PAPER

Influence of enriched $^{100}\text{Mo}$ on Mo reaction yields

Jaewoong Jang$^1$ and Mitsuru Uesaka$^{1,2}$

$^1$ Department of Bioengineering, University of Tokyo, Bunkyo, Tokyo 113-8656, Japan
$^2$ Nuclear Professional School, University of Tokyo, Naka, Ibaraki 319-1188, Japan

E-mail: jangj@korea.ac.kr

Keywords: $^{99}\text{Mo}$, $^{99m}\text{Tc}$, nuclear medicine, electron linear accelerator, cyclotron

Supplementary material for this article is available online

Abstract

In accelerator-driven $^{99}\text{Mo}/^{99m}\text{Tc}$ production, the $^{100}\text{Mo}$ enrichment level should be chosen carefully as it greatly affects the yields of the involved Mo reactions. To facilitate selecting the $^{100}\text{Mo}$ enrichment level, we defined a figure of merit called density change coefficient and developed its calculation program. Density change coefficients calculated for nine commercial enriched $^{100}\text{Mo}$ products are presented and their use in selecting the $^{100}\text{Mo}$ enrichment level is discussed.

1. Introduction

Technetium-$^{99m}\text{Tc}$, arguably the most widely used gamma emitter in nuclear medicine, is obtained via the negatron decay of its precursor, molybdenum-$^{99}\text{Mo}$ ($^{99}\text{Mo}$). Most of this precursor nuclide is produced via the fission reaction $^{235}\text{U}(n,f)^{99}\text{Mo}$ in research reactors. All but one of the major reactors, however, have only about 10 years left until the end of operation (figure 1), resulting in an increasingly unstable $^{99}\text{Mo}$ supply chain; there have been a number of $^{99}\text{Mo}$ supply shortages attributed to the aging of the reactors in the past few decades [1–3]. In response to the unstable $^{99}\text{Mo}$ supply chain, accelerator-based alternative methods of $^{99}\text{Mo}/^{99m}\text{Tc}$ production have been explored extensively, including (figure 2):

- Production of $^{99}\text{Mo}$ via the $^{100}\text{Mo}(\gamma,n)^{99}\text{Mo}$ reaction using electron linear accelerators [4–9].
- Production of $^{99}\text{Mo}$ via the $^{100}\text{Mo}(n,2n)^{99}\text{Mo}$ reaction using fast neutron generators [10–13].
- Production of $^{99m}\text{Tc}$ via the $^{100}\text{Mo}(p,2n)^{99m}\text{Tc}$ reaction using medical cyclotrons [14–25].

In all of the accelerator methods mentioned above, use of enriched $^{100}\text{Mo}$ is necessary: in the $^{100}\text{Mo}(\gamma,n)^{99}\text{Mo}$ and $^{100}\text{Mo}(n,2n)^{99}\text{Mo}$ reaction routes, the use of 99%-enriched $^{100}\text{Mo}$ can provide approximately tenfold increases in the yield and specific yield of $^{99}\text{Mo}$. In the $^{100}\text{Mo}(p,2n)^{99m}\text{Tc}$ reaction route, the use of >99%-enriched $^{100}\text{Mo}$ is required to minimize the production of Tc isotopes other than $^{99m}\text{Tc}$. Such a need for enriched $^{100}\text{Mo}$ has been known for years, and the minimum required $^{100}\text{Mo}$ enrichment levels have been reported by a number of researchers [15, 18, 23, 27].

The required $^{100}\text{Mo}$ enrichment level can be determined by considering (i) the yields of $^{99}\text{Mo}/^{99m}\text{Tc}$, (ii) the yields of impurity nuclides, and (iii) the influence of individual impurity nuclides on the radiation dose and image quality. In order to facilitate selecting the $^{100}\text{Mo}$ enrichment level, we defined a figure of merit that can be used to evaluate (i) and (ii) above. The theoretical basis, calculation results, and use of the figure of merit are presented.

2. Theory

2.1. Molybdenum hierarchy

We classify Mo into three hierarchical groups as described in figure 3: (i) Mo materials, (ii) Mo element, and (iii) Mo isotopes. The Mo materials of interest in this paper were Mo metal and Mo(VI) oxide (MoO$_3$), both of which...
are used in accelerator production of $^{99}$Mo [2, 6, 9, 11], and the former of which is used in cyclotron production of $^{99m}$Tc [2, 16, 18, 21, 23–25, 27]. We hereafter refer to Mo metal as Momet to distinguish it from the Mo element. Based on this hierarchy, we examine how the physical quantities of Mo materials and Mo elements are affected by their $^{100}$Mo content.
2.2. Amount fraction, mass fraction, and enrichment level

The amount fraction of $^{100}$Mo in naturally occurring Mo is $x_{^{100}Mo} = 0.09744$ \cite{28,29}, while the corresponding mass fraction is $w_{^{100}Mo} = \frac{0.09744 \times 100}{199.887 + 0.15 \times 15.999} \approx 0.1015$ (figure 4). Any of the two fraction quantities can be used as the enrichment level. To avoid confusion and to conform to the conventions of enriched $^{100}$Mo vendors \cite{30,31}, this paper assumes that the amount fraction of $^{100}$Mo denotes the enrichment level of $^{100}$Mo. On the other hand, the mass fraction of $^{100}$Mo, as is appropriate for describing the dependence of Mo element on the $^{100}$Mo content, will be used in explaining the influence of enriched $^{100}$Mo on the hierarchical Mo entities.

