THE POTENTIAL PRODUCTION OF MOLYBDENUM 99 IN CANDU REACTORS

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Abstract

Technicium-99m is an important isotope utilized worldwide in nuclear medicine. Its parent isotope, Molybdenum-99, is a fission product of U-235 and is produced commercially in a few reactors around the world. The CANDU®* reactor with its ability to fuel online and enhanced neutron economy has the potential to produce large quantities of Mo-99. This paper explores production strategies in the low power periphery channels in a generic CANDU reactor. A small group of channels can yield 100-115% of the world demand for Mo-99 with minor impacts on standard fuelling operations. While the infrastructure for handling and processing these quantities of material does not currently exist, this paper demonstrates the feasibility of producing large quantities of Mo-99 in a standard CANDU reactor.

1. Introduction

The production of medical isotopes is essential for imaging, diagnosis and treatment of many diseases. In particular Technicium-99m (Tc-99m) is widely used in nuclear medicine and is produced directly from the radioactive decay of Molybdenum-99 (Mo-99). Isotopes produced in nuclear reactor facilities are vital to the global medical community as was seen in 2007 and 2009 when production of Mo-99 was reduced due to reactor maintenance shutdowns. Mo-99 is a product of uranium fission and is produced in most research and power reactors as a by product of their operation. Several research reactors are fitted with dedicated isotope production targets using either high or low enriched uranium (HEU or LEU) in order to facilitate Mo-99 production. The production of Mo-99 in the irradiation targets is directly proportional to the fission rate to which the target is exposed. The fission yield of Mo-99 is approximately 6% for U-235 fission and therefore the Mo-99 yield is 6% of the fission rate [1]. The Mo-99 targets are then chemically processed to extract the Mo-99 fission product which is utilized in the generation of Tc-99m, a primary isotope used in medical imaging and diagnosis. Tc-99m has a rather short half life of 6.01h which limits its ability for transport in a pure state. However, since it is a direct decay product of Mo-99 the parent-daughter radioactive decay system has the half life of the parent isotope, Mo-99, which is 66h. This time frame makes the use of the Tc-99m generated from a Mo-99 shipment more practical providing the necessary processing and transportation time needed for the effective use of the end product. The Tc-99m can then be extracted from the decaying Mo-99 parent sample several times over the course of about a week. However, this decay rate is still relatively short and adds complexity to the production cycle since a) the isotope cannot be stockpiled or stored for prolonged periods, and b) the processing and transportation time from the production of Mo-99 by irradiation to the eventual patient application of Tc-99m means that a significant amount of Tc-99m is inevitably lost. Both of these issues demand a solution which allows for flexible and continuous production of large quantities of Mo-99 in order to ensure that treatment plans for the public are not affected.

* CANDU is a trademark of Atomic Energy of Canada.

The demand for Mo-99 for use in Tc-99m generation in North America is about 6000 6 day Curies per week, with 1/12 in Canada and the remainder in the United States. The unit of activity used by Mo-99 producers is the Curie (1 Curie = 3.7 x 10^{10} disintegrations per second or Becquerels, Bq). The producers usually calibrate a shipment value to the activity of the Mo-99 isotopes 6 days after it leaves the production facility. This quantity is the standard unit used in Mo-99 production and is referred to as 6 day Curies (6d Ci) [2]. The worldwide demand is 12000 6d Ci/week. This level of demand is equivalent to 54,000 Ci of Mo-99 at the completion of extraction processing. Including the processing time (1 day) and an extraction efficiency of 90%, the worldwide demand of 12000 6d Ci is equivalent to an end of bombardment (EOB) yield of about 77000 Curies of Mo-99 (referred to as EOB Ci) [2]. This corresponds to 160mg of Mo-99 at this end of bombardment stage [2]. The current supply of Mo-99 isotopes comes primarily from the irradiation of dedicated U-235 targets in a small number of research reactors around the globe. However, since Mo-99 is produced in any uranium fission reactor there is a possibility to expand the production sources to include power generation facilities.

In most power reactors the dedicated production and extraction of Mo-99 is prohibitive due to batch refuelling. However, CANDU reactors have online fuelling capabilities and a flexible and compact fuel design that can easily be adapted to generate Mo-99 for medical use. By optimising a few channels (3-6) in a CANDU reactor for the production of Mo-99 it is possible to generate a significant portion of the world demand even accounting for processing and delivery losses. Limited scoping studies of production of Mo-99 in a CANDU reactor have been explored previously [3]. This paper further explores the possibility of generating Mo-99 for nuclear medicine use in a generic CANDU power reactor and the implications on operability and fuel management.

2. Mo-99 Fuel Bundle Design and Fuelling Scheme

The standard fuel used in a CANDU reactor is natural uranium (fresh fuel U-235 content ~0.7%) in a 37 element bundle. This fuel configuration does generate Mo-99 but may not suitable for use in a production scheme as the ratio of Mo-99 yield to fuel volume processed is quite low especially for periphery channels of the core.

This paper explores new fuel designs for the production of Mo-99 in existing CANDU power stations using the low power channels on the periphery of the core. The bundle designs are tested using the WIMS-AECL lattice code, using both a single-cell infinite lattice and a three by three multi-cell model. In order to apply such designs to the existing fleet, these new bundles must be subjected to the same power limits as existing fuel bundles. For each design explored in this paper, the pin powers and production yield of Mo-99 are recorded.

The designs considered utilize the standard 37 element bundle with either a) 2-5% enriched LEU in the 6 elements in the second ring and the 12 elements in the third ring or b) 1-2%SEU in the 18 elements in the fourth ring. These are consistent with the standard enrichment range used by power reactors and hence would be easily available. In both cases depleted uranium (DU) with a U-235 content of 0.25%, consistent with the average level present in enrichment tailings, is used in the remaining elements. Upon the completion of the irradiation cycle, the target pins are demounted and sent for processing. Even though demountable bundle are not in common use in CANDU power reactors, they have been utilized for decades in the NRU reactor for irradiation testing and fuel development. Also demountable "carrier" bundles were examined as part of advanced fuel studies to breed thorium in CANDU reactors [4]. For the irradiation of either design, two "target" bundles for Mo-99 production are loaded together and carried within one two bundle magazine within the fuelling machine.

