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MOLYBDENUM-99 TECHNOLOGICAL DEVELOPMENT

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SHINE: Technology and Progress

K.M. Pitas, G.R. Piefer, R.V. Bynum, E.N Van Abel, and J. Driscoll
SHINE Medical Technologies
2555 Industrial Drive, 53713 Monona WI – USA

T.R. Mackie
Medical Devices
Morgridge Institute for Research
330 N. Orchard Street, 53715 Madison WI – USA

R.F. Radel
Phoenix Nuclear Labs
2555 Industrial Drive, 53713 Monona WI – USA

ABSTRACT

The SHINE system employs an accelerator-driven, low-enriched uranium solution in a geometry optimized for efficient isotope production. Neutrons produced by DT reactions in the accelerator drive fission in the subcritical solution. The process produces medical isotopes compatible with the existing supply chain while eliminating the need for HEU and reliance on aging nuclear reactors. Janesville, Wisconsin has been chosen as the site of the first SHINE facility, which will supply over half of U.S. demand for molybdenum-99. Conceptual design of the facility is complete; the environmental analysis and report required to obtain an NRC license has been completed and submitted; completion of preliminary design and submittal of a preliminary safety analysis report is anticipated in April 2013. Accelerator prototype I has demonstrated 500+ hours of operation. Prototype II is operational and undergoing system reliability testing. Target solution chemistry has been selected; target geometry has been optimized and prototyped. ^{99}Mo separation at >97% efficiency has been demonstrated.

1. Introduction

Historically, the vast majority of the world's supply of molybdenum-99 (^{99}Mo) has been produced in aging research reactors using highly enriched uranium (HEU). Despite the U.S. constituting approximately half of world demand for ^{99}Mo, commercial quantities of the isotope have not been produced domestically since 1989. Now, as foreign research reactors become increasingly unreliable at the end of their lives, and nuclear non-proliferation efforts become more important by the day, the ^{99}Mo market is seeking reliable, economic, non-HEU-based ^{99}Mo production technologies.

In response to this need, SHINE Medical Technologies (SHINE) has developed a new, LEU-based, subcritical medical isotope production system. Since the first ^{99}Mo Topical Meeting in December 2011, SHINE has made strong progress toward its goal of supplying the U.S. market with over 3,000 six-day Ci of ^{99}Mo per week by 2016. This paper first gives an overview of the SHINE technology, and then details SHINE's regulatory, organizational, and technical progress toward producing ^{99}Mo since last reported in 2011. Details are often omitted due to proprietary restrictions, and this submittal will generally only describe the status of the project rather than the outcomes of particular development activities.

2. Technology Overview

From top to bottom, the device works as follows (see **Figure 1**):

a. At the top of the device, a microwave ion source creates a deuteron beam with a nominal total current of about 50 mA, and collimates it to approximately 1 cm^2.

b. The beam is accelerated by an electrostatic accelerator to approximately 300 keV.

c. The beam passes through differential pumping sections which serve to keep the accelerator at its working pressure, while maintaining sufficient target gas pressure to produce high neutron yield.

d. The target chamber is filled with tritium gas. When the accelerated deuterium ions enter the target chamber, they impact the tritium gas, resulting in nuclear reactions that create a total neutron production of greater than 10^{13} n/s.

e. The neutrons enter a multiplier layer which increases the source neutron population by 2–3 times before it enters the target solution vessel (TSV).

f. After multiplication, the neutrons enter the LEU target solution vessel causing ^{235}U therein to fission, creating more neutrons and fission products, including ^{99}Mo. The fission process causes internal heating of the solution and the heat is rejected through the TSV walls and off-gas handling system to coolant systems, allowing steady-state operation. ^{99}Mo accumulates in the solution during the irradiation. Hydrogen and oxygen produced by the fission process are recombined into water using a simple catalytic recombiner and piped back into the TSV.

g. The TSV is surrounded by light water which reflects neutrons back into the solution vessel, cools the system, and absorbs radiation produced by the device.

After several days of irradiation, the solution is removed from the TSV and the ^{99}Mo is separated, purified, tested, and packaged for shipment to technetium-99m generator manufacturers.

