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Production of ^{99}Mo Using High-Current Alpha Beams

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ABSTRACT

It has been shown that alpha particle accelerators can be used to produce high specific activity ^{99}Mo effectively by bombarding a ^{96}Zr target using the reaction ^{96}Zr(α,n)^{99}Mo. This process generates minimal waste and does not involve the use of uranium. Further, ^{99}Mo produced by this process can be used in existing generator production. In order for this process to be practical and cost effective, it is necessary to use high-current alpha beams. We have developed, demonstrated and patented the source technology, and are working on development of the target and accelerator technology necessary to provide 100 mA_e alpha beam currents. A 100 mA_e system could produce more than 13,000 6-day Ci/year, at a specific activity of 100 kCi/g, based on ^{99}Mo production yields and specific activity of previous experiments using a low beam current.

1. Introduction

Radioisotopes are used for a large and increasing number of important medical, industrial and other applications. At the present time these radioisotopes are produced by nuclear reactors, cyclotrons, accelerators, and other special purpose devices. All of the current production techniques have specific limitations which in some cases render the needed isotopes difficult or very costly to produce, or utilize facilities that raise environmental, proliferation, or other concerns. Responding to the need for a way to produce a range of critical radioisotopes in a much more cost-effective manner, we have developed a technology for the production of a wide spectrum of radioisotopes, including ^{99}Mo, using high-intensity alpha beams.

This paper focuses on production of ^{99}Mo as the generator precursor of ^{99m}Tc, and is intended to introduce the topic. Final product specifications can be adjusted to optimize production with a wide set of specific requirements. The technology can be applied to many other isotopes.

We have completed Phase I of a plan for development of practical high-current alpha particle accelerators. Phase I included production and testing of a full-scale prototype $^{4}He^{++}$ source that supplies a continuous or pulsed beam at 85% $^{4}He^{++}$ (by current) and 32 mA_e total current (with a 6 mm aperture). It is anticipated that the prototype will be capable of supplying continuous or pulsed currents at up to 96% $^{4}He^{++}$ and 50 mA_e, with future sources capable of supplying up to 120 mA_e.

The Phase I source was operated for over 23,000 continuous hours without a single failure. The source is capable of producing high current proton, deuterium, tritium, helion, alpha, and other ion breams. For an alpha beam the normalized emittance is measured at 0.1 (-0.05 +0.15) π·mm·mrad, which while not exceptionally low, this emittance is more than adequate to provide efficient coupling to a linear accelerator.

2. General Discussion

It is possible to efficiently produce ^{99}Mo by using high intensity alpha beams. In this process a ^{96}Zr target is bombarded by energetic alpha particles to generate ^{99}Mo using the reaction $^{96}Zr(\alpha,n)^{99}Mo$. This process generates high specific activity ^{99}Mo with minimal contaminants, without the use of any uranium.

When an approximately 20 MeV alpha beam strikes a pure ^{96}Zr target, a number of possible reactions can occur. For the moment we will focus on a pure ^{96}Zr target and will include the effects of target impurities later. The cross section for the desired reaction, $^{96}Zr(\alpha,n)^{99}Mo$ reaches a peak of 180 mb at 15 MeV [1] (Figure 1). The dominate reactions are shown in table 1.

Number	Reaction	Decay Chain
1	$^{96}Zr(\alpha,\gamma)^{100}Mo$	(stable)
2	**$^{96}Zr(\alpha,n)^{99}Mo$**	**(2.75 day) $\rightarrow$ ^{99m}Tc (6.01 hour) $\rightarrow$ ^{99}Tc**
3	$^{96}Zr(\alpha,2n)^{98}Mo$	(stable)
4	$^{96}Zr(\alpha,p)^{99}Nb$	(2.6 min) $\rightarrow$ ^{99}Mo (2.75 day) $\rightarrow$ ^{99m}Tc
5	$^{96}Zr(\alpha,pn)^{98}Nb$	(51 min) $\rightarrow$ ^{98}Mo
6	$^{96}Zr(\alpha,2p)^{98}Zr$	(30.7 sec) $\rightarrow$ ^{98}Nb (51 min) $\rightarrow$ ^{98}Mo

Table 1: Dominant reaction for 16-20 MeV alpha impacts on ^{96}Zr. While there are many more possible reactions that can occur given the projected range of alpha particle energy, these are the dominant possible reactions. By limiting the beam energy to 20 MeV (or less), reactions involving more than two simple recoil particles are not observed because there is insufficient incident energy to drive them.

