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MOLYBDENUM-99 TECHNOLOGICAL DEVELOPMENT

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Mo-99 Production Utilizing Target-only Reactor Design

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ABSTRACT

Nuclear reactors have been used for years to produce the medical isotope Molybdenum-99. Most of this production is through nuclear fission, because of the specific-activity and total activity that can be produced through nuclear fission. In fact, the entire US pharmaceutical distribution of Technetium-99 (which accounts for 50% of the worlds Mo-99 demand) is based upon fission produced Mo-99. Of the five reactors throughout the world that have been routinely used to produce fission Mo-99 the lowest power level is 20 MW_{th} and the highest power level is 135 MW_{th}. This paper describes a 1 MW_{th} reactor driven by only Mo-99 LEU targets that can produce 100% of the US demand for Mo-99. This paper describes the target design, core arrangement and facility layout that will permit achieving 100% of US Mo-99 production needs. This significant reduction in reactor size permits a less costly system to build and operate.

1. Introduction

Nuclear reactors, in which the targets do not significantly impact the reactor's criticality, have been used for years to produce the medical isotope Mo-99. Most of this Mo-99 production is through nuclear fission, because of the specific-activity and total activity that can be produced through that nuclear process. In fact, the entire US pharmaceutical distribution of Technetium-99m (which accounts for 50% of the worlds Mo-99 demand) is based upon fission produced Mo-99. Of the five reactors throughout the world that have been routinely used to produce fission Mo-99 the lowest power level is 20 MW_{th} and the highest power level is 135 MW_{th}. These reactors are all located outside the borders of the US and currently utilize high enriched uranium (HEU: uranium enriched in U-235 to 20% or greater) for Mo-99 production. The two largest producers, the NRU (a 135 MW_{th} reactor operated in Canada) and the HFR (a 45 MW_{th} reactor operated in the Netherlands), each produce 1/3 of the world supply, with the remaining 1/3 being produces by the combined efforts of the other three reactors (SAFARI, OSIRIS & BR2). Both existing NRU and HFR are scheduled to shut down by 2016, with NRU activities scheduled to halt while HFR is scheduled to have a replacement system by that time. In addition, the US has passed legislations to discourage the use of HEU and incentivize the use of non-HEU produced Mo-99. For the same targets design operated in the same reactor, an LEU fueled target produces only 20% of the Mo-99 that is produced by a HEU target. A large reactor for the production of Mo-99 is both expensive and potentially more difficult to obtain operational approval for. For research and test reactors, also called “non-power” reactors, the NRC makes a distinction for reactors below and above two MW_{th}.

Proposals have been made to produce Mo-99 through a self-critical or accelerator driven solution-reactor design. Solution-reactors have been built and operated within the US, but because of fundamental issues with their design have only been operated at fairly low power level. Those power levels have been in the low 10’s of kilowatts (kW). The fundamental problem with solution-reactors is the deposition of the fission fragment energy directly into the solution. That deposition results in the dissociation of water into hydrogen and oxygen. Although downstream solutions can be incorporated to recombine these dissociation products back into water, it is there rate of formation that leads to a nuclear reactor design (whether critical or sub-critical) that is characterized by large reactivity oscillations. Only when the power density is low is the configuration safe enough to operate at a neutron multiplication level close to one. As a result, solution-reactors will be limited to a few 10’s of kW each and to meet US demand for Mo-99 up to 50 of these units will have to be in operation. The current market price of Mo-99 will not support such a solution to its production.

Proposals have also been made to produce Mo-99 through neutron activation of Mo-98 or gamma-n reaction of Mo-100. Although these methods of Mo-99 production are valid, the resulting specific-activity of the Mo-99 produced through these methods are several orders of magnitude (as much as a factor of 1000) less than is obtained from even the poorest of the uranium fission production method. As a result, the current distribution system for fission produced Mo-99 will be unable to support these approaches to Mo-99 production. In order to move these production methods into a valid supply for the US medical isotope market, Mo-99 production will have to be accomplished on site or a completely new, FDA approved, low-specific-activity Mo-99 distribution system developed.

2. Eden Radioisotope’s approach to Mo-99 Production

Eden Radioisotopes is planning to produce Mo-99 utilizing a unique production target and associated nuclear reactor with a design power-level of less than two MW_{th}. The Eden approach provides a single facility production capability which can exceed current US demand for the medical isotope Mo-99. Each production target consist of a few fuel-pin like structures assembled into a target assembly that provides the ability to chemical process the fuel without having to chop or otherwise destroy the fuel cladding. This is a significant issue since any spreading contamination could eventually result in a contamination of the technetium-99 generator manufacture facility. The target assembly is operated for a short duration (usually one week) to produce high specific-activity fission product Mo-99. With minor modifications the target assembly is modified into a driver assembly that can operate for up to 24 months but is not processed due to reduced Mo-99 specific-activity levels.