The average burnup time required for peak Mo-99 production is ~15 days and by staggering the production in multiple channels a steady amount of Mo-99 production bundles can be removed each day to provide a constant yield. Upon extraction, additional fuel handling procedures would need to be developed to remove the Mo-99 target pins from the carrier bundle and package them for processing. Depending on the desires of the production facility, the rest of carrier bundle would then either a) be sent away to spent fuel storage or b) be reloaded with new Mo-99 target pins and reused in another Mo-99 production cycle.

A plausible Mo-99 production scheme involves the use of 3-6 fuel channels with groups of 3 channels offset by about a week resulting in yields of about 2-4 Mo-99 bundles per week. This arrangement would involve an increase in the number of fuelling machine visits by 3 per week for each 3-channel group used. Since a desired output of 4 bundles using the 6 channel option is considered later in this paper, a total of 6 additional fuelling visits are necessary each week over and above the normal fuelling requirements. The production burden can be spread over multiple reactors at a multi-unit station which would reduce the fuelling requirements for each single unit. For example, two 3-channel groups each at a separate unit could be utilized to produce the continuous yield of 4 bundles per week with a lower fuelling burden at each unit.

These designs use enriched fuel as Mo-99 targets in multiple elements which increases the fission and hence pin powers of these elements. The low power periphery channels have a high margin to the maximum linear element power ratings and power limits and hence are the best locations to utilize for loading of the Mo-99 production bundles. The production bundle designs will have to adhere to the same linear ratings, bundle power and channel power limits prescribed for a standard CANDU fuel bundle. Advanced bundle designs, such as those explored for low void reactivity fuel (LVRF), which include more elements will reduce linear rating effects but also distribute the Mo-99 production over more pins increasing the number of pins to be processed and reducing the production per pin. Hence for this study the constraints are to utilize the 37-element bundle design and ensure that the powers and peaking factors are within the existing prescribed limits.

2.1 Simulation and Modelling Technique

The production of Mo-99 in a CANDU lattice was investigated using the WIMS-AECL-3.121 lattice code utilizing the ENDF-BVII-ACR cross section library. The simulations involved both an infinite lattice calculation and a multi-cell case with a three by three group of channels with the Mo-99 production channel in the middle simulating a lower power periphery channel on the edge of the core. The lattice cell is that of a standard CANDU fuel channel surrounded by moderator with a 37 element fuel bundle as per Figure 1.

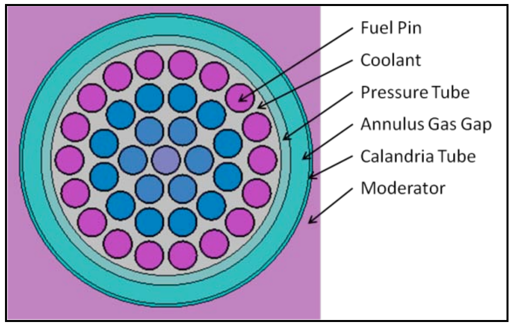

Figure 1: A 37-Element CANDU Lattice cell

The lattice cell fission energy at 100%FP is assumed to correspond to the bundle power of 900kW. Since each fuel pin contains 583.58g of fuel and there are 37 elements in the bundle the corresponding power density is 41.7W/g bundle power. All the pin powers and linear ratings are scaled according to this power density. The 900kW bundle power is the upper limit for a typical CANDU fuel bundle. For the purpose of establishing the bounding linear element rating for a given fuel design, this assessment assumes a fresh target bundle is loaded into the highest thermal flux region in the channel (bundle position 6 or 7) and burned there for the duration of the irradiation period. This is a conservative case since the bundle would initially be placed in a less central position (bundle position 1 - 4) to be burned up before progressing to the center of the channel and hence would have lower pin powers.

The multi-cell model is used to more accurately represent the real implementation of the Mo-99 bundle in the core. The lower power periphery channel design replicates the arrangement centered on channel C5 in a 380 channel CANDU reactor with reflector cells above and to the left, the Mo-99 production bundle in the center and natural uranium (NU) fuelled channels in the remaining locations, it is shown in Figure 2. The boundary conditions of the model are partially reflective with the top and left sides bordered by more water sites while the right and bottom sides are bordered by fuel sites (similar to D4 beside B6 and D4 and similar to D6 elsewhere). This configuration best represents channel C5 and its neighbouring channels and water sites.

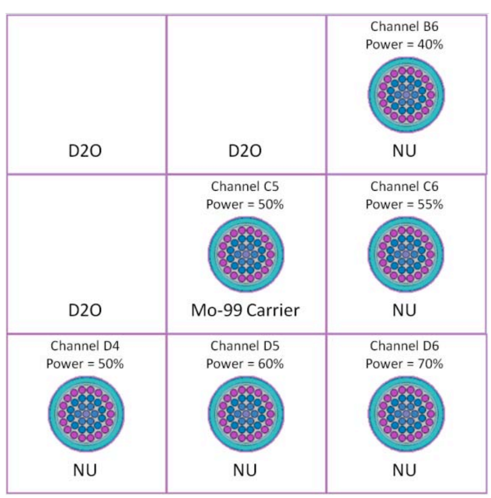

Figure 2: Mo-99 Production Multi-cell Model Representing a Low Power Periphery Channel

The mass of Mo-99 is calculated in WIMS and output for each specific fuel region. The pin power factors for each fuel ring are also output along with the cell k-infinity. These are used to evaluate the viability of the design with respect to existing limits as well as mimicking existing 37-elment bundle behaviour to the greatest extent practicable.

