Analysis of Molbdenum-99 Production Capability in the Materials Test Station

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Abstract. The United States of America currently relies on foreign suppliers to meet all of its needs for molybdenum-99 (Mo-99) used in medical diagnostic procedures. The current US demand is at least 5000 six-day curies per week. Neutronics calculations have been performed to assess whether the proposed Materials Test Station (MTS) could potentially generate Mo-99. Two target material options have been explored for Mo-99 production in the MTS: low enriched uranium (LEU) and Tc-99. For LEU, scoping calculations indicate that MTS can supply nearly half of the current US demand with only minor neutronic impact on the MTS primary mission. For the Tc-99 option, the MTS could produce about one-tenth of the US demand.

1. Introduction

The recent decision by the Atomic Energy of Canada, Limited to close the MAPLE reactors [1] has led to renewed interest within the United States of America (US) in assessing domestic sources for molybdenum-99 (Mo-99). The proposed Materials Test Station (MTS) could serve as one among several potential sources of Mo-99. It will be a spallation source driven by a 1-MW, 800-MeV proton beam, located at the Los Alamos Neutron Science Center (LANSCE) within Los Alamos National Laboratory. The MTS is designed to be an irradiation facility for testing reactor fuel and structural materials in a fast neutron spectrum similar to that seen in fast nuclear reactors [2,3].

2. Production Needed to Meet US Demand

In 2006, the US demand for Mo-99 ranged from 5000 to 7000 six-day curies (Ci) per week, with an estimated annual growth rate between 3 and 5% [4]. A six-day Ci is a unit of measure used by the industry, which is the amount of Mo-99 remaining in a Tc-99m generator six days after shipment from the producer's facility. Molybdenum-99 has a 65.94-hour half-life. To have one curie remaining after six days of decay, a producer must ship 4.54 Ci. Decay during the 30 to 40 hours of time between target removal from the irradiation facility and shipment from the chemical processing facility yields losses of 27 to 34%. Further, only about 90% of the Mo-99 in the targets is chemically recovered. Thus, for each six-day curie delivered by the processing facility, between 6.9 and 7.7 Ci must be available at the end of target irradiation. Since decay also occurs during target irradiation, the instantaneous production rate needed to ship a single six-day curie per week depends on the frequency that targets are harvested. Table I shows the instantaneous production rate of Mo-99 needed to ship a single six-day curie per week, assuming a 40-hour period for target processing and 90% chemical recovery

TABLE I: Production rates needed to ship one 6-day Ci/week vs. harvest frequency.

Harvest frequency	Instantaneous production rate needed to ship one 6-day Ci/week (mCi/h)	Equivalent ^{235}U fission heat per 6-day Ci/week (W)
Daily	52	96
Every 3 days	65	121
Weekly	97	180

efficiency. For a 3-day target irradiation, each shipped six-day curie per week requires an instantaneous production rate of 65 mCi/h. Also shown in Table I is the fission heat that must be generated by the Mo-99 production targets per shipped 6-day Ci/week if the Mo-99 is produced by fission of ^{235}U.
Two approaches have been evaluated for Mo-99 production using the MTS. The first is through the traditional ^{235}U(n,f)^{99}Mo reaction, the second is via the ^{99}Tc(n,p)^{99}Mo reaction. The first is best carried out in a thermal spectrum, while the second, which has a threshold energy of 0.58 MeV and no appreciable cross section below 4 MeV, requires a very hard neutron spectrum.

2. Materials Test Station Design

The MTS consists of tungsten spallation target sections on either side of a central fuel rodlet irradiation region, as depicted in Figure 1. Irradiation of materials test specimens takes place in materials samples cans placed up next to the spallation targets. The spallation target and fuel rodlets are cooled by liquid lead-bismuth eutectic, while the materials sample cans are cooled by heavy water. The neutron spectrum in the irradiation region is similar to that of a fast reactor, with the addition of a high-energy tail extending up to the incident proton beam energy (800 MeV). Downstream of the spallation targets and the fuel irradiation zone is the so-called “backstop” where the incident protons range out and high-energy secondary particles are attenuated.
The MTS has been designed to produce a fast neutron spectrum, and the baseline design does not include a thermal neutron spectrum region. A thermal spectrum can be introduced in the backstop, which has significant neutron flux but does not contribute in a significant way to the MTS primary mission.

