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Monte Carlo Calculations for the LINAC Irradiations

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ABSTRACT

The particle transport Monte Carlo code – MCNPX has been used to perform detailed calculations providing important input to the design and planning phases of the experiments at the LINAC facility of the CSE Division at Argonne National Laboratory. Two main experimental projects at the Argonne LINAC facility are the NorthStar photonuclear production of Mo99 via (gamma, n) reaction on Mo-100 and the mini-SHINE fission-based Mo-99 production in a uranyl-sulfate solution driven by a tantalum (or DU in the second phase of the experiments) photo-neutron target. Neutron and photon fluxes in various regions of the experimental geometry along with prompt (during the irradiation) and delayed dose rates (after the run from the activated materials), shielding requirements and yields of Mo-99 and byproducts have been calculated for both projects. The results of this work are presented here.

1. Introduction

The National Nuclear Security Administration's (NNSA) Global Threat Reduction Initiative (GTRI), in partnership with commercial entities and the US national laboratories, is working to accelerate the establishment of a reliable domestic supply of Mo-99 for nuclear medicine while also minimizing the civilian use of high enriched uranium (HEU). Argonne National Laboratory (Argonne) is supporting NorthStar Medical Radioisotopes and SHINE Medical Technologies in their efforts to become domestic Mo-99 producers.

NorthStar is utilizing the (γ, n) photo-nuclear reaction on Mo-100 enriched target for the production of Mo-99. In this approach, a high-power electron accelerator is used to produce the required flux of high energy photons through the bremsstrahlung process. Due to the small photon cross-section for the reaction and the high cost of the enriched Mo-100 material, one would want to use the highest photon flux available. Argonne and Los Alamos National Laboratory (LANL) are performing engineering, design, and proof-of-concept experiments in support of the accelerator-based production of Mo-99 using a linear accelerator (LINAC) at the Chemical Sciences and Engineering (CSE) Division of ANL.

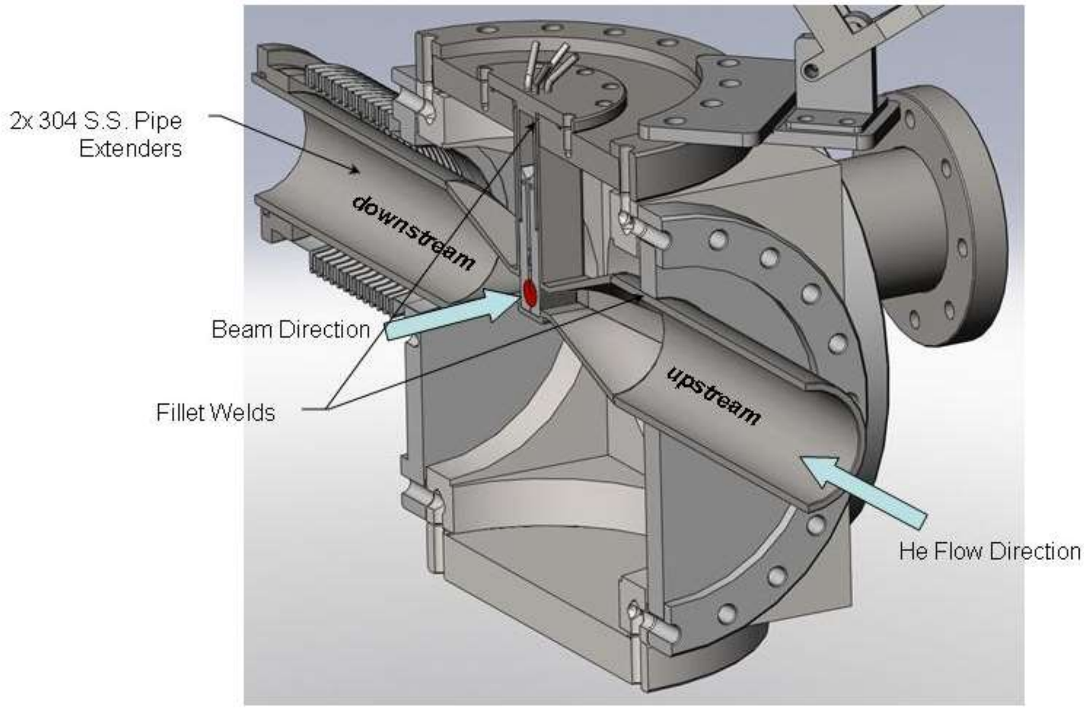
SHINE on the other hand, is currently developing the SHINE technology, which creates Mo-99 by neutron-induced fission of LEU in a sub-critical aqueous solution. SHINE can produce a primary neutron flux on the order of 10^{12} n/s/cm2 by the collision of multiple beams of deuterium (D) ions with tritium (T) gas targets and a secondary flux exceeding 10^{13} n/s/cm2 with subcritical multiplication in the aqueous solution. The D-T neutron source causes the uranium to fission, further increasing the flux by one or more orders of magnitude, and creating Mo-99 that can be extracted from the aqueous solution and purified. In essence, SHINE is an accelerator-driven sub-critical system. Argonne tasks for SHINE development are developing an understanding of the solution chemistry under operating conditions and developing the Mo-recovery and purification system. The mini-SHINE experiments are being performed using the Argonne LINAC in building 211. The experiments use an electron/photon/neutron convertor to produce neutrons that will induce fission in the target solution. The solution is 90-150 g-U/L uranyl sulfate with low enriched uranium (LEU, <20% enriched in U-235). In phase-1 experiments, we use a tantalum convertor and a target-solution volume of five liters. In Phase-2, the convertor will be depleted uranium, and the solution will be 20 L. This will generate a fission power density of up to 0.5 W/mL, equal to that foreseen for SHINE.