While not all the cross sections have been experimentally determined, computer models exist for all of them (Figure 1) and can be used for a preliminary analysis of the product quality. Examining the reactions we can see that the only undesirable radioisotopes produced (^{99}Nb, ^{98}Nb, and ^{98}Zr) have short half-lives which decay to isotopes of molybdenum (^{99}Mo and ^{98}Mo). Moreover, no significant radioactive waste material is produced during an irradiation.

The dominant undesirable products are other stable isotopes of molybdenum (^{98}Mo and ^{100}Mo), which have the effect of lowering the specific activity of the product material.

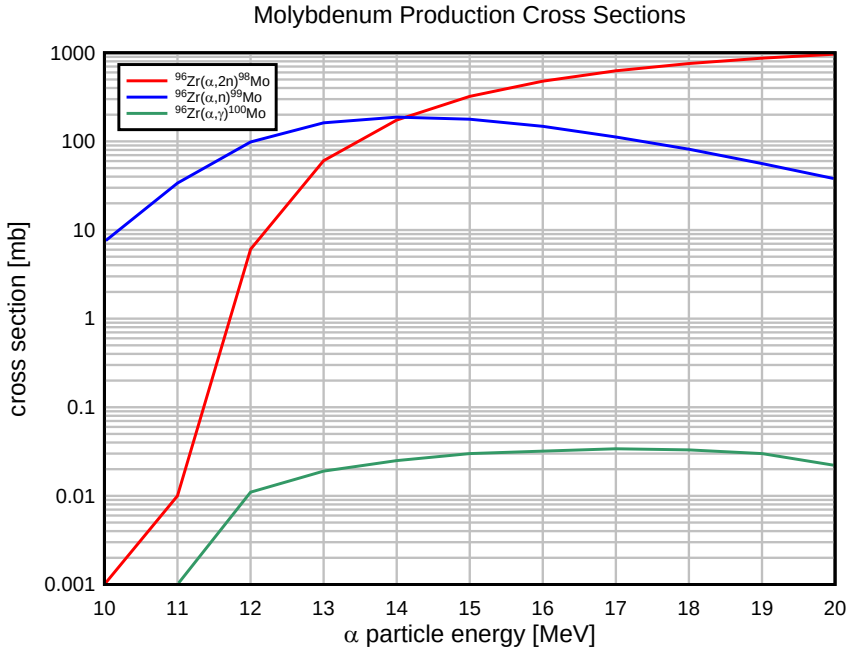

Figure 1: Logarithmic plot of the cross sections for production of molybdenum isotopes by bombardment of ^{96}Zr by alpha particles in the range of 10-20 MeV [1]. Cross sections calculated using Empire II [7].

In figure 1 the ^{96}Zr(α,n)^{99}Mo, shown in blue, is the main reaction which results in the desired product (the secondary reaction ^{96}Zr(α,p)^{99}Nb also produces ^{99}Mo though a rapid β^- decay, but has a smaller cross section). This reaction has been both simulated and experimentally measured. The cross section reaches a peak of roughly 180 mb at 15 MeV[1]. On Table 1, reactions 1 and 3 lead to direct production of stable molybdenum isotopes. Reaction 1 has a very small cross section (~0.035 mb) and leads only to trace amounts of stable ^{100}Mo. Reactions 4, 5, and 6 lead to radioisotopes of niobium or zirconium that decay relatively quickly to stable isotopes of molybdenum. The major contaminate is produced by reaction 3 (^{96}Zr(α,2n)^{98}Mo) and leads to production of stable ^{98}Mo. This reaction (over the energy range of interest) has a peak cross section of 965 mb at 20 MeV, and reduces the specific activity of the product.

3. Product Specificity and Yields

The specific activity of the ^{99}Mo produced is an important consideration. For example, ^{99}Mo can be made relatively efficiently using a proton beam and a ^{100}Mo target using the reaction ^{100}Mo(p,pn)^{99}Mo. Unfortunately, the product ^{99}Mo cannot be easily separated from the target ^{100}Mo so the specific activity of the produced ^{99}Mo does not meet current medical application requirements. Pure ^{99}Mo has a specific activity of approximately 480 kCi/g. Tests on alpha beam produced ^{99}Mo have indicated that at sufficiently low beam energy specific activities as high as 475 kCi/g have been produced. (Specific activity of 475 kCi/g was measured for incident particle energy of 11 MeV, and 220 kCi/g was measured for an incident energy of 15 MeV [8].) There is a trade-off between overall production of ^{99}Mo and specific activity.