3. Mo-99 Production with LEU Targets

In the baseline design (Figure 1), the backstop is composed of a stack of tungsten plates whose function is to range out the primary protons and attenuate high-energy secondary

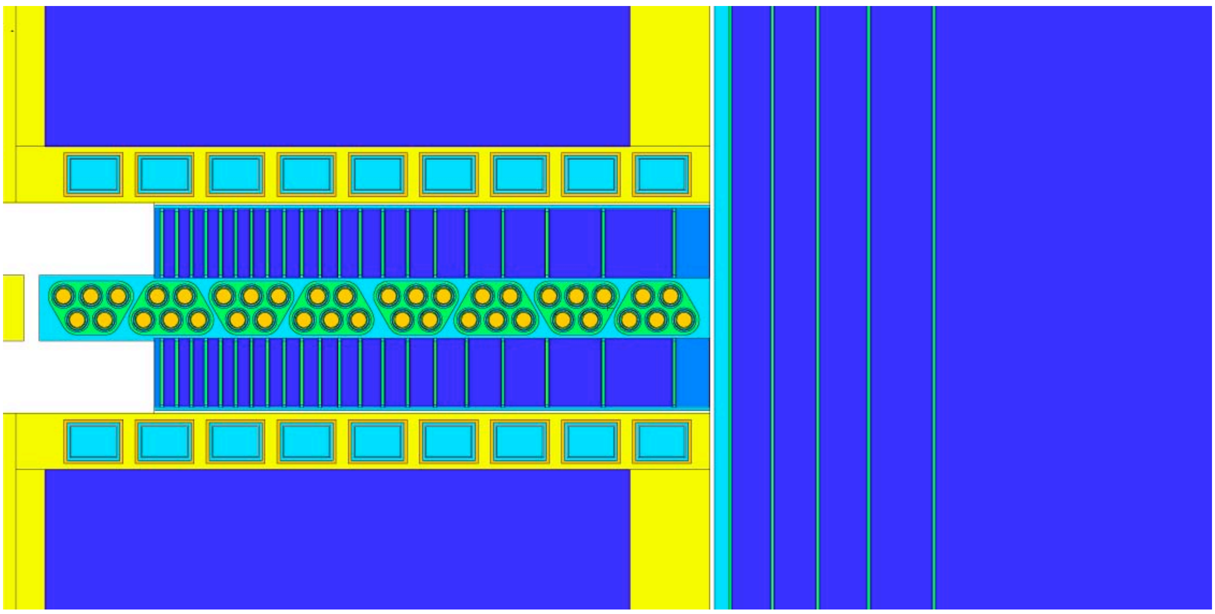

FIG. 1. MTS baseline design.

particles. The proposed modification for supporting ^{99}Mo via uranium fission is shown in Figure 2. Here the tungsten plates have been moved downstream to make room for a 12-cm-thick region of light water. Steel cylinders with a thin layer of LEU deposited on their inside surfaces are placed in the water, as depicted in Figure 2.

The unperturbed thermal neutron flux (E<0.625 eV) generated in the water region is shown in Figure 3. The peak unperturbed thermal flux exceeds 1.8×10^{14} n.cm^{-2}.s^{-1}. This thermal flux magnitude is 60% of the peak thermal flux available in the AECL NRU reactor where the North American supply of ^{99}Mo is currently produced [5].

