

Title: Fission Produced ^{99}Mo without a Nuclear Reactor

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Word Count: 2497

Financial Support: U.S. Department of Energy, National Nuclear Security Administration's (NNSA's) Office of Defense Nuclear Nonproliferation, under Contract DE-AC02-06CH11357

Running Title: Accelerator-driven Production of ^{99}Mo

ABSTRACT

Molybdenum-99, the parent of the widely used medical isotope ^{99m}Tc , is currently produced by irradiation of enriched uranium in nuclear reactors. The supply of this isotope is encumbered by the aging of these reactors and concerns about international transportation and nuclear proliferation. **METHOD:** We report results for the production of ^{99}Mo from the accelerator-driven subcritical fission of an aqueous solution containing low enriched uranium. The predominately fast neutrons generated by impinging high-energy electrons onto a tantalum convertor are moderated to thermal energies to increase fission processes. The separation, recovery, and purification of ^{99}Mo were demonstrated using a recycled uranyl sulfate solution. **RESULTS AND CONCLUSION:** The ^{99}Mo yield and purity were found to be unaffected by reuse of the previously irradiated and processed uranyl sulfate solution. Results from a 51.8 GBq ^{99}Mo production run are presented.

Key words: Molybdenum-99, fission, accelerator

INTRODUCTION

Technetium-99m ($t_{1/2} = 6$ hour), the daughter of ^{99}Mo ($t_{1/2} = 66$ hour), is used in more than 45 million diagnostic nuclear medicine procedures annually worldwide, with ~16.7 million performed in the U.S. alone (1). Despite being the largest single user of $^{99\text{m}}\text{Tc}$, the U.S. currently imports 100% of its supply (2). Current supply chains of ^{99}Mo rely on aging nuclear reactors, such as the High Flux Reactor in the Netherlands and the National Research Universal reactor in Canada (1). The major U.S. supplier, located in Canada, will cease normal production in late 2016 but will maintain the ability to produce ^{99}Mo for another 2 years, if a severe shortage occurs (3). The High Flux and National Research Universal reactors have exceeded their initial design lifetimes of 40 years, having been in operation for 55 and 59 years, respectively. In 2009-2010, both reactors were shut down for extended periods of time, causing a severe ^{99}Mo shortage (4,5). The ^{99}Mo shortage forced clinicians to ration imaging procedures, which delayed critical diagnostic tests or resulted in older, less effective techniques that in many cases increased the radiation dose to the patient (6). The U.S. ^{99}Mo market is also fragile because it relies solely on international air transportation, which has been halted in the past due to inclement weather, natural phenomena, flight delays, and terrorist threats (6).

The predominant global ^{99}Mo -production route is irradiation of highly enriched uranium (HEU, $\geq 20\%$ ^{235}U) solid targets in nuclear reactors fueled by uranium (2). Other potential ^{99}Mo production paths include $(n,\gamma)^{98}\text{Mo}$ and $(\gamma,n)^{100}\text{Mo}$; however, both routes require enriched molybdenum material and produce low specific activity ^{99}Mo , which cannot be loaded directly on a commercial $^{99\text{m}}\text{Tc}$ generator. The U.S. National Nuclear Security Administration implements the long-standing U.S. policy to minimize and eliminate HEU in civilian applications by working to convert research reactors and medical isotope production facilities to low enriched uranium (LEU, $< 20\%$ ^{235}U) worldwide (7). In 2009, the Global Threat Reduction

Initiative program began supporting multiple potential U.S. producers and national laboratories to help establish a U.S. domestic supply of ^{99}Mo by using non-HEU based technologies. Concurrently, the U.S. Congress passed the American Medical Isotopes Production Act, which was signed into law on January 2, 2013 to ensure a reliable U.S. supply of ^{99}Mo and to eliminate the export of HEU for the production of medical isotopes.

Argonne National Laboratory is performing research and development to facilitate ^{99}Mo production within the U.S to support the development of diverse technologies that will effectively and efficiently produce, recover, and purify ^{99}Mo without the use of HEU. We have facilitated development of the technology proposed by SHINE Medical Technologies, which will use a deuterium/tritium accelerator to induce subcritical fissioning of ^{235}U in an LEU uranyl sulfate solution (8). A key aspect of the SHINE technology is that medical isotopes such as ^{99}Mo can be produced subcritically, eliminating the need for a nuclear reactor (8).

Molybdenum-99 is traditionally produced via irradiation of solid uranium-aluminide dispersion targets in a nuclear reactor (2). The dissolution/digestion of these solid targets to release the ^{99}Mo is time-intensive and generates significant amounts of radioactive waste. In contrast, an aqueous uranium-salt target solution can be processed immediately after irradiation, reducing ^{99}Mo loss due to decay. Additionally, the target solution can be recycled, resulting in a significantly smaller amount of waste generated per irradiation cycle.

Several challenges currently hinder development of a solution-based commercially viable process. One challenge is that repeated irradiations will lead to buildup of long-lived fission products and transuranium elements, which could complicate purification processes and may ultimately degrade the ^{99}Mo purity required for use in medical applications. This challenge can be met by periodic “clean-up” of the solution to remove accumulated long-lived fission products.

Argonne has developed a process that uses sulfate-to-nitrate conversion followed by the nitrate-based uranium extraction process for treating the recycled solution (9). Another challenge is that radiolysis of aqueous sulfate solutions generates hydrogen and oxygen, which need to be carefully managed. Moreover, radioactive xenon, krypton, and gaseous iodine are produced during the fission process and must be captured. A third challenge is that the presence of chemically active radicals and intermediate products (e.g., hydrogen peroxide) pose complex solution chemistry complications, potentially affecting the oxidation and/or physical state of key components, including Mo.

MATERIALS AND METHODS

Herein we discuss the results of our experimental program and pilot-scale demonstration for the production of greater than 37 GBq of ^{99}Mo from recycled LEU uranyl sulfate solution, which had been used in nine production runs. The molybdenum was produced by subcritical fission induced with an electron linear accelerator.

