The Effect of $^{99}$Mo Production on the Neutronic Safety Parameters of Research Reactors

Riham M. Refeat and Heba K. Louis

Safety Engineering Department, Nuclear and Radiological Regulation Authority (NRRA), Cairo, Egypt

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ABSTRACT

One of the main purposes of research reactors is the production of radioisotopes for medicine, industry and research activities. $^{99m}$Tc is one of these radioisotopes, it is generated from the radioactive decay of $^{99}$Mo and it is supplied to hospitals as $^{99}$Mo/$^{99m}$Tc generators. The most available and efficient route for obtaining the high specific activity $^{99}$Mo required for the $^{99m}$Tc generators is the separation from fission products resulting from the irradiation of $^{235}$U targets in research reactors.

The aim of this paper is to present the neutronic calculations performed to study the effect of introducing an Irradiation Device for $^{99}$Mo Production into a research reactor core utilized for fuel irradiation. Also the operational conditions with the presence of the irradiation device for $^{99}$Mo production from low enriched fuel irradiated in a research reactor are verified. Calculations are carried out in order to evaluate all core neutronic parameters to assure that the core load satisfies all safety design criteria.

Key Words: Neutronic Calculations, Safety Design Criteria, Research Reactors, $^{99}$Mo, WIMSD-5B and CITVAP.

INTRODUCTION

Over 95% of the $^{99}$Mo required for $^{99m}$Tc generators is produced by the fission of Enriched $^{235}$U targets in nuclear research reactors. The irradiated targets are then processed and the resulting purified $^{99}$Mo solution subsequently is distributed for use in the production of $^{99}$Mo/$^{99m}$Tc generators, which is supplied to hospitals for medical uses. Researches and studies are performed on several research reactors in which the $^{99}$Mo will be produced, in order to ensure that the allowed operational safety limits of the reactor are not exceeded (1).

At the NUR research reactor a general approach is used for neutron flux optimization in the irradiation channels. The approach is essentially based upon a judicious optimization of the core configuration combined with the improvement of the reflector characteristics. Such improvements in the neutronic characteristics of the NUR reactor opened new perspectives in terms of its utilization. The method allowed increasing the thermal neutron flux for radioisotope production purposes by more than 800% (2).

At PARR-1 research reactor the neutronic and thermal hydraulic analysis has been performed for the production of 100 Ci/ 3700 GBq $^{99}$Mo using LEU annular foil target. The calculations showed that the annular targets can be safely irradiated in PARR-1 for production of required amount of fission $^{99}$Mo without compromising reactor safety (3).

The Argentinean Nuclear Regulatory Authority (ARN) has elaborated a neutronic calculation model for the Argentine RA-3 research reactor which has been converted from HEU to LEU fuel. HEU $^{235}$U targets are introduced into the core and are irradiated in order to obtain $^{99}$Mo as a fission...
A regulatory analysis of the results obtained is carried out in the framework of ARN standards for fixed experiments. It was shown that the calculated values agree with measured values within the experimental uncertainty range. Also the validation carried out proved that the model is adequate for configurations that are modified for $^{99}$Mo production (4).

In this paper analysis of the effect of introducing an Irradiation Device for $^{99}$Mo Production into a research reactor core and verification of operational conditions for this reactor with the presence of the molybdenum targets is performed.

The reactor under consideration is an open-pool type, with a thermal power of 22 MW and a maximum thermal neutron flux of $2.7 \times 10^{14}$ (n.cm$^{-2}$.s$^{-1}$) in the central irradiation position (neutron trap). The core of the reactor consists of arrays of fuel elements, reflector elements, absorber rods, gadolinium injection chambers and irradiation devices. The core is located inside the reactor pool at a water depth of 10 meters below the pool surface surrounded by a Zircalloy chimney. The chimney is open at its upper end and extended above the core. Its lower part is made of Zircalloy with double wall which defines an inner space used for the second shutdown system. The gap between Zircalloy walls is filled with compressed nitrogen in normal operation conditions. There are four gadolinium injection boxes located at each core side which would contain enough gadolinium to bring the reactor into subcritical state (5, 6).