A number of parameters can be adjusted to maximize ^{99}Mo production whilst minimizing the quantity of LEU that must be processed each irradiation cycle, for example, the number of targets, LEU layer thickness deposited on the inner walls of the targets, target tube diameter and height, and target positions in the water region. A cursory search of this multi-parameter space has led to the arrangement shown in Figure 2. Twenty-four target tubes are arranged in four rows with a lateral center-to-center spacing between 25 and 30 mm and row spacing varying between 25 and 35 mm (some of the tubes at the ends of the rows are moved from their default positions to more optimal ones). The target tubes are 14 mm outside diameter with an assumed wall thickness of 0.1 mm. The LEU layer deposited on the inside wall is 0.165 mm thick with a height of 10 cm. With a density of 19 g/cm^3, the total mass of LEU (20% ^{235}U enrichment) summed over all 24 tubes is 2.45 kg. This configuration generates 246 kW of fission heat in the targets, which yields an instantaneous ^{99}Mo production rate of 149 Ci/h. From Table I, this corresponds to a production rate of 2290 six-day Ci per week, assuming a 3-day harvest frequency.

The perturbed thermal neutron flux resulting from the presence of the LEU targets is shown Figure 4. The thermal flux is very much depressed by the large thermal fission cross section of ^{235}U, with the peak flux now down by a factor of 2.5 from the unperturbed case.

The MTS baseline design (without the thermal region and LEU targets), which can accommodate up to 4.8 linear m of fuel, is deeply subcritical. If all 40 test rodlet positions are filled with highly enriched fuel, the reactivity is only k_{eff} = 0.1. The configuration shown in

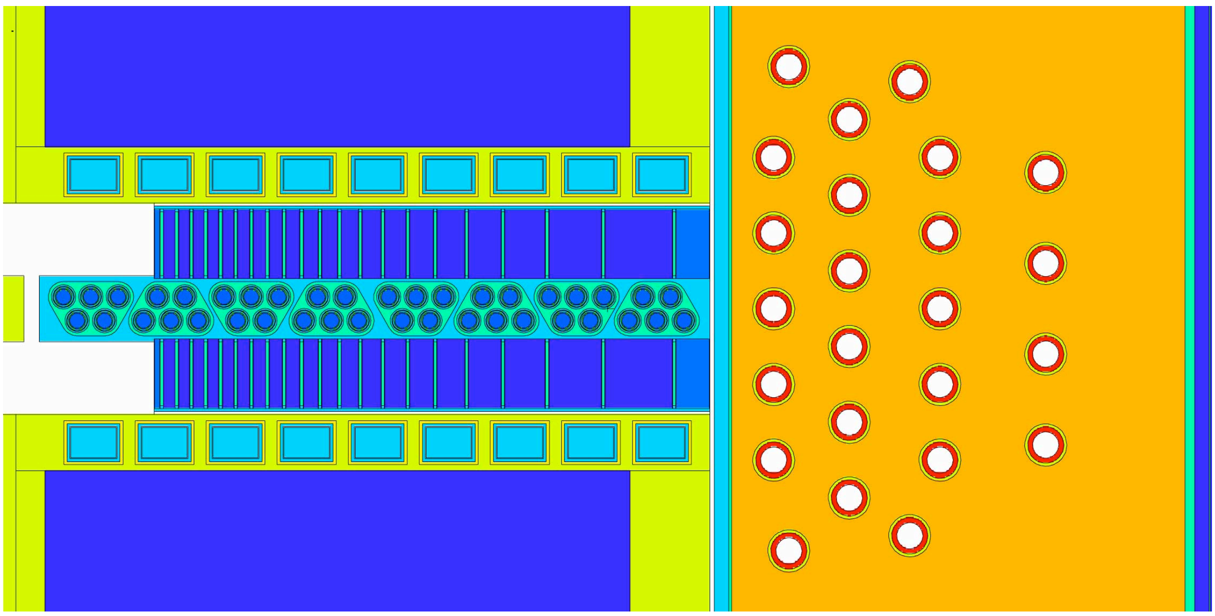

FIG. 2. MTS as modified to include downstream water region.