Figure 1 illustrates the experimental configuration. The accelerator-driven electron beam strikes a water-cooled solid Ta convertor, generating X-rays/photons via the Bremsstrahlung process with concomitant ejection of neutrons. Tantalum was chosen for its high Z value, photo-neutron production cross section, high melting point, chemical stability, and good machinability (9). Fast neutrons generated by this process are moderated to thermal energies by the LEU solution and surrounding water reflector, inducing fission of ^{235}U to produce ^{99}Mo (6.1% fission yield). The reflector water also serves to moderate the solution temperature to 80°C.

The acidic LEU solution is passed through an extraction column (TiO_2 , 110 μm particle size, Sachtopore), where ^{99}Mo along with a fraction of impurities is retained (Figure 2). The retained ^{99}Mo is stripped from the extraction column by an alkaline solution, acidified, and

passed through a concentration column (TiO₂, 40 μm particle size, Sachtopore) to obtain a solution volume acceptable for final purification (10,11). The concentrated ⁹⁹Mo undergoes final purification by the LEU Modified Cintichem (LMC) process, which removes impurities — primarily I, Ru, and Rh (12-14).

RESULTS AND DISCUSSION

To demonstrate the process, we irradiated 5 L of a recycled 0.63 M uranyl sulfate (19.8% ²³⁵U, pH 1) solution with neutrons produced from bombardment of a Ta converter with a 35 MeV electron beam of 10-kW nominal beam power. A peak fission-power density of 0.1 kW/L (1/10th of that expected in the SHINE system) was obtained. Monte Carlo N-Particle eXtended calculations predicted a theoretical yield of 66.6 GBq ⁹⁹Mo after 18 hours of irradiation at 10 kW of continuous beam power (15,16). Analysis of the irradiated solution showed 51.8±3.7 GBq ⁹⁹Mo after 20 hours, including beam tune-up time. The tune-up periods and operation at 10 kW resulted in a 10 kW-equivalent irradiation time of 15 hours (59.2 GBq theoretical yield).

Radioactive gases — mainly Xe and I isotopes — produced during the irradiation were collected and stored for decay. The concentrations of H₂ and O₂ produced during irradiation were continuously monitored and recombined to form water vapor using a heated Pt/Pd-impregnated-alumina/cordierite catalyst.

Molybdenum recovery — using a LabVIEW[®]-based control system — began 5 hours after irradiation to allow for decay of short-lived fission products. A remotely operated system was required for processing due to the high dose rates in the room after irradiation. An LEU target solution (5 L) containing 0.0023 mM Mo (stable Mo was added as Na₂MoO₄ to reflect

SHINE process conditions) was passed through a 2 cm I.D. x 10 cm long extraction column in 2 h, and ^{99}Mo was retained on the extraction column. The ^{99}Mo -loaded column was washed with sulfuric acid (pH 1) and water and stripped using 0.1 M NaOH. Uranium and most fission products passed through the extraction column. The crude ^{99}Mo product was co-eluted with iodine — predominately as ^{131}I , ^{133}I , ^{135}I — along with ^{103}Ru , ^{105}Rh , ^{125}Sn , ^{127}Sb (see the supplementary materials for a complete partitioning of radionuclides). Subsequently, the Mo product (~0.9 L) was transferred directly to a shielded cell, acidified to pH 2, and passed through a ^{99}Mo concentration column (1 cm I.D. x 1 cm long) in 1 h. The column was then washed with 0.01 M HNO_3 , followed by water, and the Mo was quantitatively stripped with 1 M NaOH to obtain 25 mL of ^{99}Mo product — a solution volume acceptable for the LMC process (<50 mL). The majority of co-eluted iodine isotopes, along with ^{103}Ru (~90%) and ^{105}Rh (~60%), were separated from the Mo in the concentration column loading step, with more than half of ^{125}Sn and ^{127}Sb partitioning in the alkaline Mo strip step (complete partitioning of fission products between process streams is included in the Supplementary Materials).

For final purification by the LMC process, the concentration column Mo product was converted to 1 M HNO_3 using 10 M HNO_3 . Any remaining iodine was removed from the ^{99}Mo product by precipitation of AgI. The key purification step was the selective precipitation of Mo with alpha-benzoin oxime. Precipitated Mo complex was washed to remove residual radioisotopes, primarily I, Ru, and Rh (minor amounts of radioactive Zr, Nb, Eu, La, Ce, and Cs were at measurable levels). The Mo precipitate was then dissolved with alkaline peroxide and passed through a three-phase column of Ag-coated charcoal (removal of organics and iodine), hydrated zirconium oxide (removal of cations), and activated charcoal (removal of organics). Purified ^{99}Mo was recovered as sodium molybdate in ~55 mL of ~0.2 M NaOH. The product

was sampled for analysis and shipped to GE Healthcare in the UK for product testing. The chemical yield of ^{99}Mo was >80%.

Figure 3 shows the gamma spectra (150-950 keV) for several important samples collected during the extraction, concentration, and purification of ^{99}Mo . A majority of the fission products and uranium were removed with the extraction column, while the concentration column removed major portions of iodine, ^{103}Ru , and ^{105}Rh . After the LMC process, only ^{99}Mo peaks were identifiable in the gamma spectrum.

The ^{99}Mo produced by using recycled LEU uranyl sulfate solution met the European Pharmacopeia purity specifications and fit into the existing supply chain by GE HealthCare (17). The product was successfully loaded onto a commercial $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generator, and then the $^{99\text{m}}\text{Tc}$ was eluted from the generator and successfully tested with two commercial radiopharmaceutical kits. The $^{99\text{m}}\text{Tc}$ eluate met the United States Pharmacopoeia standards.

CONCLUSION

To our knowledge, this is the first time greater than 37 GBq of ^{99}Mo has been produced by inducing fission in an LEU uranyl sulfate solution using an electron linear accelerator. Accelerator production of ^{99}Mo using an aqueous LEU salt solution will allow repeated use of the target solution and reduced waste generation. Under the conditions studied, the complex solution chemistry of the irradiated solution did not have a negative effect on ^{99}Mo recovery or purity, nor did it have adverse physical or chemical effects on the LEU solution. Also, long-lived radioisotopes that built up over nine previous irradiations did not affect product purity. The recovery and purification process used here produced ^{99}Mo that met purity specifications and was demonstrated to fit in the existing supply chain. Argonne phase II experiments will produce up to 740 GBq ^{99}Mo .