Two irradiation boxes will be placed inside the core for the purpose of $^{99}$Mo production replacing two fuel elements. Each irradiation box contains two target holders where the targets are loaded. The loading operation is performed inside a testing cell, and then the target holders are transferred to the auxiliary pool to be assembled in irradiation box. Irradiation boxes are transferred from the auxiliary pool to the reactor core for irradiation (1).

The reactor was operated in intervals with different operation periods. The core burnup calculations are performed up to 5 full power operation days, in order to irradiate $^{235}$U targets to produce the $^{99}$Mo as a fission product. The core configuration is shown in figure (1).

![Core Configuration Diagram](image)

*Fig. (1):* The core configuration of the reactor.
MODEL AND CODES USED

The operation follow up of the reactor is carried out using transport code on the cell level WIMSD-5B code connected by the 3D diffusion code calculation on the global reactor core level CITVAP. These codes are linked together in a computational package called “MTR_PC system” (7). Two macroscopic cross section libraries were generated (hot with xenon and cold without xenon). The libraries were generated using the cell calculation code WIMSD-5B with the updated libraries.

WIMSD (8) is a general code for reactor lattice cell calculation including burn-up calculations in wide range of reactor systems. This code solves the transport equation by collision probability or discrete ordinate method in one-dimensional geometry and calculates flux as a function of energy and position in the cell.

WIMSD first calculates spectra for a few spatial regions in the full number of energy groups of its own 69 energy group library, and uses these spectra to condense the basic cross-sections into few groups. A few group calculations are then carried out using a much more detailed spatial representation. The resulting fluxes are then expanded using the spectra of the previous calculation, so that the reaction rates at each spatial point can be calculated in the library group structure. WIMSD reads the basic microscopic cross-sections from its library tape. From these it calculates the macroscopic cross sections for each material; with resonance shielding being normally automatically calculated. In 1998, the WIMSD-5B version of the WIMSD code has been released from the OECD/NEA Data Bank (9).

CITVAP is an improved version of CITATION in the MTR_PC system. Both codes perform burn-up calculations, fuel elements management and control rod displacements with macroscopic cross sections library. Nuclear data can be given in macroscopic or microscopic cross sections library (HXS or CITATION format). These codes are used for solving problems involving the finite-difference representation of diffusion theory treating up to three space dimensions with arbitrary group-to-group scattering. X-y-z, θ-r-z, hexagonal-z, and triagonal-z geometries may be treated. The codes are designed to solve depletion problems and fuel managed for multi-cycle analysis. The adjoint flux may be determined and extensive first-order perturbation results may be obtained given microscopic data and nuclide concentrations. Static problems may be solved and perturbation results obtained with microscopic data (10).

DESIGN CRITERIA TO BE VERIFIED

For a reactor to operate in a safe manner the following design criteria should be satisfied (4).

1- Reactivity Control:

The first shutdown system (FSS) must comply with the following criteria. This is verified taking into account that the core is at the Cold-State and considering the worst case (without Xe and with molybdenum into the core):

- The safety reactivity factor (SRF) must be at least 1.5.
- The shutdown margin (SMA) must be at least 3000 pcm.
- The core must be subcritical with a shutdown margin (SM-1) of at least 1000 pcm, with any of the control plates out of the core.
- The core must be subcritical with the non compensating control plates out (SMA-NC) of the core.

Similarly, the second shutdown system (SSS) must comply with the following criteria, under the same condition as the first shutdown system.
• The shutdown margin (R2SS) must be at least 1000 pcm with the control plates compensating the excess of reactivity during the operation recommended.
• It can be noticed that at the occurrence of a single failure, the reactor can still be shut down with a margin (R2SS-1) of at least 500 pcm, being the recommended value.

2- Fixed Experiment Reactivity Worth:

The Reactivity Worth (RW) of any fixed experiment should satisfy the following conditions:
• The maximum allowed reactivity for any fixed experiment (Rexf) is 1200 pcm.
• The maximum allowed reactivity for all fixed experiments (Rexp) is 3000 pcm.

CALCULATION RESULTS AND DISCUSSION

A- Verification of the Safety Design Criteria:

1- First Shutdown System (FSS)

The main neutronic parameters of the core are listed in tables 1 and 2 below. Table 3 shows the values of calculated parameters for FSS conditions.

Table (1): Calculation for the Neutronic Parameters of the Core.