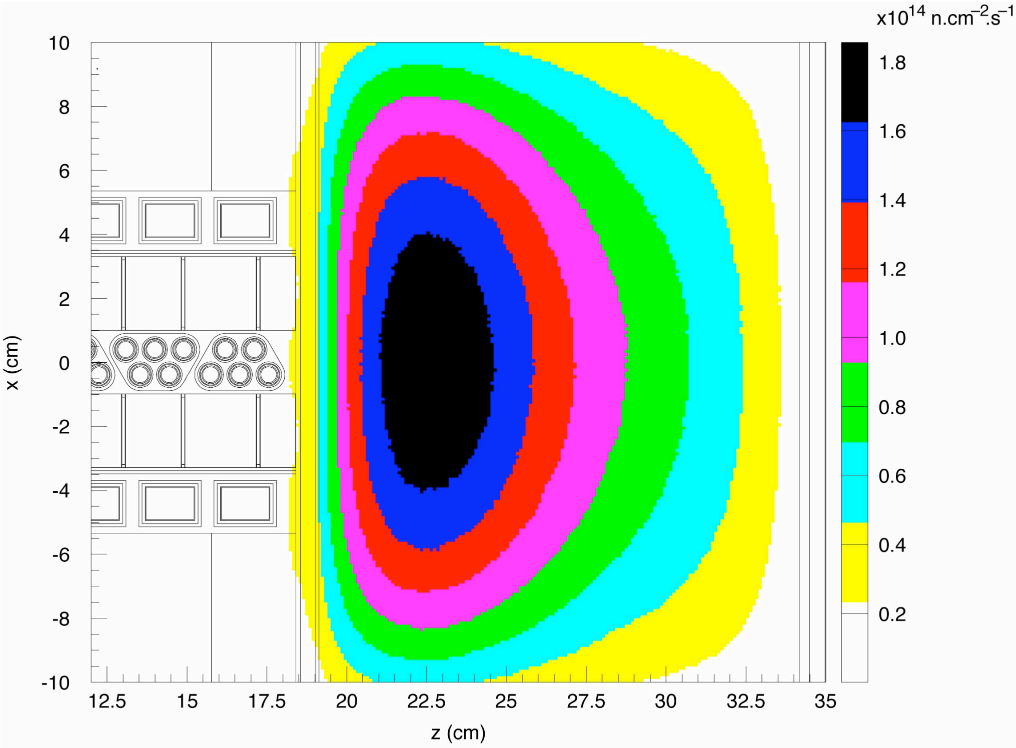

FIG. 3. Unperturbed thermal flux in the downstream water region.

Figure 2 is also deeply subcritical. This system, including the 2.45 kg of LEU, has $k_{eff} = 0.4$. Of course, analyses must be performed to assess the potential for criticality under credible postulated accident scenarios prior to implementation of this Mo-99 production scheme.