ACKNOWLEDGEMENTS

Thanks to Kurt Alford, James Bailey, James Byrnes, Roman Gromov, Lohman Hafenrichter, Andrew Hebden, James Jerden, Charles Jonah, Brad Micklich, Kevin Quigley, John Schneider, Zaijing Sun, and Kenneth Wesolowski for their contributions.

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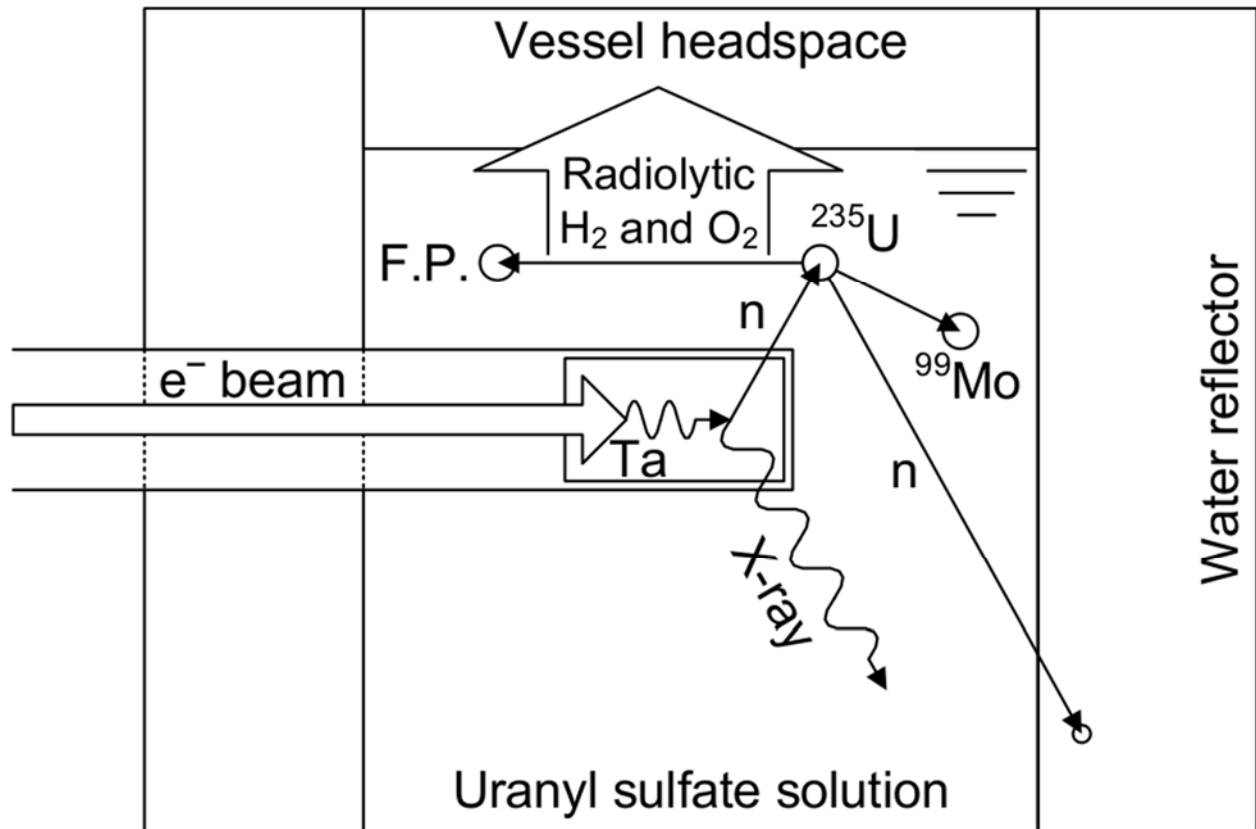


FIGURE 1. Schematic for neutron generation in a Ta target to induce fission in ^{235}U for production of ^{99}Mo . (F.P. stands for fission products.)

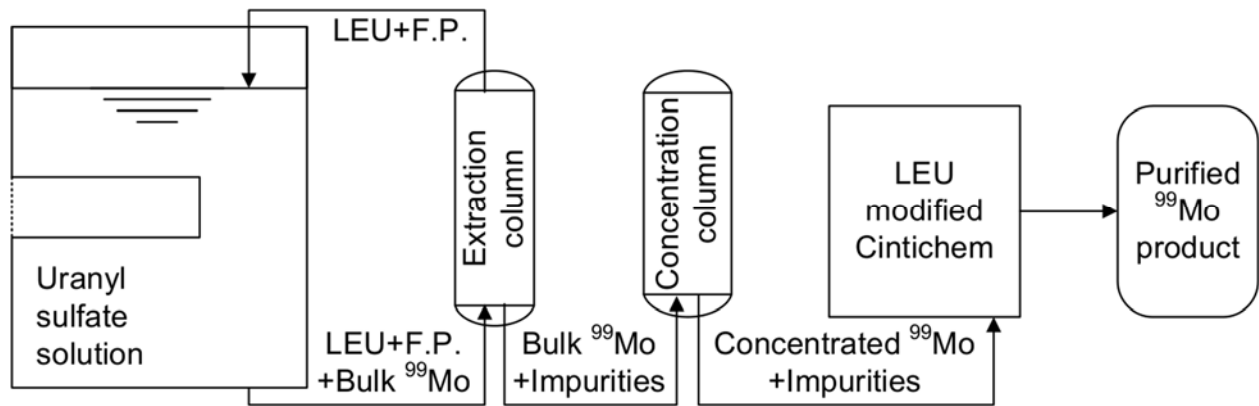


FIGURE 2. Key components of the ^{99}Mo recovery and purification process.