<table>
<thead>
<tr>
<th>State</th>
<th>Rexc-Cold</th>
<th>Rexc-Hot</th>
<th>PPF</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOC</td>
<td>5472</td>
<td>1695</td>
<td>2.15</td>
</tr>
<tr>
<td>EOC</td>
<td>4430</td>
<td>1058</td>
<td>2.15</td>
</tr>
</tbody>
</table>

Table (2): The RW of the FSS with and without single failure and without non compensating control plates.

<table>
<thead>
<tr>
<th>State</th>
<th>CR1 out</th>
<th>CR 2 out</th>
<th>CR 3 out</th>
<th>CR 4 out</th>
<th>CR 5 out</th>
<th>CR 6 out</th>
<th>CR-NC out</th>
<th>NONE R1SS</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOC</td>
<td>10198</td>
<td>11671</td>
<td>11534</td>
<td>10300</td>
<td>11843</td>
<td>11593</td>
<td>9474</td>
<td>14497</td>
</tr>
<tr>
<td>EOC</td>
<td>10199</td>
<td>11805</td>
<td>11649</td>
<td>10431</td>
<td>11992</td>
<td>11705</td>
<td>9576</td>
<td>14674</td>
</tr>
</tbody>
</table>

Table (3): The calculated parameters for FSS conditions

<table>
<thead>
<tr>
<th>State</th>
<th>Rexc</th>
<th>SM1SS</th>
<th>SRF</th>
<th>SM-1</th>
<th>SMA-NC</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOC</td>
<td>5472</td>
<td>9463</td>
<td>2.88</td>
<td>5164</td>
<td>4440</td>
</tr>
<tr>
<td>EOC</td>
<td>4430</td>
<td>10244</td>
<td>3.3</td>
<td>5769</td>
<td>5146</td>
</tr>
</tbody>
</table>

2- Second Shutdown System (SSS):

Table 4 shows the Gadolinium Reactivity Worth. The values of the calculated parameters for SSS conditions are presented in table 5.
Table (4): Gadolinium RW

<table>
<thead>
<tr>
<th>Face empty</th>
<th>BOC</th>
<th>EOC</th>
</tr>
</thead>
<tbody>
<tr>
<td>None</td>
<td>12955</td>
<td>12734</td>
</tr>
<tr>
<td>FC-1</td>
<td>9333</td>
<td>9050</td>
</tr>
<tr>
<td>FC-2</td>
<td>9390</td>
<td>9112</td>
</tr>
<tr>
<td>FC-3</td>
<td>9661</td>
<td>9386</td>
</tr>
<tr>
<td>FC-4</td>
<td>7732</td>
<td>7471</td>
</tr>
</tbody>
</table>

Table (5): The calculated parameters for SSS conditions

<table>
<thead>
<tr>
<th>R2SS</th>
<th>R2SS-1</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOC</td>
<td>EOC</td>
</tr>
<tr>
<td>12955</td>
<td>12734</td>
</tr>
</tbody>
</table>

3- Fixed Experiment Reactivity Worth:

Table 6 shows the absolute values of reactivity worth for all the fixed experiment which includes the cobalt irradiation device (CID) and the two molybdenum irradiation device (MID1 and MID2).

Table (6): Absolute values of fixed experiment reactivity worth

<table>
<thead>
<tr>
<th>CID</th>
<th>MID1</th>
<th>MID2</th>
<th>Rexp</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOC</td>
<td>EOC</td>
<td>BOC</td>
<td>EOC</td>
</tr>
<tr>
<td>946</td>
<td>956</td>
<td>237</td>
<td>230</td>
</tr>
<tr>
<td>247</td>
<td>239</td>
<td>1430</td>
<td>1425</td>
</tr>
</tbody>
</table>

Table 7 shows the compliance of the design criteria listing the worst case of each value involved.