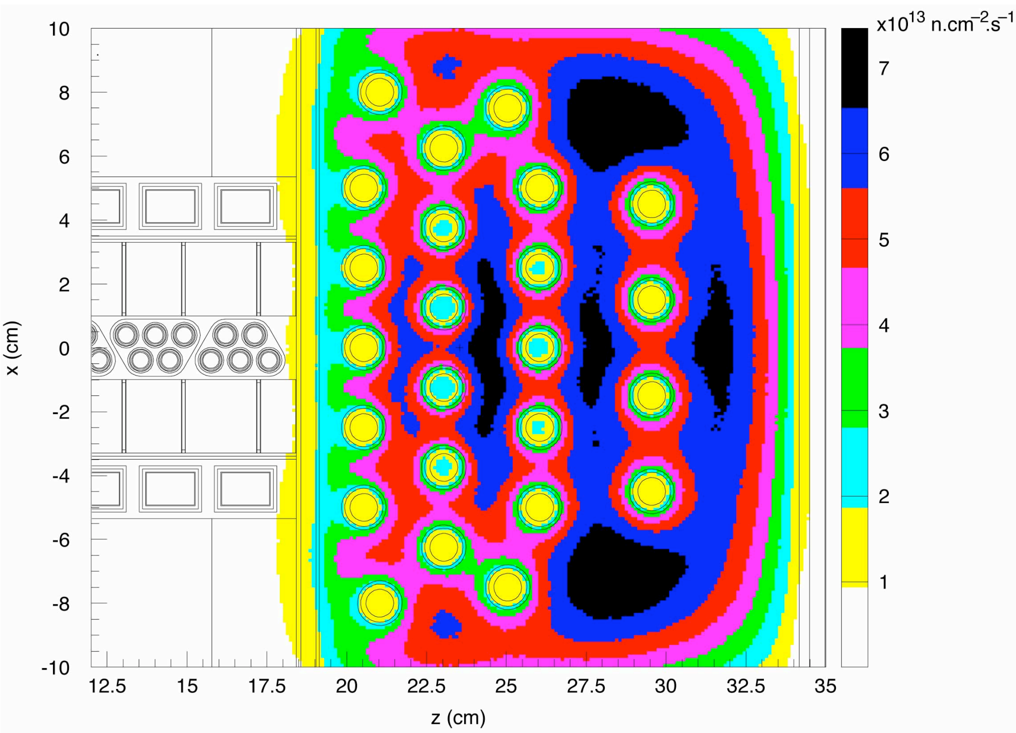

FIG. 4. Thermal flux in the downstream water region as perturbed by the presence of the LEU targets.

There are a number of test fuel rodlets located near the backstop that would likely be negatively impacted by this modification of the baseline design, but these are not the "prime" positions for fast spectrum fuel irradiation and could potentially be sacrificed in order to support a Mo-99 production mission. The penalty in fuel rodlet irradiation volume would be approximately 10 to 15% (4 to 6 rodlets out of 40 total).

4. Mo-99 Production with Tc-99 targets

An unconventional method for ^{99}Mo production would utilize the high-energy tail of the neutron spectrum in the MTS to excite the ^{99}Tc(n,p)^{99}Mo reaction. This option has the advantage of avoiding the generation of a high-level waste stream. Technetium does not exist in nature, but there is plenty available in spent nuclear fuel, with high isotopic purity of atomic mass 99. Many issues would need to be addressed before proceeding down this path, and this option is presented here only to assess whether there is any merit, from a neutronics standpoint, for further study.

Figure 5 shows the presumed cross section for the ^{99}Tc(n,p)^{99}Mo reaction. Below 20 MeV, the presumed cross section follows the JENDL 3.2 evaluation. Above 45 MeV, the presumed cross section is derived from LAHET [6] predictions using default physics models. Data between 20 and 45 MeV were adjusted to provide a smooth transition. Also shown in Figure 5 are available experimental data from threshold to 20 MeV taken from CSISRS/EXFOR [7], which shows generally good agreement with the JENDL 3.2 evaluation.

Molybdenum-99 production targets, in the form of plates composed of pure ^{99}Tc, were placed in the backstop region. These plates replaced the central 8 cm × 8 cm region of the first four backstop plates. At an assumed mass density of 11 g/cm^3, the total ^{99}Tc mass in the 406-cm^3 volume occupied by the plates is 4.47 kg. The calculated instantaneous production rate of ^{99}Mo for this configuration is 27.1 Ci/h. If harvested every 3 days, this would supply about