2.3. Mass fraction of Mo element

The mass fraction of Mo element in Mo$_{met}$ and MoO$_3$ can be expressed as

$$w_{Mo}(w_{^{100}Mo}) = \frac{n_{Mo}}{n_{Mo,^{100}Mo} + n_{O,MoO_3}}$$

$$= \left( \frac{n_{Mo}}{n_{Mo,^{100}Mo}} \right) \left( \frac{n_{Mo}}{n_{Mo,MoO_3}} + \frac{n_{O}}{n_{O,MoO_3}} \right)^{-1}, \quad (1)$$

where $n$ denotes the amount of substance (number of moles), $\overline{M}$ the weighted-average molar mass, $M$ the molar mass, and $A$ represents the mass numbers of the naturally occurring isotopes of the chemical elements.

For nonenriched MoO$_3$, for example, (1) becomes

$$w_{Mo}(0.1015) = \frac{(1)(95.949 \text{ g mol}^{-1})}{(1)(95.949 \text{ g mol}^{-1}) + (3)(15.999 \text{ g mol}^{-1})} \approx 0.66656.$$

If the MoO$_3$ is enriched in $^{100}$Mo and now has a $^{100}$Mo mass fraction of 0.990, the mass fraction of Mo element will increase to

$$w_{Mo}(0.990) = \frac{(1)(99.887 \text{ g mol}^{-1})}{(1)(99.887 \text{ g mol}^{-1}) + (3)(15.999 \text{ g mol}^{-1})} \approx 0.67544.$$

In other words, the mass of Mo element per mass of MoO$_3$ will increase by a factor of

$$w_{Mo}(0.990) \approx \frac{0.67544}{0.66656} \approx 1.0133. \quad (2)$$

By contrast, $w_{Mo}$ in Mo$_{met}$ is unaffected by its associated $^{100}$Mo mass fraction:

$$w_{Mo}(0.990) \approx \frac{(1)(99.887 \text{ g mol}^{-1})}{(1)(99.887 \text{ g mol}^{-1}) + (0)(15.999 \text{ g mol}^{-1})} = 1.0000. \quad (3)$$

Equations (2) and (3) are plotted in figure 5 along with other postenrichment $^{100}$Mo mass fractions. The mass fraction of Mo element in MoO$_3$, although slowly, increases with increasing $^{100}$Mo mass fraction. An
increased mass fraction of a Mo element means an increase in its mass density, which further increases the $^{100}$Mo mass density and in turn contributes to improving the $^{100}$Mo reaction yields. These cascading effects will be explained in the next subsection.

2.4. Density change coefficient

The mass density of $^{100}$Mo can be written as

$$\rho_{^{100}\text{Mo}}(w_{^{100}\text{Mo}}) = w_{^{100}\text{Mo}}/\rho_{\text{Mo-mat}}$$

where the subscript Mo-mat is the placeholder for Mo materials. Because of the dependence of $w_{^{100}\text{Mo}}$ on $w_{^{100}\text{Mo}}$, when $w_{^{100}\text{Mo}}$ is changed, $\rho_{^{100}\text{Mo}}$ is affected not only by the change in $w_{^{100}\text{Mo}}$, but also by $w_{\text{Mo}}$. For instance, $\rho_{^{100}\text{Mo}}(0.1015) = (0.1015)(0.66656)(4.690 \text{ g cm}^{-3}) \approx 0.3173 \text{ g cm}^{-3}$,

which, if the $^{100}$Mo mass fraction is increased to 0.9900, will become

$$\rho_{^{100}\text{Mo}}(0.9900) = (0.9900)(0.67544)(4.690 \text{ g cm}^{-3}) \approx 3.136 \text{ g cm}^{-3}.$$

The $^{100}$Mo mass density will then increase by a factor of

$$\frac{\rho_{^{100}\text{Mo}}(0.9900)}{\rho_{^{100}\text{Mo}}(0.1015)} = \frac{3.136 \text{ g cm}^{-3}}{0.3173 \text{ g cm}^{-3}} \approx 9.884.$$

