ABSTRACT

Production of $^{99}$Mo in 1-neutron-exchange reactions between natural isotopes $^{100}$Mo + $^{98}$Mo → $^{99}$Mo + $^{99}$Mo and $^{100}$Mo + $^{98}$Mo → $^{99}$Mo + $^{98}$Mo + n, is proposed. A 1,000 MeV mixed beam of 33-fold ionized natural $^{98}$Mo$^{++}$ and $^{100}$Mo$^{++}$, emitted by ATLAS Linac, is injected into strong focusing Precetron (‘Exyder’), 1 m in DIA, $V = 10^5$ cm$^3$ to form self-colliding beam $migma$ configuration dynamically stabilized by non-linear electron-cloud method with densities at or above space charge limit, $n ≥ 10^{12}$ cm$^{-3}$, with Luminosity $\sim 10^{36}$ - $10^{39}$ cm$^{-2}$ s$^{-1}$ confinement $\tau >> 20$ s. $^{99}$Mo, non-interacted $^{100}$Mo + $^{98}$Mo and other reaction products are magnetically transported into orthogonal magnetic spectrometer automatically separating $^{99}$Mo into collector. Production of 1 mg of $^{99}$Mo a day to provide 1,000 medical procedures (“1K Molytron”) requires injection of $i = 1,000$ pµa (particle microampere) $^{100}$Mo + $^{98}$Mo potentially available at IMP Hangzhou, China. Proposed steps: (1) Proof-of-Principle (POP) test using ATLAS $^{98}$Mo + $^{100}$Mo injection $i = 1$ pµa into Exyder to produce 1 µg of $^{99}$Mo/day, providing for 1 medical procedure (“Micro-Molytron’). (2) Build booster with replica of IMP injector for ATLAS that would deliver $i=10,000$ pµa to generate 10 mg $^{99}$Mo/day for 10,000 procedures at electricity cost of $1 per procedure (“10K Molytron”) (3) Build 10 units of ‘10K Molytrons’ manufacturing plants to meet entire global daily $^{99}$Mo demand of 100,000 procedures. Est. capital cost: $30 million/unit.

1. INTRODUCTION

Medical isotopes are soft radiation sources that illuminate pathology of interest in the body’s interior. We propose to upgrade the proven Precetron technology used in light stable isotope production, for the task of production of artificial short-lived isotope Molybdenum 99, $^{99}$Mo, half-life 66 hours. Precetron is the only particle beam system that had stably operated for hours at the beam densities 10 times the “space charge limit.”

$^{99}$Mo decays into Technetium-99, Tc, which has half-life of 6 hours and is the medical radiation source. The parent/daughter pair, $^{99}$Mo/$^{99}$T, produces useful diagnostic radiation for up to 6 days. It is used in 80% of all diagnostic imaging procedures, approximately 40 million times a year, mostly for imaging of blood flow to the heart and the spread of cancer to bones. Strength of a $^{99}$Mo isotope radiation source is measured in 6-days Curies, “6-day-Ci”. One 6-day-Ci performs 20 medical procedures. One gram of

* Single set of parameters considered.
$^{99}\text{Mo}$ equals 50,000 6-day Ci, hence can do 1,000,000 procedures. [For definitions and numerical data relevant to Molytron see APPENDIX].

Mo-99 is currently produced from uranium-235, $^{235}\text{U}$, via fission of $^{235}\text{U}$ induced by slow (‘thermal’) neutron: $n + ^{235}\text{U} \rightarrow ^{99}\text{Mo} + ^{134}\text{Xe} + 2.5 n + 215\text{ MeV}$. Natural uranium consists of 0.7% $^{235}\text{U}$ and 99.3% uranium 238, $^{238}\text{U}$. Both, $^{235}\text{U}$ and $^{238}\text{U}$ are fissionable but the former has 1,000 times greater cross section; it is the energy source in both, electricity producing nuclear reactors and nuclear defense technologies.