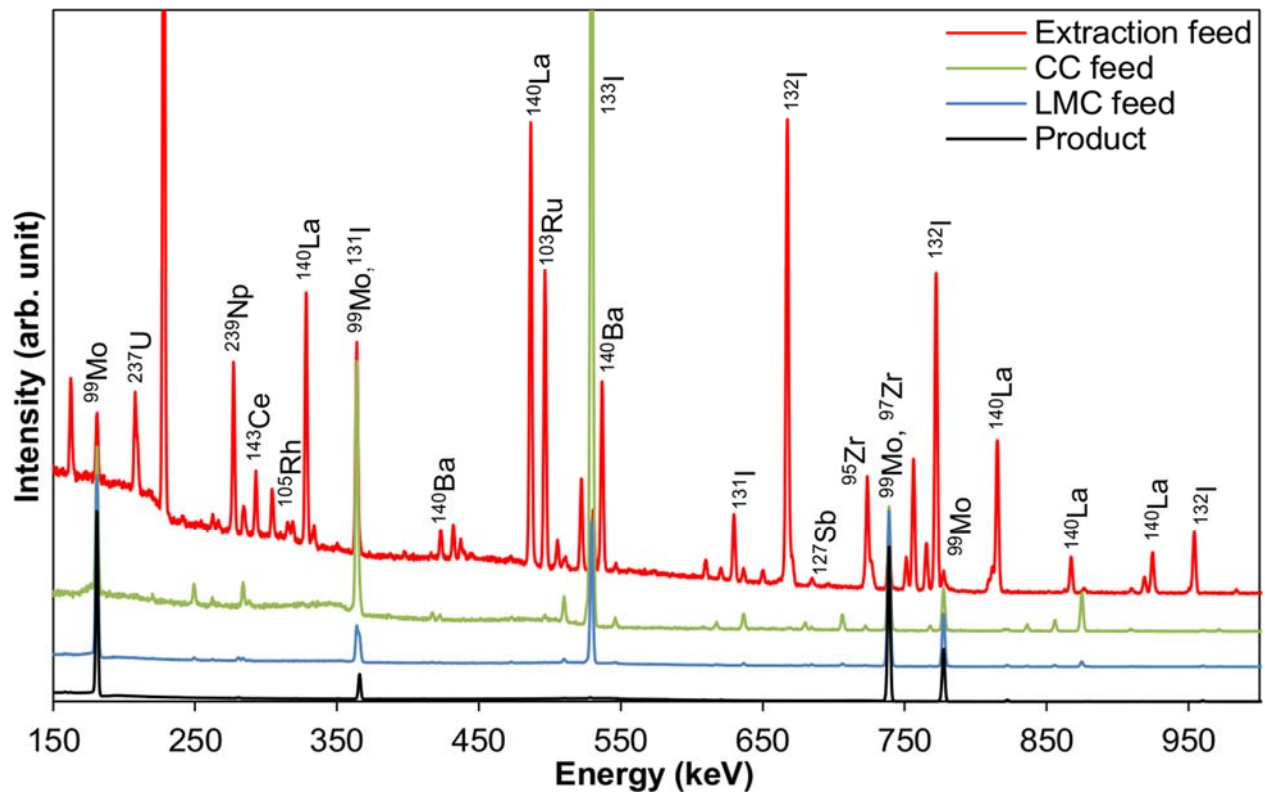


FIGURE 3. Spectra shown for irradiated target solution/extraction column feed (red), concentration column feed (green), LMC feed (blue), and final product (black).

Materials and Methods

Experimental Details

The irradiation was performed by impinging a LINAC electron beam on a water-cooled tantalum target, which was located in a dry well of the stainless steel target solution vessel (TSV) holding 5 L of low enriched uranium (LEU) solution as uranyl sulfate. Figure S1 shows the irradiation vessel and key components such as the Ta target, cooling water reflector, electron beam, condensers, and LEU solution located within a lead shielded cell. Irradiated uranyl sulfate solution was passed through a titania extraction column, washed with 0.05 M H₂SO₄, then H₂O, and a pH-adjustment NaOH wash step was applied. The retained Mo was stripped from the column using 0.1 M NaOH. All operations were performed at 80°C. A LabVIEW[®]-based control system was used to remotely control all of the solenoid valves used to pass the irradiated target solution, wash solutions, and strip solutions through the extraction column located in a negative-pressure secondary containment glovebox. The ⁹⁹Mo-rich extraction column product was acidified with HNO₃ to pH 2 and passed through a second titania column (concentration column). This was followed by 0.01 M HNO₃ and H₂O washes, with final Mo stripping from the concentration column using 1 M NaOH. All concentration column operations were performed at 80°C. The ⁹⁹Mo-rich product, now in 1 M NaOH, was acidified to 1 M H⁺ with 10 M HNO₃ and further purified using the LEU Modified Cintichem (LMC) process (13). Concentration column operations and the LMC process were performed in a hot cell using manipulators.

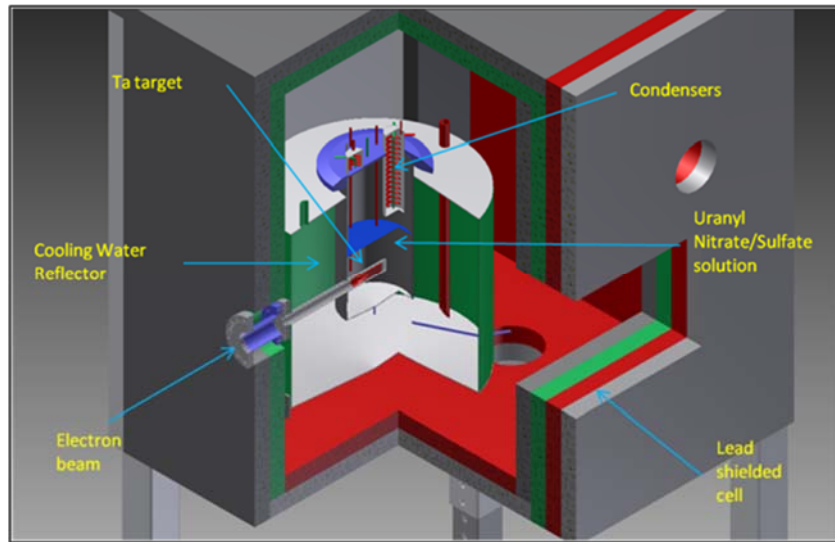


FIGURE S1. Irradiation vessel enclosed within Pb shielded cell.

