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Mo-99 Production Using a Subcritical Assembly

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ABSTRACT

Fission-based molybdenum-99 (^{99}Mo) production can be achieved using an aqueous low-enriched uranium (LEU) solution driven by a low-cost, efficient deuterium-tritium (D-T) neutron source. Differential pumping along the accelerator column will maintain a sufficiently high pressure tritium gas target to achieve a neutron production rate of greater than 10^{13} neutrons/s. The neutron source will sit at the center of a nested annular vessel system containing the LEU solution and moderator in a geometry optimized for ^{99}Mo production. Hydrogen and oxygen from radiolysis of water will be recombined and volatile radioactive gas will be removed continuously during operation. ^{99}Mo will be extracted from the aqueous solution in batches using established techniques with the goal of supplying half of U.S. demand.

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 their end of life 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, in partnership with the Morgridge Institute for Research, has developed the Subcritical Hybrid Intense Neutron Emitter (SHINE). The SHINE device creates ^{99}Mo via fission of uranium-235 (^{235}U) in a subcritical aqueous solution driven by an innovative, accelerator-based neutron generator developed by Phoenix Nuclear Labs.

The motivation for this configuration was realized by analyzing the strengths and weaknesses of both accelerator and aqueous reactor technologies. Accelerators are generally very safe, stable platforms for isotope production, but lack high yield and specific activity. Aqueous reactors provide high yield and specific activity, in addition to having a simplified extraction process over solid targets, but are subject to large variations in power level due to void, chemical and temperature effects which could potentially lead to control problems.

The SHINE system combines the strengths and minimizes the weaknesses of both approaches. By operating subcritically, the control and safety of the accelerator system is imparted to the subcritical medium. Changes to the power level based on void, chemistry, and temperature are bounded in a subcritical system and relatively small. However, due to subcritical multiplication and a uranium target, high yield and specific activity are obtained.