A bundle fuelling scheme is also explored as part of this study and optimized for the peak Mo-99 production level which occurs around 15 days of irradiation. The bundle fuelling scheme is designed considering a typical axial power profile of a CANDU reactor to determine the favourable dwell times for each stage and bundle positions for the Mo-99 production bundles. The concept of using a few channels fuelled in a staggered manner to yield constant output is also explored.

3. Bundle Designs and Results

The Mo-99 production bundle must meet the same safety and design constraints of regular fuel. Specifically the design must maintain a maximum linear rating of 60kW/m. Also the k-infinity value should be close to that of standard NU to allow for existing operational practices to be maintained. This reactivity comparison is made in the multi-cell model where the overall k-infinity is compared to that of a standard fully NU fueled configuration and kept within 10-15mk of this multi-cell reference value. The limit for the infinite cell cases is within ~60-75mk which was backed out from the 10-15mk multi-cell model limit for each design. While the impacts of the Mo-99 target pins on safety analysis were not directly examined in this work, it should be noted that given the very small number of channels involved, the similarity in k-infinity and axial power profile, and the limits on the channel/bundle/pin power imposed; the resultant impact on accident analysis should be small. However, direct confirmation would still be required if these designs were to be implemented in operating reactors.

The design utilizes several target pins which can generate a large amount of Mo-99 per bundle. The channels on the periphery of the core operate at lower powers than those in the central regions allowing for higher levels of fuel enrichment and a larger number of Mo-99 target pins without exceeding the linear element rating limit. The lower power characteristics of the periphery channels reduce the importance of the linear power rating limit and redirect the focus to the k-infinity constraint. The bundle designs and limits dictate that the target pins be located in either the second and third rings or just the fourth ring. Each target pin is fuelled with low or slightly enriched uranium (LEU, <20% U-235; SEU, <2% U-235). In order to deal with the increased reactivity of this fuel over natural uranium the remaining elements of the bundle are fueled with depleted uranium. A demountable bundle design similar to that proposed in Reference 4, would be ideal for operating either of the designs. The Mo-99 target pins would be demountable from the bundle after irradiation is complete and processed onsite to extract the Mo-99. In order to assess the performance of the designs, the typical radial power profile across a standard 37 element NU bundle is examined in Figure 3. The power profile is lowest in the center and higher in the outer rings. The flux profile is also of a similar shape.

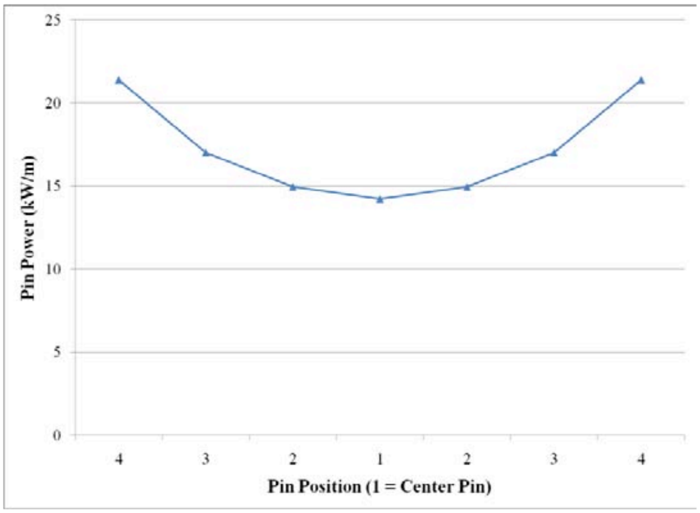

Figure 3: Pin Power Profile of a CANDU 37 element NU Bundle

The first proposed design places enriched target elements in both the second and third rings taking advantage of their lower power levels providing a larger margin to the linear rating limit. The third ring sits mid-range in the flux and power distribution allowing for adequate flux exposure for Mo-99 production but maintaining lower powers than in the outermost ring. The second ring provides more target pins at a lower flux level and hence should not be limiting in terms of the linear element rating. The design utilizes both the second and third ring to provide more production capacity and to distribute the total power of the bundle over the increased number of target elements, 18 pins. Given the bundle flux profile, higher enrichments can be used without bundle reactivity becoming an issue.

The second fuel design places the enriched targets only in the 18 pins in the outer (fourth) ring. This is the highest flux area where the most fission will occur and more Mo-99 will be produced. However, the outer ring's higher flux requires the enrichment level to be reduced compared to the first design to avoid exceeding the cell reactivity and linear rating limits.

Using an infinite cell model of the Mo-99 bundle designs an enrichment that satisfies the linear power rating limit and maintains a suitable k-infinity value close enough to a standard NU CANDU lattice cell is investigated. The goal is to optimize Mo-99 production in the cells while keeping within the power and reactivity limits. The cell power is set at 50%FP (100%FP power density is 41.7W/g, or 900kW/bundle), which is representative of periphery channels in the core. The linear rating limit of 60kW/m and a reactivity constraint of keeping the infinite cell k-infinity within 60-75mk of a standard CANDU lattice cell were utilized in this study. Multiple target pin enrichments were evaluated for both designs in an effort to satisfy the constraints and maximize Mo-99 yield.

For the first fuel design with targets in the second and third ring, enrichments of 2%-3% U-235 were tested. All trials met the linear rating limit but enrichments greater than 2.0% exceeded the infinite cell reactivity requirement on k-infinity. For the 2.0% U-235 enrichment in the target elements and depleted uranium in the remaining elements the infinite lattice had a k-infinity of

1.21774 for fresh fuel (+63.8mk from NU case). The maximum pin powers in the second and third ring were 34.8kW/m and 44.6kW/m respectively. The Mo-99 yield is shown in Figure 4.