Gas Analysis System

The LEU solution remained inside the TSV during irradiation, as shown in Figure S2. The headspace gas of the TSV, which was held slightly negative (1-3 in. H₂O) by the gas collection system, was recirculated through a H₂-O₂ recombining catalyst using an explosion proof pump (Air Dimensions Inc., model R221-FV-RA1-AHK). The catalyst re-combines the H₂ and O₂ produced from the radiolysis of water. This was done to maintain the concentration of hydrogen in the headspace of the TSV below the operating limit of 1%, four times lower than the lower flammability limit of H₂ of 4%. The catalyst was heated to 115°C to facilitate the removal of water generated in the bed during the recombination of hydrogen and oxygen. This operating temperature also prevented condensation of water vapor — due to humidity in the headspace gas — on the catalyst surface. Previous experimental runs showed that when the humidity was too high, humidity-induced condensate formed and inhibited the H₂-O₂ recombining activity of the catalyst. A condenser placed before the recirculating pump also served to drop the humidity of the headspace gas prior to entering the catalyst. Part of the safety basis for these experiments was to keep the concentration of hydrogen at $\leq 1\%$. When necessary, air was added to the TSV headspace to control the concentration of hydrogen. The air was added using a calibrated mass flow controller (OMEGA Engineering, Inc., model FMA5502-ST).

A diaphragm pump (KNF Neuberger, Inc., Model 810) was used to transport the gas from the headspace of the TSV to the gas analysis system. The gas was pulled from the headspace, analyzed, and returned to the vessel. Headspace gases were analyzed using a

Pfeiffer PrismaPlus[®] QMG 220 Residual Gas Analyzer (RGA), which was equipped with a quadrupole mass filter and a secondary electron multiplier (SEM) detector.

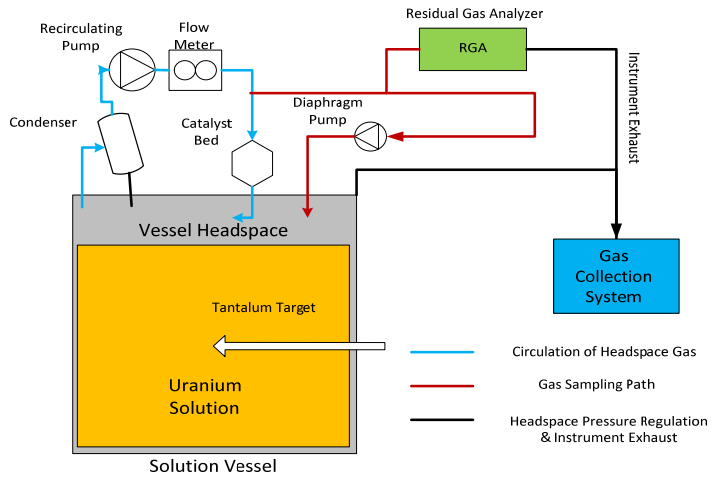


FIGURE S2. Diagram of the target solution vessel and analytical and gas collection systems.

Gas Collection System

The gas collection system (Figure S3) maintained the TSV at sub-atmospheric pressure, which was a safety requirement for the experiments. The TSV was kept sub-atmospheric to prevent the release of fission gases ^{135}Xe , ^{85}Kr , and volatile radioisotopes of iodine.

The gas collection system was also designed as an outlet for potential excess gases, thus preventing over-pressurization of the TSV. Waste gases from the analytical system were exhausted to this system. Gases were transported to high pressure cylinders using two chambers. Chamber #1 was connected to the TSV and analytical systems. It housed a pressure transducer (OMEGA Engineering, Inc., model MMA030V10H3C0T3A6CE) and a diaphragm pump (GAST Manufacturing Corporation, model D0AP704AAEMD).

The inlet of the pump was open to the chamber volume. The outlet of the pump was connected to Chamber #2. The pump inside Chamber #1 was actuated when the pressure reached 11.3 psig. This evacuated Chamber #1 and filled chamber #2. Chamber #2 contained a pressure transducer (OMEGA Engineering, Inc., model MMG5.0KV10P2C0T3A6CE) and a scuba pump (NARDI Compressor, model Atlantic 100). The pump inlet was open to the chamber volume. The outlet was connected to high-pressure cylinders. When the pressure inside Chamber #2 reached 4 psig, the scuba pump actuated and filled the high-pressure cylinder.

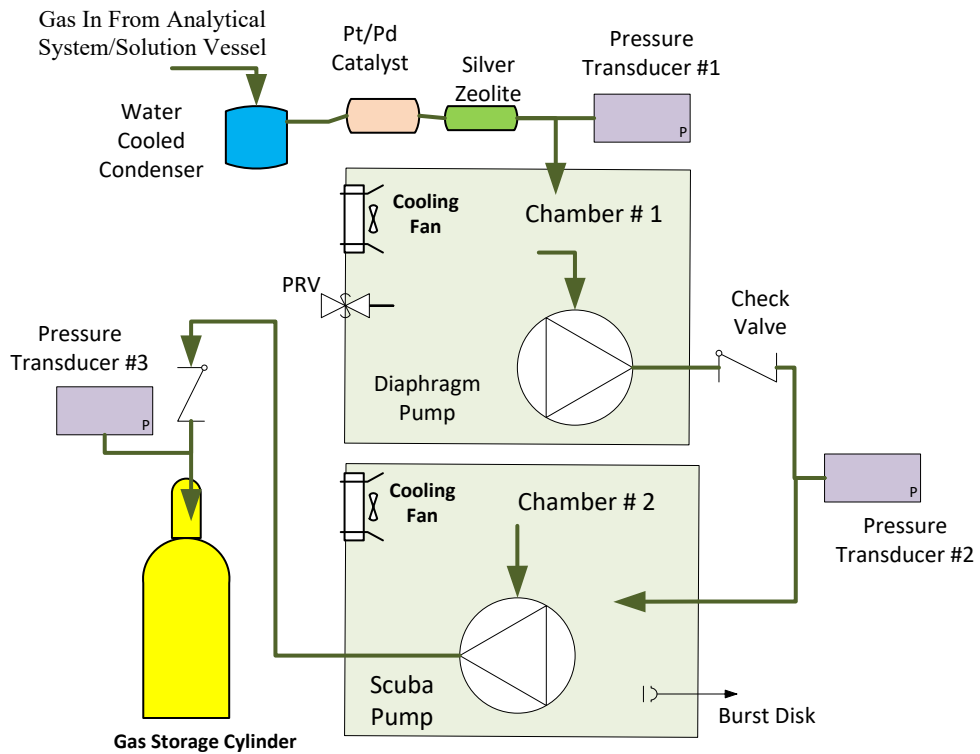


FIGURE S3. Diagram of the gas collection system.

