with light-ion beams on high atomic number (Z) targets and the other for heavy-ion beams on low-Z targets.

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## Introduction

Concepts are being developed for advanced exotic beam facilities that will use powerful driver accelerators for the production of short-lived rare isotopes. One of these projects was originally known as the Exotic Beam Facility [1] and is now known as the Rare Isotope Accelerator project, RIA [2]. Multi-beam drivers capable of producing high power beams of both very light and very heavy ions are now technically feasible [3]. A challenge for such facilities is the development of production targets to be used for a variety of reaction mechanisms with beam powers of over 100 kilowatts.

This paper presents engineering concepts developed at Argonne for using liquid lithium coolant for two types of targets, one for use with light-ion beams on high atomic number (Z) targets and the other for heavy-ion beams on low-Z targets. The high-Z target is used in a 2-step, neutron-generator configuration that can be optimized to maximize the yield of neutron-rich fission products in a nearby secondary uranium target. The 2-step geometry has the advantage of separating the primary beam power from the secondary ISOL target [4]. A large fraction of the primary beam power is dissipated in an appropriate target material such as tungsten that has a high neutron multiplicity when irradiated by high-energy light ions such as protons, deuterons, or helium-3. As discussed below, a low-Z primary target has a harder neutron spectrum at forward angles and, hence, may be preferable for the production of certain isotopes via a higher energy fission process. If the primary target material is either porous or fabricated with internal cooling channels, liquid lithium is an excellent medium for removing the deposited power. The secondary target can be a porous form of uranium carbide, as currently used for direct irradiation at existing ISOL facilities [5]. In the 2-step geometry the secondary target can be an annular shape surrounding the primary target. The secondary target is thus

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heated by a combination of fission power, heat from the primary target, and possibly an external heater, as necessary for operation at the required temperature above 2000 Celsius.

Low-Z targets for heavy beam fragmentation are typically beryllium or graphite. Lithium, being even lower in Z, is also appropriate for this application. Small diameter, high power heavy ion beams such as uranium produce very high power densities in these targets, more than a megawatt per cubic centimeter. Again, flowing liquid lithium is an excellent medium for carrying away this power.

These concepts and the required hardware are described in more detail in the following sections.

## Target requirements for next-generation exotic beam facilities

A next-generation exotic beam facility such as RIA, which is based on a high-power, multi-beam driver, will use a variety of isotope production mechanisms. Figure 1 illustrates some target concepts that are being developed for use with the driver beams that range from ions as light as protons to as heavy as uranium. Preliminary engineering analysis of these concepts indicates that they are all compatible with primary beam power of 100 kW or more. Two of the schemes shown in Fig. 1 use liquid lithium as the target for fragmentation reactions and one uses liquid lithium as the coolant for a neutron-generator configuration, both as discussed above. The fourth scheme is a large-area, tilted foil to be used as an ISOL-type target for direct irradiation with light ions. The tilted-foil concept reduces the thickness of the target material for isotope diffusion and thermal conductivity relative to the thickness as seen by the beam for isotope production. The large area of the tilted foil also reduces the power density (in power per unit area) to a level consistent with radiation cooling (~100 watts per square centimeter). A fifth scheme for a high power ISOL target, not illustrated here, uses stacks of thin refractory-metal foils. Development of this type of target has been carried out by the RIST collaboration [6]. High power targets of refractory metal foils are also being tested at TRIUMF and progress reports were given at this conference [7, 8].

## Advantages of liquid lithium cooling

The advantages of liquid metals as coolants in a variety of applications have been recognized for many years. A closed loop of liquid lithium to provide a windowless target for a high power (d,n) neutron generator was proposed in 1977 [9]. A high-capacity lithium loop was constructed and successfully operated for 2 years at Hanford to evaluate systems and components for potential future use in a fusion-materials test facility [10]. Advanced fast fission reactors, such as EBR-I and EBR-II at Argonne and the Phenix and Super Phenix reactors in France, use liquid sodium coolant [11]. A lead-bismuth alloy has been used for many years as the coolant in several Russian reactors [12]. Liquid gallium systems have been designed and operated at the Argonne Advanced Photon Source for the cooling of silicon crystals irradiated with high brightness X-ray beams [13]. The Spallation Neutron Source project at Oak Ridge National Laboratory will use a liquid mercury target designed for 2 MW of 1-GeV protons [14].