In particular, as the beam energy is increased more ^{99}Mo is produced and the specific activity decreases. The latter is due to a corresponding increase in ^{98}Mo production due to an increase in the ^{96}Zr(α,2n)^{98}Mo reaction (Figures 1 and 2). The expected operating range for the system is approximately 20 MeV to generate ^{99}Mo with a specific activity of 100 kCi/g. Production estimates in this paper are based on an alpha energy of 20 MeV and a specific activity of 100 kCi/g.

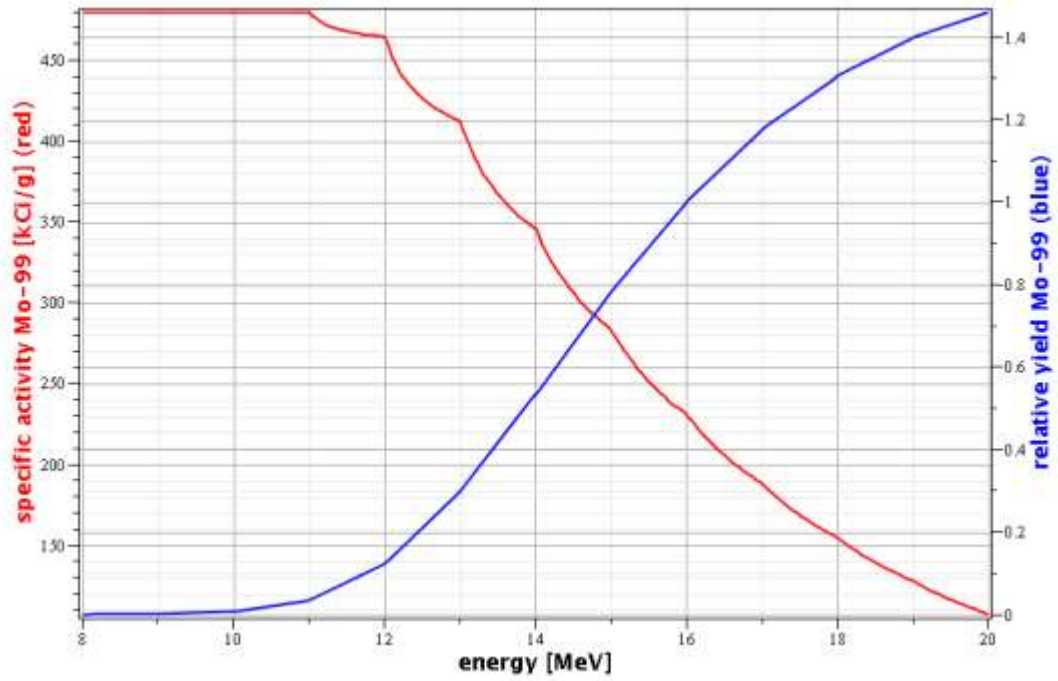

Figure 2: Tradeoff between yield (blue) and specific activity (red) is shown. As the beam energy is increased the yield increases but the specific activity is reduced as more ^{98}Mo is also produced in the target. Yield is plotted relative to actual test measurements made for a 16 MeV alpha beam.

Production of ^{99}Mo is directly proportional to the beam current. We are proposing the use of 100 mA_e alpha particle accelerators for the production of ^{99}Mo. The yield from a single machine can be estimated based on the cross section data (**Error! Reference source not found.**). The true yield from the device also depends, however, on the processing cycle for the targets. For example, if the targets are processed daily, a single accelerator produces 54.2 6-day curie/day or 380 6-day curie/week. If, however, the targets are processed only once a week, then a single accelerator produces 202 6-day curie/week, with other weekly processing cycles leading to values between these two limits (Figure 3).