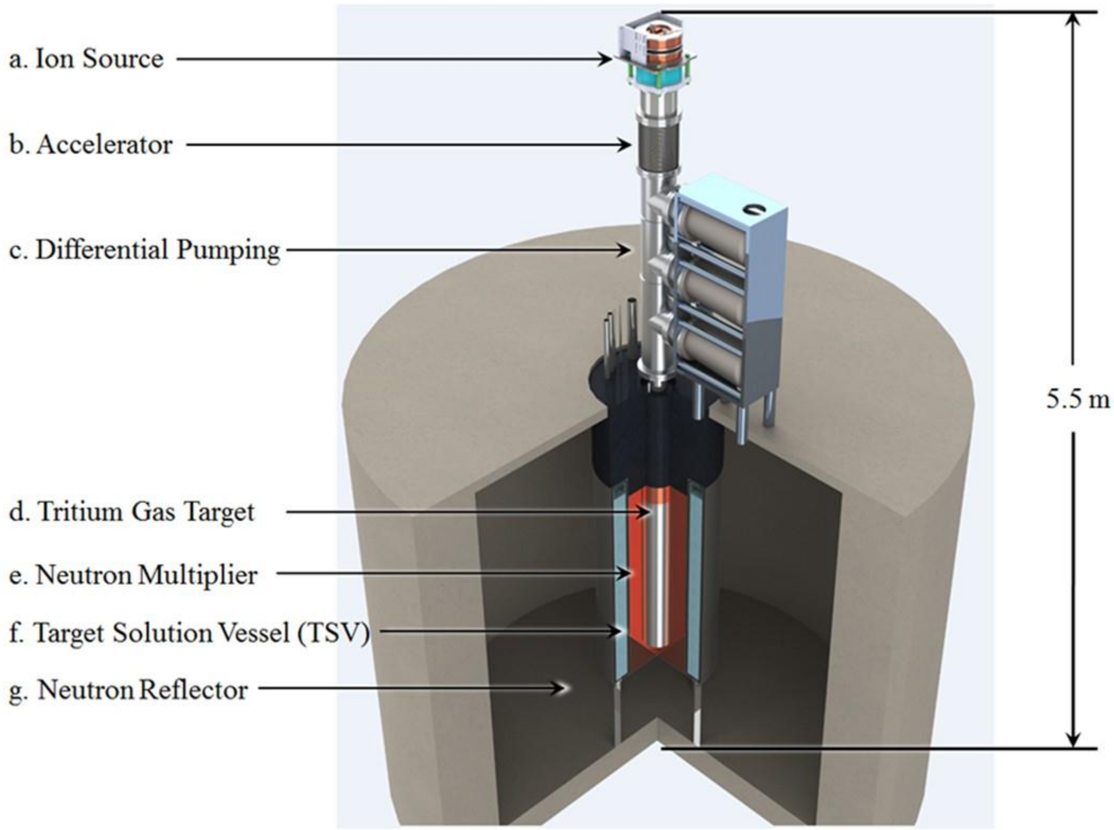

Figure 1. *The SHINE medical isotope production device.*

3. Facility Design and Licensing

Facility Design

Preliminary design of the SHINE facility is the cornerstone of the construction permit application, and has occupied the majority of SHINE resources over the past 12 months. Preliminary design includes system descriptions, mass balances physical layouts, shielding, structural design and seismic analysis, safety related system designs, selections of appropriate codes and standards, a preliminary integrated safety analysis and preliminary accident analysis, and many other elements needed to support a construction permit application.

Since the last meeting, conceptual design of the SHINE facility has been completed and preliminary design will culminate around the end of April, 2013 with the completion of the Preliminary Design Report. This document forms the basis for the Preliminary Safety Analysis Report (PSAR) that will be submitted to the NRC in support of our construction permit application.