If the change in $w_{\text{Mo}}$ by $w_{^{100}\text{Mo}}$ had not been taken into account, (5) would have been

$$\frac{(0.9900)(0.66656)(4.690 \text{ g cm}^{-3})}{(0.1015)(0.66656)(4.690 \text{ g cm}^{-3})} = \frac{3.095 \text{ g cm}^{-3}}{0.3173 \text{ g cm}^{-3}} \approx 9.754,$$

leading to an underestimation. Note, also, that (5) is actually the product of (2) and (6):

$$\left[\begin{array}{c}
0.67544 \\
0.66656
\end{array}\right] \left[\begin{array}{c}
0.9900(0.66656)(4.690 \text{ g cm}^{-3}) \\
(0.1015)(0.66656)(4.690 \text{ g cm}^{-3})
\end{array}\right] = \left(\begin{array}{c}
1.0133 \\
0.9754
\end{array}\right) \approx 9.884,$$

in which the role of (1) is clearly shown. Under the same conditions, the $^{100}$Mo mass density increment of Mo$_{\text{net}}$ will be

$$\frac{\rho_{\text{Mo}_{\text{net}}}(0.9900)}{\rho_{\text{Mo}_{\text{net}}}(0.1015)} = \frac{(0.9900)(1.0000)(10.28 \text{ g cm}^{-3})}{(0.1015)(1.0000)(10.28 \text{ g cm}^{-3})} \approx 9.754.$$

The dependence of mass densities of Mo element and $^{100}$Mo on the $^{100}$Mo mass fraction are summarized in figure 6. As shown in the bottom-right panel, the mass density of Mo element $\rho_{\text{Mo}}$ in MoO$_3$ increases with the $^{100}$Mo mass fraction. This positive slope of $\rho_{\text{Mo}}$, then improves the rate of increase in the $^{100}$Mo mass density $\rho_{^{100}\text{Mo}}$, as can be seen from (4). Consequently, MoO$_3$ can have a greater $^{100}$Mo mass density increment than Mo$_{\text{net}}$ when enriched in $^{100}$Mo. However, it should be noted that the absolute $^{100}$Mo mass density in Mo$_{\text{net}}$ remains greater than that in MoO$_3$ by a factor of 3.2. Namely, the ratio between the $^{100}$Mo mass density in nonenriched Mo$_{\text{net}}$ and that in MoO$_3$ is
Equation (7) shows that the $^{100}\text{Mo}$ reaction yields of Mo$_\text{met}$ are greater than those of MoO$_3$ regardless of the $^{100}\text{Mo}$ enrichment level.

Next, we examine how the number densities of the hierarchical Mo entities are affected by the $^{100}\text{Mo}$ mass fraction. The number density of a substance $i$ is related to its mass density by

$$N_i = \frac{\rho_i}{\rho_i} N_A,$$

where $N_A$ is the Avogadro constant. Inserting (4) into (8), the number density of $^{100}\text{Mo}$ is written as

$$N_{^{100}\text{Mo}}(w_{^{100}\text{Mo}}) = \frac{w_{^{100}\text{Mo}} w_{^{100}\text{Mo}} (w_{^{100}\text{Mo}})}{M_{^{100}\text{Mo}}} \frac{\rho_{^{100}\text{Mo}}} {N_A},$$

which, in terms of the $^{100}\text{Mo}$ amount fraction, is equivalent to

$$N_{^{100}\text{Mo}}(x_{^{100}\text{Mo}}) = x_{^{100}\text{Mo}} x_{^{100}\text{Mo}} N_{^{100}\text{Mo}} (x_{^{100}\text{Mo}})$$

or

$$N_{^{100}\text{Mo}}(x_{^{100}\text{Mo}}) = x_{^{100}\text{Mo}} x_{^{100}\text{Mo}} N_{^{100}\text{Mo}} (x_{^{100}\text{Mo}})$$

provided that the Mo material is Mo$_\text{met}$ or MoO$_3$, either of which has $x_{^{100}\text{Mo}} = 1$.

Equations (8) and (9) are plotted in figure 7 for the hierarchical Mo entities. Because $^{100}\text{Mo}$ is the heaviest Mo isotope among the naturally occurring ones, increasing $w_{^{100}\text{Mo}}$ increases $M_{^{100}\text{Mo}}$ and thereby $M_{^{100}\text{Mo}}$. Correspondingly, increasing $w_{^{100}\text{Mo}}$ decreases $N_{^{100}\text{Mo}}$ and $N_{^{100}\text{Mo}}$. Meanwhile, $N_{^{100}\text{Mo}}$ increases linearly (note the logarithmic $y$-axis) with increasing $w_{^{100}\text{Mo}}$.