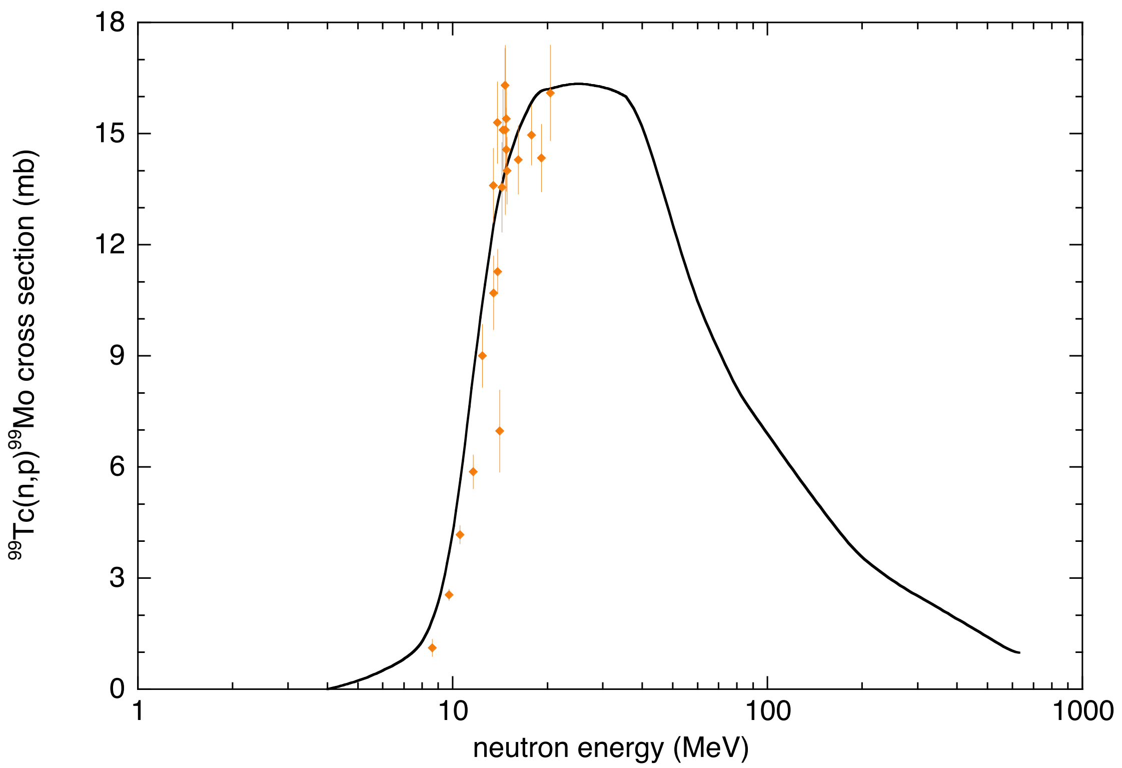

FIG. 5. Presumed cross section for the ^{99}Tc(n,p)^{99}Mo reaction.

500 six-day Ci per week or one-tenth of the current US demand. From this initial neutronic assessment, the ^{99}Tc target option does not look attractive when compared to the production rate obtained using LEU targets.

5. Conclusions

The viability of producing Mo-99 in the proposed Materials Test Station has been investigated as a means of contributing to a diversity of domestic production sources. Two target material options were evaluated: ^{99}Tc and the traditional ^{235}U. Assuming targets are harvested every three days, the ^{99}Tc option produced about 500 six-day Ci per week with 4.5 kg of target material irradiated. The ^{235}U target material, in the form of 2.5 kg of LEU, produced 2300 six-day Ci per week, or nearly half of current US demand. Further optimization is possible that could produce higher yields or reduce the amount of LEU that must be processed.

From a neutronics standpoint, the inclusion of a light water region for Mo-99 production using LEU targets does not significantly impact the primary mission of fast spectrum fuels and materials irradiations. It may, however, impact some operational aspects, such as the regulatory framework. These impacts have yet to be assessed.

From this initial neutronics assessment, it appears that the proposed Materials Test Station could be modified in a manner that would allow it to contribute in a meaningful way, in concert with a diversity of other sources, to supply Mo-99 to the North American continent.

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