Plant Licensing

SHINE has had substantial engagement with the NRC, including a number of pre-application meetings aimed at increasing understanding of SHINE's unique technology, and clarifying NRC expectations for the SHINE license application. Pre-application meeting topics have included:

- Technological approach overview,
- The content and structure of SHINE's license application,
- The application submission and review schedule,
- Facility design,
- The physics and design of the subcritical assembly,
- The subcritical assembly start-up and operational strategy,
- The security plan,
- Postulated accidents,
- Site selection,
- Environmental monitoring,
- Waste management,
- Transportation of raw materials, products, and waste
- Human health impacts of the facility.

The pre-application meetings also included a site visit by the NRC. During the site visit, the site selection process was discussed in greater detail, as well as the results of the Janesville and alternate site investigations.

On March 26th, 2013 SHINE submitted part one of the Construction Permit application (including an Environmental Report) to the NRC. The Environmental Report involved over 18 months of planning, data collection, and analysis of the selected site location in Janesville, WI and the surrounding area, culminating in an approximately 2,000-page document. This document serves as the basis of the NRC environmental review needed to grant a Construction Permit.

Part two of the Construction Permit application, including the majority of the Preliminary Safety Analysis Report (described previously) is currently undergoing final review. It will be submitted to the NRC by April 30th, 2013.

4. Organizational Progress

Since 2011, SHINE and Morgridge have worked to build an incredibly skilled and dedicated workforce, and has grown our in-house teams from 12 at the time of the last workshop to approximately 40 skilled individuals. Key experience has been added in the areas of nuclear operations and NRC licensing. In these particular areas, we have added:

- A plant manager—30 years of nuclear industry experience,
- An engineering manager—30 years of nuclear design/construction/operation experience, and 10 years of experience in first-of-a-kind plants,
- Licensing staff—approximately 100 years of combined nuclear industry licensing experience.

The team presently consists of nearly two dozen engineers, a half dozen quality and safety staff, and approximately another dozen administrative and support staff.

5. Technical Progress

Accelerator Development

Since 2011, significant testing and validation has been done on the particle accelerator that will create the neutrons to drive the ^{99}Mo facility, including reliability testing on prototype I (the unit presented at the last workshop) and commissioning of prototype II—the first plant-scale accelerator. Prototype II is a full-power unit designed to accommodate the needs of the SHINE facility.

Figure 2—Prototype II neutron source is a plant-scale device

Overall, reliability testing has been conducted on four distinct technological platforms: the microwave test stand (MST), prototype I (P-I), the ion source test stand (ISTS), and prototype II (P-II).

Operational reliability for the MTS, P-I, and the ISTS all meet or exceed the SHINE production facility requirements. Additional reliability information, such as mean time to service and maintenance downtime, are more difficult to determine from available data given the current development stage. However, preliminary results from the operation of the P-I and ISTS systems are encouraging. Demonstrated operational reliability parameters are summarized in Table 1.

	MTS	**P-I**	**ISTS**	**P-II**
Accumulated Run Time (hours)	100	500	200	20
Longest Continuous Operation (hours)	40	8	122	1
Mean Time to Shutdown (hours)	4	1	40	1
Uptime During Operation (%)	>99%	98%	>99%	>95%

Table 1: Operational reliability data on prototype neutron sources

While insufficient data has been collected at this point to completely ensure operability for months and years in the plant, significant degradation of the accelerator components has not been seen in the tests run so far. Therefore, major issues with accelerator reliability are not anticipated. Nonetheless, prototype II will be run extensively to provide as much reliability data as possible.

Prototype II

While most of the operational experience so far has been based in the predecessors to prototype II, preliminary results from this full-scale production system are encouraging. Prototype II was designed to deliver a 50 mA beam at 250-300 keV to a deuterium gas target for plant reliability testing, and beams at this power level are routinely created and focused. Figure 2 shows prototype II at the Monona facility, and Figure 3 shows a highly focused 50 mA beam passing through a ¼" diameter hole at the entry to the gas target.