Mo-99 is currently produced in old research reactors fueled with ‘Highly Enriched Uranium’ (HEU) which contains more than 20% $^{235}\text{U}$. Since HEU could be enriched to weapons grade (> 40%), production of Mo-99 from $^{235}\text{U}$ raises serious concern in the control of nuclear weapon proliferation. US Government has closed all $^{99}\text{Mo}$ producing reactors in the United States and seeks methods and means for producing Mo-99 without reactors and without HEU and. Research on production of $^{99}\text{Mo}$ from LEU (Low Enriched Uranium, about 3%) are actively pursued by two US companies.

2. REACTIONS

Molybdenum-99 will be created in Molytron in 1–neutron–exchange endoergic reaction:

$$^{100}\text{Mo}^{\text{+33}} + ^{100}\text{Mo}^{\text{+33}} \rightarrow ^{99}\text{Mo}^{\text{+33}} + ^{99}\text{Mo}^{\text{+33}} - 2.35\text{MeV} \quad (1)$$

and 1–neutron–knockout reactions would take place e.g.

$$^{100}\text{Mo}^{\text{+33}} + ^{98}\text{Mo} \rightarrow ^{99}\text{Mo}^{\text{+33}} + ^{98}\text{Mo} + n - 8.3\text{MeV}. \quad (2)$$

There are 7 natural isotopes of M, with masses: 92, 94, 95, 96, 97, 98 and 100, all are natural isotopes found in mines with occurrence 9.5% to 24% (Fig. 1A).

![Graph showing the masses and natural occurrence of 7 isotopes of Molybdenum.](image1)

**Figure 1A.** Masses and natural occurrence of 7 isotopes of Molybdenum. 1B. 12 kinds of collisions between 7 isotopes in Fig. 1A producing Mo-99.
There are 12 permutations between 7 Mo isotopes all generating $^{99}$Mo (Fig. 1B). Examples are:

\[
\begin{align*}
^{100}\text{Mo} + ^{97}\text{Mo} &\rightarrow ^{99}\text{Mo} + ^{97}\text{Mo} + n - 8.3\text{MeV} \\
^{100}\text{Mo} + ^{98}\text{Mo} &\rightarrow ^{99}\text{Mo} + ^{99}\text{Mo} - 2.35\text{MeV} \\
^{100}\text{Mo} + ^{96}\text{Mo} &\rightarrow ^{99}\text{Mo} + ^{97}\text{Mo} - 1.5\text{MeV} \\
^{100}\text{Mo} + ^{96}\text{Mo} &\rightarrow ^{99}\text{Mo} + ^{96}\text{Mo} + n - 8.3\text{MeV}
\end{align*}
\]

3. OVERALL OPERATION OF MOLYTRON

Molytron has four major components (Fig. 2):

(i) Injector
(ii) EXYDER Strong focusing Precetron /Autocollider
(iii) Beams transporter magnet system
(iv) Magnetic extractor/separator of $^{99}$Mo

![Figure 2. Four components of the system Molytron. Input $^{98}$Mo + $^{100}$Mo; separated outputs: $^{99}$Mo, + non-reacted $^{98}$Mo + $^{100}$Mo + all other fragments/reaction products.](image)

Five stages of physical processes involved (Fig. 3) are:

- **Stage 1.** $180^\circ$ collision between $^{98}\text{Mo}^{33}$ and $^{100}\text{Mo}^{33}$ at COM energy 2,000 MeV, lab Mo energy 1,000 MeV.
- **Stage 2.** Formation of compound nucleus $^{198}\text{X}^{84}$ with 1-neutron exchange.
- **Stage 3.** Two $^{99}\text{Mo}$ newly formed emitted at $180^\circ$ along with other fragments and reaction products from stage 1.
- **Stage 4.** Beams of $^{98}\text{Mo}$ and fragments pass through beam transport quadrupoles and are magnetically transported into stage 5.
- **Stage 5.** Magnetic separator: dispersion spectrometer separates $^{99}\text{Mo}$ from fragments.