Extensive Monte Carlo modeling studies have been performed to support experimental work being conducted at the CSE LINAC facility for both NorthStar and SHINE projects. A general purpose Monte Carlo particle transport code called MCNPX [1] was used for modeling. Main interests for these calculations were Mo-99 production yields (both photo-nuclear and fission-based), photon/neutron fluxes, prompt and delayed (from activated materials) dose rates around the target area and shielding requirements to name a few. This paper presents a short summary of a number of selected simulation studies for each Mo-99 production project.

2. NorthStar Experiments

The target assembly for the NorthStar experiments was designed and implemented by Los Alamos National Laboratory. A closed-loop gaseous-helium-cooled target is illustrated in **Figure 1**. The target assembly consists of a target holder and a cradle that holds 25 12 mm diameter 1 mm thick disks with 0.5 mm gaps between them for helium to pass through. Most of the target assembly was manufactured from 304 stainless steel, except for the target holder and the cradle, which were made from Inconel-718 for superior thermal performance.

Photo-nuclear-based Mo-99 production for NorthStar experiments were simulated with MCNPX. Both natural and 100% Mo-100 enriched targets were studied. Natural disks were chosen for heating studies, while enriched disks will be used during production phase experiments. Modeling results of heat distribution and Mo-99 production profiles for natural molybdenum disks irradiated with 35 MeV electron beam are presented in **Figure 2** and **Figure 3**, respectively. The simulation results are normalized per kW of beam power. **Figure 2** shows that at 35 MeV the maximum heat load falls on disk number four - around 0.93 kW/cm^3 per kW of beam, while **Figure 3** reveals that the production is maximized at disk number six – 3.8E+10 Mo-100(γ, n)Mo-99 reactions/cm^3-sec per kW. Heating and production profiles were compared to independent simulations performed at Los Alamos National Laboratory and they were in very good agreement.