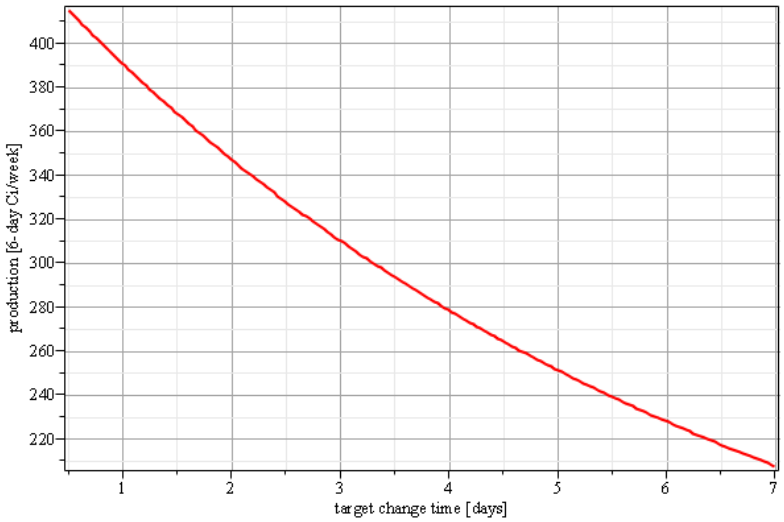

Figure 3: Estimated ^{99}Mo production over a week for a 100 mA_e accelerator system as a function how often the targets are changed.

4. Impact of Target Material

In reality, the target material is not pure, but rather contains some mixture of zirconium isotopes and possibly other contaminant. Enriched ^{96}Zr is readily available on the stable isotope market at greater than 99.99% ^{96}Zr, but it is helpful to consider the use of less pure zirconium in order to understand the effects of impurities.

Zirconium has five stable isotopes (90, 91, 92, 94, and 96), with ^{96}Zr (2.80 % of natural zirconium) being the heaviest. Table 2 summarizes all the possibly significant reactions between the alpha beam and a zirconium target at a 20 MeV impact energy. Reactions 1-6 have been discussed in detail above, with reactions 2 and 4 (bold) leading to production of ^{99}Mo. For this discussion we are primarily interested in reactions that lead to waste disposal issue and/or reactions that lead to potential contamination of the product material with other radioisotopes of molybdenum.

Examining Table 2, reactions that lead to other radioisotopes of molybdenum are indicated by a dark grey background. Reactions 21 and 26, lead to production of the long lived ^{93}Mo. These reactions can be completely suppressed by ensuring that the target does not contain ^{90}Zr or ^{91}Zr. Reactions that lead to potential waste disposal issues are indicated by a light grey background. Reactions 16, 17, 22, 24, and 29 lead to long-lived radioisotopes of niobium or zirconium. These reactions can be suppressed by removing ^{90}Zr, ^{91}Zr, and ^{92}Zr from the target material.

While high purity ^{96}Zr (> 99.99%) is available, it is possible to use lower and less expensive target material. For example 97% enriched ^{96}Zr is less expensive and contains 3% ^{94}Zr with trace about of ^{92}Zr (less than 0.01%) and essentially no ^{91}Zr or ^{90}Zr. The overall tradeoff is a small reduction in yield (~3%), a small decrease in specific activity (still greater than 100 kCi/g) and no substantial production waste or contamination.

Number	Target	Reaction	Product	Decay	Product	Decay	Product
1	^{96}Zr	α,γ	^{100}Mo				
2		**α,n**	**^{99}Mo**	**2.75 d**			
3		α,2n	^{98}Mo				
4		**α,p**	**^{99}Nb**	**15 s /2.6 m**	**^{99}Mo**		
5		α,pn	^{98}Nb	2.9 s / 51 m	^{98}Mo		
6		α,2p	^{98}Zr	30.7 s	^{98}Nb	2.9 s / 51 m	^{98}Mo
7	^{94}Zr	α,γ	^{98}Mo				
8		α,n	^{97}Mo				
9		α,2n	^{96}Mo				
10		α,p	^{97}Nb	1.23 h / 53 s	^{97}Mo		
11		α,pn	^{96}Nb	23.4 h	^{96}Mo		
12		α,2p	^{96}Zr				
13	^{92}Zr	α,γ	^{96}Mo				
14		α,n	^{95}Mo				
15		α,2n	^{94}Mo				
16		α,p	^{95}Nb	35 d / 2.61 d	^{95}Mo		
17		α,pn	^{94}Nb	20k y / 6 m	^{94}Mo		
18		α,2p	^{94}Zr				
19	^{91}Zr	α,γ	^{95}Mo				
20		α,n	^{94}Mo				
21		**α,2n**	**^{93}Mo**	**3500 y / 6.9 s**	**^{93}Nb**		
22		α,p	^{94}Nb	20k y / 6 m	^{94}Mo		
23		α,pn	^{93}Nb				
24		α,2p	^{93}Zr	1.6M y	^{93}Nb		
25	^{90}Zr	α,γ	^{94}Mo				
26		**α,n**	**^{93}Mo**	**3500 y / 6.9 s**	**^{93}Nb**		
27		α,2n	^{92}Mo				
28		α,p	^{93}Nb				
29		α,pn	^{92}Nb	700 y / 62 d	^{91}Zr		
30		α,2p	^{92}Zr				

Table 2: Summary of reactions between a 20 MeV alpha beam and a zirconium target. Decay times with two values indicate the ground state and the metastable state. Bold entries on a white background produce ^{99}Mo. Entries with a dark grey background (21 & 23) lead to other radioisotopes of molybdenum. Entries with a light grey background (16, 17, 22, 24 and 29) produce radioisotopes that may complicate waste disposal.