Lithium has several physical properties that make it generally useful as a coolant [15]. First, the melting point is 181 °C so that it flows well at 200 °C where it has a very low vapor pressure ($<10^{-8}$ torr). The boiling point, where the vapor pressure is 1 atmosphere, is 1336 °C. Due to its good heat capacity, ~4 J/g-K, and relatively wide working temperature range, lithium can carry away high heat loads at fairly low mass flow rates. For example, 100 kW of power can be carried away by liquid lithium at a flow rate of 200 g/s with a temperature rise of 125 K. The pump tested at Hanford [10] had 100 times this flow capacity. The liquid lithium pump at the Argonne ALEX facility has a similar large capacity [16]. Pumps for a wide range of flow capacity for liquid lithium and other metals are readily available; specific examples are discussed in the next section.

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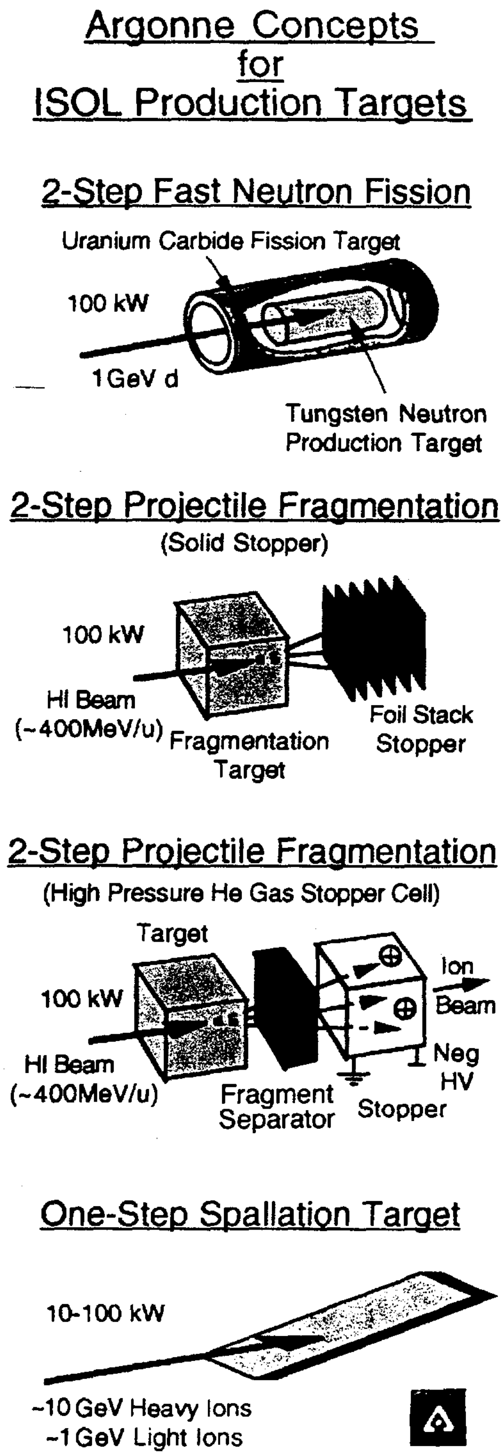


Fig. 1 Several concepts for high power targets.

In some applications, such as the 2-step neutron generator target discussed below, the lithium is used as a heat exchange fluid. The primary beam power is deposited in another material such as tungsten. In these applications the high heat transfer rate of the liquid metal is one of its important attributes. Heat-transfer rates up to 10 $kW/cm^2$ at reasonable flow rates and at pressures less than 1 atmosphere are attainable. It is the high heat transfer coefficient of liquid gallium that made it attractive for cooling synchrotron radiation optical components [13].

Liquid lithium has an excellent combination of properties for use as a target for heavy ion fragmentation. Its low atomic number gives more atoms in the productive range of the primary beam, as well as, lower multiple scattering of the secondary isotopes than higher atomic number targets. At achievable linear flow velocities, even with power densities over 1 $MW/cm^3$, the peak temperatures and corresponding vapor pressures are low enough to be used as windowless targets directly in the beam line vacuum.