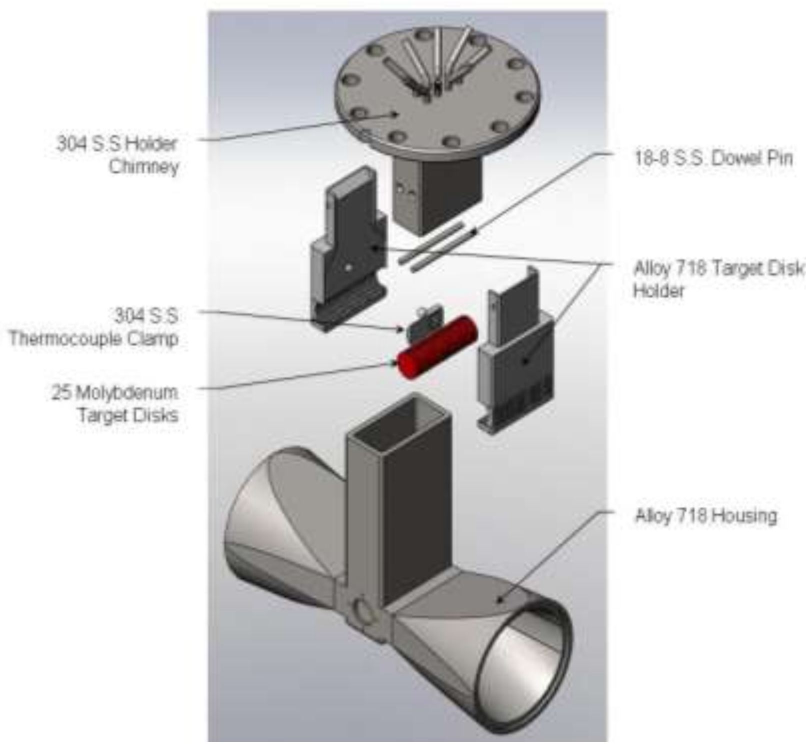


Figure 1. Helium cooled Mo-99 production target assembly for NorthStar experiments. Designed and implemented by Los Alamos National Laboratory

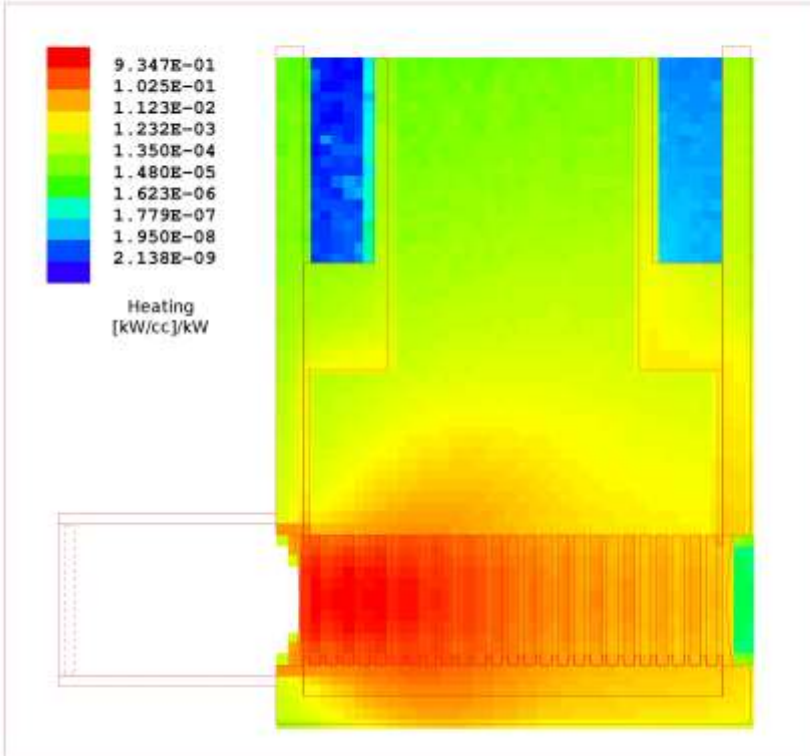


Figure 2. Power density distribution [kW/cm^3] per kW of beam power for a 35 MeV beam and natural molybdenum disks. Simulated with MCNPX.