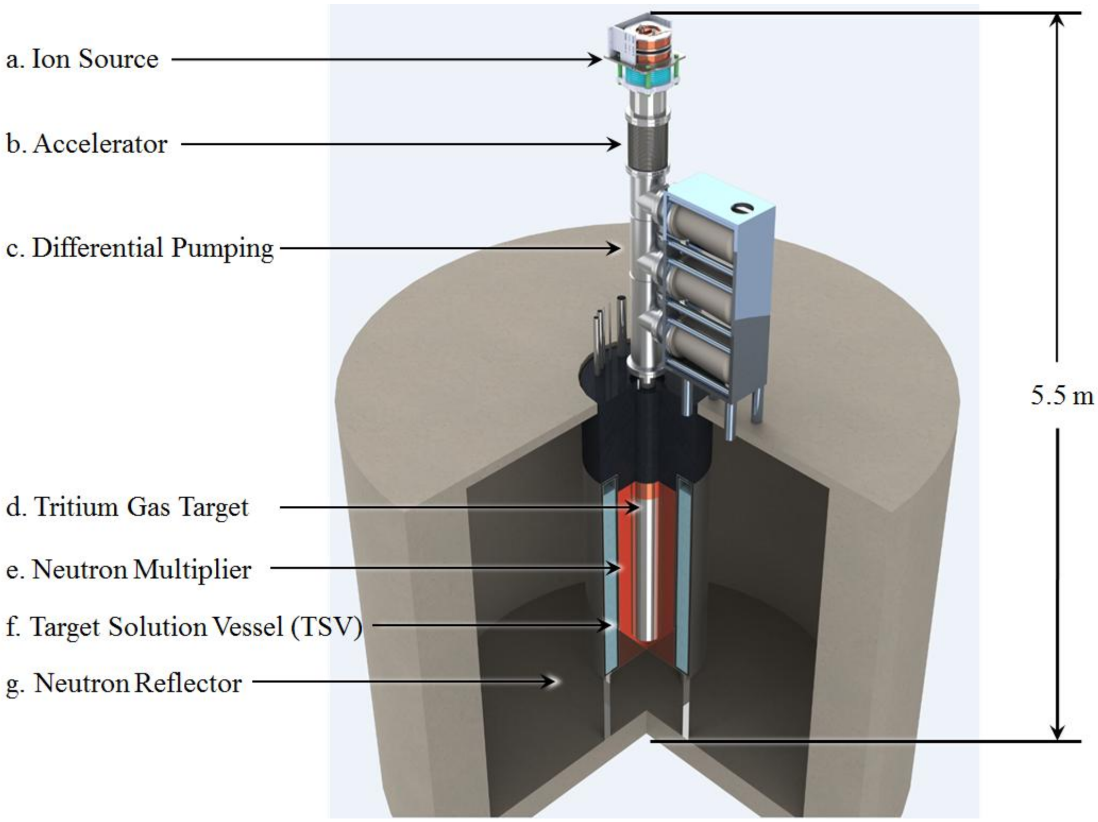

Figure 1. *The Subcritical Hybrid Intense Neutron Emitter (SHINE).*

2. How It Works

a. At the top of the device (see **Figure 1**), a microwave ion source creates a deuteron ion 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 the 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 entering 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 produces internal heating of the solution and the heat is rejected through the vessel walls and off-gas handling system to secondary coolant systems to allow for 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 LEU target vessel is surrounded by layers of heavy and light water which reflect neutrons back into the solution vessel, cool the system, and absorb 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 ^{99m}Tc generator manufacturers.

3. Key Characteristics

<u>Aqueous, LEU Target</u>
Producing ^{99}Mo without the use of HEU removes the threat of nuclear proliferation and complies with U.S. and international efforts to eliminate civilian use of HEU. Also, because the LEU is in an aqueous solution, ^{99}Mo can be extracted from the target without a wasteful target dissolution process. Once the ^{99}Mo is removed, the target solution may be re-used.

<u>Low Environmental Impact</u>
Typically, production of ^{99}Mo is performed in research reactors, where a small fraction of the neutrons produced by the reactor strike a target. Most of the fission occurring in the reactor does not contribute to isotope production, but does contribute to nuclear waste production. In the SHINE system, nearly every fission that occurs will contribute to isotope production, resulting in hundreds of times less nuclear waste per unit of medical isotopes than in the case of conventional reactors.

In addition, the SHINE process uses a very small amount of energy compared to other accelerator-based ^{99}Mo production methods, and therefore has a smaller environmental impact.

For example, The SHINE production facility is estimated to consume approximately 1 MW of electrical power. Considering that:

- 18.6% of power in the U.S. comes from nuclear[1];
- average transmission and distribution losses are about 7%[2]; and
- the thermal efficiency of a PWR is about 33%[3],

approximately 600 kW of fission power is required to power the plant. In comparison, other accelerator technologies will require ten to fifty times this amount of power, and will create a correspondingly higher amount of nuclear waste from power consumption alone. Interestingly, the SHINE plant will produce less nuclear waste from the direct production of isotopes (approximately 550 kW thermal) than it will from electricity usage, and will produce better than ten times less nuclear waste than other particle accelerator approaches in total.

^{99}Mo That Fits into the Existing Supply Chain

Because the SHINE process produces ^{99}Mo using uranium, just as today's producers do, ^{99}Mo produced by SHINE will have the same characteristics as ^{99}Mo that is produced today. This means that it can be used by downstream supply chain participants with minimal to no change in their current practices, ensuring quick adoption by major marketers and distributors.

Co-production of Other Isotopes

In addition to ^{99}Mo, other isotopes with established, smaller, but still significant markets are produced including other imaging isotopes such as xenon-133 (^{133}Xe), and the rapidly growing therapy isotope iodine-131 (^{131}I). Iodine-125 (^{125}I) and other neutron-capture-based isotopes can be produced if desired by taking advantage of SHINE's neutron flux.

Low Cost

The SHINE production system is based on an inexpensive accelerator that operates at high efficiency. In addition, the increased production level provided by multiplication in the subcritical solution and its reuse lead to a low production cost per unit of medical isotopes. Finally, use of the existing supply chain minimizes the cost of reaching customers.

4. Safety Considerations

The SHINE facility will use a new type of medical isotope production technology which combines a fusion neutron source with subcritical multiplication. The new technology will take advantage of the safety aspects of the subcritical system, combined with a neutron source that can be cycled on and off in a short time period. The result is a system that is always "shut down" from a neutronics perspective, and can be turned off from a thermal perspective in less than about one second. Given that the system does utilize the fission process for part of the production unit, the safety of the system must be clearly and completely understood. Some of these important safety considerations are detailed below.

