Hybrid Accelerator-Heavy Water System for Production of a Reliable, Domestic Supply of Molybdenum-99 without the Use of Highly Enriched Uranium

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ABSTRACT

Advanced Medical Isotope Corporation licensed a "hybrid" accelerator-based technology from the University of Missouri to provide a minimum of 50% of the United States Mo-99 demand from a subcritical solution of Low-Enriched Uranium. An electron beam device strikes a high density target. The photons "shine" into a stainless steel tank holding D_2O and low-enriched uranium salt. The photons eject neutrons from deuterium atoms, initiating fission in the LEU target material, which provides a "boost" to the neutron population. The Mo-99 is extracted continuously while recycling the solution back into the reaction chamber. After six days the batch is completed and the Mo-99 is then extracted from the collection columns to be purified. The patented concept has been refined by a series of tests conducted at the Idaho Accelerator Center and by hundreds of independently confirmed MCNPX computer runs. This accelerator design and technique can meet the US production requirements.

Introduction

In recent years, the "^{99}Mo/^{99m}Tc Shortage-Crisis" has caused major interest in producing ^{99}Mo in the U. S. This included a number of charged particle accelerator approaches being pursued. This paper presents the results of four years of intense analyses and computer calculations to come up with an optimized system design to produce very high activity (3000 6-day Ci/week) and high specific activity (50,000 Ci/g) ^{99}Mo.

Advanced Medical Isotope Corporation (AMIC) is a medical isotope production company located in Kennewick, Washington. As part of its mission to deliver critical isotopes to doctors and patients, AMIC has licensed and enhanced a "hybrid" accelerator-based technology from the University of Missouri that will produce fission product Mo-99 from a subcritical solution of Low-Enriched Uranium (LEU). AMIC has analyzed many other approaches to producing Mo-99 and has concluded that the hybrid system presented in this paper is superior to any other accelerator technique.

The purpose for developing this ^{99}Mo/^{99m}Tc method and material is to support effective medical diagnostic applications in U. S. hospitals. This approach started with a concept based upon a University of Missouri PhD Thesis [1] to create a large thermal neutron flux for an electron driven accelerator connected to a vessel containing a D_2O solution. The system described in this paper adds Low Enriched Uranium (LEU) to the D_2O solution so that very large thermal flux amplification occurs. This amplification (subcritical multiplication) is of the order of a factor of 100 and comes from the $1.0/(1.0-k_{eff})$ effect. A significant advantage to design of this system is that it is self-limiting and cannot go critical. It can also be quickly shut down by simply switching off the electron beam - both major advantages over conventional reactor production methods.

Mo-99 Production System

Our production system consists of utilizing a commercially available electron beam device to strike a dense target of tungsten or tantalum. The photons generated by the interaction of the electrons and the metal "shine" into a stainless steel tank holding heavy water, D_2O, containing low-enriched uranium salt, such as uranyl nitrate. The photons eject neutrons from deuterium atoms in the D_2O that are subsequently thermalized in the same medium. These neutrons initiate fission in the LEU target material, which in turn provides a subcritical "boost" to the neutron population by contributing fission neutrons into the tank. The Mo-99 is extracted continuously during irradiation while recycling the solution back into the reaction chamber. After six days the batch is completed and the Mo-99 is then extracted from the collection columns to be purified. This layout is depicted in Figure 1.

