# **R&D OF LIQUID LITHIUM STRIPPER AT FRIB\***

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## Abstract

Charge stripping is one of the most important processes for the acceleration of intense heavy ion beams, and the charge stripper greatly affects the performance of the accelerator facility such as the final beam power. In this paper, the design method and the achieved performance of the liquid lithium stripper recently developed for FRIB will be reported.

## **INTRODUCTION**

The Facility for Rare Isotope Beams (FRIB) at Michigan State University is a 400 kW continuous wave (CW) high intensity heavy ion linear accelerator (linac) facility for the study of rare isotopes that don't exist on the earth [1]. A charge stripper is used in the FRIB driver linac as a critical device that allows increasing the energy gain of ion beams by a factor of 2. Charge stripping occurs at energies of 17-20 MeV/u, where the beam power will be 40 kW during full power operations. No solid materials that are available today can serve as a long-life stripper under these severe thermal and radiation conditions. Therefore, FRIB has developed a state-of-the-art self-replenishing liquid lithium charge stripper (LLCS) to overcome the technological bottlenecks [2].

In this paper we describe how we designed the FRIB LLCS system and report achieved performance. It would be worth noting that the process described in this paper could generally apply to other liquid lithium systems or even other fluid systems in which performance testing under operating conditions with the actual fluid is costly (both time and money) and simulation experiments are highly desired to test critical performance before the actual system is built.

## **DESIGN PROCESS**

#### Conceptual Design

The LLCS concept was proposed by Nolen [3]. There were critical but unknown performance of the LLCS concept in the beginning; how could a stable and uniform thin (0.5-1 mg/cm<sup>2</sup>) liquid lithium film in a high vacuum environment be formed?; could the film withstand the foreseen extreme thermal load (56 MW/cm<sup>3</sup>) imposed by the full power uranium beam?; and could charge stripping charac-

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teristics of the lithium film be acceptable for further acceleration of stripped beams? The first one: formation of a stable and uniform thin liquid lithium film was considered to be the most critical. To validate such a liquid lithium film, a series of R&Ds began at Argonne National Laboratory (ANL).

There are various methods to form a liquid film. One method is that when a round liquid jet impinges on the edge of a solid surface, a thin film is formed in the downstream (e.g. tap water from a faucet impinging on a spoon). This method was pursued at ANL.

It is not common to build a liquid lithium system until critical system performance is validated and there remain only validations that can be performed with liquid lithium. This is because liquid lithium systems are normally complicated thus expensive. The systems need heaters to melt solid lithium (the melting point of lithium is 180.5°C) and keep it liquid, heater controllers and temperature sensors to keep the temperature within a desired operating range. Tanks or vacuum chambers in liquid lithium systems are usually filled with argon or maintained under high vacuum to provide an inert environment. Otherwise the liquid lithium will be contaminated. For example, tanks / vacuum chambers must not be filled with air or nitrogen because lithium reacts with them, which could cause a lithium fire. Thus, dedicated sub-systems that provide an inert environment in the tanks / vacuum chambers are also required. To mitigate and prevent abnormal events, safety controls are required too.

To efficiently test concepts, usually water is used as a simulant of liquid lithium. The law of similarity guarantees that two different types of flow become similar when these flows are properly scaled and dimensionless numbers that are relevant to these flows (e.g. the Reynolds number) are the same. The dimensionless numbers that are relevant in this case are the Reynolds number and Weber number. The Reynolds number is defined as

$$\operatorname{Re} = \frac{LU}{\nu},\tag{1}$$

where L is the characteristic length, U is the characteristic speed, and v is the kinematic viscosity.

The Weber number is defined as

We = 
$$\frac{\rho L U^2}{\sigma}$$
, (2)

where  $\rho$  is the density of the liquid,  $\sigma$  is the surface tension between the liquid and the gas.

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To use the law of similarity, one must know a ballpark estimate of the Re and We numbers of the lithium flow that needs to be achieved. As seen in Eq. (1) and (2), the lithium temperature that determines  $\nu$ ,  $\rho$  and  $\sigma$ , the characteristic length (required film thickness in this case), and the characteristic speed (required jet speed in this case) determine the Re and We numbers. The lithium temperature should be as low as technically possible to reduce its chemical activity. The required film thickness comes from the requirement of the charge stripping performance, and the required jet speed should be high enough to efficiently remove the heat deposited by the beam.