We now generalize the influence of $w_{^{100}\text{Mo}}$ on $\rho_{^{100}\text{Mo}}$ and $N_{^{100}\text{Mo}}$. Using (9), the $^{100}\text{Mo}$ densities before and after $^{100}\text{Mo}$ enrichment are interrelated by

$$\rho_{^{100}\text{Mo}}(0.1015) \text{ in Mo}_{\text{met}} = \frac{(0.1015)(1.000) (10.28 \text{ g cm}^{-3})}{(0.1015)(0.66656)(4.690 \text{ g cm}^{-3})} \approx 3.288,$$

and the corresponding ratio for $w_{^{100}\text{Mo}} = 0.990$ is

$$\rho_{^{100}\text{Mo}}(0.990) \text{ in Mo}_{\text{met}} = \frac{(0.990)(1.000) (10.28 \text{ g cm}^{-3})}{(0.990)(0.67544)(4.690 \text{ g cm}^{-3})} \approx 3.245.$$
where \( w_{\text{Mo-100}}^{\text{bef}} \) and \( w_{\text{Mo-100}}^{\text{aft}} \) denote the pre- and post-enrichment \(^{100}\text{Mo}\) mass fractions, respectively. Assuming that \( w_{\text{Mo-100}}^{\text{bef}} \) is fixed to be the mass fraction of nonenriched \(^{100}\text{Mo}\), and expressing \(^{100}\text{Mo}\) as a Mo isotope, (11) reduces to

\[
D_{\text{Mo-A}}(w_{\text{Mo-100}}^{\text{aft}}) = \frac{w_{\text{Mo-A}}(w_{\text{Mo-100}}^{\text{aft}})}{w_{\text{Mo-A}}(0.1015)} \cdot \frac{w_{\text{Mo}}(w_{\text{Mo-100}}^{\text{aft}})}{w_{\text{Mo}}(0.1015)},
\]

which we call a density change coefficient (DCC).

Using (10), the DCC can also be defined in terms of the \(^{100}\text{Mo}\) amount fraction:

\[
D_{\text{Mo-A}}(x_{\text{Mo-100}}^{\text{aft}}) = \frac{x_{\text{Mo-A}}(x_{\text{Mo-100}}^{\text{aft}})}{x_{\text{Mo-A}}(0.0974)} \cdot \frac{M_{\text{Mo-mat}}(0.0974)}{M_{\text{Mo-mat}}(x_{\text{Mo-100}}^{\text{aft}})},
\]

As implied by \( w_{\text{Mo}} \) in (12) and \( M_{\text{Mo-mat}} \) in (13), the DCC is dependent on the molecular composition of a Mo material and, therefore, should be calculated separately for \( \text{Mo}_{\text{mat}} \) and \( \text{MoO}_3 \).

3. Methods

3.1. \textit{enrimo}: A DCC calculation program

Despite the concise forms of the DCC, its calculation can be error-prone and time-consuming. To automate DCC calculations and to facilitate data exchange, we developed a Perl program called \textit{enrimo}.

The DCC calculation algorithm of \textit{enrimo} is described in figure 8. As of v1.05, the following conditions can be customized via the command-line options: (i) Mo materials of interest, (ii) the Mo isotope to be enriched (in addition to enriched \(^{100}\text{Mo}\), enriched \(^{98}\text{Mo}\) can be examined for the \(^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}\) reaction route), (iii) the fraction type to refer to the enrichment level, (iv) the range of enrichment levels, (v) the minimum depletion level applied to all of the associated nuclides, and (vi) the order of nuclide depletion in the process of isotopic enrichment. Also, an input file can be used to specify the minimum depletion levels of individual nuclides and the calculation precision.

Once the calculation conditions are specified, \textit{enrimo} prepares the data necessary for DCC calculations and enters the main module. The core task of the main module is to redistribute the fraction quantities of the Mo isotopes according to the given \(^{100}\text{Mo}\) enrichment level. Based on the redistributed fraction quantities, the molar mass of the Mo material under investigation is recalculated, which in turn is used for the DCC calculation via (13). If the mass fraction has been set to represent the enrichment level, (12) is used instead. Finally, the precision of calculation results are adjusted according to the user specifications, and the product nuclides of photon, neutron, and proton reactions on \(^{92,94-98,100}\text{Mo}\) are associated with the calculated DCCs.

Data files are generated each time a series of calculations for a Mo material is completed. The supported output formats are plain text (.dat); LaTeX tabular environment (.tex); comma-separated values (.csv);...
Microsoft Excel 2007 (.xlsx); JavaScript Object Notation (.json); and YAML (.yaml). Considering the typical use frequencies of these formats, we have set .dat and .xlsx as the default output formats. More detailed descriptions of enrimo are documented in its source code, which is available in [32].