The “point-design” presented here considers a single ad-hoc set of instrumental and operating parameters at each stage, hence should be taken as “first-cut” only, open to an order-of-magnitude performance improvements or decline.
Figure 3. 5 stages of physical processes in MOLYTRON.

4. ENERGY OF INJECTED Mo

This design is based on the ATLAC injector because it has highest injection current in the USA; but it could be one of the existing high Z high energy injection systems consisting of ion source and accelerator like those at BNL, CERN and IMP in Hanzhou (China). Design and operation of the injector is not part of this study.
Minimum ion energies determined by the Coulomb barrier. Since $Z = 42, A = 100$ Mo has a high Coulomb threshold in LAB frame (stationary target)

$$T_{\text{LAB}}^{\text{Coul}} = 1.2 \frac{Z^2}{A^{1/3}} = 456 \text{MeV}. \quad (7)$$

Corresponding to COM energy of

$$E_{\text{COM}}^{\text{Coul}} \geq \frac{456}{4} = 114 \text{MeV}. \quad (8)$$

Unlike in classical colliding beams, in Precetron single beam collides nearly head-on, at an average intersecting angle of $165^\circ$ with itself. Hence the laboratory and COM frames of reference are nearly the same by taking

$$T_{\text{LAB}}^{\text{Coul}} = 57 \text{MeV}. \quad (9)$$

There is no cross section information for the Mo+Mo reactions at any energy which defines the first task: measurement of the Mo cross sections (Eqns. 1-6) vs. energy.
The only reliable indication as to what expect for cross section for the reactions (Eqns. 1-6) is the measured cross-section measured \( \sigma = 2 \text{ barn} \) at 14 MeV in production of \( ^{99}\text{Mo} \) in Reaction \( ^{100}\text{Mo} + \text{n} \rightarrow ^{99}\text{Mo} + 2\text{n} - 8.34\text{MeV} \) (10)

Let us estimate cross section using a simple fireball model in which \(^{100}\text{Mo}\) nucleus is as a bag containing 58 neutron and 42 proton; and \(^{98}\text{Mo}\) containing 56 neutron and 42 proton. If the bag is broken, 56 neutron jet will be released against \(^{100}\text{Mo}\) and 58 n jet against \(^{98}\text{Mo}\), both groups to undergo Reaction (10), with \( \sigma = 2 \text{ b} \), provided, however, energy of each nucleon is 14 MeV.

To break the bag requires overcome the nucleon separation energy (BE per nucleon) of 8.5 MeV. It would be necessary to have COM energy equal to or greater than the 100 nucleons \( x \times 8.5 = 850 \) plus a kinetic energy of each of 14 \times 100 = 1,400 total 2,250 MeV or self-colliding beam energy \( T_{\text{LAB}} = 1,125 \text{ MeV} \) par Mo beam. \(^{98}\text{Mo}\) of 1,300 MeV has been reached at ATLAS. In this case, the cross section could reach \( \sigma = (114)2 \text{ b} = 228 \text{ b} \), which is 30 times the geometric, 7.8 b. Order of magnitude greater cross section at the energies above nucleus dissociation fireball has been observed at RHIC at BNL. We chose the initial kinetic energy of injected Mo ions and cross section to be

\[
T_{\text{LAB}}^{\text{Min}} = 1,000\text{MeV}.
\]

\[
\sigma^{(99}\text{Mo}) \sim 10b.
\]

From injector, 33 times ionized beam of \(^{98}\text{Mo}^{33+}\) of the kinetic energy of 1,000 MeV thus resulting in 100 MeV\(\rightarrow\)1,000 MeV collisions at COM energy of 2,000 MeV which is sufficiently close to all nucleon separation energy from Mo nucleus.