The Mo-99 production nuclear reactor consists of a limited number of forced water cooled target or driver assemblies heavily reflected by radial beryllium reflectors. The radial beryllium reflector is used to; 1) minimize the number of target and driver assemblies to achieve criticality;

2) flatten the radial power distribution to achieve higher average-to-peak power levels; and 3) provide a significant background neutron source to enable a fairly quick approach to critical after Mo-99 target assembly change out. The nuclear reactor is operated as a pool type reactor to permit the routine extraction of targets assemblies without having to disassemble a reactor vessel.

Mo-99 is produced in approximately 6.1% of all U-235 fission events, while stable non-Mo-99 molybdenum is produced in approximately 18% of all fissions in the time scale of Mo-99 production. (This number increases to 24% when molybdenum-95 yield is included, but molybdenum-95 has a 65 day half-life precursor zirconium-95 that minimizes molybdenum-95 yield during the limited time associated with Mo-99 production). Mo-99 builds up with reactor operational time, reaching approximately 43 curies per kW of target power after seven days and 50 curies per kW of target power after 14 days. As a result, Mo-99 specific-activity is almost twice as high for seven day reactor operation as it is for 14 day reactor operation. As a result, the target is normally removed from the reactor after one week and processed. For the original Centichem targets with very thin HEU coatings, iodine and noble gases were cryogenically condensed prior to opening the fuel element, for LEU pellet designs this appears not to be an issue since few fission products are deposited outside of the fuel matrix and the fission product concentration results in little diffusion of fission products to fuel pin open volumes. The uranium and all fission products are dissolved in an acid cocktail and additives added to prevent several elements from precipitating when the molybdenum is eventually precipitated. The molybdenum precipitate is then separated from the acid cocktail by filtration. The molybdenum is taken back into solution and passed through a series of purification steps. The Mo-99 decays at a rate of 1% per hour, and process speed is vital to its efficient production.

3. Detailed Mo-99 Reactor Design (patents are pending on these designs)

Figure 1 shows a perspective view of the Mo-99 production target fuel pellet and target assembly that will be developed to meet Mo-99 production demands. The fuel pellet consists of uranium dioxide enriched to 19% U-235. The uranium dioxide thickness must be sufficient to provide a pellet that can be handled and operated in the reactor while providing an inner flow path sufficient for chemical processing, our current design calls for 1 mm thickness. Clearly a single hollow pellet can be used, but the use of a 180 degree sectioned pellet relieves the hoop stress that develops from radial temperature gradients in the fuel while assuring the retention of the pellets when compared to 120 degree or 90 degree sections. The fuel pellets are eventually configured into a hollow tube configuration and stacked to fill the desired fueled length of the fuel pin. The fuel pin provides a protective clad for the fuel and is made of 316-stainless-steel. The fuel pellets are retained in the fuel pin by use of non-fueled pellets, relieve springs and end tubing at the ends of the fuel pin. The overall length of the loaded fuel pin with the end tubing attached is approximately 60 cm (24 inches). When constructed, the loaded fuel pin provides a fuel arrangement that is cooled through the clad when operated in the reactor and provides an internal chemical flow path that is utilized when processing the target in the hot cell. Although any number of fuel pins can be grouped into an assembly, from a single pin to as many as is necessary to meet the daily Mo-99 demand, we believes that processing one target daily should provide approximately 25% of the market demand. A 25% market share is required for this reactor driven production to be self supporting. For a target to meet 25% of the daily market demand, each target must operate at an average power of approximately 50 kW for one week.

As such, seven fuel pins each with an outside diameter of 3/8 inches are grouped to form a target assembly. The outside diameter of these fuel pins are chosen to meet heat rejection requirements based on nucleate boiling limitations. More pins with a smaller diameter could be assembled, but that would add unnecessarily to the cost of the target fabrication. Fewer pins with a larger diameter could be assembled, but that would add to the leakage of the core due to the resulting overall reduction in average core density. The vented target fuel pins are connected to common plenums at the upper and lower ends of the target assembly. Connections are provided on the target assembly at both the upper and lower ends to connect the target assembly to a chemical processing loop for extraction of the uranium dioxide and fission products without loss of cladding integrity. Those connections are capped during reactor operations, the cap serve as an assembly handling connector as well as a lower spacer for positioning assemblies in the reactor.