## Components of liquid lithium systems.

**Materials and corrosion.** Studies, such as those at Hanford [10], have shown that stainless steel can be used as the primary structural material for liquid lithium cooling loops operating at temperatures up to about 400 °C. More recent considerations of corrosion and chemical compatibility of stainless steel and vanadium alloys for use in fusion reactors were reported in [17]. Corrosion loss rates of stainless steel in contact with liquid lithium are about 2 and 20 microns per year at 400 °C and 500 °C, respectively. The loss rates of the vanadium alloy reviewed in [17] are about a factor of ten lower than stainless steel. Titanium is also expected to have low loss rates. On the other hand, solubility of graphite in lithium seems to preclude its use as a window material in liquid lithium targets. Based on limited available data [18, 19, 20], beryllium may be useful as a window material, but corrosion loss rates need to be more carefully measured to determine its useful lifetime in these applications. Refractory materials, such as tungsten, are expected to have long lifetimes in contact with liquid lithium at these temperatures.

**Pumps.** Practical pumps for closed-loop liquid lithium cooling applications are available in a large range of capacities. Pressure-versus-flow-rate load-line curves for several types of pumps are shown in figure 2. The available pumps are useful for heat loads as small as a few watts to as large as several megawatts. Some of the pumps are available commercially [21,22] and others are adapted from commercially available linear actuators [23]. Liquid-metal pumps can be either standard mechanical rotary types [21], as used in the ALEX facility at ANL [15], or Lorenz-force pumps [22,23], such as used in the Hanford tests [10]. Rotary pumps must use motor windings, bearings and other mechanical components compatible with the elevated operating temperatures (~200-400 °C) and chemical properties of the specific liquid metals to be pumped. Lorenz-force pumps can be DC, single-phase AC, or three-phase AC. Several DC pumps for liquid gallium have been developed at the Argonne Advanced Photon Source [24]. These pumps use permanent magnets to apply a magnetic field perpendicular to the tubing that contains the liquid gallium and a high-current DC power supply to provide an electrical current perpendicular to both the tubing and the magnetic field. The resulting Lorenz force pushes the liquid gallium through the tubing. Single-phase AC conduction pumps are very similar except the applied magnetic fields and currents are provided via two sets of AC coils, one producing a transverse current and the other a transverse magnetic field. One of the pumps used in the Hanford lithium loop [10] was of this type. Three-phase AC pumps can be configured in an annular, linear-induction geometry [10,23]. These pumps have a very simple configuration with a set of three-phase AC coils producing both an AC magnetic field and an induced current that interacts with it to provide the Lorenz force on the fluid. The large, main-loop pump at Hanford [10] was of this type.

