TECHNICAL ASSUMPTION FOR Mo-99 PRODUCTION
IN THE MARIA REACTOR

FEASIBILITY STUDY

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ABSTRACT

The main objective of U-235 irradiation is to obtain the Tc-99m isotope which is widely used in the domain of medical diagnostics. The decisive factor determining its availability, despite its short life time, is a reaction of radioactive decay of Mo-99 into Tc-99m. One of the possible sources of molybdenum can be achieved in course of the U-235 fission reaction.

The paper presents activities and the calculations results obtained upon the feasibility study on irradiation of U-235 targets for production of molybdenum in the MARIA reactor. The activities including technical assumption were focused on performing calculation for modelling of the target and irradiation device as well as adequate equipment and tools for processing in reactor. It has been assumed that the basic component of fuel charge is an aluminium cladded plate with dimensions of 40×230×1.45 containing 4.7 g U-235. The presumed mode of the heat removal generated in the fuel charge of the reactor primary cooling circuit influences the construction of installation to be used for irradiation and the technological instrumentation. The outer channel construction for irradiation has to be identical as the standard fuel channel construction of the MARIA reactor. It enables to use the existing slab and reactor mounting sockets for the fastening of the molybdenum channel as well as the cooling water delivery system. The measurement of water temperature cooling a fuel charge and control of water flow rate in the channel can also be carried out be means of the standard instrumentation of the reactor.

1.0. INTRODUCTION

The Institute of Atomic Energy is research institution which research activity includes the wide field of nuclear physics and nuclear technology with their applications. The main nuclear facility of the IAE is the research reactor MARIA. The multipurpose high flux reactor MARIA is a water and beryllium moderated, water cooled reactor of pool type with pressurised fuel channels containing concentric multi-tube assemblies of fuel elements. The operational power is 30 MW and thermal neutron flux in the centre of the core is around 2.5×10^{14} n/cm^2s.

During the latest years in MARIA reactor were being irradiated many different target materials among them sulphur, potassium chloride, tellurium dioxide, cobalt, potassium bromide, samarium, iridium and others mainly for the Radioisotope Center POLATOM located in Świerk on the same site as reactor site. As it is seen from the attached histogram, Fig.1.1. the MARIA reactor after a long period of upgrading in the years 1985-1992, was gradually increasing the production.
Based on the reactor technical conditions and its staffing one can predict that MARIA reactor will be operated until the years 2015-2020. Above conditions allow to initiate different actions including development of the new technologies dedicated for radioisotope production. The main program is focused on xenon gas loop for iodine-125 production and fuel loop for uranium targets irradiation for molybdenum-99 production.

![Graph showing target materials irradiated in MARIA reactor in the years 1978-2006](image)

**Fig.1.1.** Target materials irradiated in MARIA reactor in the years 1978-2006

On the January 1st, 2007 the Research Developing Center for Radioisotopes POLATOM was joined the IAE structure at the right of Research Enterprise. Based on infrastructure and qualified personnel IAE and Radioisotope Center to produce molybdenum-99 using LEU targets will be developed and correspond to achieved results of investigation will be implemented to production Mo-Tc generators. The different action of this project are and will be taken in the future. Among them are: neutronic, thermal-hydraulic and dose rate calculations, target fabrication and irradiation device development, arrangement of the hot cell, irradiated target disassembling, radiochemical separation and purification process, wastes management as well as licensing of process and facilities.

### 2.0. TECHNOLOGICAL ASSUMPTIONS FOR Mo-99 PRODUCTION IN MARIA REACTOR
In the technology analysis for production of molybdenum being a fission product of enriched uranium 235 in the MARIA reactor it has been assumed that the basic component of fuel charge is an aluminium cladded plate with dimensions of 40×230×1.45 containing 4.7 g U-235. The presumed mode of the heat removal generated in the fuel charge of the reactor primary cooling circuit influences the construction of installation to be used for irradiation and the technological instrumentation [1]. The outer channel construction for irradiation has to be identical as the standard fuel channel construction of the MARIA reactor. It enables to use the existing slab and reactor mounting sockets for the fastening of the molybdenum channel as well as the cooling water delivery system. The measurement of water temperature cooling a fuel charge and control of water flow rate in the channel can also be carried out be means of the standard instrumentation of the reactor.

In the channel location analysis it has been assumed that the channel will be placed in the core at the position providing the thermal neutron flux density of \(2.0 \times 10^{14} \text{n/cm}^2\text{s}\). The number of target plates is limited by the permissible temperature of the transported fuel charge which cannot exceed 300°C. In this condition the maximum amount of plates to be in the molybdenum channel is eight pieces. It means that amount of uranium-235 will be limited to 37.6 g in irradiation cans. The details of calculations are presented in the chapter 3.

2.1. Basic equipment and technological operations

Molybdenum channel

Internal structure of the molybdenum channel (Fig.2.1) though preserves the construction principle of the Field’s tube significantly differs from the structure of reactor fuel channel. In the middle channel tube dividing the water flow the two cans with four uranium plates in each one can be placed.