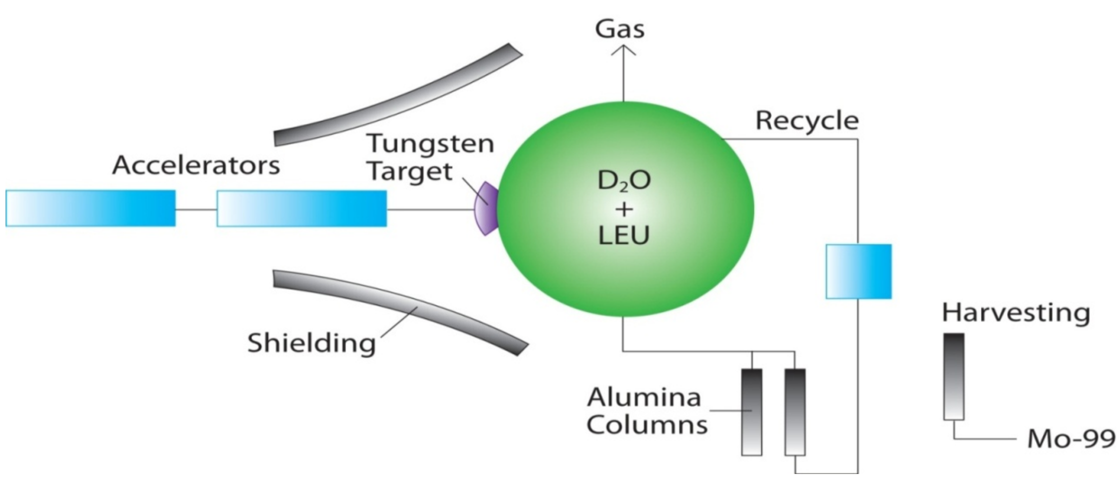

Figure 1. Mo-99 Production System.

The AMIC designed system has two IBA Rhodotron TT-300 accelerators in series. The beam from the first accelerator feeds the second accelerator. This configuration maximizes production efficiency without entering into realms that requires new technology development for single larger accelerator approaches. This system yields a very monochromatic beam, which can be controlled easily to optimize accelerator location. Production can be scaled-up with a second set of accelerators delivering photons to the same or another reaction vessel.

Our recently optimized beam design parameters are 24 MeV, 350 kW, and 14.6 mA; however, the system design is flexible and can function well in a range between 15 to 40 MeV and 150 to 400 kW. Achieving thermal flux levels equal to 1.23 E+12 is necessary to reach the Mo-99 production goals of 1500 6-day Curies per system (25% of US needs) at a specific activity of 50,000 Ci/g. Optimization of the MCNXP calculations achieves more than twice that flux, which would produce at least 3000 6-day Curies/week (50% of US needs).

The "Proof of Concept" Experiments

In July and August 2009, a series of experiments were performed at the Idaho Accelerator Center. In these experiments electron beams were directed on dense metal material to create photons. A small tank of D_2O was set up next to the photon source and the resulting neutron flux was measured using metal foils of gold, indium and other metals. Cadmium covers were placed over the metal foils in some configurations to help estimate the epithermal neutron flux. In addition, neutron reflector material was placed in various thicknesses around the D_2O tank and the effect on the resulting neutron flux recorded using the metal foils.

The experimental results matched calculated estimates and thereby provided us with a confirmation of the reliability and applicability of these models. These series of experiments proved that it is possible to generate a significant, sustained neutron flux in D_2O using photons from a commercially available electron beam accelerator.

Further experiments are planned with small quantities of LEU/D_2O to validate our model calculations and strengthen the predictions for the capacity of the commercial system. In particular, these tests will quantify the subcritical multiplication/amplification $(1.0/(1.0\text{-}k_{eff}))$ effect. Specific activity, purity and by-product profiles are expected to be comparable to existing commercially available ^{99}Mo.

Modeling Calculations

The MCNPX calculations produced thermal and total neutron flux values as high as 2.6e12 n/cm^2-s and 6.8e12 n/cm^2-s, respectively. Given the MCNPX flux and ^{235}U, ^{239}Pu and ^{238}U fission cross sections, ^{99}Mo activities were calculated with the CHAIN [2] computer code.

The system design involves the use of two IBA electron TT 300 Rhodotron accelerators connected in series to give 24 MeV electron energies with a maximum current of 16 mA (384 kW power). To achieve optimization both MCNPX k_{eff} and flux calculations were performed. An important aspect in easily obtaining k_{eff} values close to 1.0 is the use of several different reflector materials surrounding the vessel containing the D_2O/U solution. These and other scenarios were extensively analyzed with the computer codes. As an example, Figure 2 shows results of 6-day Ci/week ^{99}Mo produced for a 6 day irradiation versus a HDPE reflector/moderator thickness.