3.2. DCC calculation conditions
Two enriched isotope vendors use gas centrifuges for $^{100}$Mo enrichment [33, 34]. When $^{92,94–98,100}$Mo]Mo(VI) fluoride is fed into a gas centrifuge, the light stream becomes the waste and the heavy stream containing $^{100}$Mo becomes the product, leading to the depletion of lighter Mo isotopes in the process of $^{100}$Mo enrichment. To emulate this centrifugal enrichment of $^{100}$Mo, the $^{92,94–98}$Mo nuclides were depleted in ascending order of mass number in our DCC calculations. DCCs were calculated for nine commercial enriched $^{100}$Mo products listed in table 1.

4. Results and discussion
The overall dependence of $^{92,94–98,100}$Mo DCCs on the $^{100}$Mo enrichment level is shown in figure 9. In its most basic sense, the DCC of a Mo isotope is the factor by which the amount of the Mo isotope concerned is modified by a change in the $^{100}$Mo enrichment level. Because the amount of a Mo isotope is directly proportional to the yields of the involved Mo reactions, a DCC can be deemed as a scale factor for reaction yields. In other words, a DCC greater than 1.0 means that the yields of the involved reactions will increase, and a DCC less than 1.0 means that the yields of the involved reactions will decrease.

DCCs calculated for the nine commercial enriched $^{100}$Mo products are presented in table 2, some of which are also plotted in figure 10 for showing their relative magnitudes. As expected from (1), the $^{100}$Mo DCCs were greater in MoO$_3$ than in Momet, meaning that the $^{100}$Mo increment ratio is higher in MoO$_3$ than in Momet. As shown in (7), however, it should be noted that the absolute $^{100}$Mo mass density is always greater in Momet than in MoO$_3$ by a factor of about 3. On the other hand, the $^{92,94–98}$Mo DCCs were almost the same in the two target materials.

The importance of $^{92,94–98,100}$Mo DCCs differs by the $^{99}$Mo/$^{99m}$Tc production methods. In both the $^{100}$Mo($\gamma,n$)$^{99}$Mo and $^{100}$Mo(n,2n)$^{99}$Mo reaction routes, the major impurity radionuclides are niobium (Nb) radioisotopes produced from $^{92,94–98}$Mo [5, 6, 9, 11]. Because Nb isotopes can be separated from $^{99}$Mo and $^{99m}$Tc by chemical means [6, 35], the practical importance of $^{100}$Mo enrichment is its influence on the yield and specific yield of $^{99}$Mo, putting emphasis on the $^{100}$Mo DCC. For instance, if the $^{99}$Mo yield of $^{100}$Mo($\gamma,n$)$^{99}$Mo obtained from a nonenriched Mo$_{\text{met}}$ target was $Y_{Mo-99}(0.0974) = 100.00$ GBq,
using 99.01\% enriched 100\textsuperscript{Mo} will provide a 99\textsuperscript{Mo} yield of
\[ Y_{\text{Mo-99}}(0.990\ 1) = D_{\text{Mo-100}}(0.990\ 1) Y_{\text{Mo-99}}(0.097\ 4) = 9.765\ \times\ 100.00\ \text{GBq} = 976.58\ \text{GBq}. \]

Similarly, if the 99\textsuperscript{Mo} specific yield of 100\textsuperscript{Mo(n,2n)}99\textsuperscript{Mo} obtained from a nonenriched MoO\textsubscript{3} target was
\[ S_{\text{Mo-99}}(0.097\ 4) = 10.000\ \text{GBq Mo-g}^{-1}, \]
using 99.54\% enriched 100\textsuperscript{Mo} will result in
\[ S_{\text{Mo-99}}(0.995\ 4) = D_{\text{Mo-100}}(0.995\ 4) S_{\text{Mo-99}}(0.097\ 4) = 9.946\ \times\ 10.000\ \text{GBq Mo-g}^{-1} = 99.468\ \text{GBq Mo-g}^{-1}. \]
Table 2. DCCs of $^{92,94-98,100}\text{Mo}$ calculated for the commercial enriched $^{100}\text{Mo}$ products listed in Table 1.