It would be worth noting that a similar approach was taken during the conceptual design of the liquid lithium target of the International Fusion Materials Irradiation Facility (IFMIF) [4].

The water simulation experiments at ANL were aimed at experimentally obtaining a so-called stability diagram in the Re-We domain, in which stable regimes of a flow (there could be more than one stable regime) are depicted and visualized. Figure 1 shows the water film stability diagram obtained at ANL [5]. Once the diagram is obtained, one can expect the conditions under which a stable liquid lithium film can be made. Therefore, one can proceed with the next step: the proof of principle test. The experiments also revealed that not all the water that impinged on the deflector formed the free jet, but some remained on the deflector, and it interacted the film when it dripped off the deflector, disturbing the film. To resolve this issue, the original circular edge shape of the deflector was made concave such that the water on the deflector is guided along the edge toward the lowest points that are located away from the jet impingement point. Figure 2 shows photos of the water film made during the experiments [6].



Figure 1: Film stability diagram in the Re-We domain obtained with water as a simulant of liquid lithium. Another simulant, FC-3283, was also used. The inserted labels are scales to the parameters of lithium thin film [5].

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Figure 2: Photo of water film formed during the experiments [6]. The left shows the original deflector and the right shows the improved deflector.

## Proof of Principle Test

This proof of principle (PoP) test stage can be still considered as the conceptual design stage. In some cases, PoP tests may fail. Then, the concept must be re-considered and improved.

Usually at this stage, a liquid lithium system is made to test the concept(s) as a PoP test. But in general, it is still not worth building a full scale final system because the concept has not yet been tested with the real fluid: liquid lithium. At this stage, the most critical concepts that are yet to be validated and are the key to determine the success, should be focused.

After stable water film jets were formed and a stability diagram was obtained at ANL, a liquid lithium system was built to demonstrate a stable liquid lithium film. The system built was not equipped with a circulation pump. Instead, the system was equipped with a pressure vessel where liquid lithium was able to be pressurized by argon, which drove the flow. Because of this configuration, the test could not continue for more than 20 minutes; the vessel needed to be re-charged with liquid lithium by transferring the lithium back into the pressure vessel and then the vessel needed to be re-pressurized. This approach was taken because an optimal drive pressure to form a stable liquid lithium film jet was still unknown. The development of a circulation pump for continuous operations was spared for a future task.

The investigations at ANL revealed that the hydrodynamic instability could be overcome by flowing the liquid lithium film jet at >50 m/s (corresponding drive pressure of 1 MPa). A roughly 10 µm thick, 1 cm wide, stable lithium film jet was successfully formed in vacuo [5, 6]. The high flow velocity was not only for the hydrodynamic stability but also necessary to carry away the intense beam power to avoid boiling or excessive vaporization. The resolution for the liquid dripping issue was further improved by adding so-called wicks to the lowest points of the deflector to facilitate the drainage of remaining lithium. Fig. 3 shows the liquid lithium free jet that was made with

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the improved deflector [7]. With this result, it was concluded that a stable liquid lithium free jet was able to be made with this method.

The film thickness was measured with a 30 keV electron beam by measuring a transmission rate of the beam [6]. It was found that the film has a thickness distribution both in the streamwise and spanwise directions. The thickness where beams are expected to pass through the film was 0.632 mg/cm<sup>2</sup> (12.3 mm).

With these results, the first and most critical performance: formation of a stable and uniform liquid lithium film in a high vacuum environment, was validated.



Figure 3: Liquid lithium film jet formed with improved deflector [7]

Next, the thermal performance of the lithium jet was tested with the LEDA (Low-Energy Demonstration Accelerator) proton source borrowed from Los Alamos National Laboratory (LANL) [7]. A 65 keV proton beam was used. At this energy the protons stop within the first 1.5 mm of the lithium film. The experiment demonstrated that the velocity of >50 m/s was sufficient to carry away 300 W of the thermal power deposited in the lithium film within a 1 mm diameter beam spot and a thickness of the first 1.5 µm over the total thickness of 10 µm. The estimated peak volumetric heat input from the proton beam was approximately 65 MW/cm<sup>3</sup>, more than the FRIB average power density deposition (56 MW/cm<sup>3</sup>). Also noted is the 300 W power deposition was more than half of the total FRIB's power deposition of 450 W. This experiment did not include the radiation damage that the heavy ions would have on a solid carbon foil, but with a self-replenishing liquid the lattice damage is not an issue. Figure 4 shows a photo of the lithium film receiving the 300-W 65-keV proton beam [7]. With this result we confirmed the superior thermal performance of the windowless liquid lithium free jet; however, the final unknown performance: actual charge state distributions after stripping remained unproven and had to wait until the tests in the FRIB linac.