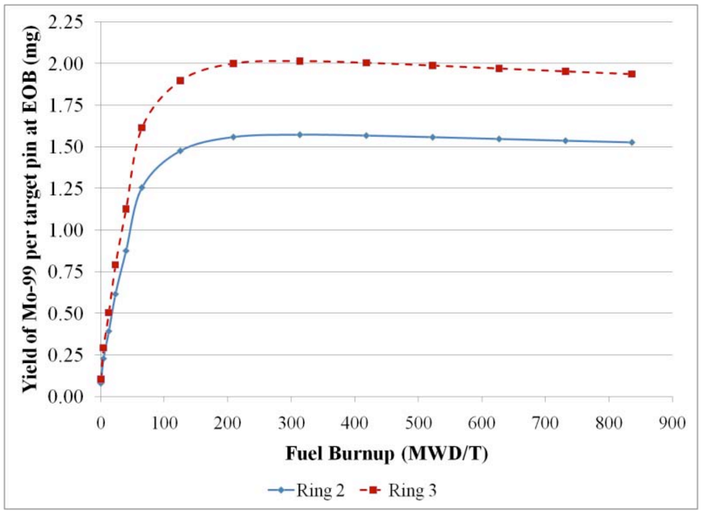

Figure 4: Mass of Mo-99/pin Vs Burnup (2.0% U-235 in R2 and R3)

The peak yield for Mo-99 was reached at 15 days of irradiation corresponding to a cell burnup of 313MWd/T. The Mo-99 production in the targets was 1.6mg/pin in the second ring and 2.0mg/pin in the third ring. The utilization percentages for the targets, given the fuel mass of 583.58g per pin, were $2.69\text{x}10^{-4}$% and $3.45\text{x}10^{-4}$% for the second and third ring pins, respectively. The total production in the bundle was 33.6mg of Mo-99 for an overall utilization of $3.20\text{x}10^{-4}$%. This yield corresponds to 21% of the world weekly demand for Mo-99 for each bundle produced. For a 4 bundle per week output this production strategy would yield 134.4mg of Mo-99 or $6.453\text{x}10^{5}$ EOB Curies. This amounts to about 84% of the world demand for Mo-99, albeit for a more diluted target pin than those currently in use at research reactors.

For the second design with targets in the fourth ring only, the higher power flux is distributed over all 18 of the target pins resulting in more fission raising the k-infinity value for the same enrichment. Therefore in order to maintain the reactivity within the limit the enrichment level must be reduced from the previous case. The linear power limit is the secondary constraint in this configuration. Enrichment was reduced to 1.5% U-235 resulting in a fresh fuel infinite cell k-infinity value of 1.22949 (71.6mk from the NU case). The maximum pin power for the targets in the fourth ring was 43.8kW/m. The Mo-99 yield for this design peaks after 15 days of irradiation (313MWd/T cell burnup) at 2.0mg/pin. The total yield for this design was 35.8mg of Mo-99 per bundle for a utilization $3.41\text{x}10^{-4}$%. This yield corresponds to 22% of the world weekly demand for Mo-99 for each bundle produced. Given the aforementioned 4 bundle per week extraction level this production strategy would yield 143.2mg of Mo-99 or $6.8790\text{x}10^{5}$ EOB Curies. This amounts to about 89% of the world demand for Mo-99.

The design with the targets in the fourth ring produces more Mo-99 for a lower level of enrichment in the target and has slightly larger margin to the linear element rating limit (~1kW/m better than the first design). The design is also mechanically simpler since only the outer ring needs to be demounted for processing.

Upon the end of bombardment the target ring(s) is (are) demounted and sent for processing. The target ring(s) can then be replaced with new LEU Mo-99 target pins, re-using the 19 depleted uranium elements in the bundle for another cycle. However, the re-use is limited to a few cycles as excessive irradiation of the depleted uranium will produce Pu-239 through neutron absorption that will affect the reactivity of the fuel. Alternatively, the production facility may choose send the carrier bundle directly to spent fuel storage to avoid the risks of extra fuel handling and its effects on the probability of fuel failure.

It should be noted at this point that a standard NU bundle does produce Mo-99 during the course of its irradiation in the core. However, as mentioned in Section 2, the yield available from the NU bundle is not high enough to warrant the processing efforts required to extract the Mo-99. In the case of a periphery channel the Mo-99 yield is about 0.8-1.3mg/pin (~0.6-1.2 Ci/g of fuel) depending on the fuel ring, with the largest production at the outer edge. This yield is almost half that of the two proposed designs described earlier. Production of Mo-99 with an NU target bundle may be more feasible in a high power channel where yields would be about 2mg/pin (~2.2Ci/g of fuel) making them on par with the production levels proposed here. However, the study of Mo-99 production in a high power central channel in a CANDU is beyond the scope of this specific paper and needs to be further investigated.

3.1 Multi-cell Periphery Channel Model

The multi-cell periphery channel case was run for the two designs to check the overall k-infinity value and to ensure that the Mo-99 production level and linear rating of the fuel are maintained. The multi-cell model used for the periphery channel, described in Figure 2, puts the Mo-99 production bundle at the center of a 3x3 matrix of channels that represent the arrangement surrounding channel C5 in a generic 380 channel CANDU reactor with empty moderator cells in the upper left and regular NU fuelled channels below and to the right. The channel powers in this model range from 40-70%FP with the Mo-99 production channel at 50%FP (100%FP power density is 41.7W/g or 900kW/bundle).

In order to ensure consistency with a fully NU periphery channel multi-cell lattice, the model was run entirely with NU. The overall K value for the NU periphery channel multi-cell was 1.1280 for fresh fuel. The maximum pin power factor (PPF) in the model was 1.149 corresponding to a pin power of 28.6kW/m at a burnup of 32MWd/T.

This periphery channel multi-cell model more realistically describes the operating environment for a high yield Mo-99 production bundle. The Mo-99 production increases over the infinite lattice case due to the increased moderation available at the periphery of the core. This effect also increases the pin powers. The successful cases for the two designs discussed previously were assessed in this multi-cell lattice.