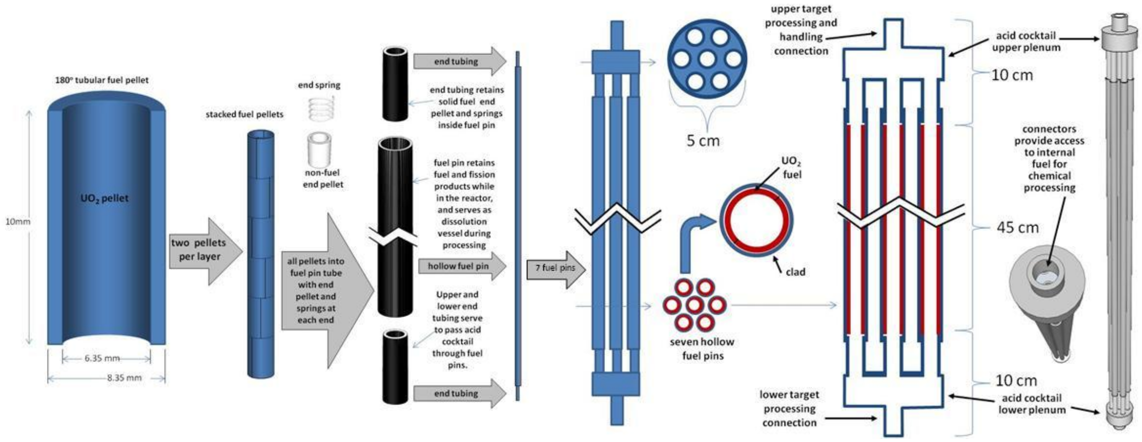

Figure 1: Target fuel pellet, pin and assembly

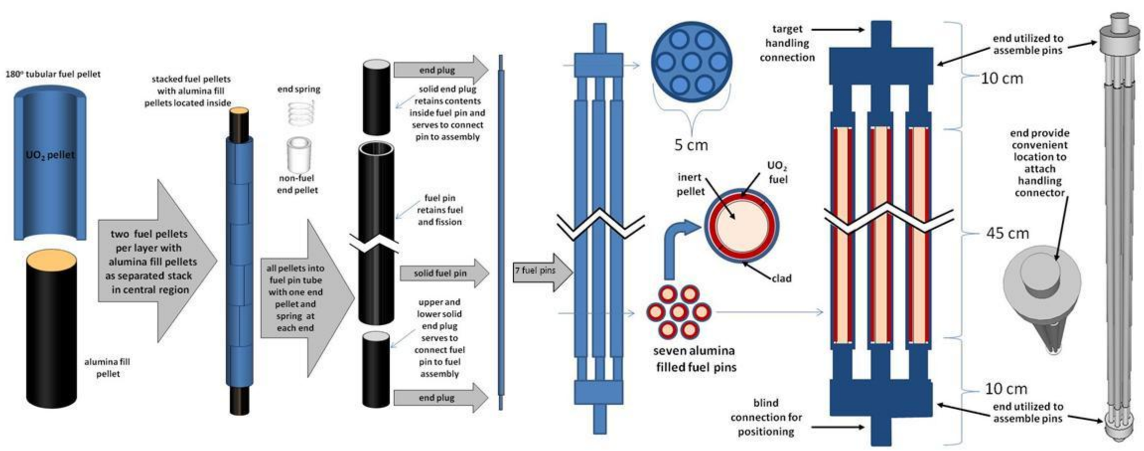

Figure 2: Driver fuel pellet, pin and assembly

Figure 2 shows a cutaway and perspective view of the driver fuel pin and driver assembly. The driver fuel pin is very similar to the target fuel pin with the inclusion of an alumina fill pellet to help maintain the uranium dioxide in the proper configuration over many months of operation and solid end rods where flow path providing tubing was used in the target fuel pin. The driver assembly looks nearly identical to the target assembly, but does not provide a flow path for process fluid through the assembly. The driver assembly provides the same fuel loading, fuel to moderator ratio and fuel enrichment as does the target assembly. Replacing a target assembly with a driver assembly or vise versa does not substantially modify the reactor performance. Total UO_2 within either fuel assembly is calculated at 750 grams. At today's cost for natural uranium, conversion services and enrichment services, the cost of enriched UO_2 is calculated at slightly less than $8,000 per target or driver assembly. Pellet fabrication could push the fuel cost up to $10K per assembly and each finished assembly is estimated to cost less than $15K.