Decay Heat

Once the accelerator is shut down, the radioisotopes in the solution will generate a small amount of decay heat. Immediately upon shutdown, the solution will generate about 6% of its initial power, and after 20 minutes, the solution will generate less than 1% of its initial power. The power generated within the solution will continuously decrease at all later times.

Since the facility is operating at very low power, and since the aqueous target and reflector offer a large heat capacity, the decay heat for an entire week is calculated to result in a temperature rise of less than 20°C after shutdown if all cooling capability is completely lost. This minor temperature rise is small enough that it will not pose safety concerns in terms of system integrity or stability. Furthermore, any increase in temperature drives the system further from criticality, increasing system safety margins even if no other controls are functional.

Radiolytic Gas Effects

Another aspect of the safety considerations for the facility will be the impact of radiolytic bubbles. It is well known that the generation of fission products in water causes some of the water molecules to decompose into elemental hydrogen and oxygen. This hydrogen and oxygen will form small bubbles within the solution, which will then rise to the surface to be collected and recombined in the catalytic recombiner (as mentioned previously).

Given that the radiolytic bubbles will displace uranium solution, they will result in a loss of reactivity of the system. Therefore, the loss of these bubbles will result in an increase in reactivity. From a safety perspective, it is important to understand the amount of feedback in the system if all of the radiolytic bubbles were quickly removed from the solution (e.g. through a large increase in pressure, chemistry effects, etc.).

The change in system reactivity and thermal power was studied for a range of possible void fractions. The system was studied in an operational state and the limiting case of an instantaneous loss of all void in the system. The results are plotted in the figure below.

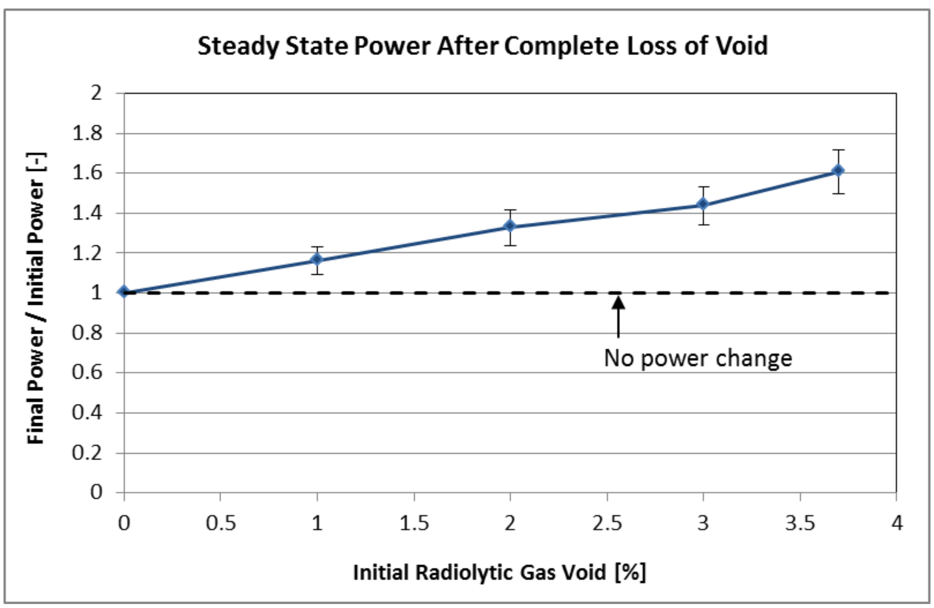

Figure 2. *The effects of a complete loss of void on the steady state power of the system for a range of possible operating void levels. Note that upon collapse of any expected void level in the system, the system always remains in a stable, subcritical state (evidenced by bounded steady state power levels).*

As shown in the above figure, the calculations reveal that a collapse of any void within the expected range of the system will result in an operational power level that is bounded (the system remains subcritical). For all of the systems illustrated in the figure, a complete loss of void would result in power increasing by a factor less than two (2). Given the low power of the system, this is a relatively small change in power that is expected to be easy to manage from a heat removal standpoint.

Moreover, the power level change shown in the above figure is the limiting case. Due to strong negative reactivity feedback effects, the actual power feedback would be significantly less than this. As the power of the system increased, the LEU solution temperature would also increase, and lead to an inherent decrease in system reactivity.