The first simulation had Mo-99 targets of LEU at 2.0% U-235 in rings two and three and depleted uranium in the remaining elements. The multi-cell k-infinity value for this trial was 1.14467 for fresh fuel which is 12.9mk from the NU value. The maximum pin powers were 42.0kW/m in the second ring and 51.6kW/m in the third ring. These pin powers are 5-6kW/m higher than in the infinite lattice case due to the higher moderation available at the periphery of the core. The multi-cell k-infinity is close to the NU value because the Mo-99 production bundle makes up only 1/6 of the fuel in the multi-cell case. The pin power and reactivity limits have been maintained to a sufficient degree for the multi-cell simulation. The Mo-99 production peaks at 15 days of irradiation (438MWd/T multi-cell burnup) with a yield of 2.1mg/pin and 2.6mg/pin in the second and third rings respectively. This results in utilizations of about $3.67x10^{-4}$% and $4.49x10^{-4}$%. The total production in the bundle was 44.3mg of Mo-99 for an overall utilization of about $4.22x10^{-4}$%. This yield corresponds to 27.7% of the world weekly demand for Mo-99 for each bundle produced. Therefore, a production cycle of 4 bundles per week would yield 177.2mg of Mo-99 or $8.512x10^5$ EOB Curies. This amounts to about 111% of the world demand for Mo-99.

The second simulation utilized Mo-99 targets of SEU at 1.5% U-235 in the fourth (outer) ring and depleted uranium in the remaining elements. The multi-cell fresh fuel k-infinity value for this trial was 1.14423 which is 12.6mk from the NU value. The maximum pin power in the fourth ring was 51.3kW/m. The Mo-99 production peaked again at 15 days of irradiation (438MWd/T multi-cell burnup) with a yield of 2.6mg/pin and a utilization of about $4.48x10^{-4}$%. The total production in the bundle was 47.1mg of Mo-99 which corresponds to 29.4% of the world weekly demand for Mo-99 for each bundle produced. The 4 bundle per week yield would be 188.3mg of Mo-99 or $9.043x10^5$ EOB Curies, about 118% of the world demand for Mo-99.

The second design (targets in ring 4) provides the best Mo-99 production utilization and greatest linear rating margins while using a lower enrichment level that is less costly.

In both designs, the margins for the cell reactivity and the linear element ratings can be increased by lowering the enrichment of the Mo-99 targets. However, this increased margin is at the cost of Mo-99 yield so a balance between the desired margin and production yield must be achieved.

3.2 Proposed Bundle Shift Scheme

The Mo-99 production bundle designs are both effective when used with a multi channel staggered bundle shifting scheme. By staggering the channels over a sub region of the core, it is easy to achieve the required burnup for all the production bundles while maintaining a steady production of Mo-99 bundles each week. In particular, it is important that the target bundles in the center of the core are recovered as quickly as possible to avoid excessive Mo-99 decay.

The lattice calculations discussed above are performed at the peak flux and power value which is not consistent across the axial length of the core. Therefore, to study the actual Mo-99 production as a function of fuelling scheme and residence time, the axial power profile for a standard CANDU reactor (Figure 5) is included in the fuel shuffling calculations along with the optimal full power time (about 15 days corresponding to an infinite cell burnup of 480MWd/T). This allows the design of a fuel shuffling pattern which is optimal for producing Mo-99.

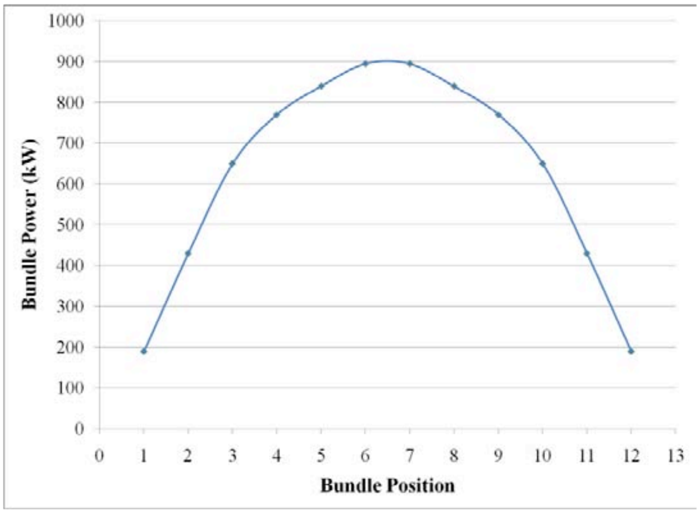

Figure 5: Axial Bundle Power Profile of a CANDU reactor [5]

The proposed fuelling cycle begins from a standard full NU channel and inserts the Mo-99 production bundles in pairs. Utilizing a series of 2 and 6 bundle shifts the Mo-99 bundles are passed through the core via multiple 7 day stages. Once developed, the cycle involves 3 steps and maintains 2 Mo-99 bundles in the channel at all times. The cycle results in an average flux time for the two bundles of 19.1 full power days (FPD) corresponding to an average burnup of 392MWd/T (infinite lattice case). A depiction of this shuffle scheme is described in Table I and Figure 6.