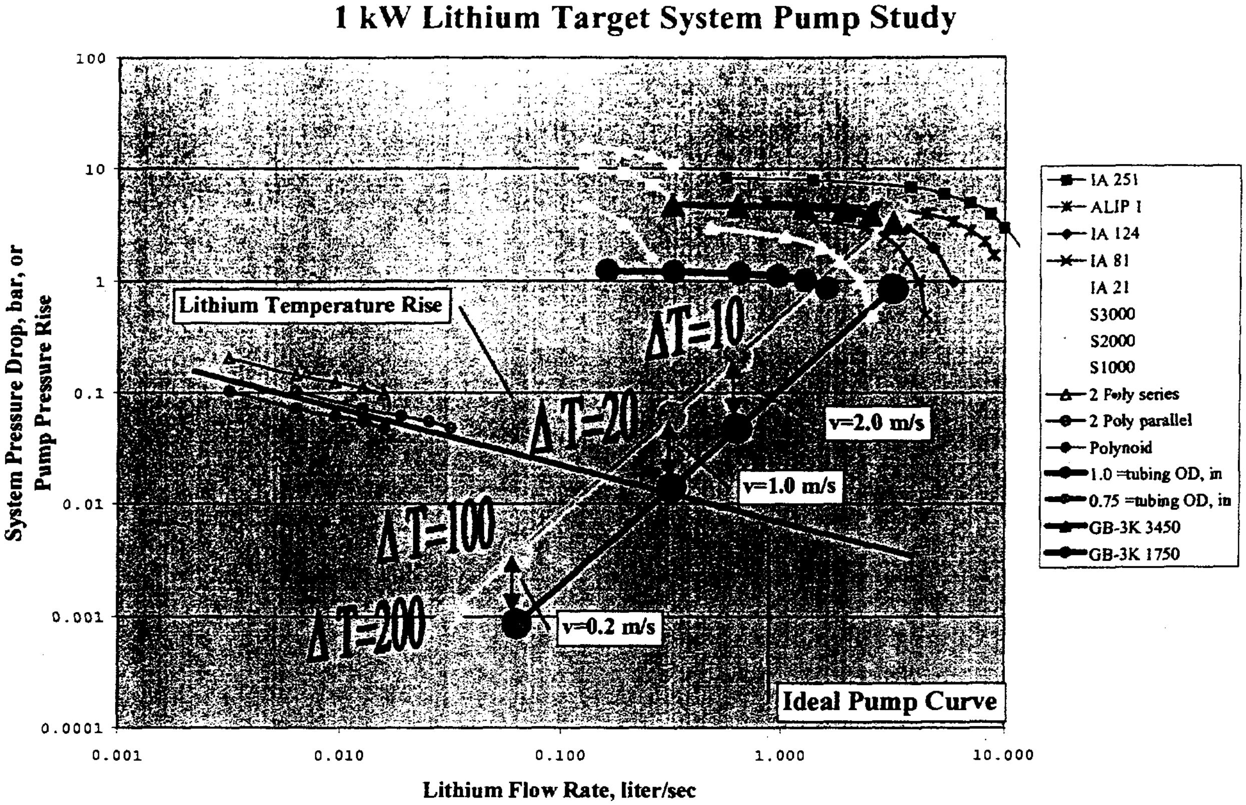


Fig. 2. Pump load curves, pressure drop vs. flow rate, for several types of liquid lithium pump.

## Contaminant control.

Liquid lithium loops used in targets of exotic beam facilities must have provisions for the control of both stable and radioactive contaminants. In the Hanford test experiments [10] a chemistry loop with both cold and hot trap branches was used for this purpose. It was demonstrated, for example, that hydrogen could be maintained at the 100 weight parts per million or less through the trapping of lithium hydride by a cold trap consisting of a stainless steel mesh. The hot trap, consisting of hot titanium foils, was used to remove reactive contaminants by gettering. The build up of contaminants such as tritium was not considered to be a problem.

## Safety.

Working with alkali metals in the laboratory requires following well established safety procedures. Lithium can react violently with water and will spontaneously combust if exposed to air at high temperatures. However, with proper precautions, many liquid-lithium cooling loops have been properly designed and operated safely. Table 1 lists standard procedures such as are used at the ALEX facility at Argonne [15].