Table (7): Verification of the system conditions

<table>
<thead>
<tr>
<th>Acceptance Value</th>
<th>Calculation Results at</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>BOC</td>
<td>EOC</td>
</tr>
<tr>
<td>(SRF) &gt; 1.5</td>
<td>2.88</td>
<td>3.3</td>
</tr>
<tr>
<td>(SMA) ≥ 3000</td>
<td>9463</td>
<td>10244</td>
</tr>
<tr>
<td>(SM-1) ≥ 1000</td>
<td>5164</td>
<td>5769</td>
</tr>
<tr>
<td>(SMA-NC) &gt; 0</td>
<td>4440</td>
<td>5146</td>
</tr>
<tr>
<td>(R2SS) &gt; 1000</td>
<td>12955</td>
<td>13122</td>
</tr>
<tr>
<td>(R2SS-1) &gt; 500</td>
<td>7732</td>
<td>7856</td>
</tr>
<tr>
<td>(Rexp) ≤ 1200</td>
<td>946</td>
<td>956</td>
</tr>
<tr>
<td>(Rexp) ≤ 3000</td>
<td>1430</td>
<td>1425</td>
</tr>
</tbody>
</table>
B. Effect of the $^{99}$Mo on the neutron flux and power density:

The effect of the $^{99}$Mo on the neutron flux and power density due to the presence of a less number of fuel plates was analyzed (cold with CR fully withdrawn). Figure (2) shows the distance dependence of the neutron thermal flux along a line perpendicular to the fuel plates inside the core ($y$-axis of the core), while figure (3) shows the distance dependence of the neutron thermal flux along a line parallel to the fuel plates inside the core ($x$-axis of the core). In both figures the continuous line presents the neutron thermal flux in case of $^{99}$Mo insertion while the dashed line presents the neutron thermal flux in case without $^{99}$Mo device.

In figure (2) there are two peaks because as shown in figure (1) there are two $^{99}$Mo devices along the perpendicular path inside the core. From figures (2) and (3) it is observed that, the flux peak is relatively smaller with the $^{99}$Mo irradiation device inserted.

The effect of the $^{99}$Mo on the power density is calculated. Figure (4) shows the distance dependence of the power density along a line perpendicular to the fuel plates inside the core, while figure (5) shows the distance dependence of the power density along a line parallel to the fuel plates inside the core. In both figures the continuous line presents the power density in case of $^{99}$Mo insertion while the dashed line presents the power density in case without $^{99}$Mo device.

From figures (4) and (5) it is observed that, the power density peak is relatively smaller with the $^{99}$Mo irradiation device inserted.

Fig. (2): The neutron thermal flux along a line perpendicular to the fuel plates inside the core with and without $^{99}$Mo device.
Fig. (3): The neutron thermal flux along a line parallel to the fuel plates inside the core with and without $^{99}$Mo device.

Fig. (4): The power density along a line perpendicular to the fuel plates inside the core with and without $^{99}$Mo device.
Fig. (5): The power density along a line parallel to the fuel plates inside the core with and without \(^{99}\)Mo device.

CONCLUSION

Detailed analysis for a research reactor core utilized for fuel irradiation to produce \(^{99}\)Mo was performed. The reactor core was modified and two \(^{99}\)Mo irradiation devices were introduced. The analysis was performed using the transport code WIMSD-5B on the cell level connected by the 3D diffusion code calculation CITVAP on the global reactor core level.

The operational conditions with the presence of an irradiation device for 99Mo production were verified. The analysis performed showed that the core load satisfies all neutronic safety criteria. It was also shown that the flux and power density peaks are relatively smaller with the \(^{99}\)Mo irradiation device inserted compared to the case without the \(^{99}\)Mo irradiation device.

ABBREVIATIONS

BOC: Begin of subcycle
CID: Cobalt irradiation device
CR: Control rod
EOC: End of subcycle
FE: Fuel element
FSS: First Shutdown System
MID1: Molybdenum irradiation device in position 1
MID2: Molybdenum irradiation device in position 2
NUR: An open pool research reactor at Centre de Développement des Techniques Nucléaires in Algiers.
PARR1: Pakistan Research Reactor-1
PPF: Power peaking factor
RW: Reactivity worth
R1SS: Reactivity of the first shutdown system
R2SS: Reactivity of the second shutdown system
Rexc: Reactivity excess
Rexp: Reactivity of a fixed experiment
Rexp: Total reactivity of fixed experiments
SM: Shutdown margin
SM-1: Shutdown margin with the most effective plate out
SM1SS: Shutdown margin of the first shutdown system
SMA-NC: Shutdown margin with the non-compensating control plates out
SRF: Safety reactivity factor
SSS: Second Shutdown System

REFERENCES