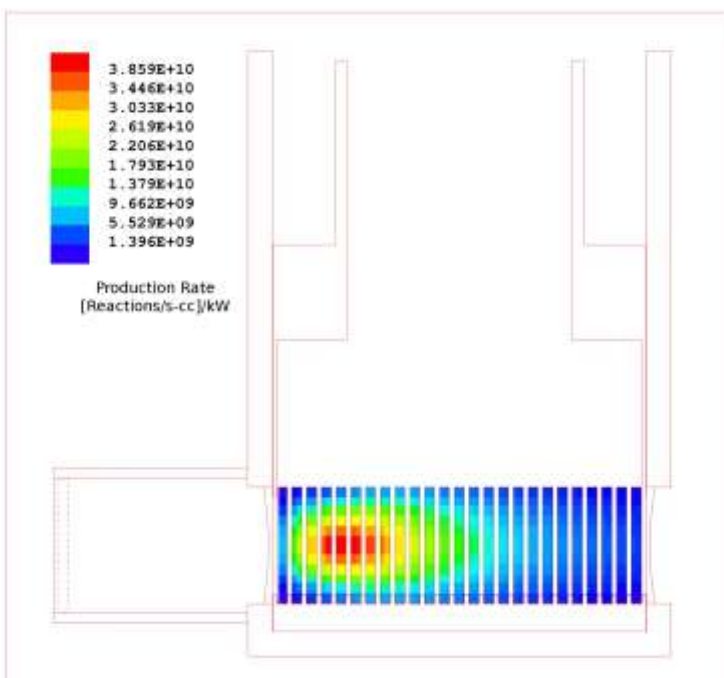


Figure 3. Mo-99 production profile [reactions/cm^3-sec per kW] for a 35 MeV beam and natural molybdenum disks. Simulated with MCNPX

Disk by disk production yields of Mo-99 are summarized in **Figure 4** for the next phase of the NorthStar experiments planned at Argonne LINAC facility. Two electron beam energies – 35 MeV and 42 MeV and three target-disk enrichments - E1 (Trace – 97.39% of Mo-100), E2 (Isoflex – 99.03% of Mo-100) and E3 (Isoflex – 95.08% of Mo-100) were considered for these calculations. The results were scaled for 500 μA (17.5 kW) and 240 μA [10.08 kW] beam currents for 35 MeV and 42 MeV, respectively. Natural molybdenum targets with short (2 hour) irradiations were considered for thermal stability studies, while mixed targets (first 6 disks - enriched, rest - natural) were chosen for 24 hour and 7 day production experiments. Total end of bombardment (EOB) Mo-99 activities for each case are shown in **Table 1.**