**Table I. Safety Guidelines for Lithium Target Systems**

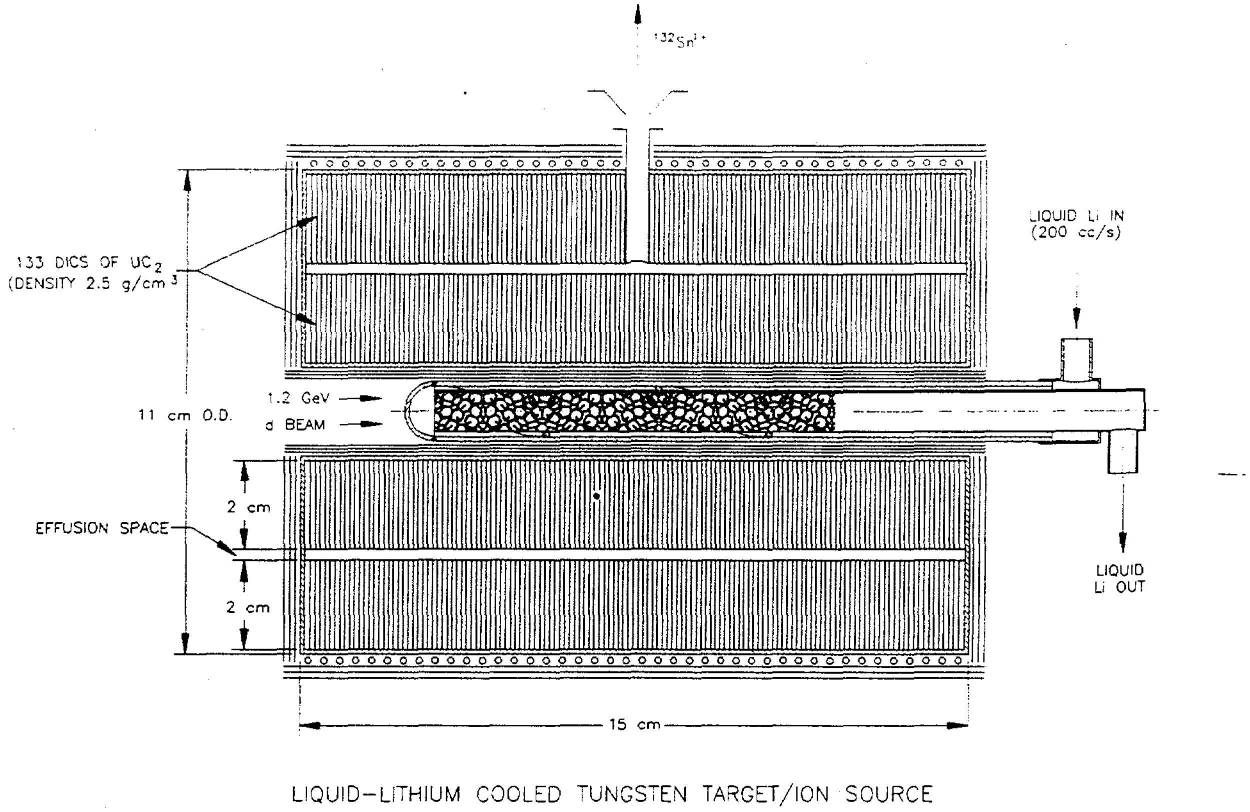
- Alkali metal safety training of all persons assigned to the operations
- Emergency planning including posted shutdown procedures for equipment
- Emergency equipment and supplies including protective clothing, respiratory protection, and fire fighting items
- Protective equipment to prevent injury to personnel if a leak or spray is possible
- Secondary containment of lithium
- Physical containment or ventilation/filtration system for protection of the environment in the event of a leak preventing a copious release of oxide and hydroxide smoke
- Leak, fire detection, and fire suppression systems
- Safeguards for prevention of entry of water or other incompatible materials into the lithium system
- Built-in protection for apparatus and equipment that is run unattended, including automatic shutoff in event of malfunction
- Posting on the entry doors of rooms containing more than one pound of lithium.

## Liquid-metal-cooled, Two-Step Target

A high flux of neutrons can be generated by using a high-Z target such as tungsten irradiated by high-energy light ions such as protons or deuterons [25]. The geometry for producing the most useful flux of secondary neutrons depends on the specific application. Producing the most neutron-rich fission fragments requires a compact coaxial geometry such as shown in Fig. 3. Using liquid-metal cooling of the core of this target is useful in keeping the assembly compact while thermally decoupling the primary and secondary target components. The high neutron multiplicity of tungsten, combined with its high density, make it a good choice for a compact primary target [25], while lithium is a good choice of coolant for the reasons stated above. The studies summarized in reference [26] show that $10^{14}$ fissions per second are generated in the secondary uranium carbide target with ~100 kW of protons or deuterons at 1-GeV beam energy. The fissions induced by secondary neutrons in the few-MeV energy range are the most effective in producing the most neutron-rich fission products. The optimal primary/secondary target geometries for producing fission products for elements outside the standard asymmetric fission mass regions will be different since such isotopes are produced better by fission induced by higher energy neutrons. The forward-peaked high-energy neutrons produced via the breakup of energetic deuteron beams on low-Z targets can be used for this purpose [4, 27-32]. At low deuteron beam energies the 2-step geometry has been used in ISOL laboratories [33] because fission yields are increased due to the larger useful uranium target volumes.