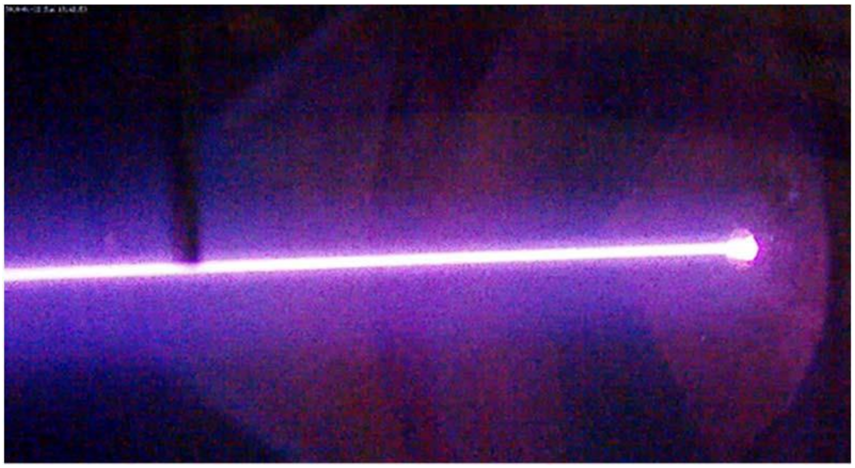

Figure 3—50 mA ion beam collimated through a ¼" aperture at the entry point of the gas target

The purpose of prototype II is to assess issues that may limit the operational lifetime of the neutron drivers at full power in the plant from the standpoint of beam operations. This testing is just getting underway, and the results are expected to lead to enhancements that will ultimately lead to a test platform that is identical to the plant configuration.

Neutron output in prototype II has not yet been measured at full power, but preliminary neutron results are expected in the next month. A license ammendment from the State of WI will be required in order to operate prototype II for extended time periods with deuterium gas as a target. In the meantime, a non-neutron deriving gas such as helium will be used. Long-term operational reliability from prototype II is expected by late this year.

Tritium purification

The accelerator creates neutrons by colliding a deuterium beam with tritium gas. An inherent side effect of this process is that the two species will eventually become mixed, and reduce the yield of the target system. In order to ensure high yield, the mixed species must be separated into individual species again real-time.

SHINE intends to use the TCAP process developed by Savannah River National Laboratory (SRNL) to ensure continual purification of the tritium gas target in the accelerator. TCAP is a simple, elegant process and has been used in production for a long time. SRNL has entered into a WFO agreement with SHINE to provide a TCAP unit for evaluation and it is expected that they will provide the initial units to kick off plant operations.

In addition, SRNL is helping to develop the tritium process systems to safely move the gas around the plant.

<u>Subcritical Assembly and Off-Gas System Development</u>

Subcritical Assembly

The SHINE subcritical assembly consists of the neutron multiplier, the target solution vessel (TSV), and the subcritical assembly support structure (SASS). Each of these components has been geometrically optimized and their key parameters (dimensions/materials) are, for the most part, finalized.

The neutron multiplier serves two purposes: first, it multiplies the neutron population through various nuclear reactions, and second, it degrades the energy of the D-T source neutrons so they better couple to the TSV solution. Since the last presentation, the neutron multiplier geometry and materials have been selected by an optimization analysis that maximizes TSV yield while preserving affordability.

The TSV consists of an annular uranyl sulfate target that surrounds the gas target of the DT accelerator. Neutrons created by the accelerator are emitted from a nearly isotropic line source and enter the target solution, inducing fission. The distributed nature of the particle source is a significant advantage in creating an evenly distributed heat profile throughout the uranyl sulfate solution.

Figure 4—Irradiation cell containing the accelerator and subcritical assembly

Since the last meeting, tremendous progress has been made in finalizing the design parameters of the TSV, including physical dimensions and materials. In particular, analyses have been performed in the areas of thermo-hydraulics, inherent safety, and start-up and operational strategies. Steady-state system responses have been modeled using MCNP5 and radioisotope production has been modeled using MCNP5/ORIGEN-S. Modeling suggests that the production of up to 1,000 6-day Ci Mo-99 per device per week is possible given achievable accelerator output parameters. Modeling of the TSV is supported by in-house staff, the Los Alamos (LANL) and Argonne National Laboratories (ANL), and the University of Wisconsin.