Figure 3 shows a perspective view of the core support tubes and partial tubes in various stages of assembling a core support structure. The core support tubes are sized to fit the specific design of the target and driver assemblies. The core support structure is made from Zircaloy, since it is a onetime cost and not a recurring cost like the target assemblies. For our chosen target and fuel assemblies, the core support structure consist of nineteen (approximate five cm diameter) tubes and eighteen partial tubes connected to a upper and lower core support plate. The tubes are assembled to force the coolant flow up through the production targets and driver fuel assemblies. The core support structure provides a radial beryllium reflector to minimize fuel requirement. The beryllium reflectors also provide the structure that finishes defining the outer eighteen holes in the core support structure. Six of those holes are utilized for reactor criticality control and twelve of those holes provide additional reflector plugs and potential locations for additional production target assemblies.

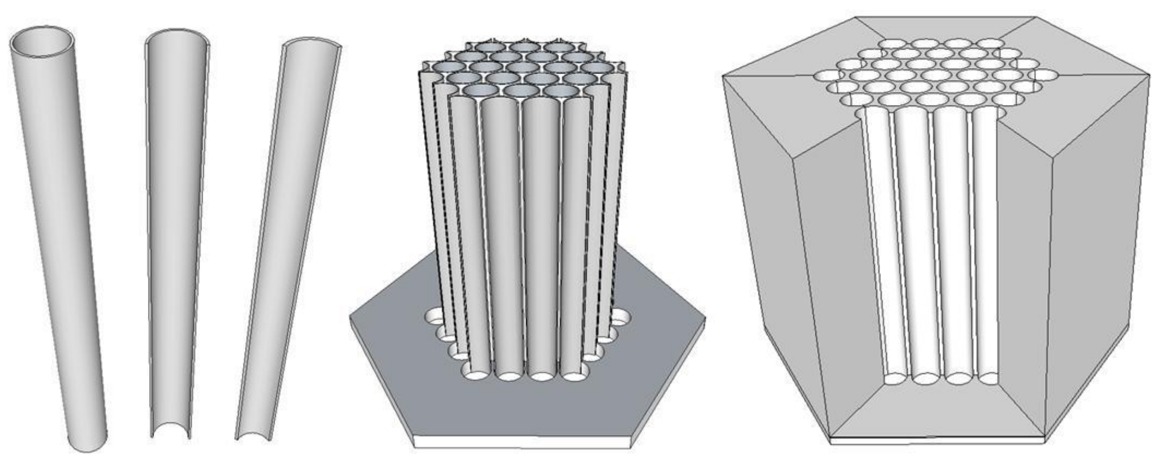

Figure 3: The core support structure in various stages of assembly, from tube to nearly completed unit with one of the six beryllium reflectors still to be installed.

Figure 4 shows a perspective view of the core support structure with target assemblies, driver assemblies and beryllium reflector plugs installed; the core support stand; and the two assembled into the operating unit that is operated at the bottom of the deep open pool. The core support stand supports the core sufficiently elevated to permit operation of the reactivity control system.

In this drawing the front three plates are removed from the stand to reveal alignment of the stand and core. The core support stand also supplies the support platform for all target assemblies, driver assemblies and cylindrical beryllium plugs. The target and driver assemblies are shown without end caps to permit a visual difference to be observed. Six holes in the support platform align with holes in the core support structure; those are the holes for the reactor control elements. Beneath those six holes are control element snubbers, design to slow the control element drop (in the event of a reactor scram) by hydraulic flow restriction just prior to final metal to metal contact. The coolant flow is introduced below the support platform through the sides of the core support stand (three shown in drawing), flowing through holes in the support platform then up and around all driver and target pins. Each reactor control element consists of a poison section followed by a reflector section. The poison section of the elements is expected to be natural enriched boron carbide filled cylinder, while the reflector follower is anticipated to be beryllium.