5. EXYDER

Centerpiece of the system is the strong focusing autocollider “Precetron” also known as Exyder. There is significant literature on Precetron and Exyder. Advantages of colliding beams over beam-on-target reactions are: (i) reaction rate is proportional to beam current-squared vs linear beam current; (ii) COM energy is the sum of the energies of two beams vs. \( \frac{1}{4} \) of the energy of a single beam. (iii) Moreover, in self-colliders, COM energy \( 2E \) is obtained with one single energy.

Referring to Figs. 6, 7, a mixed beams of \(^{100}\text{Mo}^{33+}\) and \(^{98}\text{Mo}^{33+}\) of kinetic energy \( T \) are injected into the center of symmetry of axially symmetric focusing magnetic field produced by pair of (superconducting) magnet coils of Precetron \(^{1-3}, \text{Exyder}^{14}, \) in which a single a beam collides with itself \((z\text{-axis is horizontal).} \) Unlike in the original Precetron\(^{3,4}, \) the magnetic field of EXYDER will be strong focusing\(^4, \) designed by J. P. Blewett especially for Precetron shortly before his death\(^8,9,11,12 \) (Figs. 12, 13). Strong focusing systems confine 100 times higher beam currents than week focusing systems. Salient property of Precetron is that all orbits, injected with canonical angular momentum \( p = 0, \) intersect in center of symmetry irrespective of particle momentum (energy) \( (\text{Figs. 6, 7).} \) Reaction (1) takes place in a disc shaped reaction chamber between an axially symmetric focusing magnetic field. A fraction of the beam ions, \( f, \) is trapped by the combined collisional and Lorentz dissociation.
Figure 5A. Layout of the injected beam transport system, chamber and superconducting magnet of Precetron MIGMA IV, proposed to be the model for MOLYTRON with strong-focusing magnet. 5B. Midplane cross section of the reaction chamber of Precetron MIGMA IV showing self-colliding orbits formation from dissociated beam of D$_2^+$. 

Injected into center, dissociates $\rightarrow$ D$^*$, D$^0$
Canonical Ang. Momentum $= 0$ is the key to single particle orbit stability.
Production of isotopes $^3$T and $^3$He in 1-neutron and 1 proton exchange reactions in auto-colliding 0.725 MeV $^2$D beam in Precetron (model MIGMA IV), $^2$D + $^2$D $\rightarrow$ $^3$T + p + 4 MeV and $\rightarrow$ $^3$He + n + 3.4 MeV, had been routinely running and described in detail$^{3,4,10}$ (Figs. 5, 7, 8). The nonlinear Van de Pol stabilization technique is described in Figs. 9 and 10. The result is unprecedented steady operation at beam densities 10 times the space charge limit with a confinement time of 24 s, limited only by vacuum (Fig. 11). Measurement and theory are found in Refs. 2-13, and Figs. 9 and 10.

Figure 6. Point-design of Molytron, based on the design and operating experience with Precetron$^{3-6}$. TOP. 1000 MeV beam of $^{98}$Mo$^{3+}$ + $^{100}$Mo$^{3+}$ from ATLAS injector enters radially into EXYDER reaction chamber 0.1 m$^3$ sandwiched between superconducting coils. Reaction products and non-interacted $^{98}$M and $^{100}$Mo propagate axially left, $-z$, and right, $+z$, through strong-focusing system, one on each side. Next, ions enter orthogonal fringe magnet spectrometer that physically separates $^{99}$Mo from all other ions at the collector. MIDDLE. Axial section showing relation of the injection tube with the reactor and extractor system. BOTTOM. Angular dispersion of $-z$ ions is transposed by the fringe field into linear separation by approx. 20 cm of $^{99}$Mo from $^{98}$Mo and $^{100}$Mo, as explained in Fig. 14.
Figure 7. Midplane cross section of the reaction chamber showing auto-colliding orbits of 34 and 40 times ionized Mo nuclei. Top view of the EXYDER reaction chamber. The 450 MeV Mo+33 beam enters the chamber from the left through port #1 and is trapped by 1 electron stripping Mo+33→Mo+34. 6 electron stripped orbit is shown as Mo+40. The unstripped beam flows to the beam dump through port #2 for recuperation. Mo trapping rate of 90% is projected at density n~10^{12} cm^{-3}.