The uranium charge is cooled with a stream of water flowing upward the channel which enables to use the standard mode of connecting the system of delivery of the cooling water to the channel. Simultaneously, dynamic pressure of the flow tends to lift up the cans with uranium plates. To protect against it the pressure pad holds the cans in the lower position and the movement of plates is prohibited by a mechanical blocking device which constitutes an integral part of the can. In the channel upper part the pressure pool is rested on the central screw. The central screw is mainly aimed to provide the sealing of the molybdenum channel during the work when the pressure in cooling circuit is 1.7 MPa.

Can for the uranium plates irradiation

The can is aimed to keep plates at the stable position and to form the flow of water around them during the molybdenum channel work. An aluminium can must also provide appropriate conditions of the uranium plates reloading.

In the upper part of the can there is a mechanical blocking device of the plates position and the characteristic for the MARIA reactor gripping head adjusted to the lifting magnet and mechanical grapple, which belong to the reactor accessories. The construction of the lower part of the can is based on the need to prevent its movement during reloading operation of the plates. The total height of the can to be placed in the molybdenum channel is 280 mm and its diameter 50 mm.
Channel discharging and fuel plates reloading

Based on the irradiation analysis [2] the molybdenum channel can be discharged at least 10 hours after reactor shutdown. Only after this time the channel can be unsealed the central screw and the pressure pad pulled out and the two cans with fuel plates removed one after the other. Each can during all the technology stages must be placed under shielding of water layer with a thickness of at least three meters. Under these conditions, the can is being transported into the reactor sluice on the reloading stand.

The stand must be equipped with appropriate technological outfit. This outfit is aimed to make unable any movement of the can with fuel plates and the inner container and it has to be equipped with mechanical tools enabling gripping and embedding of the individual plates in the sockets of the inner container.

Before starting the unsealing operation of the channel the transport container is being prepared for loading. It is assumed that the reloading will be accomplished under layer of water in the reactor sluice to which the transport container will be sunk. A stable position of the container during loading of the plates will be provided by an appropriate hoisting device which rests on the sluice walls in the way shown in Fig. 2.3. The construction of the hoisting device is confined by the size of the container outer diameter which is 900 mm and the inner spacing of the sluice walls which is 1500 mm.
Fig. 2.3. Transport container in the reactor sluice

The transport container will be transported to the special new constructed hot cell in the vicinity of reactor building. Other analysed in feasibility study way of transport is underwater transfer of irradiated samples from sluice region directly through technological pool into receiving hot cell (see Fig. 2.4). There irradiated target will be disassembled for chemical processing.

2.2. Proposed localization of molybdenum channel

The core configuration with the foreseen positions for the molybdenum installation was depicted in Fig. 2.2. All indicated positions match the physical and technological conditions which were defined by expected parameters of irradiated uranium targets. The final choice of the place will be done after performing a series of activation measurements in the proposed positions.
2.3. **Time of accomplishing the technology**

After reactor scram and a few hours cooling period of the molybdenum channel will be evacuated from reactor core. At first the transport container will be transported on the reactor upper surface and it will be prepared to be lowered into the reactor sluice. The shielding slabs to the pool are taken out and the bulk head for water way between the pool and sluice is lifted. Also the inner container is being removed from the transport container and set on the reloading platform.

After 10 hours cooling period the following operations and their duration time are carried: unsealing the molybdenum channel (10 min), taking out the central screw of the pressure pad (10 min), taking out one can from the channel and transferring it to the reactor sluice (5 min), reloading the plates from the can into the inner container (15 min), placing the inner container inside the transport container and revolution of the ball shielding (5 min), lifting the transport container and its decontamination (30 min). Total time above technological operation is assumed for 2 hours.

3.0. **PHYSICAL PARAMETERS OF URANIUM TARGETS AT MARIA REACTOR**

In this chapter the calculation results of the basic irradiation and cooling parameters of uranium charge containing 37.5 grams of the U-235 isotope are presented. Calculation has been accomplished by means of ORIGEN code [3] using the spectrum indexes calculations performed by means of WIMS [4]. The following data have been obtained:

\[
\begin{align*}
\text{THERM} & = 0.8076; \\
\text{RES} & = 0.1369; \\
\text{FAST} & = 1.8687
\end{align*}
\]

3.1. **Production, cooling and selection of the Mo-99 manufacturing parameters**

The main objective of U-235 irradiation is to obtain the Tc-99m isotope which is widely used in the domain of medical diagnostics. The decisive factor determining its availability, despite its short life time, is a reaction of radioactive decay of Mo-99 into Tc-99m. One of the possible sources of molybdenum can be achieved in course of the U-235 fission reaction. Variation of molybdenum activity during the irradiation of U-235 charge versus time when the process parameter is to be determined by thermal neutron flux at the nuclear reactor is shown in Fig. 3.1.

![Fig. 3.1. Variation of Mo-99 activity during irradiation time](image1)

![Fig. 3.2. Variation of Mo-99 activity during the cooling time](image2)
As follows from the plot the molybdenum activity rises proportionally with increasing the neutron flux while its variation versus irradiation time depends on the approaching to the saturation state and in the considered range of 120-240 h, it changes by ~ 10%. Variation of Mo-99 activity during the cooling time is depicted in Fig. 3.2.