## Windowless target for heavy ion fragmentation

Several laboratories around the world use the heavy-ion fragmentation reaction mechanism to produce secondary beams of exotic isotopes. There are many papers at this conference and its predecessors based on this method. To date, the intensities of the primary heavy ion beams, usually in the 100 to 1000 MeV per nucleon energy range, are typically low enough to permit use of low-Z solid targets such as beryllium and carbon. At the highest intensities in use today rotating wheels of the target material are used in order to spread out the beam power. This is especially important in systems such as SISSI at GANIL [34], where very small diameter beam spots are used, thereby increasing the deposited power density. However, for future facilities, at even higher primary beam currents and power densities, a windowless liquid lithium target may be a very attractive solution.



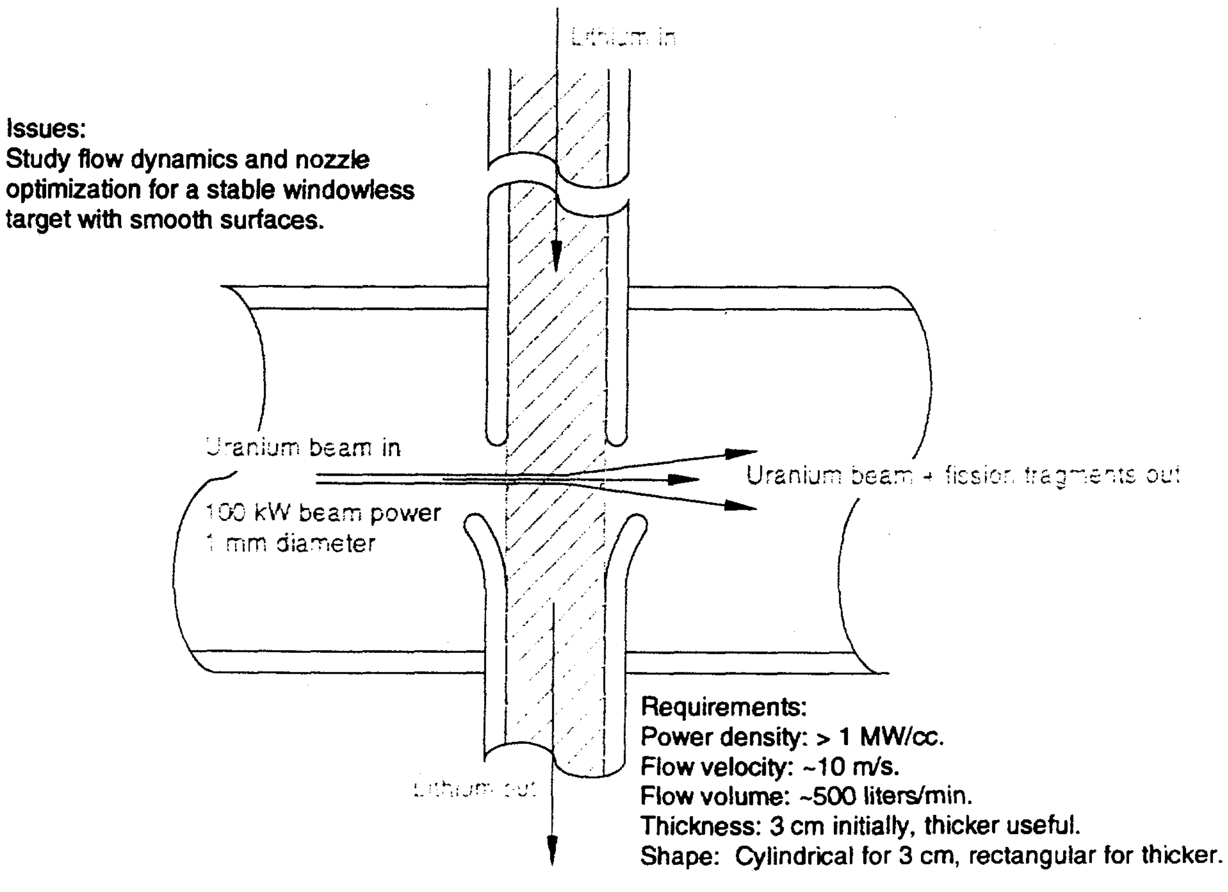
LIQUID–LITHIUM COOLED TUNGSTEN TARGET/ION SOURCE

Fig. 3. A schematic drawing of a 2-step, neutron-generator target. A high-energy deuteron or proton beam passes through the primary, neutron-generating high-Z target in the center. The secondary, uranium carbide target is cylindrically symmetric and surrounds the primary target. Spallation and breakup neutrons induce fission reactions in the secondary target. The primary target is thermally decoupled from the secondary target and is cooled by a liquid lithium heat exchanger fluid in this example. The secondary target is coupled to an ion source as indicated schematically at the top.