| Vendor | $\%_{\text{Mo}=100}$ | $^{92}\text{Mo}$ | $^{94}\text{Mo}$ | $^{95}\text{Mo}$ | $^{96}\text{Mo}$ | $^{97}\text{Mo}$ | $^{98}\text{Mo}$ | $^{99}\text{Mo}$ | $^{100}\text{Mo}$ |
|--------|------------------------|------------------|------------------|------------------|------------------|------------------|-----------------|-----------------|------------------|
| Trace  | 97.39                  | 0.000 3          | 0.000 3          | 0.000 3          | 0.001 0          | 0.102 1          | 9.667 9         |
|        | 97.42                  | 0.034 8          | 0.018 8          | 0.017 6          | 0.021 1          | 0.038 0          | 9.616 2         |
| ISOFLEX| 99.01                  | 0.005 9          | 0.006 3          | 0.006 3          | 0.008 0          | 0.021 8          | 9.765 8         |
|        | 99.03                  | 0.005 2          | 0.007 3          | 0.005 4          | 0.008 0          | 0.021 4          | 9.767 7         |
|        | 99.05                  | 0.005 2          | 0.005 2          | 0.006 1          | 0.007 0          | 0.021 4          | 9.769 5         |
|        | 99.27                  | 0.003 9          | 0.003 1          | 0.002 4          | 0.008 0          | 0.018 6          | 9.790 3         |
|        | 99.54                  | 0.000 4          | 0.000 5          | 0.000 5          | 0.000 1          | 0.000 2          | 9.895 7         |
|        | 99.815                 | 0.000 2          | 0.000 3          | 0.000 2          | 0.000 3          | 0.000 5          | 9.946 8         |
|        | 99.86                  | 0.000 3          | 0.000 5          | 0.000 3          | 0.000 3          | 0.000 5          | 9.978 4         |

Figure 10. DCCs of $^{92,94-98,100}\text{Mo}$ in (a) Momet and (b) MoO$_3$. The percentages indicate $^{100}\text{Mo}$ enrichment levels. Note that the y-axes are not uniformly scaled in order to accommodate $^{98,100}\text{Mo}$ exhibiting relatively high DCCs.
Table 3. Product radionuclides (PRNs) associated with proton reactions on $^{92,94–98,100}$Mo. Listed are PRNs whose half-lives are longer than 10 min and shorter than one year, and reactions whose TENDL-2017 [36] peak cross sections below 25 MeV$^2$ are greater than 0.1 mb. The decay data were retrieved from NuDat 2.7 [37]. Detailed studies of proton reactions on Mo isotopes can be found in [14,17–19].

| PRN | Principal decay mode | Proton reaction | $^{92}$Mo | $^{94}$Mo | $^{95}$Mo | $^{96}$Mo | $^{97}$Mo | $^{98}$Mo | $^{100}$Mo |
|-----|----------------------|-----------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| 98Zr | 83.4 d | $^{98}$Y | (p,γp) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99Nb | 2.0 h | $^{99}$Zr | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99mNb | 1.1 h | $^{99}$Zr | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99Nb | 14.6 h | $^{99}$Zr | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99mNb | 60.9 d | $^{99}$Zr | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99Nb | 10.2 d | $^{99}$Zr | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99Nb | 35.0 d | $^{99}$Mo | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99Nb | 3.6 d | $^{99}$Nb | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 96Nb | 23.4 h | $^{96}$Mo | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 97Nb | 72.1 min | $^{97}$Mo | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 91Mo | 15.5 min | $^{91}$Mo | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 93Mo | 6.9 h | $^{93}$Mo | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 99Mo | 66.0 h | $^{99}$Mo | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) | (p,α) |
| 93mTc | 2.8 h | $^{93}$Mo | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 93mTc | 43.5 min | $^{93}$Tc | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 94Tc | 4.9 h | $^{94}$Mo | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 94Tc | 52.0 min | $^{94}$Mo | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 95Tc | 20.0 h | $^{95}$Mo | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 95Tc | 61.0 d | $^{95}$Mo | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 96Tc | 4.3 d | $^{96}$Mo | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 96Tc | 51.5 min | $^{96}$Tc | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 97mTc | 91.0 d | $^{97}$Tc | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 99Tc | 6.0 h | $^{99}$Tc | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |
| 100Tc | 14.0 min | $^{101}$Ru | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) | (p,γ) |

Notes.

a Above which energy the amounts of Tc impurities rapidly increase [27].

b The excitation function can be found in [14].

c The excitation function can be found in [19].

As shown above, the $^{100}$Mo DCC can quantitatively estimate the yield and specific yield of $^{95}$Mo for enriched $^{100}$Mo, which in turn can be used for selecting the $^{100}$Mo enrichment level in the $^{100}$Mo($γ,n$)$^{99}$Mo and $^{100}$Mo($n,2n$)$^{99}$Mo reaction routes.

On the contrary, the $^{92,94–98}$Mo DCCs as well as the $^{100}$Mo DCC play important roles in the $^{100}$Mo($p,2n$)$^{99}$Tc reaction route, because the $^{92,94–98}$Mo DCCs are directly proportional to the amounts of Tc isotopes other than $^{99m}$Tc (table 3). These Tc impurities, which cannot be chemically separated from $^{99m}$Tc, increase radiation dose [15] and result in image quality degradation [20]. Therefore, an enriched $^{100}$Mo product having low $^{92,94–98}$Mo DCCs as well as a high $^{100}$Mo DCC is necessary in the $^{100}$Mo($p,2n$)$^{99m}$Tc reaction route.