Table I: Inputs and Outputs for Channel Fuelling Scheme

STAGE	DAY	INPUT	OUTPUT	STEP
0	**0**	0	0	**0**
1	**0**	NU/NU/MO-1/MO-2	NU/NU/NU/NU	**S1**
2	**7**	NU/NU	NU/NU	**S2 (A)**
3	**14**	NU/NU	NU/NU	**B**
4	**21**	NU/NU/MO-3/MO-4/NU/NU	MO-1/MO-2/NU/NU/NU/NU	**C**
5	**28**	NU/NU	NU/NU	**A**
6	**35**	NU/NU	NU/NU	**B**
7	**42**	NU/NU/MO-5/MO-6/NU/NU	MO-3/MO-4/NU/NU/NU/NU	**C**
8	**49**	NU/NU	NU/NU	**A**
9	**56**	NU/NU	NU/NU	**B**
10	**63**	NU/NU/MO-7/MO-8/NU/NU	MO-5/MO-6/NU/NU/NU/NU	**C**
11	**70**	NU/NU	NU/NU	**A**

	BUNDLE POSITION												
STAGE	1	2	3	4	5	6	7	8	9	10	11	12	STEP
0	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	0
1	NUO2	NUO2	MO-99 1	MO-99 2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	S1
2	NUO2	NUO2	NUO2	NUO2	MO-99 1	MO-99 2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	S2 (A)
3	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	MO-99 1	MO-99 2	NUO2	NUO2	NUO2	NUO2	B
4	NUO2	NUO2	MO-99 3	MO-99 4	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	C
5	NUO2	NUO2	NUO2	NUO2	MO-99 3	MO-99 4	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	A
6	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	MO-99 3	MO-99 4	NUO2	NUO2	NUO2	NUO2	B
7	NUO2	NUO2	MO-99 5	MO-99 6	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	C
8	NUO2	NUO2	NUO2	NUO2	MO-99 5	MO-99 6	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	A
9	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	MO-99 5	MO-99 6	NUO2	NUO2	NUO2	NUO2	B
10	NUO2	NUO2	MO-99 7	MO-99 8	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	C
12	NUO2	NUO2	NUO2	NUO2	MO-99 7	MO-99 8	NUO2	NUO2	NUO2	NUO2	NUO2	NUO2	A

Figure 6: Proposed Fuelling Scheme for Mo-99 Production in a CANDU reactor

The scheme exposes each bundle to three separate flux cycles during their residence time in the reactor. The three 7 day stages consist of a pre-burn period in a lower flux location (bundle position 3 or 4), a high flux first stage (bundle position 5 or 6) and a high flux second stage. Upon completion of the second high flux stage the bundles are extracted and sent directly for processing. This ensures that the Mo-99 produced has very little time to decay before it reaches the processing facility. The first target bundle experience stages at 72.6%FP, 93.8%FP and 100%FP while the second experiences 86.0%FP, 100.00%FP and 93.8%FP, resulting in flux times of 18.6FPD (377MWd/T burnup) and 19.6FPD (405MWd/T burnup).

The natural uranium bundles that are used as spacers, once extracted, can be reused in a subsequent Mo-99 production cycle until they reach the standard burnup level or they can be considered spent and sent to the spent fuel storage bay. The effects on fuelling result in one fuelling of 2 or 6 bundles every 7 days for a single Mo-99 channel. For the proposed output level of 2-4 bundles per week, this production scheme would involve a group of 3-6 Mo-99 production channels and require approximately 3-6 additional fuelling operations per week. Multiple production channels offset by one stage will yield a steady supply of Mo-99 bundles. A group of 3 channels started 7 days after each other will provide a consistent 2 bundles/week starting 21 days after the initial channel is started. The three channel output is shown in Table II.

Table II: Multi Channel Mo-99 Production Results

Mo-99	Days of Irradiation								
Bundle Yield	0	7	14	21	28	35	42	49	56
Channel 1	Start	0	0	2	0	0	2	0	0
Channel 2	n/a	Start	0	0	2	0	0	2	0
Channel 3	n/a	n/a	Start	0	0	2	0	0	2
Total	0	0	0	2	2	2	2	2	2

Since there is a desire to maintain a steady level of production over the week, the channels can be offset by a few more days to produce the bundles at different times of the week. For instance, a Monday, Wednesday and Friday output scheme could be employed, allowing a more constant production level of Mo-99 throughout the week. However, this delay in processing must be balanced with the loss of Mo-99 yield and hence could not be extended more than 3-4 days.

A longer term cycle is only feasible up to about 40-50 days of irradiation since any more than this will result in the levels of activity and heat generation in the bundle from the decay of fission products being too high for the bundle to be removed from the underwater spent fuel bay and taken for chemical processing via a hot-cell. The short term extraction of fuel (less than 50 days in core) is much more suitable for immediate processing due to the lower activity and heat load of these bundles and the maximized yield of Mo-99.

The total number of additional fuelling visits in a week is 3 over and above the normal fuelling operations for a three channel group scheme. In situations where the fuelling machine is busy or unavailable, additional dwell time of a few days would not significantly affect the Mo-99 production level. The usage is small compared to the 15-20 fuelling visits performed each week but the impact on fuelling operators and the fuelling machine should be taken into consideration.

In both the cases described in Section 3.1, the suggested production level is 4 bundles per week so two sets of 3 staggered channels could be used to provide 100-120% of the global demand for Mo-99. The processing station would take in 18 pins from each of the 4 bundles being extracted at the end of each 7 day stage. For the design with the targets in the fourth ring, the total mass of fuel to be processed would be about 42kg providing ~188mg of Mo-99 resulting in a utilization of about 4.48×10^{-4} % from the 6 channels.

The proposed Mo-99 production bundles are close in performance to standard fuel and the extra bundle shifting should not increase the operator burden significantly. Additionally, in a multi unit station the channels or channel groups can be distributed over multiple units reducing the burden further on the individual fuelling machines. For both designs described, the worldwide weekly Mo-99 demand is more than satisfied using a relatively small number of channels.