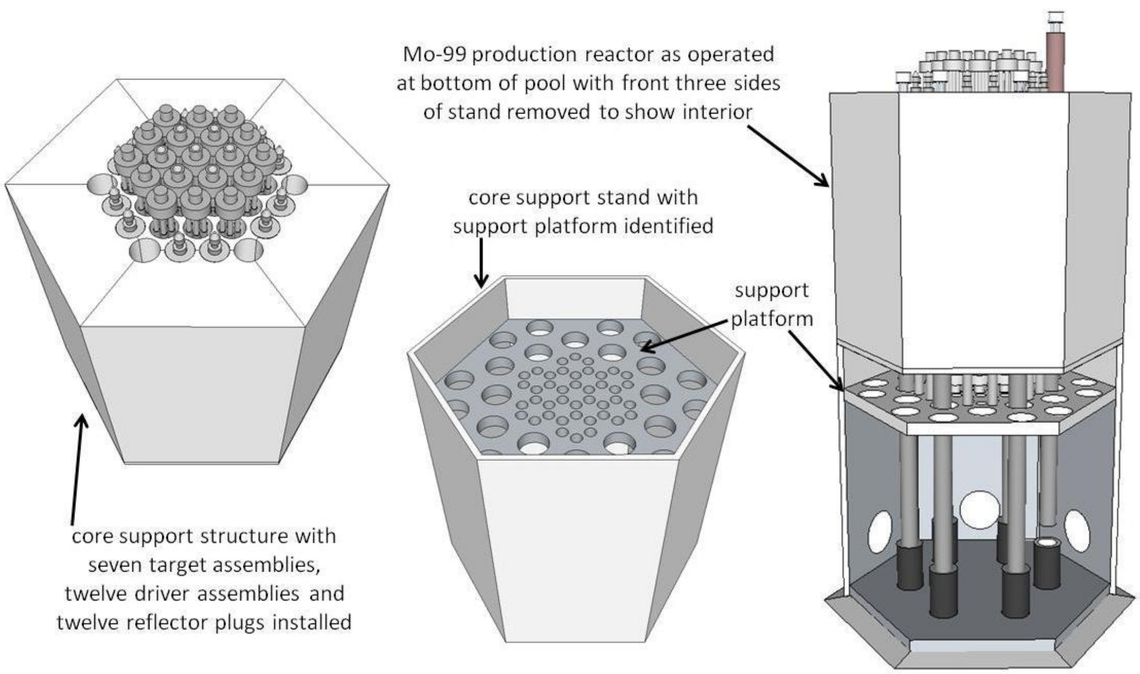

Figure 4: Core support structure with target and driver assemblies installed with core support stand.

Figure 5 shows the two extremes for reactor configuration, the smallest core arrangement and the largest core arrangement. For the smallest core arrangement, nineteen assemblies are installed into the core support structure. Only when all nineteen assemblies are being used as production target assemblies will it be necessary to expand the number of assemblies into the last row of the core support structure for additional production capability. As minimum production capabilities, one target assembly and eighteen driver assemblies can be configured for the production of Mo-99. The one target assembly would be processed once a week to meet 5% of the US demand. The upper production limit for the nineteen assembly core configuration would be to use all nineteen assemblies as target assemblies. Under this “target-only” reactor configuration four targets assemblies would be processed for each of four days and three targets would be processed on the fifth day, with all nineteen of the targets assemblies spending seven days operating in the reactor. Under this configuration the Mo-99 production facility would meet 100% of the current US Mo-99 medical isotope demand. Additional demand can be met by processing more targets daily with slightly less irradiation time each, or by adding additional targets in the outer beryllium plug ring. This remains as an operational flexibility for the facility.

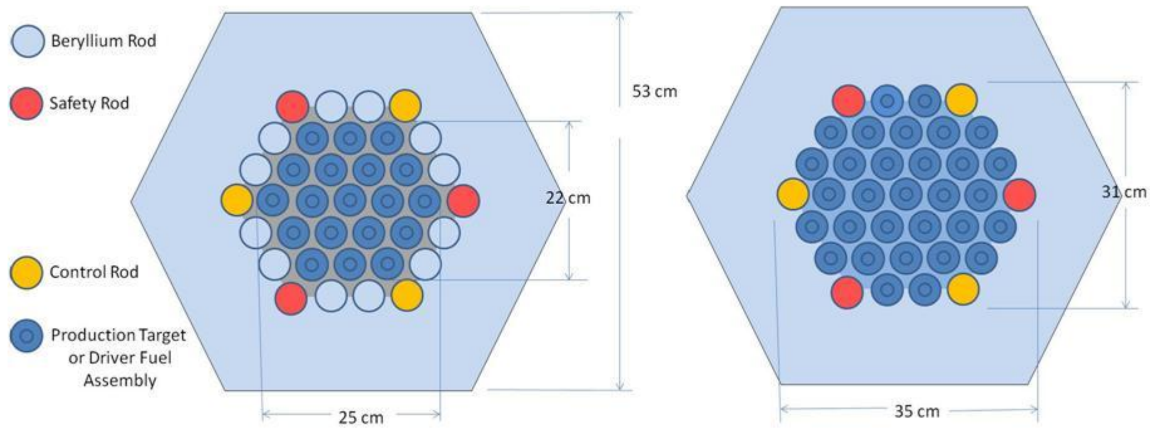

Figure 5: The Mo-99 production reactor can be configured to go critical on nineteen assemblies or accommodate up to thirty-one assemblies to produce 150% of current US demand.