In their studies [15] and [20], Hou et al. reported that at proton beam energies below 20 MeV, the contents of $^{94–97}$Mo contribute more than the content of $^{96}$Mo to the dose increase and image quality degradation. Recent studies by [24, 25] have also reported comparable results. In this regard, the $^{94–95}$Mo DCCs calculated in this paper suggest that if proton beams of $<20$ MeV are used, the 97.39% enriched $^{100}$Mo product is preferable to the 97.42%, 99.01%, 99.03%, 99.05%, and 99.27% ones. Quantitatively speaking, the $^{95}$Mo DCC of the 97.39% enriched $^{100}$Mo product in the form of Mo$_{net}$ is smaller than those of the 97.42%, 99.01%, 99.03%, 99.05%, and 99.27% enriched $^{100}$Mo products by factors of 8–58.7, while the $^{100}$Mo DCCs differ only by factors of 1.001–1.019. This means that while the yield of the $^{100}$Mo($p,2n$)$^{99m}$Tc reaction remains almost unchanged, all the yields of the proton reactions on $^{95}$Mo, for example $^{95}$Mo($p,2n$)$^{94}$Tc and $^{95}$Mo($p,n$)$^{95}$Tc, can be reduced by factors of 8–58.7. The same is true for the yields of the proton reactions on $^{94,96,97}$Mo. The low $^{94–97}$Mo DCCs of 97.39% enriched $^{100}$Mo are also highlighted in figure 10.

The $^{98}$Mo DCC should also be considered if the proton beam energies are greater than 20 MeV, which is the threshold for the $^{98}$Mo($p,3n$)$^{96}$Tc and $^{98}$Mo($p,3n$)$^{96}$Tc reaction routes [14, 36]. $^{96m}$Tc deexcites to $^{96}$Tc with 98% probability [37], and $^{96}$Tc is reported to be one of the major impurities affecting the radiation dose and image quality [15, 20, 24, 25]. Therefore, the 99.815% and 99.86% enriched $^{100}$Mo products, whose $^{98}$Mo DCCs as well...
as $^{92-97}$Mo DCCs are significantly lower than the other enriched $^{100}$Mo products, are preferable for proton beams of $>20$ MeV.

5. Conclusion

A figure of merit called the DCC can quantify the influence of enriched $^{100}$Mo on Mo reaction yields. DCCs can be calculated for various $^{100}$Mo enrichment levels using the dedicated program enrimo.

The main advantage of using DCCs is that the changes in Mo reaction yields resulting from a change in the $^{100}$Mo enrichment level can be easily estimated. For example, the $^{100}$Mo DCC of 99.01% enriched $^{100}$Mo in the form of $^{97}$Mo is 9.765 $\times$ 8, meaning that 9.765 $\times$ 8 times greater $^{99}$Mo reaction yields can be obtained. Likewise, the $^{92-97}$Mo DCCs of 97.39% enriched $^{100}$Mo in the form of $^{97}$Mo, or 0.000 3–0.001, suggest that the $^{92-97}$Mo reaction yields can be reduced by factors of 1000–3333.

In determining the required $^{100}$Mo enrichment level, the $^{100}$Mo DCC alone can be useful in the $^{100}$Mo($^{\gamma}$,$n$)$^{99}$Mo and $^{100}$Mo($p$,$2n$)$^{99}$Mo reaction routes, where the yield and specific yield of $^{99}$Mo are the primary concerns. In contrast, the $^{92-94}$Mo DCCs as well as the $^{100}$Mo DCC need to be considered in the $^{100}$Mo($p$,$2n$)$^{99}$Mc reaction route, where the production of chemically inseparable Tc impurities must be minimized.

Complete DCC data are available as supplementary materials online at stacks.iop.org/JPCS/3/055015/mmedia. The source code of the DCC calculation program enrimo is available in an open-source repository [32].

ORCID iDs

Jaewoong Jang @ https://orcid.org/0000-0002-8466-0502

References

[1] Ponsard B 2010 Mo-99 supply issues: status report and lessons learned Proceedings of the 14th International Topical Meeting on Research Reactor Fuel Management, RRFM 2010 (Marrakesh, Morocco) (Brussels: European Nuclear Society)

[2] International Atomic Energy Agency 2013 Non-HEU Production Technologies for Molybdenum-99 and Technetium-99m (IAEA Nuclear Energy Series No. NF-T-5.4) (Vienna: International Atomic Energy Agency)

[3] National Academies of Sciences, Engineering, and Medicine 2016 Molybdenum-99 for Medical Imaging (Washington, DC: National Academies Press) (https://doi.org/10.17226/23563)

[4] Galea R, Wells R G, Ross C K, Lockwood J, Moore K, Harvey J T and Isensee G H 2013 Phys. Med. Biol. 58 2737–50