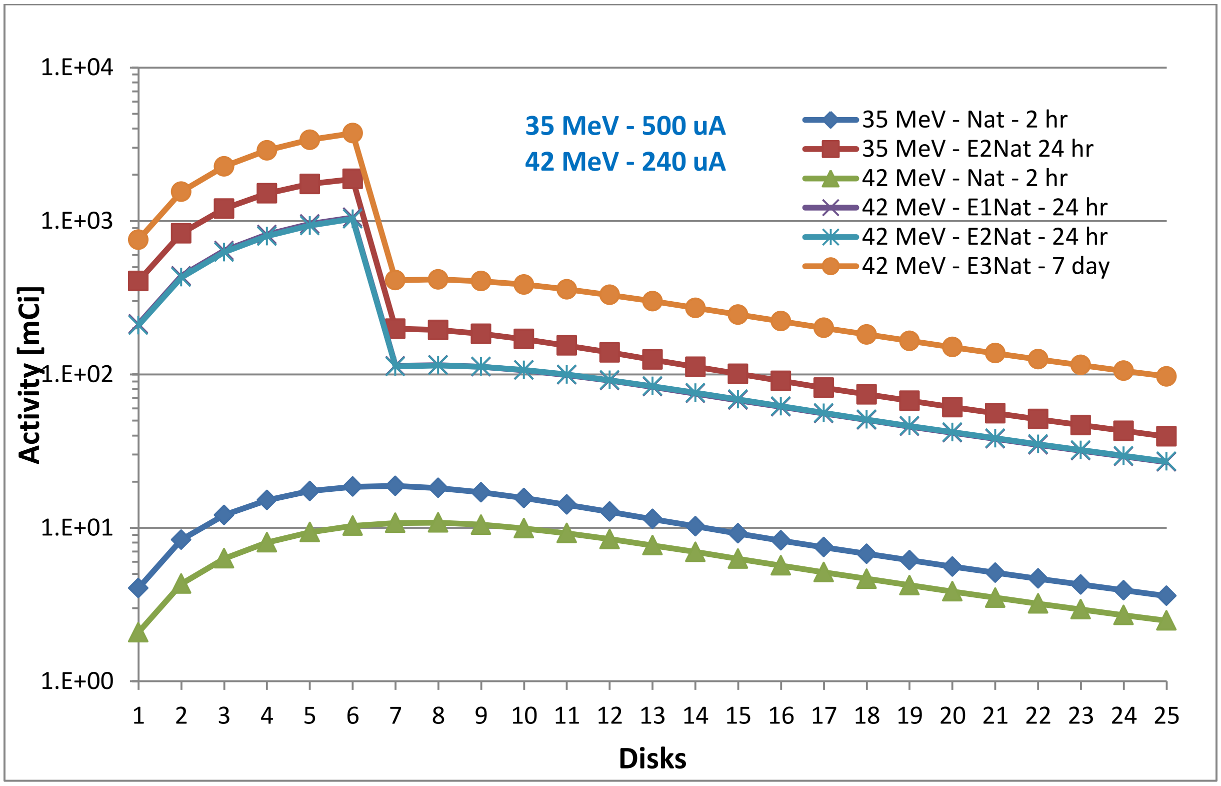


Figure 4. Disk by disk end of bombardment Mo-99 activities [Ci] for different target configurations and beam energies. Simulated with MCNPX

Table 1. Total EOB Mo-99 activities for each target configuration and irradiation parameters

Energy [MeV]	Target Configuration	Current [μA]	Duration [hours]	Activity [Ci]
35	Natural	500	2	**0.26**
35	E2-Natural	500	24	**9.56**
42	Natural	240	2	**0.16**
42	E1-Natural	240	24	**5.35**
42	E2-Natural	240	24	**5.39**
42	E3-Natural	240	156	**19.20**

3. MCNPX Modeling for Mini-SHINE Experiments

3.1 Volumetric Study

MCNPX calculations were performed to study fission versus total power-deposition characteristics as a function of the uranyl nitrate solution volume for the mini-SHINE system. Calculations were done for 5L solution Phase-I Ta target (benchmark case) and 5, 10, 20 and 30 L solution volumes with the Phase-II DU target. The Phase-I case assumed 6mm FWHM Gaussian beam profile, while Phase-II calculation were performed with a more spread-out 1.8cm FWHM beam. All the simulations were done at 30 MeV beam energy. Maximum beam powers of 10 kW and 20 kW were chosen for Phase-I and Phase-II cases, respectively.

Figure 5 demonstrates the peak fission power density as a function of the solution volume. The plot suggests that the maximum fission power density in the solution increases from about 0.25 W/cc for 5 L to around 0.4 W/cc for 20 L volume and therefore gets closer to the plant-relevant level (0.5 W/cc).