4. Mo-99 Production Calculations

Figure 6 shows the Mo-99 inventory and specific-activity as a function of time for one kilowatt (kW) of reactor thermal power. The total inventory of Mo-99 is directly proportional to the reactor power since the process that produces Mo-99 (fission) is the identical process that produces the reactor power, and the specific inventory of fission produced Mo-99 is only a function of reactor time. The maximum local surface power density is limited by Departure from Nucleate Boiling (DNB) which for unpressurized system is approximately 140 watts/cm^2. Picking a design peak power level of 100 watts/cm^2 (for a safety margin on DNB) and a peak to average power density of 1.62 (to be discussed later), the average surface power density for the Mo-99 production reactor will be 62 watts/cm^2. For a single target assembly consisting of seven fuel pins each 3/8 inch OD and 45 cm long, the total surface area is 943 cm^2 and the total power is 58 kW. However, the reactor shuts down daily for approximately two hours to accomplish target exchange which reduces the effective power to 53 kW. This power results in a Mo-99 production after one week of 2280 curies at discharge from the reactor. Mo-99 is sold on a six-day-curie measurement, which is defined as the Mo-99 activity six days from shipment. Thus the 2280 curies must be reduced by the process recovery time as well as the six days. For simplicity, the process recovery time is taken as one day, resulting in 0.171 six-day-curies per curie discharged from the reactor. Each production target assembly produces 390 six-day-curies and, if a process recovery rate of 90% is achieved, 350 six-day-curies supplied to meet market demand. Processing nineteen production target assemblies per week, the total Mo-99 product to market is anticipated to be 6,650 six-day-curies per week. The total US market for Mo-99 in 2012 was 6,000 six-day-curies per week.

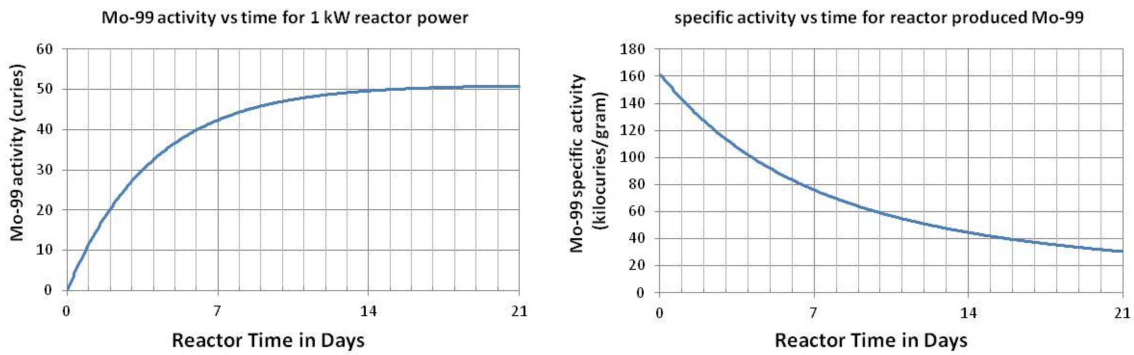

Figure 6: Mo-99 activity and specific-activity as a function of reactor operational time

5. Mo-99 Reactor Calculations

Utilizing the driver and target cores previously described, a bare un-reflected reactor could be constructed using 75 assemblies. The peak to average power ratio of a bare right circular reactor is 3.64 and, if the reactor peak power is limited to 100 watt/cm^2, would have a design power level of two MW_{th}. By operating the reactor with a water reflector, a situation that automatically occurs for all pool reactors, the number of elements can be reduced to approximately 37, and the peak to average power significantly improved. Initial calculations show a peak to average power ratio of 1.92 for a reactor element on 15 mm pitch, and with the same peak power limit as before that reactor would have a design power level of 1.85 MW_{th}. Figure 7 shows the radial and axial thermal flux distribution for the water reflected reactor using the described fuel assemblies. The power distribution is proportion to the thermal flux distribution. By using a radial reflector constructed of beryllium, the size of the reactor can be significantly improved even as compared to the water reflected reactor. Figure 8 shows the radial and axial thermal flux distribution when a beryllium reflector is used. The peak to average power distribution ration for this system is 1.5 and the reactor power with the same limitations as before is 1.2 MW_{th}. When all nineteen fuel assemblies making up this reactor are target assemblies, this reactor is a “target-only” reactor.