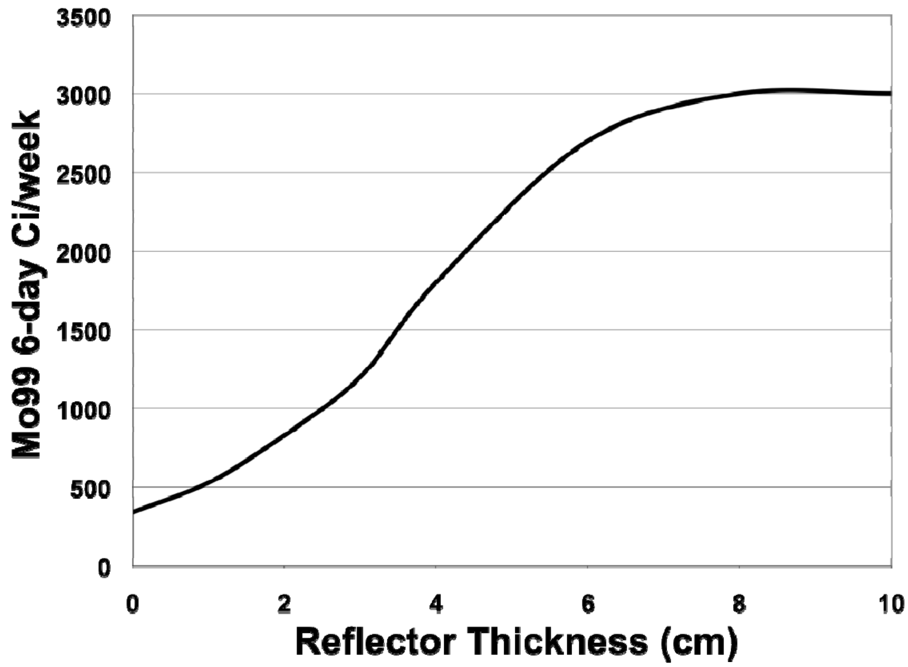

Figure 2. ^{99}Mo 6-dayCi/week production.

These calculations have been verified independently by several different technical organizations. For example, Ion Beam Applications (Belgium) has produced a very detailed report [3] confirming these type of results. Also there have been several MU-AMIC ^{99}Mo patents detailing this approach [4-7].

Commercial System Design and Schedule

The project is divided into five system modules: 1) Accelerator and reaction vessel, 2) Mo-99 separation and purification, 3) Uranium nitrate recycle, 4) Waste management, and 5) Airborne emissions treatment. Each system has been through a pre-conceptual design. Conceptual design and targeted engineering module demonstration projects are planned to reduce any risks of schedule delay. Figure 3 outlines the integrated system design.