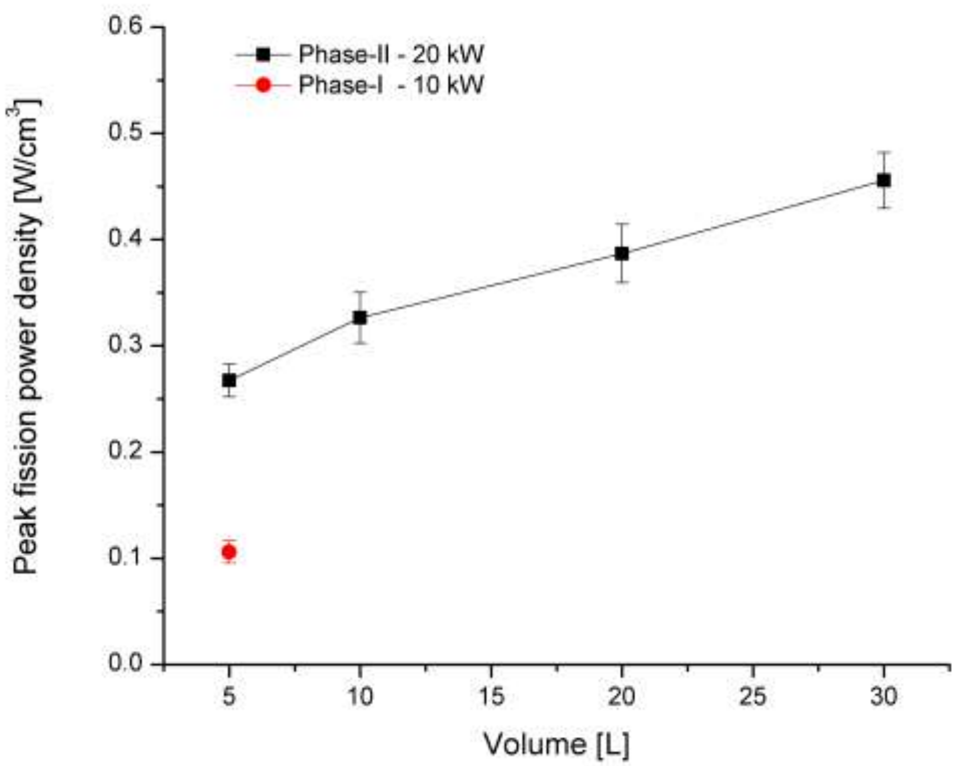


Figure 5. Peak fission power density as a function of volume for Phase-I and Phase-II targets

Total power deposition in the solution consists of gamma/electron cascades (low LET radiation) plus power deposited by fission fragments (high LET). The results of this study show (**Figure 6**) that the fraction of the power coming from fission increases significantly with volume. At 5L volume these fractions are 38% and 50% for Phase-I and Phase-II targets, respectively. It grows up to 73% for 20L and reaching 81% for 30L solution. This means that with larger volumes than 5L, mini-SHINE will be closer to SHINE subcritical system conditions. Radiolysis and radiolytic gas formations depends on LET of the radiation, and most of the radiolysis and gas formation for the SHINE system is caused by high LET fission tracks.

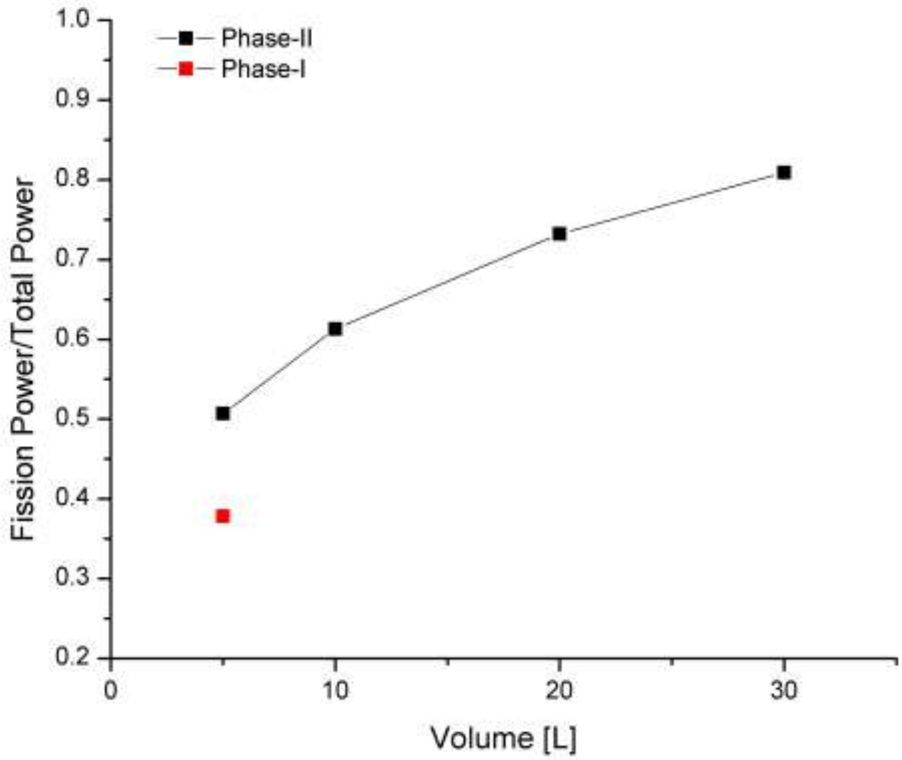


Figure 6. Ratio of fission power to total power as a function of volume for Phase-I and Phase-II targets.