For a 100-kW uranium beam, as proposed for RIA [2, 3] on a liquid-lithium target with a 1-mm diameter beam spot, the power density in the target is over 1 MW/cm$^3$. The power density in a solid beryllium target, under the same conditions, is four times higher due to the higher density of beryllium. The power density, the circumference of the wheel, and the thermal conductivity of the beryllium determine the temperature rise in a rotating wheel of beryllium. To be a viable option, the rotating wheel must have a large circumference, about one meter or more, and be internally cooled by a liquid or gas at a distance within about a centimeter of the beam spot. Thermal radiation from the surface of a rotating wheel to an external surface would not keep the temperature below the melting point of beryllium. The dependence of properties such as the thermal conductivity of beryllium on radiation damage induced by the heavy ion beam would have to be measured to determine the long-term behavior or lifetime of such a wheel. Similar reservations apply to the use of a rotating graphite wheel. Even though thermal radiation can be effective in cooling graphite, its much lower thermal conductivity would lead to an unacceptable internal temperature drop between the beam-heated portion of the graphite and the radiating surface. The thermal conductivity of graphite also decreases further with beam-induced radiation damage [35]. In summary, rotating wheels of either beryllium or graphite are viable solutions for high-power heavy-ion fragmentation targets up to some power density, but a windowless flowing liquid-lithium target is probably the best choice for the highest intensities. Resistance to thermal shock and radiation damage are very desirable features of the liquid targets.

A concept for a windowless liquid-lithium target is shown in Fig. 4. The concept is similar to the lithium target system constructed and tested at Hanford [10] except that it is windowless on both sides rather than just one side. The Hanford target was designed for a high-power deuteron beam to enter on the windowless side, but only for neutrons to exit through the side with the window. High power heavy-ion beams that must both enter and exit the fragmentation target necessitate the use of windowless liquid targets. It is the thermal-conductivity-limited temperature drop between the outside surface of a window and the inner, liquid-metal-cooled surface that leads to this conclusion. At lower power densities a hybrid target, e.g. with liquid lithium flowing between beryllium windows, is a viable alternative. The 3-cm thick lithium target flowing at 5-20 m/s, as shown in Fig. 4, is appropriate for a 100-kW, 400 MeV per nucleon uranium beam as discussed above. Figure 5 shows the simulated peak temperature within the beam spot area for the examples of 10 and 20 m/s flow velocities. Lower velocities, leading to peak temperatures as high as ~600 K where the lithium vapor pressure is ~$10^{-6}$ torr, can be used with minimal evaporation of lithium into the beam line. The computer code HIJET, developed to study the behavior of moving liquid metal targets during high-power beam irradiation [36], was used for these simulations. This code is part of the general multipurpose HEIGHTS computer simulation package, which was designed to study the various effects of energetic beams or plasma particles in composite and heterogeneous target systems [37]. Hydrodynamic studies and nozzle development for windowless liquid-lithium targets are part of the R&D plan described below.

Concept for 3 cm thick windowless flowing liquid lithium target



**Issues:**
**Study flow dynamics and nozzle optimization for a stable windowless target with smooth surfaces.**

**Requirements:**
**Power density: > 1 MW/cc.**
**Flow velocity: ~10 m/s.**
**Flow volume: ~500 liters/min.**
Thickness: 3 cm initially, thicker useful.
Shape: Cylindrical for 3 cm, rectangular for thicker.

Fig. 4. Concept for windowless liquid-lithium target for fragmentation of heavy ion beams.