3.3 Simulated Periphery Channel Production Cycle

The second bundle design with the fourth ring as targets was selected as the most attractive option at this time since the demounting of a single ring is viewed as being superior and the margin to the max linear rating is higher. Hence this option was selected for more detailed analysis of the full Mo-99 production cycle following the bundle shift scheme proposed in the previous section. The fuel tested is the 1.5% enriched SEU in the fourth ring. The study in section 3.1 was performed with the 100%FP power density set at 41.7W/g for 100%FP; this corresponds to the bundle being freshly inserted into bundle position 6 or 7 in channel C5 which is at 50%FP. In these bundle positions, axial flux is at 100% and the bundle will experience the highest power density (50%FP * 41.7W/g = 20.85W/g). However, the a fresh bundle will never be inserted directly into the middle of the channel but will be burned up for some time in earlier bundle positions (1-5) for a sufficiently enough that the pin powers are lower than those described in Section 3.1 and thus will provide adequate margin to the 60kW/m limit.

This simulation uses the multi-cell model, described in Section 3.1.1, and follows a Mo-99 production bundle through as per the shifting strategy proposed in section 3.2. The bundle begins at position 3, with bundle power (BP) at 72.6% of maximum (power density of 15.14W/g) for 7 days then moves the bundle to position 7 at 93.8% of maximum BP (power density of 19.57W/g) for 7 days and then shifts the bundle to position 9 at 100% BP (power density of 20.85W/g) for 7 more days after this point the bundle is removed from the core and the Mo-99 production yield is measured. The maximum pin power during the trial is mapped along with the Mo-99 cell power density and the Mo-99 production yield and is provided in Figure 7.

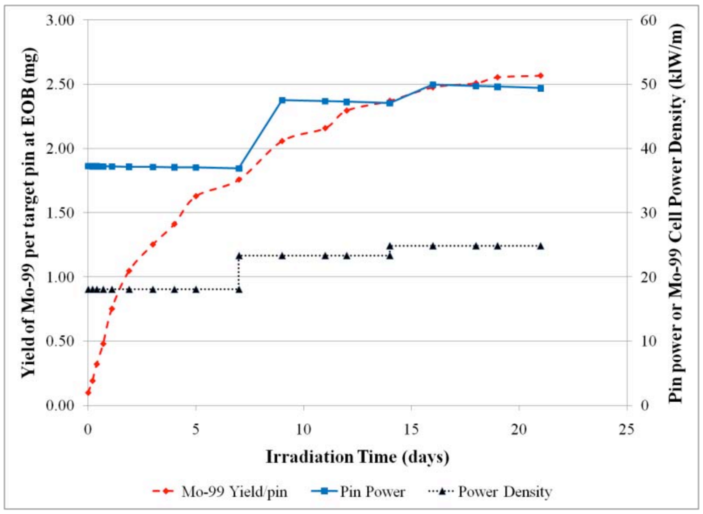

Figure 7: Pin Powers and Mo-99 Yield for Simulated periphery Channel Mo-99 Production

The pin powers during the trial are maintained below the linear rating limit of 60kW/m with a maximum pin power of 49.9kW/m at an irradiation time of 16 days (383MWd/T burnup). The total cycle involved three stages of 7 days each for a total irradiation time of 21 days corresponding to a multi-cell burnup of 529MWd/T. The Mo-99 production yield at the end of the cycle is 2.6mg/pin for a full bundle yield of 46.2mg or 2.219×10^5 EOB Curies.[†]

The companion bundle goes through a similar cycle, starting at bundle position 4 and moving to positions 6 and 8 subsequently. The power levels experienced by this bundle are 86.3% (17.94W/g), 100% (20.85W/g) and 93.5% (19.57W/g) for bundle positions 4, 6, and 8 respectively. The maximum pin power during the cycle for this bundle was 49.8kW/m at 16 days (430MWd/T burnup). The total cycle was again 21 days with a burnup of 568MWd/T. The Mo-99 production yield at the end of the cycle is 2.5mg/pin for a full bundle yield of 44.6mg or 2.142×10^5 EOB Curies. The yield is 3.5% lower than its companion bundle since it is exposed to the 20.85W/g location first resulting in slight Mo-99 decay while in the 19.57W/g location.

Therefore, the 2 bundle set travels through the channel in three stages of 7 days each for a total irradiation time of 21 days and an average burnup of 548MWd/T and produces a total of about 90.8 mg of Mo-99 (4.361×10^5 EOB Ci) which would satisfy 57% of weekly worldwide demand. This is 96.4% of the peak value calculated in Section 3.1.1 since it does not spend the full

[†] Mo-99 Yields may be slightly affected by the burnup of the surrounding fuel which is not fully accounted for here.

irradiation time at the maximum power. Using the staggered production strategy described in Section 3.2 this would be the weekly yield for a 3-channel group. Therefore with two production groups the cycle could yield almost 113% of the weekly world demand for Mo-99.

4. Mo-99 Production Issues

The production of Mo-99 in CANDU reactors is a promising opportunity as seen in the possible yields available. It has the potential of producing vast quantities of Mo-99 in a very reliable fashion for a period of decades and as such addresses many of the issues in current production methods. However, there are issues of operability and durability within the process that should be addressed in subsequent work. The design concerns include three areas; fuel handling, possible reuse of the carrier fuel and fuelling requirements. In terms of Mo-99 production challenges post-irradiation, there is the issue of volume and throughput as well as the additional active waste generated as compared to the existing research reactor production methods.

These periphery channel designs will require a fully demountable bundle to allow the second and third or the fourth rings to be removed and processed. There has been extensive use of demountable bundles for fuel experiments in NRU and some demountable bundle designs were explored by AECL for use in Thorium breeding cycles that could be modified and adopted for Mo-99 production [4]. The durability and reliability of these designs to withstand full scale CANDU fuelling loads and stresses would have to be assessed. The bundles would be removed from the core to the spent fuel bay at which point the targets could be demounted within the bay or moved to a Hot Cell facility and then the targets could be processed in an onsite facility. A cost benefit analysis of this process would need to consider these additional costs related to the handling and processing procedures and facilities.