Mini-SHINE Phase-2 Target Design Study

Previous MCNPX simulations [2] suggested twice as high neutron flux from the solid DU target then the solid Ta target, so, to use this advantage, the target must consist of mostly DU. The only configuration that has chance of improved heat dissipation would be a target consisting of the thin disks separated by the narrow water channels. A layout of this concept is shown in Figure 5. In this concept, all disks have a diameter of 50 mm; disks 1-10 are 2 mm (1.5 mm DU and 0.05 mm Zr, and disks 11-17 are 6-mm thick (5 mm DU and 1 mm Zr).

Figure 7 shows the energy deposition inside the target. The CSSDA range of a 35-MeV electron in uranium is 12.35 g/cm^2. At a mass density of 19.3 g/cm^3, this means that the maximum penetration of the source electrons into the uranium converter is about 0.644 cm (neglecting any slowing down in the Zircaloy cladding or the water coolant), or near the front of the fourth DU disk. Because disks 11-17 are three times thicker than disks 1-10, energy deposition rises from disk 10 to 11.

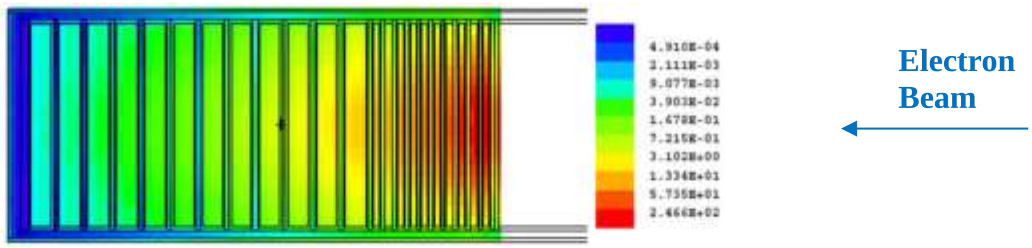


Figure 7. Energy deposition (watts/cm^3/kW) in the depleted uranium photo-neutron converter, for the irradiation conditions given in the text

This target design was placed in the middle of the 20 L uranyl nitrate solution of the mini-SHINE experimental model and further modeling studies were carried out using 35 MeV incident electron beam. The electrons were modeled as a Gaussian beam with FWHM = 1.8 cm, three times the width of the beam used with the Ta target in Phase-1. The geometry of the target and solution is shown in **Figure 8**. The solution container is surrounded by a water reflector of thickness about 13 cm.

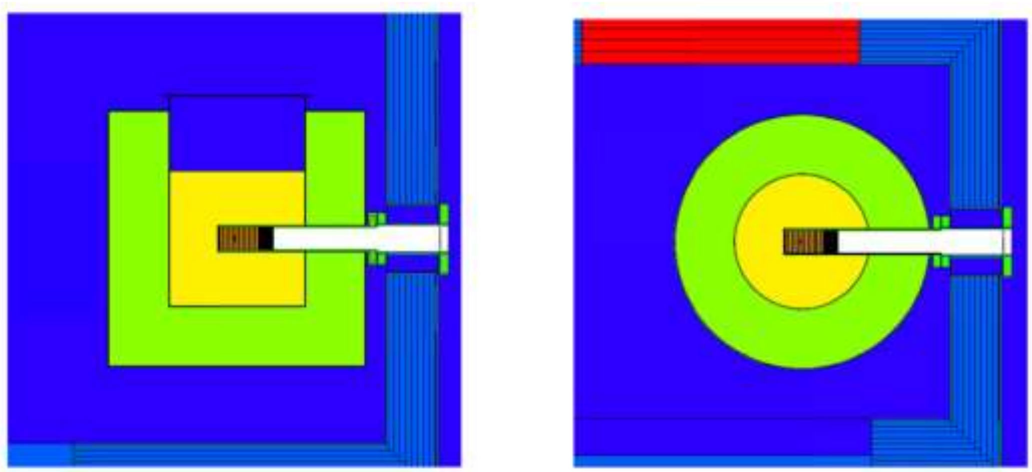


Figure 8. (left) Side view of MCNPX model used for DU target and uranyl nitrate solution calculations. The beam is incident from the right. (right) Top view of the MCNPX model through thee beamline. The beam is incident from the right