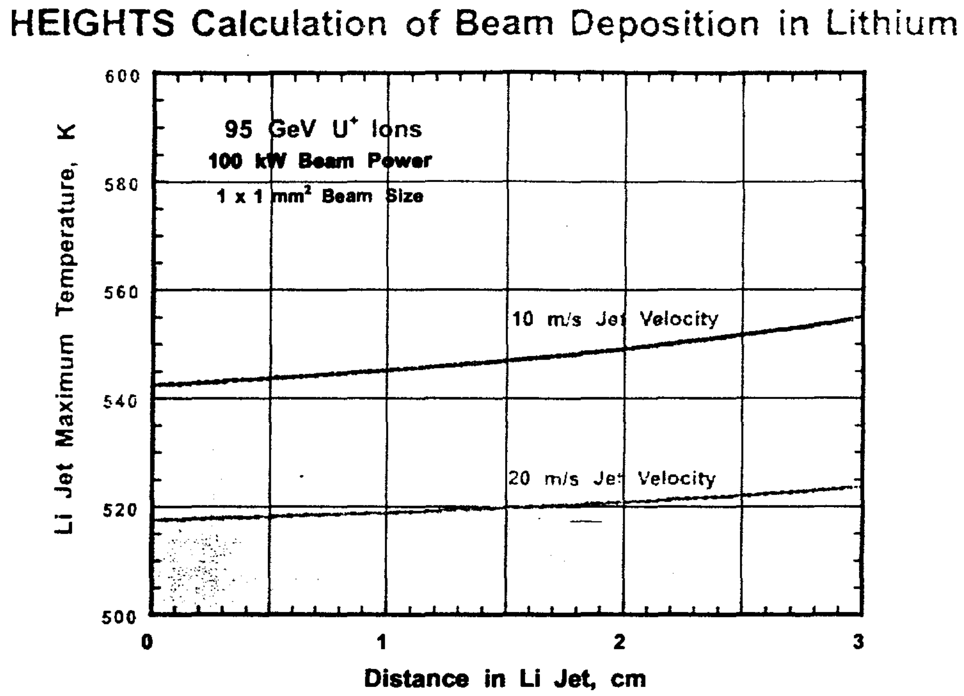


Fig. 5. Peak temperature within a flowing lithium target due to beam energy deposition at the two indicated lithium flow velocities.

## R&D plans

This paper presented the rationale and preliminary engineering concepts and simulations for liquid-lithium-cooled targets for use in the future at high intensity exotic beam facilities such as RIA. These concepts require further design optimization and testing, both off-line and in-beam, to develop an experience base prior to their use at full power. The present plan in the United States for such R&D during the coming three years is outlined in Table II. An additional need is to develop concepts for fragmentation targets for use with lower-Z beams at ~400 MeV per nucleon. For such beams the use of pure lithium is not desirable because its low density leads to target thicknesses of 10 cm or more. Such large thicknesses reduce the resolving power of the fragment separators that must sort and purify the secondary fragment beams. Options for reducing the target thickness by using a hybrid Be/Li target, a rotating wheel target, or a higher density liquid metal such as gallium will have to be evaluated.

**Table II. RIA R&D Projects for Liquid-Lithium-Cooled Targets**

1. Small liquid lithium loop for use with 3-kW, 200-MeV/u Ca, Ar, and lighter beams at MSU/NSCL.
    - Designed by MSU/ANL collaboration.
    - Commission at ANL Technology Development Division liquid lithium “ALEX” facility.
    - Install and use on-line at the NSCL A1900 after the intensity upgrade.
    - Parameters: 1-kW power in target, 1-3 cm Li, 2 thin beryllium windows, 5 mm wide channel, 1 m/s flow velocity, 100 cc/s volume flow (1.5 gal./min.).
2. Develop windowless liquid lithium target for use with 100-kW, 400-MeV/u uranium beams.
    - Use existing ANL “ALEX” facility to test hydrodynamics of 3-cm thick target at 10-m/s flow velocity.
3. Develop and test liquid-lithium-cooled tungsten target for 2-step, neutron generator configuration.
    - Design for 100-kW, 1.2-GeV deuteron beam.
    - Test with beam power at ORNL/ORELA facility or LANL/LANSCE facility.

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