[5] Mang era K, Ogimoto K, Ziriba R, Fitzpatrick J, Brown J, Pellerin E, Barnard J, Saunders C and de Jong M 2015 J. Radioanal. Nucl. Chem. 305 79–85

[6] Sekimoto S, Tatenuma K, Suzuki Y, Tsuruguchi A, Tanaka A, Tadokoro T, Kani Y, Morikawa Y, Yamamoto A and Ohtsuki T 2017 J. Radioanal. Nucl. Chem. 311 1361–6

[7] Martin T M, Harahsheh T, Munoz B, Hamoui Z, Clanton R, Douglas J, Brown P and Akabani G 2017 J. Radioanal. Nucl. Chem. 314 1051–62

[8] Jang J, Yamamoto M and Uesaka M 2017 Phys. Rev. Accel. Beams 20 104701

[9] Takeda T et al 2018 J. Radioanal. Nucl. Chem. 318 811–21

[10] Nagai Y et al 2013 J. Phys. Soc. Jpn. 82 064201

[11] Tsukada K, Nagai Y, Hashimoto K, Kawabata M, Minato F, Saeki H, Motoishi S and Itoh M 2018 J. Phys. Soc. Jpn. 87 043201

[12] Leung K-N, Leung J K and Melville G 2018 Appl. Radiat. Isot. 137 23–7

[13] Capogni M et al 2018 Molecules 23 1872

[14] Celler A, Hou X, Bénard F and Ruth T 2011 Phys. Med. Biol. 56 5469–84

[15] Hou X, Celler A, Grimes I, Bénard F and Ruth T 2012 Phys. Med. Biol. 57 1499–515

[16] Gagnon K, Wilson J S, Holt C M B, Abrams D N, McEwan A J B, Mitlin D and McAuliffe S A 2012 Appl. Radiat. Isot. 70 1685–90

[17] Gagnon K, Wilson J S and McAuliffe S A 2012 Nucl. Med. Biol. 39 923–5

[18] Esposito I, Vecchi G, Pupillo G, Taibi A, Ucelli L, Boschi A and Gambaccini M 2013 Sci. Technol. Nucl. Ins. 2013 972381

[19] Quim S M, Sudar S, Scholten B, Koning A J and Coenen H H 2014 Appl. Radiat. Isot. 85 101–13

[20] Hou X, Tanguay J, Vuckovic M, Buckley K, Schaffer P, Bénard F, Ruth T J and Celler A 2016 Phys. Med. Biol. 61 8199–213

[21] Martini P, Boschi A, Cicoria G, Ucelli L, Pasquali M, Duatti A, Pupillo G, Marengo M, Loriggiola M and Esposito J 2016 Appl. Radiat. Isot. 118 302–7

[22] Anderson J D, Thomas B, Selivanova S V, Berthelette E, Wilson J S, McEwan A J B and Gagnon K 2018 Nucl. Med. Biol. 60 63–70

[23] Martini P et al 2018 Appl. Radiat. Isot. 139 325–31

[24] Uzunov N et al 2018 Phys. Med. Biol. 63 185021

[25] Meléndez-Alafort L, Ferro-Florès G, De Nardo L, Bello M, Puisasco M, Negri A, Zorz A, Uzunov N, Esposito J and Rosato A 2019 Med. Phys. 46 1437–46

[26] OECD Nuclear Energy Agency 2018 The Supply of Medical Radioisotopes: 2018 Medical Isotope Demand and Capacity Projection for the 2018–2023 Period (Supply of Medical Radioisotopes series) (Paris: OECD)

[27] International Atomic Energy Agency 2017 Cyclotron Based Production of Technetium-99m (IAEA Radioisotopes and Radiopharmaceuticals Reports 2) (Vienna: International Atomic Energy Agency)

[28] Mayer A J and Wieser M E 2014 J. Anal. At. Spectrom. 29 85–94
[29] Meija J et al 2016 Pure Appl. Chem. 88 293–306
[30] Trace Sciences International Molybdenum isotopes accessed March 27, 2019
[31] ISOFLEX Stable isotopes of molybdenum available from ISOFLEX accessed March 27, 2019
[32] Jang J 2019 enrimo - Investigate the influence of an enriched Mo isotope (v1.05) Zenodo (https://doi.org/10.5281/zenodo.2628760)
[33] Trace Sciences International Modes of production accessed March 2, 2019
[34] ISOFLEX Stable isotopes produced in gas centrifuges accessed March 2, 2019
[35] Ueno K, Sasaki M and Ishimori T 1969 J. Nucl. Sci. Technol. 6 203–6
[36] Koning A J and Rochman D 2012 Nucl. Data Sheets 113 2841–934
[37] National Nuclear Data Center 2019 NuDat 2.7 accessed April 25, 2019 (http://www.nndc.bnl.gov/nudat2/)