Tallies were made of the energy deposited and fission rate in each disk, the energy deposited in the remaining cells surrounding the target region, and the fission rate inside the uranyl nitrate solution. **Figure 9** shows fission rates in the uranyl nitrate solution. Because of the nature of this tally, it has no meaning inside the part of the geometry covered by the target. The total energy deposited in a uranylnitrate solution is about 155 watts/kW. For 20 kW of incident electron beam power, the average energy deposition will be about 0.15 W/cm^3. The maximum energy deposition in the solution will be about 0.02-0.025 W/cm^3/kW, or about

0.4-0.5 W/cm^3 for 20 kW of incident electron beam. This new design will allow experimental studies of radiolysis of fissioning solutions of uranium salts and optimize the Mo-99 separation scheme under plant-relevant conditions.

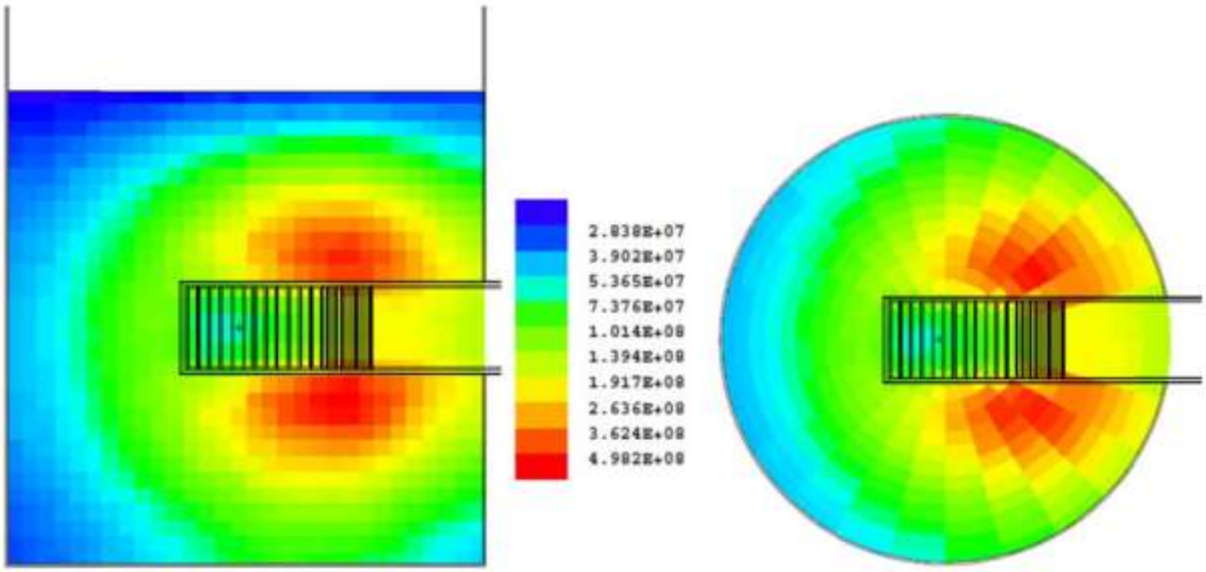


Figure 9. Fission rates (# fissions/cm^3/kW) in the uranyl nitrate solution, for the irradiation conditions given in the text. Left - side view. Right - top view.

4. Conclusions

A number of selected MCNPX modeling results for NorthStar and mini-SHINE experiments were highlighted in this paper. These and other modeling studies proved to be invaluable tools to aid the design and planning of these experiments. These calculations also provided bases for radiation safety and handling of activated targets. Besides Mo-99 yields and important design parameters, dose rates during and after the irradiation (from activated materials) as well as shielding requirements and detailed time dependent inventory of the activated products were part of the main interests of these calculations. Future Mo-99 production experiments for both NorthStar and SHINE projects will continue to be supported by MCNPX modeling.

5. Acknowledgments

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6. References

[1] D. Pelowitz (Ed.), MCNPX User's Manual, Version 2.6.0, LA-CP-07-1473, Los Alamos National Laboratory, 2008.

[2] V. Makarashvili, S. Chemerisov, B. Micklich, "Simulations of a LINAC-based photo-neutron source" *Nuclear Instruments and Methods – A,* Vol. **696**, pp. 136-140, 2012