In order to efficiently utilize resources, thought should be given to the reuse of the non target elements in the carrier bundles. The carrier elements are DU and hence are unsuitable for use in the standard fuel channels in the core. However, the carrier bundles in both designs can be reloaded with new Mo-99 target pins and be sent back into the Mo-99 production channel for another cycle. The amount of recycles that the carrier bundle elements can undergo would be limited to the standard burnup level or irradiation time of a normal CANDU fuel bundle and the level of Pu-239 produced within the DU elements. The standard NU bundles that are used as spacers in the Mo-99 generation channels proposed in Section 3.2 can be reused either within the Mo-99 channels or after a single use be sent to another channel in the core to undergo the remaining 96% of their fuel burnup. The carrier elements and the NU spacer bundles could also be disposed of directly after a single cycle to the spent fuel bay, which avoids the risks associated with extra fuel handling. The long term prospect for mechanical fuel damage increases with the amount of handling, in particular for demountable designs; hence the potential for reuse may be restricted. A reliability and safety assessment should be considered to determine the potential for reuse of the carrier elements and spacer fuel since this would reduce the impact of this process on the quantities of waste generated.

The final concern for in reactor production is the added burden on the fuelling machines to perform the required bundle shifts for the Mo-99 production channels. The proposed shift size of 2-6 bundles is within the capability of the fuelling machine for periphery channels. As described earlier a three channel staggered strategy will yield a constant 2 bundles every 7 days which could be spread out over the week. A three channel group of Mo-99 production channels would require 3 shifts per week or about one every other day. For the proposed 6 channel group this amounts to 6 additional visits per week. This demand on the fuelling machine would be over and above the standard fuelling movements needed to maintain the core reactivity in the reactor. As discussed earlier, production within a multi-unit station could split up the 3-channel production groups or the individual production channels over multiple reactors reducing the burden on each individual fuelling machine and providing backup production in the event of an outage. The excess burden on the fuelling machines will have to be investigated to ensure that the system can handle the demand without significant maintenance costs and wear to the fuelling machine.

Although the Mo-99 produced in this process represents quantities in excess of global demand from just one CANDU reactor, the Mo-99 is diluted in a larger quantity of material (i.e. fuel) than the current research reactor based production methods. Preliminary assessments show that the Mo-99 yield per gram of fuel from this proposed strategy is about 5-10% of that of conventional research reactor targets. Therefore, to produce the equivalent quantities of Mo-99, processing facilities would have to be able to handle on the order of 10 times the volumes as compared to existing facilities. While new processing facilities would be needed for such a production scheme, it remains to be demonstrated if such volumes can be processed in a sufficiently short time period so as to make the entire delivery cost effective.

5. Conclusion

The potential use of a standard CANDU reactor to supplement the production of Mo-99 was shown to be physically viable and a valid approach to addressing the production of Mo-99 isotopes. However, further investigation into the economic challenges of the proposed system is necessary to determine the possibility of a practical production strategy. The proposed designs for a periphery channel were shown to produce sizable quantities of Mo-99 while maintaining the necessary linear power rating and reactivity limits. Both the designs were simulated using the WIMS-AECL-v3.121 code for an infinite lattice and an appropriate 3 by 3 multi-cell arrangement. The design with the second and third rings enriched to 2.0% U-235 and the remaining elements made up of depleted uranium produced 44.3mg of mo-99 per bundle. Using three channels in staggered production the yield is 2 bundles/week resulting in 88.6mg of Mo-99 or 55% of the world weekly demand. The design with the fourth ring containing SEU Mo-99 target pins enriched to 1.5% U-235 and depleted uranium in the remaining elements produced 47.1mg of Mo-99 per bundle. Using three channels in staggered production the proposed yield is 2 bundles/week resulting in 94.2mg of Mo-99 or 59% of the world weekly demand.

A detailed simulation of the proposed three-stage two-bundle-in-core shift strategy with the fourth ring design was found to maintain an acceptable Mo-99 production level of 45.4mg/bundle and did not exceed the prescribed limits. Utilizing 6 channels, the weekly output of 4 bundles would yield 181.6mg of Mo-99 or about 113% of the world weekly demand. Both the bundle designs proposed are demountable and the carrier elements can be reused over multiple cycles if necessary. The challenges of the demountable bundle design, fuel reuse and fuelling machine demand as well as the impacts on safety should be considered in detail in future studies. A cost benefit analysis accounting for the additional fuelling, facility, handling costs would need to be performed as part of the next phase of this investigation to determine if this strategy is economically viable.

6. References

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Appendix 1:

A1.1 Calculation of Specific Activity of Mo-99

$$S_a = \lambda N = \frac{ln2}{\tau_{1/2}} \frac{N_A}{M_a}$$

Where for Mo-99,
$\tau_{1/2} = 2.749d = 237{,}510s$, $\lambda = 2.918 \cdot 10^{-6}\ s^{-1}$,
$N_A = 6.022 \cdot 10^{23}\ atm/mol$, $M_A = 98.91\ g/mol$

Resulting in a specific activity of 1.777×10^{16} Bq/g or 4.802×10^{5} Curies/g
(NB: 1 Curie = 3.7×10^{10} Bq)

Therefore, the end of bombardment (EOB) specific activity is $4.8\text{x}10^{5}$ Ci/g

World weekly EOB demand is 77,000 Ci which is 0.160g or 160mg of Mo-99 at EOB.

The processing efficiency for Mo-99 is about 90% and takes about a day. This results in a post processing total of 70.65% of the EOB value, approximately 54,000Ci for world weekly demand.

The post processed specific activity would then be, 3.391×10^{5} Ci/g

The 6 day curies value is computed using $N(t) = N_0 e^{-\lambda t}$,
With t = 6 days $\frac{N(6\ d)}{N_0} = 0.2203$

Therefore, the weekly world demand for Mo-99 is 1.198×10^{4} 6dCi, or about 12,000 6dCi and the specific activity of Mo-99 after processing is = 7.470×10^{4} 6d Curies/g. [2].