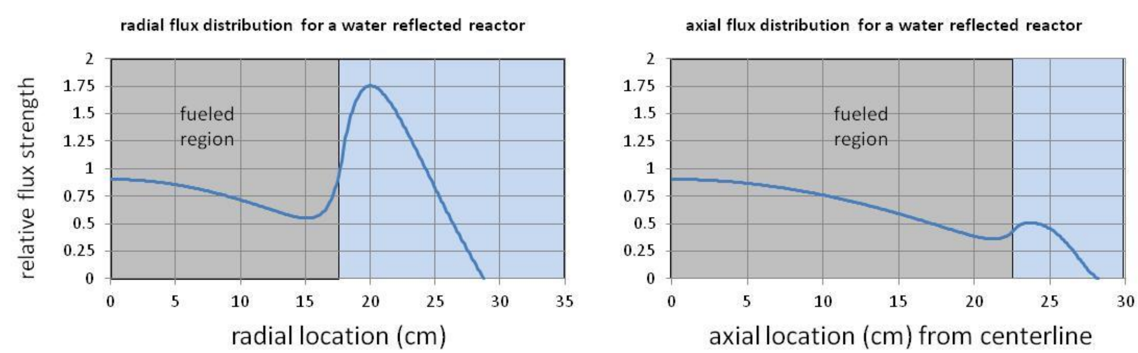

Figure 7: Radial and Axial thermal flux distribution for a water reflected reactor with described fuel assemblies and a fuel pitch of 15 mm

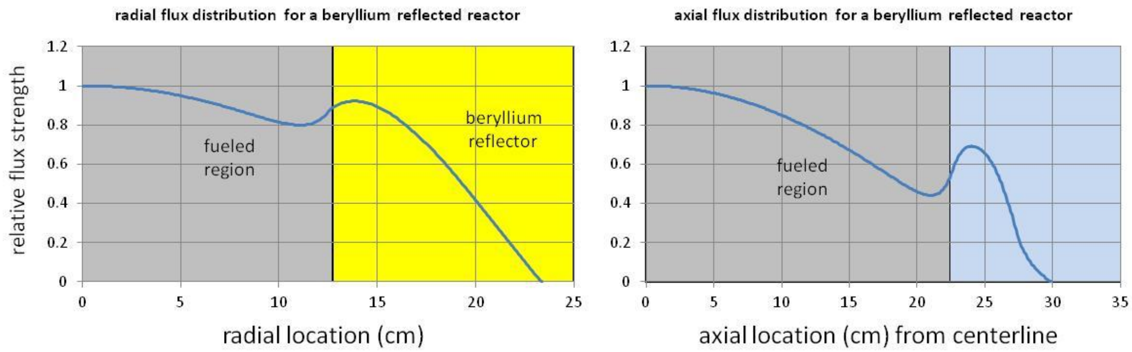

Figure 8: Radial and Axial thermal flux distribution for a beryllium radial reflected reactor with a fuel pitch of 15 mm

6. Mo-99 Reactor Daily Startup Issue

The Mo-99 production reactor could undergo “refueling” every day, during that “refueling” one or more target assemblies are replaced with new target assemblies. As a result of these target assemblies change-outs some uncertainty will exist on the fuel loading of the reactor and a determination must be made on the control element position for criticality. The normal procedure to accomplish that determination is to use a neutron source and approach criticality through a series of sub-critical neutron multiplication steps. The time that each measurement takes is driven by the neutron source strength. The sub-critical neutron multiplier (M) is related to keff by the simple equation M=1/(1-keff). As the reactor approaches criticality, the sub-critical neutron multiplier (M) approaches infinity, and by plotting one-over-M (1/M) versus the removal of the reactivity control elements, the control element position for criticality is easily obtained. Although some uncertainty may exist on what the un-multiplied source strength is, that uncertainty becomes irrelevant as the reactor approaches criticality. For the Mo-99 production reactor, the sub-critical neutron source is provided by the beryllium reflector and the fission product decay gamma of the target assemblies and driver assemblies that were not changed out. Beryllium has a gamma-n reaction with a threshold gamma energy level of 1.66 MeV. Several significant yield fission products exist with gamma emission that exceed this threshold and decay half-life that are relatively long when compared to the target change-out time, among those are; Kr-87 with a half-life of 1.27 hours and a maximum gamma energy of 2.55 MeV; Kr-88 with a 2.84 hour half-life and a maximum gamma energy of 2.39 MeV; and La-142 with a 1.54 hour half-life and a maximum gamma energy of 2.54 MeV.

7. Mo-99 Production Facility

The Mo-99 production facility will consist of one reactor, one heavily shielded hot cell, various hot and cold labs and associated office locations for all employees. Figure 9 shows our first conceptual first-floor layout of the two-story plus basement facility. The reactor will be operated around the clock, seven days per week. The hot cell will be operated Monday through Friday on a single shift schedule. The hot cell and the reactor are co-located within the same building and a target transfer system joins the two for rapid target transfers.