MCNPX Calculations

A detailed model of the experimental setup was developed and simulated with MCNPX, a Monte Carlo particle transport code developed by Los Alamos National Laboratory. The main goal of this particular simulation study was to obtain fission-rate profiles and calculate the end-of-bombardment (EOB) activity of ^{99}Mo based on the total number of fissions in the target solution. The model used for the MCNPX simulations included a section of the vacuum beam pipe housing a cylindrical (with a conical cut in the middle) tantalum photo-neutron generator at the end. The tantalum convertor was positioned at the center of the target solution contained in the 304SS target vessel. The target solution was modeled as 5 L of uranyl sulfate at 130 g-U/L, including 19.8% LEU. The model also included the empty dry wells and the water reflector tank as part of the TSV assembly.

The electron beam was sampled with a 6-mm FWHM Gaussian distribution in both the X- and Z-direction inside the vacuum beam pipe traveling mono-directionally along the Y-axis towards the tantalum convertor. The energy of the beam was 35 MeV. All results were normalized per 1 kW of incident beam power. Fission-rate profiles obtained from this simulation are presented in Figure S4. The results show a peak fission rate density of $3.43\text{E}+8$ fissions/s/cm³ per kW of beam power. This is almost entirely due to slow neutron-induced fission of ^{235}U . Fast fission of ^{238}U and photo-fissions (^{235}U and ^{238}U) are only 0.14% and 0.56% of ^{235}U fission, respectively. The total fission rate of ^{235}U was determined to be $6.42\text{E}+11$ fissions/s per kW of beam power.

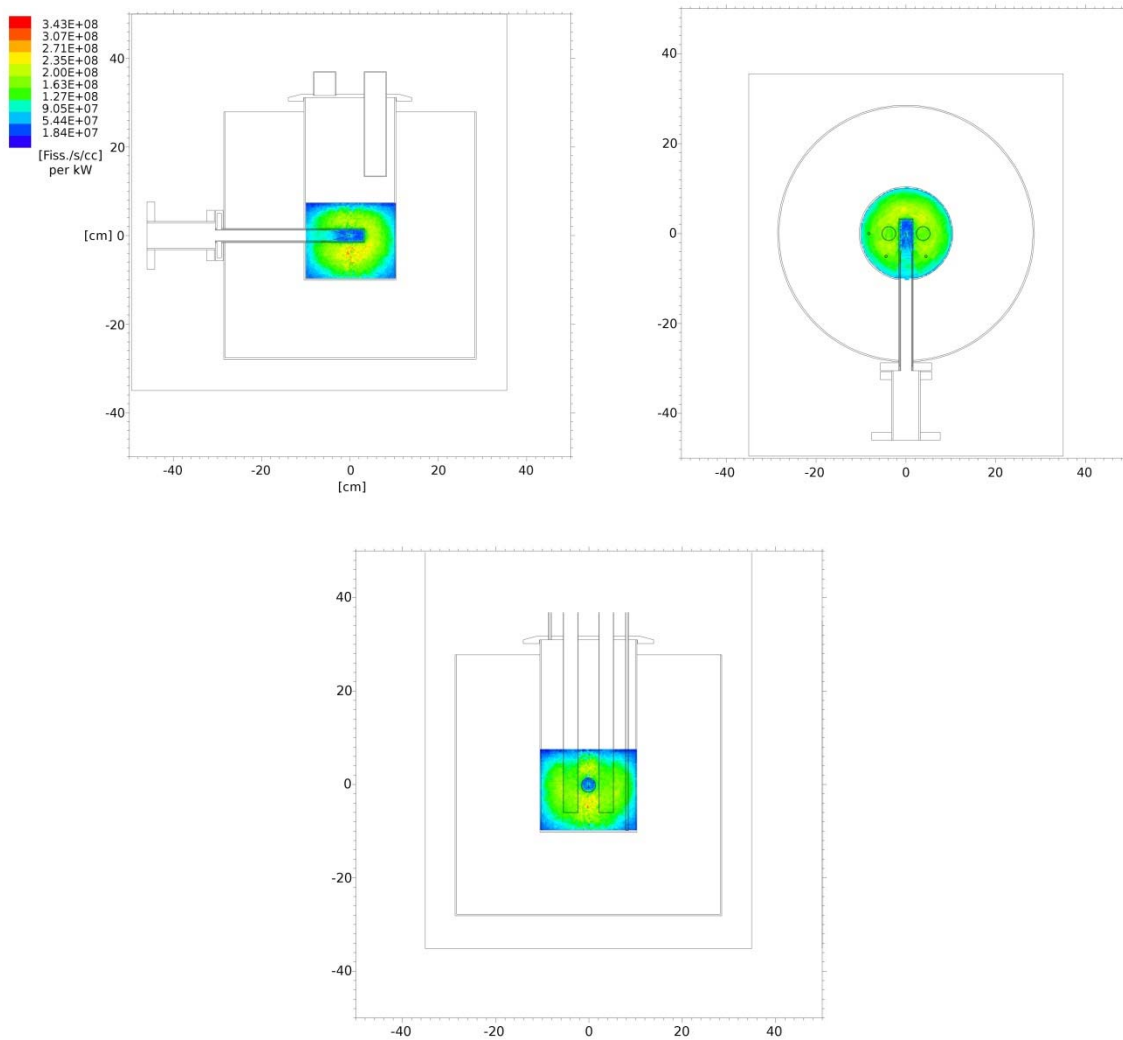


FIGURE S4. Uranium fission profiles [fissions/s/cm³] in the target solution simulated with MCNPX. YZ plane is in the top left, XY plane is in the top right, and XZ plane is in the bottom. The electron beam is traveling along the Y-axis in this geometry. *Note: because of the nature of this tally, results in the parts of the geometry where there is no uranyl sulfate have no meaning.*