Figure 4: Lithium film jet receiving 300-W (65 MW/cm<sup>3</sup>) 65-keV CW proton beam [7]

#### Detailed Design

Based on the achievements at the PoP test stage, the detailed design was initiated. It is obvious that every component necessary to meet requirements for operations must be designed and fabricated at this stage. The main focus was the lithium circulation pump development and safety control measures. There were many other less critical but important considerations. A few selected important notes were added to the last part of this section.

The pump was designed based on the design developed by Smither [7,8]. This pump is a DC electromagnetic (EM) pump with permanent magnets, utilizing linear pressure build-up along the flow inside the pump. Since flow fluctuations that could be generated with an AC pump were a concern, a DC pump was selected. This pump is not for a high flow rate because of the magneto-hydrodynamic pressure drop due to a strong electro motive force (EMF). However, the EMF produces an induced voltage across the pump, which is proportional to flow rate. By measuring the induced voltage we can measure a flow rate without a separate flow meter. Performance test of the pump was performed with liquid lithium.

During the detailed design phase, a great amount of effort was made to design safety control measures to prevent and mitigate liquid lithium hazards. Liquid lithium reacts with air, and the reactions could lead to a fire depending on the conditions. It vigorously reacts with water producing hydrogen. Therefore the reaction with water must be prevented. The LLCS system at FRIB has three engineered safety control measures: a secondary containment vessel (SCV), an argon blanket in the SCV, and a beamline gate valve system (BGVS). Figure 5 shows a sketch of the FRIB LLCS system which includes the above three engineered safety control measures. The SCV is to provide an enclosed barrier between the lithium loop and the accelerator tunnel and to contain any lithium leaks within the controlled space. The argon blanket in the SCV is to provide an inert atmosphere in the SCV. With the SCV and argon blanket, in case lithium leaks out of the loop, it won't react with air. In case the vacuum chamber develops a vacuum leak, it

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will only end up being filled with argon. The BGVS consists of two beamline gate valves (upstream and downstream of the vacuum chamber) and redundant vacuum sensors. The BGVS is to isolate the vacuum chamber when a loss of vacuum is detected. The concern is vacuum leaks in the beamline outside of the LLCS system, which could introduce air into the LLCS vacuum chamber. Also, the BGVS does not allow operators to open the beamline gate valves when the linac tunnel is unsecured. This way a possibility of air leaks is eliminated when workers can be present around the LLCS system.

Maintainability is an important consideration. The nozzle and deflector are considered to be major components that need regular maintenance because they interact with high-speed liquid lithium flow, therefore they could be eroded. Corrosion is expected to happen, but because the operating temperature (220 °C) is close to the melting point (181 °C), the corrosion rate is expected to be very low. The nozzle and deflector are mounted onto the vacuum chamber using ConFlat flanges. The gaskets must not be made of copper because of its poor compatibility with liquid lithium. Steel gaskets are used in the FRIB LLCS system. Copper gaskets can be used where liquid lithium is not present.

As a main vacuum pump, a cryopump was selected. Among advantages of using cryopumps in a liquid lithium system, most notable is that they can operate in a closed environment unlike turbo molecular pumps, which always need an auxiliary backing pump, and cannot create a physical boundary between atmosphere and the lithium environment. Use of a turbo pump in a liquid lithium system would require a reliable gate valve that isolate the lithium environment from atmosphere in case of pump failures.



Figure 5: FRIB liquid lithium charge stripper system. The liquid lithium loop is completely enclosed by a secondary containment vessel as a safety control. Noted by red are safety controls.

# ACHIEVED PERFORMANCE

Overall, the developed LLCS has shown satisfactory thermal and stripping performance so far. The safety system has never been tripped without a few false positives

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during the optimization process in the early commissioning stage. The EM pump has been working well too. It has demonstrated continuous stable operations (for example, see [9]). Here we report achieved thermal and stripping performance of the LLCS. For details, please refer to our previous papers [2, 10, 11].