The hot cell will be constructed to provide six independent extraction boxes, six independent purification boxes, and three independent shipment packaging boxes. Figure 10 shows our first conceptual layout of the hot cell boxes. Full market demand can be met with only 2/3 of these hot-cell boxes in operation, the additional boxes will permit redundancy as well as isotope product development. It is anticipated that additional (non-Mo-99) isotope production efforts may require an additional operational shift in the hot cell facility. The hot cell will provide 140 density-inches (density-inches: a measurement of shielding thickness and specific density of the shielding material) of shielding between the target extraction location and the extraction process operator, 20 density-inches of shielding between the extraction box and purification box and 120 density-inches of shielding between the purification and packaging boxes.

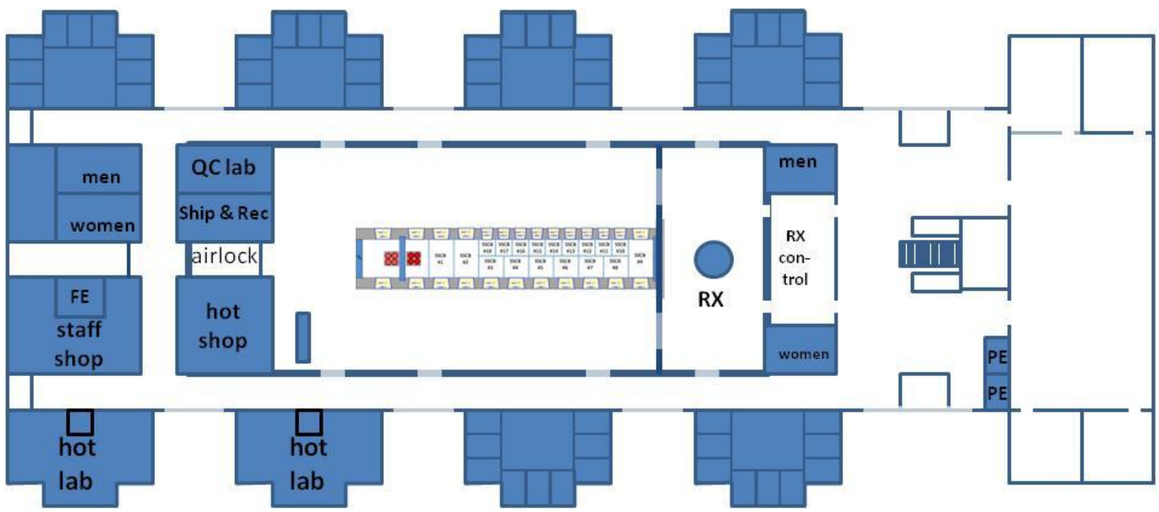

Figure 9: The Mo-99 production facility is expected to house the hot cell, reactor and all offices within a two-story plus basement complex, providing offices and working locations for upwards of 150 personnel

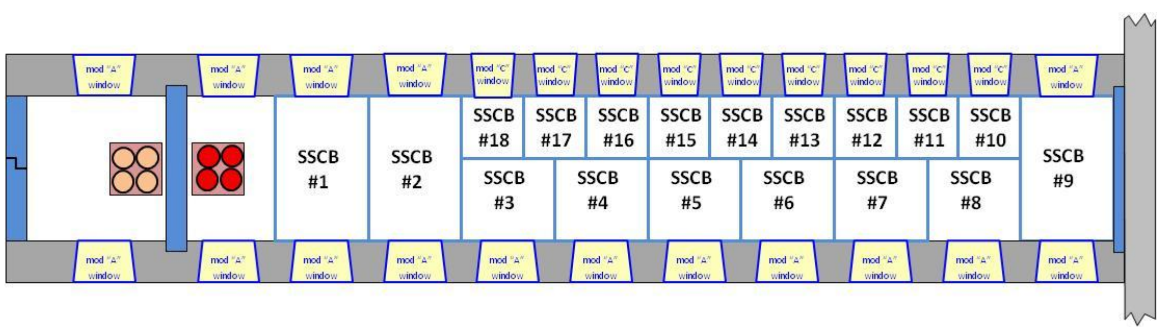

Figure 10: Looking down from above the hot cell provides up to six independent process lines, a target receiving box, an asset recovery box, a waste packaging box and two remote elevators for movement of the waste to and from decay storage location in basement

8. Summary

Eden Radioisotopes intends to move forward with a Mo-99 production facility based upon meeting a 6,000 six-day-curie per week demand utilizing this unique reactor design. This reactor is sized to meet the production needs of the medical Mo-99 industry. With today's market price for Mo-99, this facility can be self sustaining with as little as 25% of the current US Mo-99 market, thus redundancy of production facilities is still an option for this design.