The EOB activity of ^{99}Mo was calculated based on equation (1) using the fission rate obtained from the MCNPX simulation. A comparison of ^{99}Mo EOB activity for experimental and theoretical conditions is shown in Figure S5. This result predicted 66.6 GBq of ^{99}Mo EOB activity after 18 hours of irradiation with a 35 MeV, 10 kW electron beam.

$$A = FR \times P \times Y \times (1 - e^{-(\ln 2/T)t}) \quad (1)$$

A – EOB activity

FR – Fission rate = $6.42\text{E}+11$ fissions/s per kW

P – Fission power

Y – Cumulative fission yield of ^{99}Mo = $6.11\text{E}-2$

T – Half-Life of ^{99}Mo = 65.94 h

t – Irradiation time

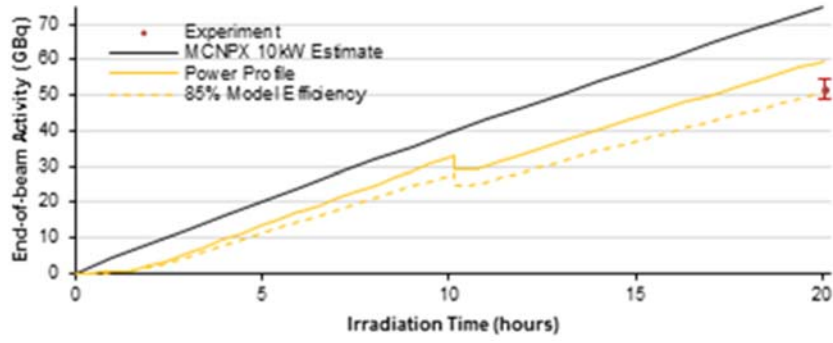


FIGURE S5. Comparison of theoretical results to experimental results for ^{99}Mo production EOB.

Gamma Counting

All generated samples were gamma counted using a HPGe detector (Ortec), and spectra were analyzed using GammaVision software (Ortec). The detector was calibrated for efficiency with an Eckert & Ziegler multinuclide standard. Due to the complex spectra of multiple radionuclides present in most of the samples, only certain radionuclides could be clearly identified (Table S1). To better detect ^{99}Mo activity in the column effluent and acid wash, it was assumed that transient equilibrium had occurred during gamma counting of all samples, so the half-life of ^{99}Mo (65.9 h) was used as the half-life for $^{99\text{m}}\text{Tc}$. The $^{99\text{m}}\text{Tc}$ activity represents Mo behavior, where some ^{99}Mo was found in the column effluent and acid wash samples (based on $^{99\text{m}}\text{Tc}$ activity), but large errors are associated with those values. Errors associated with the gamma counter are $\pm 5\%$. Due to its short half-life, ^{132}I was tracked via the half-life of its parent, ^{132}Te ; however, its activity is indicative of tellurium chemistry and not that of iodine. The activities of the daughters in other mother/daughter pairs (Zr/Nb-95, Zr/Nb-97, and Ba/La-140) are generally not indicative of the daughter or the parent when the time after separation has occurred is short. Signals were seen for these isotopes, but correcting to EOB is highly inaccurate. As a result most daughter activities are not shown. It is important to note that ^{131}I activity that grew in from decay of $^{131\text{m}}\text{Te}$ between the time of separation and counting needs to be taken into account.

TABLE S1. List of radionuclides, their gamma energy, half-life, and branching ratios.

Nuclide	Energy (keV)	Half-life	Branching Ratio
¹³² Te	49.82	78.2 h	14.40%
¹³² I	522.68	78.2 h	16.09%
¹⁴⁷ Nd	91.1	11.0 d	27.90%
⁹⁹ Mo	181.07	65.9 h	6.00%
^{99m} Tc	140.5	65.9 h	87.2%
²³⁷ U	208.01	6.75 d	21.90%
²³⁹ Np	277.6	56.5 h	14.10%
¹⁴³ Ce	293.27	33 h	42.80%
¹⁰⁵ Rh	318.9	35.4 h	19.20%
¹⁵¹ Pm	340.06	28.4 h	22.40%
¹⁴⁰ Ba	423.7	12.7 d	3.15%
¹⁰³ Ru	497.08	39.25 d	91%
¹³³ I	529.87	20.8 h	87%
⁹¹ Sr	555.56	9.5 h	61.50%
¹³¹ I	636.97	8.04 d	7.30%
⁹⁷ Nb	657.92	72.1 m	98.20%
¹³⁷ Cs	661.66	30 y	85.10%
¹²⁷ Sb	685.5	92.4 h	35.70%
⁹⁷ Zr	743.36	16.9 h	98%
⁹⁵ Zr	756.73	64 d	54.50%
⁹⁵ Nb	765.81	34.97 d	99.80%
^{131m} Te	793.77	30 h	13.80%
¹³⁶ Cs	818.51	13.2 d	99.70%
¹²⁵ Sn	1088.9	9.6 d	4.20%
¹⁵⁶ Eu	1230.69	15.2 d	8.80%
¹³⁵ I	1260.41	6.57 h	28.90%
¹⁴⁰ La	1596.21	40.27 h	95.40%

Supplementary Text

Extraction Column

Table S2 shows gamma counting results for the extraction column fractions. Column effluent (CE) fractions are collected 30 minutes apart. The acid wash (AW#1) fraction was collected after 3.75 minutes of washing the extraction column with 0.05 M H₂SO₄. The strip fractions were collected 9 minutes apart. Activities highlighted in yellow had 1 σ values greater than 20%. About 8% of the ¹³¹I activity after separation is due to decay of ^{131m}Te.

TABLE S2. Gamma counting results for the extraction column.