Potential advantages of accelerator production of ^{99}Mo include a simpler post-irradiation chemical separation process and generation of virtually no nuclear waste material. Given the assumption that the target is 100% pure ^{96}Zr, exposure to the alpha beam generates the desired ^{99}Mo along with ^{98}Mo, ^{100}Mo, ^{99}Nb, and ^{98}Nb, plus the remaining ^{96}Zr in the target. The niobium radioisotopes decay quickly to ^{99}Mo and ^{98}Mo. The ^{98}Mo and ^{100}Mo are stable and simply reduce the specific activity of the final product. The ^{96}Zr is stable and is recycled for use as target material. Relatively simple chemistry [2] can be used to separate the

zirconium, niobium, and molybdenum from each other, reducing the chemical processing expense, time and losses. Further, minimal radioactive waste is generated and any potential material (such as niobium removed in chemical processing) decays to stable isotopes within five hours of end of irradiation.

5. Accelerator Issues

Efficient generation of ^{99}Mo by alpha beam irradiation requires an efficient, high-current, accelerator system. Fundamentally, a beam of singly charged helium ($^4He^+$), or doubly charged helium ($^4He^{++}$) could be used to drive such reactions. However, acceleration of alpha particles ($^4He^{++}$) is at least fourfold more efficient due to the higher mass-to-charge ratio leading to a smaller more efficient accelerator.

The main challenge in generating a high current $^4He^{++}$ beam is the source system. Typical ion sources (duoplasmatron, simple electron cyclotron resonance, etc.) can generate high current $^4He^+$ beams, but the $^4He^{++}$ current is typically limited to only a few percent of the total current [3]. Advanced ion sources, such as an ECRIS (electron cyclotron resonance ion source), can generate reasonable fractions of $^4He^{++}$, but are limited to small beam currents (typically much less than 1 mA)[4]. As described above, we have completed development of a full-scale fully functional $^4He^{++}$ Source that is currently supplying a CW beam at 85% $^4He^{++}$ (by current) and 32 mA_e total current (with a 6 mm aperture). The current version of the Source will be capable of supplying CW currents at up to 96% $^4He^{++}$ and in excess of 50 mA_e. A larger version of the Source is will be able to supply in excess of 120 mA_e.

Very high current linear accelerators (above ~20 mA_e) require the use of advanced design techniques in order to dissipate the heat generated in the accelerator. A 100 mA_e alpha beam at 20 MeV carries 1 MW in the beam. If the accelerator were only 10% efficient, then 9 MW of power would need to be disputed in the accelerator. Practical design of a 100 mA_e system can be implemented using advanced hybrid cooling (Alpha Source intellectual property), advanced accelerator structures, and/or with superconducting technology. In recent years all of these fields have made significant advances making high current industrial linear accelerators practical and cost effective.

The basic accelerator design will consist of a magnetic LEBT (low energy beam transport) that couples the ion source to the accelerator. A room temperature RFQ (radio frequency quadrupole) will accelerate the particles to about 8 MeV. The final acceleration from 8 MeV to 20 MeV can be accomplished use super-conducing cavities, or using H-mode structures with permanent magnet quadrupole focusing [5].

It is also necessary to have a target system that can dissipate the heat generated by the beam. This could be done by using multiple targets and/or spreading the beam over a larger area [6]. A proprietary high-current target for ^{99}Mo production is currently under development.

6. Conclusions

Efficient production of ^{99}Mo using a high intensity alpha beam is not only practical, it can provide a reliable, distributed source of ^{99}Mo without significant radioactive waste and without using uranium. Based on current use of ^{99}Mo, roughly 18 100 mA_e machines could

produce the entire United States demand for ^{99}Mo. In addition, deployment of seven 100 mA_e machines would fill the supply gap created when Chalk River Nuclear Plant is decommissioned in October 2016. Such a system would provide a robust, redundant production system that could be implemented over time, and would provide a reliable, cost-effective, domestic supply of ^{99}Mo.

7. References

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