Target solution composition has also been determined, and will consist of aqueous uranyl sulfate. The key parameters of uranyl sulfate concentration and pH have been selected and included in the neutronics, thermohydraulics, and production models. Solution composition decisions have been, in part informed by LANL and ANL. Material corrosion test programs are being developed to supplement and validate existing literature for the materials of construction to ensure vessel corrosion is minimal during the facility lifetime. Corrosion programs are being supported by the Oak Ridge National Laboratory (ORNL) with support from LANL and ANL.

Significant preliminary safety analysis work has performed to ensure the integrity of the TSV during all potential abnormal events. Neutronics transient analysis models are being developed with the support of the national laboratories and will be used to support the safety analysis during final design through modeling each abnormal event. The negative feedback coefficients of the subcritical assembly lead to a stable and self-regulating system.

Another development since the last meeting is the design of the SASS. The SASS is a robust physical structure that surrounds the TSV, providing physical support and creating an additional barrier against the release of fission products from physical insult or corrosion. Cooling water is pumped between the SASS and the TSV in a closed loop to ensure any leakage is contained, and periodic sampling of this water is to be performed to verify the integrity of the TSV.

Offgas System
An important auxiliary system tied to the subcritical assembly is the off-gas control system. Its primary responsibility is the recombination of hydrogen and oxygen created by radiolysis of water during TSV operation. Recombination of water is done through a passive, catalytic recombiner, however many other components are required for the off-gas system to function properly. In addition, it may be used to trap radioiodine if substantial quantities are found in the TSV off-gas stream.

Since the last workshop, SHINE has completed the design and construction of a prototype offgas system and created a plant-like connection to a prototype target solution vessel. The purpose of the prototype is to measure recombination efficiencies of hydrogen and oxygen under conditions that will be similar to those that will be found in the plant, determine if chemicals in the TSV solution can impact recombiner efficiency, and test various hydrogen sensors under for compliance with plant requirements. Experiments on this system are underway now, and are serving to inform designs for the eventual ^{99}Mo facility.

<u>Chemical Separation and Purification</u>
After an irradiation cycle, target solution is transferred from the TSV to an extraction process so that the relatively small amount of desired ^{99}Mo can be separated from the acidic solution, which also contains a large excess of uranyl ions, as well other numerous other fission products. The target solution is pumped to a holding tank and then on to hot cells where extraction and purification are performed.

SHINE Medical Technologies with the help of ANL, LANL, the Wisconsin Institute for Medical Research (WIMR) and the Morgridge Institute for Research (MIR), has demonstrated successful extraction of molybdenum from irradiated uranyl sulfate solutions with very high efficiency (greater than 95%). This technological achievement is particularly impressive considering the conditions: the mass to mass ratio of uranium to molybdenum is greater than one million, and the chemical speciations of both the uranium and molybdenum need to be controlled through all steps, balancing the generally reducing environment of the

TSV and the oxidizing conditions of the solubilizing acid. Additionally, other fission products need to be monitored and controlled, requiring a multi-step approach.

The molybdenum is initially separated from the large excess of uranyl ions using established separation techniques employing ion exchange columns. From there the molybdenum is further purified for pharmaceutical use using the LEU-modified Cintichem process. The Cintichem process was developed, patented, and put into commercial use back in the 1980's in Tuxedo, NY. It uses a series of well-established precipitations and chromatographic clean-up steps to remove other fission by-products that may travel with the molybdenum through the initial extraction. Radionuclides of iodine, transition metals, p-block semi-metals, and even potentially transuranics are removed before the molybdate moves to final packaging, quality control testing and labeling. The ^{99}Mo is then shipped by air to customers.

6. Summary

The MIR/SHINE team has made exceptional progress since the last meeting on all fronts toward establishing a domestic supply of ^{99}Mo and other medical isotopes. The team has grown, and incorporated experienced players from the nuclear industry with talent from many other areas required to ensure successful plant operation. A major regulatory filing has been completed and the second half of that filing will occur within a month, kicking off the Construction Permit application review by NRC. A tremendous amount of work has gone into facility design, and preliminary design will be complete this month. Important technical progress also continues to be made in partnership with the Morgridge Institute for Research, the University of Wisconsin and the National Laboratories, ensuring that our medical isotope plant will be ready for commercial operations by mid-2016.