Radio Nuclide	CE#1 (KBq/g)	CE#2 (KBq/g)	CE#3 (KBq/g)	CE#4 (KBq/g)	AW#1 (KBq/g)	Strip#1 (KBq/g)	Strip#2 (KBq/g)	Strip#3 (KBq/g)	Strip#4 (KBq/g)	Strip#5 (KBq/g)
⁹⁵ Zr	38.3	2.2	2.6	2.6	2.6	0.8	1.1	0.5	0.5	0.5
²³⁷ U	333	629	740	740	444	18.5	18.5	11.1	7.4	3.7
¹⁵⁶ Eu	7.4	18.5	22.2	22.2	18.5	3.7	3.7	1.5	1.5	1.5
¹³⁷ Cs	3	7	6.7	5.9	5.9	0.4	0.4	0.2	0.1	0.1
²³⁹ Np	7000	14800	15900	15900	10700	111	74	74	74	37
⁹⁹ Mo	111	259	259	185	259	8140	5920	2775	1295	629
¹⁰³ Ru	115	270	303	315	344	44.4	104	85.1	59.2	40.7
¹³² I	1040	2700	3400	3500	5900	74	37	37	37	37
^{131m} Te	555	740	1110	2100	1110	296	259	148	148	74
¹³¹ I	48.1	88.8	99.9	115	170	4800	6700	4400	2000	851
¹³³ I	1480	2960	3700	3700	3330	77700	111000	74000	32200	14100
¹³⁶ Cs	1.9	5.2	6.3	10.4	7	0.7	0.1	0.1	0.4	0.4
¹⁴⁰ Ba	337	1300	1600	1700	1300	37	33.3	18.5	14.8	7.4
¹⁴³ Ce	6300	13700	14100	14400	9600	111	296	185	111	111
^{99m} Tc	111	259	259	185	259	8100	5900	2800	1300	629
¹³² Te	1000	2700	3400	3500	5900	74	37	37	37	37
¹⁰⁵ Rh	2400	5600	5900	6300	5200	1100	1200	962	222	237
¹²⁵ Sn	18.5	51.8	37	74	74	18.9	28.1	20.7	15.9	13
¹²⁷ Sb	30	66.6	66.6	70.3	51.8	133	315	318	255	204
¹⁴⁷ Nd	444	1000	962	962	666	7.4	7.4	3.7	3.7	3.7
¹⁵¹ Pm	925	1700	2600	1500	1500	1100	1100	740	370	333

Concentration Column and LEU Modified Cintichem Process

The distribution of isotopes and corresponding 1σ values for the concentration column (CC) are shown in Table S3. After acidification of the CC product, the solution was filtered with a polypropylene filter (40 mm, 0.3 μm , Zenpure) and the radionuclides found on the filter and in the concentration column fractions are listed in Table S3. The product from the CC was fed into the LMC process for final purification. Activities detected in the LMC process are shown in Table S4.

TABLE S3. Isotope partitioning for the concentration column at EOB.

Radio-nuclide	Feed (MBq)	Eluent (MBq)	HNO ₃ Wash (MBq)	H ₂ O Wash (MBq)	LCM Liquid Waste (MBq)	⁹⁹ Mo Product (MBq)	Filter (MBq)
	1 σ , %	1 σ , %	1 σ , %	1 σ , %	1 σ , %	1 σ , %	1 σ , %
⁹⁹ Mo	52500	96.2	7.4	55.5	104	54400	722
	2.7	21	13	2.6	2.7	2.3	2.3
¹⁰³ Ru	118	102	1.2	0.2	1.1	11.8	6.7
	2.2	1.9	2.4	3.4	2	9.3	1.9
¹³¹ I	7100	3800	198	53.3	29.6	1040	105
	1.9	1.9	1.9	2	1.9	2.8	2
¹³³ I	121000	65500	3400	910	490	17200	46
	1.9	1.9	1.9	1.9	1.9	1.9	74
¹³⁶ Cs	3.7	0.7	0.03	0.01	0.04	1.1	0.01
	13	(MDA)	(MDA)	(MDA)	7.6	26	(MDA)
¹⁰⁵ Rh	270	163	3.7	0.7	0.7	74	18.5
	21	13	15	20	(MDA)	28	24
¹²⁵ Sn	33.3	N/A	1.1	0.4	2.1	37	0.37
	28		19	19	4.3	26	25
¹²⁷ Sb	200	3	0.1	0.04	14.4	115	0.2
	3.6	30	(MDA)	(MDA)	3.1	4	(MDA)
⁹¹ Sr	2220	681	22.2	7.4	11.1	520	N/A
	(MDA)	16	17	20	25	(MDA)	
¹³⁵ I	182000	109500	5800	1500	860	28500	N/A
	2.7	2.3	2.4	2.3	4.6	5.8	

N/A – data not available (more than 12 half-lives), MDA – minimum detectable activity value reported

TABLE S4. Activities detected in LMC fractions at EOB.

Radio-nuclide	RF-1 (MBq)	RFW (MBq)	LMC product (MBq)
	1 σ , %	1 σ , %	1 σ , %
⁹⁵ Zr	1.85	0.15	0.15
	MDA	8.8	4
⁹⁵ Nb	3.33	0.037	0.33
	11	16	2.2
⁹⁹ Mo	48800	115	47000
	2.3	3.6	2.3
¹⁰³ Ru	7.4	7.4	0.15
	14.5	1.9	3.1
¹³¹ I	744	333	0.19
	3	1.9	26
¹³³ I	12600	5550	2.22
	1.9	1.9	2.9
¹²⁵ Sn	33.3	22.2	0.11
	19	3.7	MDA
¹²⁷ Sb	107	122	0.037
	4.9	3.2	MDA

The purity specifications for the final ^{99}Mo product are given in Table S5.

TABLE S5. Radionuclidic purity in ^{99}Mo product solution from the LMC process at 36 h after EOB.

Ratio	Determined value 36 h after EOB	Product Specification	Within specification
$^{131}\text{I}/^{99}\text{Mo}$	5.28E-06	$\leq 5 \times 10^{-5}$	YES
$^{103}\text{Ru}/^{99}\text{Mo}$	4.91E-06	$\leq 5 \times 10^{-5}$	YES
$\Sigma\alpha/^{99}\text{Mo}$	<6E-12	$\leq 1 \times 10^{-9}$	YES