Burnup Effects

Since the LEU solution will naturally change composition during the irradiation process, it is important to understand the effects of that change in solution composition on the neutronics and thermal hydraulics of the system. Using neutronics codes, the isotopic concentrations of various solutions have been studied over the course of one week of irradiation.

One of the important figures of merit will be the amount that the system remains subcritical during irradiation since this will impact many other safety calculations for the system. The figure below presents the calculated results.

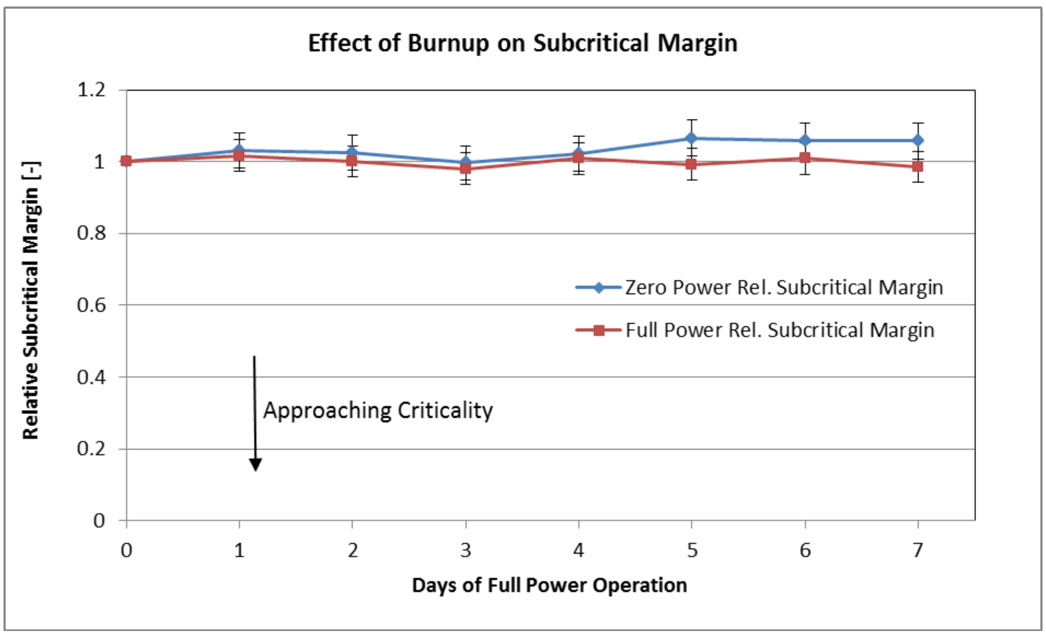

Figure 3. *The neutronics impacts from one (1) week of continuous operation at full power. The figure shows the small impact that burnup is expected to have on the neutronics response of the system during full power and zero power conditions. All results are normalized to the initial subcritical margin for the respective case (zero power or full power).*

The figure above shows the calculated subcritical margin of the system is relatively insensitive to the burnup on the time scale of typical ^{99}Mo production schedules (1 week). The total variation in the subcritical margin due to fission and transmutation is calculated to be less than 15% of the initial margin. Therefore, the system is expected to be capable of running at steady-state full power with little or no manual reactivity manipulations for one continuous week.

Reactivity changes beyond the period of a one week irradiation are not considered relevant because the system will be drained in order to separate the medical isotopes and will then be re-characterized when the tank is refilled for the next run. As the system is filled, the multiplication factor will be measured and the height will be set to obtain a constant initial multiplication factor as compared to previous weeks.

5. Conclusions

Fission-based ^{99}Mo production using an aqueous LEU subcritical device driven by a deuterium-tritium neutron source has numerous safety, environmental and commercial advantages. SHINE Medical Technologies intends to supply half of U.S. demand for ^{99}Mo using this technology.

6. References

[1] U.S. Energy Information Administration, "Electric Power Monthly," November, 2011, <http://www.eia.gov/cneaf/electricity/epm/epm_sum.html>.

[2] U.S. Energy Information Administration, "Frequently Asked Questions: How much electricity is lost in transmission and distribution in the United States?" June, 2011, < http://www.eia.gov/tools/faqs/faq.cfm?id=105&t=3>.

[3] J. J. Duderstadt and L. J. Hamilton, *Nuclear Reactor Analysis*. Ann Arbor, MI: John Wiley & Sons, 1976.