Figure 8. Precursor of MOLYTRON: Precetron MIGMA IV. Accelerator injection energy: D_{2}^{+} of 1.45 MeV. Superconducting NiTi magnet: 6 Tesla on coil, 3.2 Tesla midplane. Vacuum: 10^{-11} torr (static); 10^{-9} (beam-in). Volume is 5 liters, baked for 24 hours at 450°C.
Figure 9. Nonlinear stabilization technique to exceed space charge limit. MIGMA end plates geometry: M = MIGMA; E1, E2 = end plates at z = +3.75 cm; Z1 = half thickness of MIGMA. (B) Precession observed in RF spectrum. Frequency spectrum of observed in Precetron. The observed peak corresponds to the cyclotron frequency of 28 MHz. Two peaks symmetrically placed about it are the sidebands due to precession. The observed separation from the central peak is equal to the precession frequency. $f_P R = \Delta f = 450 \, kHz$. (C) MIGMA particle density vs. r, measured by amplitude of Schotky Noise RF signal.

Figure 10. Critical test: Axial bounce frequency of electrons vs. negative voltage at the plates, $-U_p$, with low $D_2^+$ beam injection, $I = 0.035 \, mA$. (B) The resistance term $p/\alpha$ as a function of the applied electric potential to end plates E1 and E2 [Phys Rev. Lett. 70, 1818 (1993)]. While Landau damping is by linear increase in amplitude, the non-linear damping is achieved by the increase in frequency via increase of voltage. Resistance changes sign from negative (instability) to positive (stability). [Lashinsky and Dewan IEEE Trans. Autom. Contr. 12, 244 (1967); 14, 212 (1969)].
Figure 11. Ion confinement time and ion energy distribution inside reaction chamber. Energy spectrum of the \( D^+ \) observed in Precetron prevents thermalization! Mixed Mo beams will have similar features. (A) Longest ion energy confinement time ever observed at ion kinetic energy below 1 MeV; \( \tau_{1/e} = 24 \pm 4 \) sec. (B) Energy spectrum of the \( D^+ \) expected at observed in Precetron prevents thermalization. Mixed Mo beams will display same features.

Figure 12. Conceptual design by J. Blewett\textsuperscript{8,9} of EXYDER-Alternating Gradient Strong Focusing Precetron. Orbits C and A are defocused and over-focused respectively, hence are lost (axially).
6. TRANSMITTER

The transmitter is a standard system of 2 strong focusing quadrupoles.

7. COLLECTOR OF $^{99}$Mo

After passing through decelerator, the mixed beam enters magnetic Orthogonal magnetic mass spectrometer with vertical space dispersion at the exit of which Mo-99 is collected on one spot (Fig. 14).
Figure 14. Orthogonal magnetic spectrometer selector/extractor of Mo-99, physically separated from all other ions. Schluter, Phys. Rev. Lett. Elevation (A) and top view (B).

8. PARAMETERS OF MOLYTRON

a. Luminosity, L, is a colliding beam reaction rate for $\sigma = 1$ (Fig. 15A).

$$L = 2n^2vV$$  \hspace{1cm} (12)

where $n$ = Mo ion particle density $[\text{cm}^3]$; $v$ = velocity of Mo ion in LAB frame; and $V$ = volume of reactor chamber $[\text{cm}^3]$.

b. $^{99}$Mo production rate/gram s$^{-1}$ (Fig. 15B)

$$R_M = \frac{2n^2vV\sigma}{6 \times 10^{21}}$$  \hspace{1cm} (13)

where $s$ = cross section for production of $^{99}$Mo.

c. Daily rate in mg/day (Fig. 15D)

$$D_M = \frac{2n^2vV\sigma}{6 \times 10^{21}} \times 86,400$$  \hspace{1cm} (14)
d. $^{99}$Mo production rate particles/sec (Fig. 15C)

$$R_p = 2n^2\nu V\sigma$$ (15)

**Figure 15A.** Luminosity, $L$, vs. Mo ion particle density for two values of reactor volume, $V=10^4$ cm$^3$ and $10^5$ cm$^3$. Reference line is visual aid for comparison of Diagrams A-D. **Fig. 15B.** $^{99}$Mo production rate [g/s] vs Mo ion beam particle density.