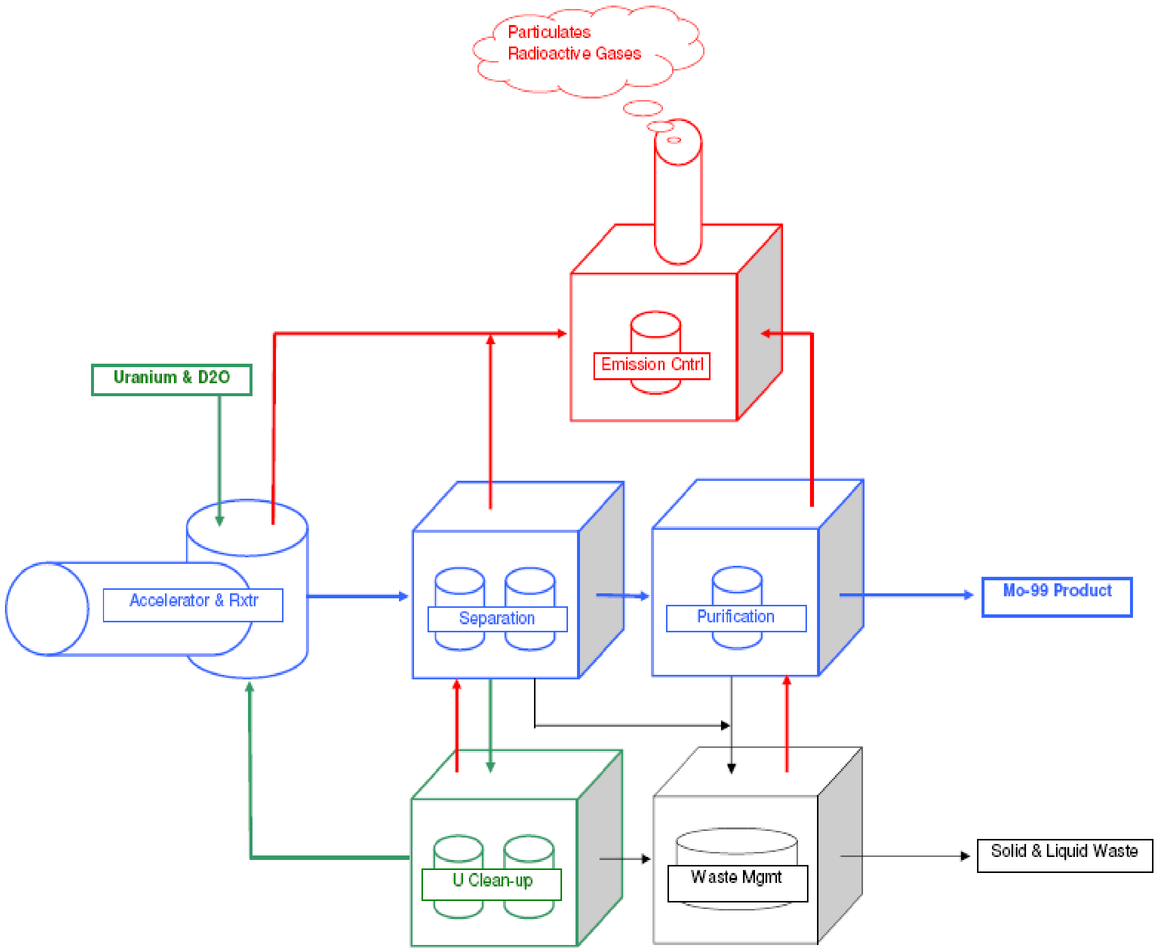

Figure 3. Conceptual Layout of Production Facility Production Processes.

The integrated system design was done to primary functional design criteria:

- Ensure a stable non-critical system under all scenarios
- Ensure employee safety and insignificant environmental impacts
- Maximize the production rates
- Produce the lowest cost and most reliable system that satisfies, at least, 50% of the US needs

The top level project schedule to achieve a working commercial system is summarized in Figure 4:

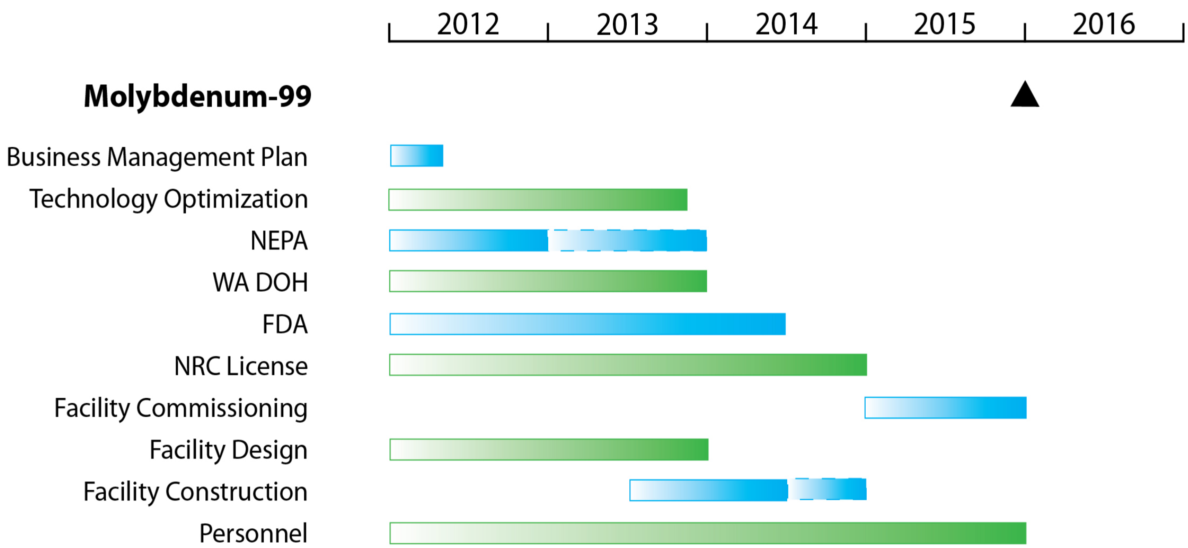

Figure 4. Commercial Facility Schedule

Conclusions

The results given in this paper indicate that large quantities of high specific activity ^{99}Mo can be produced with a single electron driven subcritical (LEU) reactor system. Because of its small-footprint/lower cost it is conceivable to have one or more of these systems operational and capable of delivering a significant fraction of the U.S. domestic need within just a few years.

References

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