# Thermal Performance

As already mentioned, to simulate the thermal condition that will be created by the full power uranium beam, we irradiated the lithium film with a proton beam produced with the LEDA source. To test the thermal performance with a heavy ion beam, we irradiated the lithium film with a high intensity argon beam [2, 11]. The thermal performance of the film was measured with an <sup>36</sup>Ar<sup>10+</sup> beam at 17 MeV/u and a peak current of 12 particle- $\mu$ A, the highest peak current allowed within the present accelerator operational envelope. The beam duty cycle was set at 5.4% with the repetition rate of 10 Hz, resulting in the instantaneous peak beam power of 7340 W during each 5.4 ms period. The peak power loss in the lithium film  $(0.6 \text{ mg/cm}^2, \text{dE/dx})$  $=405 \text{ keV}/\mu\text{m}$ ) was 50 W. The estimated volumetric power deposition in the lithium during the high-power test reached 6.2 MW/cm<sup>3</sup>, or 11% of the FRIB full power operation value (56 MW/cm<sup>3</sup>). Since it took approximately 20 µs for the flowing lithium at 50 m/s to completely cross the beam spot of 1 mm, it was considered that the longest time constant of any thermal and fluid dynamic responses of the lithium flow was 20 µs. Thus the 5.4 ms long beam, which was 270 times longer than the longest time constant, may be considered well-representing a continuous beam. Figure 6 shows photos of the liquid lithium film jet stripping the high-intensity argon beam. During this test the beam parameters after the stripper were stable and the LLCS system parameters such as temperatures and vacuum pressure were also stable.



Figure 6: Lithium film jet stripping high-intensity pulsed argon beam (duty cycle 5.4%). Heat deposition to the lithium was estimated to be 50 W (6.2 MW/cm<sup>3</sup>). The LED light was turned off on the right photo to take a clear beam spot.

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As of now, we have conducted charge stripping tests with <sup>36</sup>Ar<sup>10+</sup>, <sup>48</sup>Ca<sup>10+</sup>, <sup>124</sup>Xe<sup>26+</sup>, <sup>238</sup>U<sup>36+</sup>. Table 1 summarizes the beam energies used. The root-mean-square (rms) beam radius was estimated to be 0.5 mm at the stripper. The commissioning tests included measurement of LLCS film thickness, measurement of charge states after the LLCS. Most of results have been reported in our papers [2, 11]. Charge stripping experiment data were compared with ETACHA simulations. And the measured data showed a reasonable agreement with simulations. The film stability was a great concern for stable beam operations, but it turned out that the film was stable enough for acceleration of stripped beams (the fluctuations of the beam energy after the stripper was less than 0.1% of the incoming beam energy) [2]. The lithium stripper has been used for a user operation [11]. Figure 7 shows new measurement data: 20 MeV/u <sup>48</sup>Ca charge state distribution after 1 mg/cm<sup>2</sup> lithium film. Measured charge state distribution of the same beam after a 1.5 mg/cm<sup>2</sup> carbon foil was also shown together.

Table 1: Heavy Ions Used During LLCS Commissioning With Heavy Ion Beams

<sup>36</sup> Ar <sup>10+</sup> 17 MeV/u & 20 MeV/u <sup>48</sup> Ca <sup>10+</sup> 20 MeV/u <sup>124</sup> Xe <sup>26+</sup> 17 MeV/u		Energy	Ions
<sup>48</sup> Ca <sup>10+</sup> 20 MeV/u <sup>124</sup> Xe <sup>26+</sup> 17 MeV/u	u	17 MeV/u & 20 MeV/u	$^{36}Ar^{10+}$
<sup>124</sup> Xe <sup>26+</sup> 17 MeV/u		20 MeV/u	$^{48}Ca^{10+}$
		17 MeV/u	$^{124}\mathrm{Xe}^{26+}$
<sup>238</sup> U <sup>36+</sup> 17 & 20 MeV/u		17 & 20 MeV/u	238U <sup>36+</sup>



Figure 7: 20 MeV/u <sup>48</sup>Ca charge state distribution after 1 mg/cm<sup>2</sup> lithium film. Charge state distribution of the same beam after 1.5 mg/cm<sup>2</sup> carbon foil is also shown.

#### SUMMARY

In this paper, the design process and achieved performance of the FRIB liquid lithium charge stripper was presented. The developed LLCS has shown satisfactory thermal and stripping performance so far. As FRIB ramps up its power towards the final goal of 400 kW, the performance of the LLCS at higher beam powers will be tested.

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