**Figure 15C.** $^{99}$Mo mass production rate in g per s vs Mo ion particle density for two values of reactor volume, $R_M$. **Fig. 15D.** $^{99}$Mo particle production rate per second, [ps$^{-1}$], vs. particle density, $R_p$.

9. CONCLUSIONS

Basic Molytron operating parameters are:
- 1 mg $^{99}$Mo yields 1,000 medical procedures.
- To produce 1 mg $^{99}$Mo/day with trapping efficiency $f=0$, the injected current of natural Mo required is $i = 160$ pμA [particle microamp].
• Closest to this requirement, albeit higher by a factor of 10, is \( i = 1,000 \, \text{pμa} \) injector at IMP Lanzhou, China. IMP could produce 10 mg \(^{99}\text{Mo}\)/day and facilitate 10,000 procedures/day (‘10K Molytron’), which is 1/10 of the global daily \(^{99}\text{Mo}\) demand.

We propose the 3-phase program as follows:

1. Beam-on-target cross section measurements are a function of Mo energy from 100 to 1,000 MeV of the reactions \(^{100}\text{Mo} + {}^{98}\text{Mo} \rightarrow {}^{99}\text{Mo} + {}^{99}\text{Mo}\) and \(^{100}\text{Mo} + {}^{98}\text{Mo} \rightarrow {}^{99}\text{Mo} + {}^{98}\text{Mo} + n\), to be carried out at the Fragments Spectrometer of ATLAS (see Fig. 4).

2. Proof-of-Principle (POP) experiment using ATLAS injector which could deliver \( i = 1 \, \text{pμa} \) Mo; assuming \( f = 0.1 \) and \( \sigma = 10 \, \text{barn} \), it should generate 1 pμg \(^{99}\text{Mo}\) /day for 1 medical procedure/day.

3. Following POP, a “10K Molytron” manufacturing plant should be built using a 10-fold boosted replica of IMP injector. Est. cost: $40 million.

4. Build ten ‘10K Molytron’ manufacturing plants to be distributed globally to meet the entire global demand of 100,000 procedures for \(^{99}\text{Mo}\).

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REFERENCES

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APPENDIX A

Vital numerical data relevant to MOLYTROM design rounded to ±20%, are:
1. Half-life of Mo-99 is 66 hours i.e. nearly 3 days.
2. Mo-99 is supplied to hospitals on weekly intervals and it is sold in units of “6 Day Curies” or 6-D Ci; 1 Ci is 22% of 1 6-D Ci.
3. 1 g of Mo-99 provides approximately → 50,000 6-D Ci.
4. One Ci provides enough technetium-99 for approximately 20 diagnostic procedures.
5. From (3) and (4), 1 g of Mo-99 provides → 1,000,000 procedures.
6. Market price of Mo-99 is $500 - $1,000 per 6 D Ci per twenty procedures.
7. From (4) and (6), one procedure costs $24 - $50.
8. From (3) and (6), it follows that market price of Mo-99 is $25,000,000 - $50,000,000 per gram. (Capital cost is not included into the price as the reactors are 100% subsidized by local governments).
9. Global production of Mo-99 is 40 g/year or 100 mg/day.
10. A 40,000,000 medical procedures take place annually, 20 million of which within the United States, 20 million all other countries of the world i.e. 95% of the population use 50%; hence, the ultimate potential foreign market is 800 million procedures/ year at $20 per procedure, i.e. $16 billion a year.