The choice of the operation conditions for the irradiation installation at the MARI A reactor an irradiation of the fuel sample containing 37.5 g of U-235 is performed at the neutron flux 1.8×10^{14} n/cm^2s for 120 hours. Assuming, that due to the reactor location the uranium sample is transferred to next stages of chemical treatment with the time delay of 12 hours from the moment of reactor scram, then the molybdenum activity in compliance with the curve on the Fig. 3.2 is 2.5×10^{14} Bq.

Bearing in mind the variations described on the plot 3.2 the most favourable option in these conditions is to irradiate the uranium charge during 150 hours at the neutron flux of 2.0×10^{14} n/cm^2s. It means that the activity at the discharging moment will be 3.4×10^{14} Bq.

For the further analyses the option to produce molybdenum within 150 hours at the neutron flux of 2.0×10^{14} n/cm^2s was chosen. Based on the calculations the main parameters for the uranium charge in these conditions are presented in Table 3.1.

Table 3.1. Specification of the irradiation parameters of uranium charge for the Mo-99 production

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Amount</th>
<th>Amount</th>
</tr>
</thead>
<tbody>
<tr>
<td>HEU target</td>
<td></td>
<td>LEU target</td>
</tr>
<tr>
<td>U-235 uranium mass in the fuel charge</td>
<td>g</td>
<td>37.6</td>
</tr>
<tr>
<td>Enrichment</td>
<td>%</td>
<td>93</td>
</tr>
<tr>
<td>Thermal neutron flux</td>
<td>n/cm^2s</td>
<td>2.0×10^{14}</td>
</tr>
<tr>
<td>Power during operation</td>
<td>kW</td>
<td>270.0</td>
</tr>
<tr>
<td>Time of irradiation</td>
<td>h</td>
<td>150</td>
</tr>
<tr>
<td>Mo-99 concentration after irradiation</td>
<td>mg</td>
<td>2.08</td>
</tr>
<tr>
<td>Mo-99 concentration after 36 h cooling time</td>
<td>mg</td>
<td>1.43</td>
</tr>
<tr>
<td>Mo-99 activity after activation</td>
<td>Bq</td>
<td>3.35×10^{14}</td>
</tr>
<tr>
<td>Mo-99 activity after 36 h cooling time</td>
<td>Bq</td>
<td>2.47×10^{14}</td>
</tr>
<tr>
<td>Total activity of the charge after 10h cooling time</td>
<td>kCi</td>
<td>151.0</td>
</tr>
<tr>
<td>Total activity of the charge after 36 h cooling time</td>
<td>kCi</td>
<td>77.0</td>
</tr>
<tr>
<td>Thermal power after activation</td>
<td>kW</td>
<td>12.1</td>
</tr>
<tr>
<td>Thermal power after 10 h</td>
<td>W</td>
<td>900.0</td>
</tr>
<tr>
<td>Thermal power after 36 h</td>
<td>W</td>
<td>410.0</td>
</tr>
</tbody>
</table>

3.2. Hazard for the operation personnel

The underwater transport of the cans containing the half of the charge mass from the channel to the loading stand enables to preserve the proper thickness of water shielding layer and to handle the operations with nuclear fuel. The greatest threat for the personnel is caused by inner container loading operation in the 10-th hour after reactor scram.

Applying a conservation principle it has been assumed that loading of the inner container containing the full uranium charge is to be conducted with the minimum water shielding layer of 3 m. The hazard sources in the 10-th hour after reactor scram have been defined in compliance with the calculation by means of the ORIGEN code. The dose rates on the water surface were calculation by means of the Microshild [5] code and they are presented in Fig.3.3.
Fig. 3.3. Dose rates on the water shielding surface shield thickness – 3 m.

The threat caused by the handling operations of the reloading during 0.5 h results in dose absorption of 3mR (at the 10-th hour after reactor scram) and it is within the limits of the admissible threat.

3.3. **Generation and removal of the heat**

As follows from the table the maximum thermal power during the operation of molybdenum channel is 270 kW, which for the surface of the eight plates determines the heat flux on the surface to be 208 W/cm². This value is not far away from the admissible one being permitted by the fuel manufacturer which is 220 W/cm² and it doesn’t create any danger for the fuel integrity.

These aren’t any problems associated with heat removal when assuming that the installation is connected to the reactor cooling system, which is able to take out the heat from an individual channel operating on 1.8 MW power level. However, some changes and adaptations are to be done as refers to the currently applied measurement and control devices (it concerns the flow rates capacity) which in the case of molybdenum channel will be operating within the range of high inaccuracy (the fringe of measurement range).

All analyses of the molybdenum channel operation together with the thermal-hydraulic parameters will become a subject of safety analysis documentation (assessment analyses to be transmitted to the Nuclear Regulatory Body) and the Installation Safety Analysis Report.

3.4. **Radioactive waste during first stage chemical treatment**

The technical analysis assumes that the first stage of chemical treatment will be performed in the reactor MARIA site. The uranium plates recovered from irradiated target will be dissolved in stainless steel dissolver using nitric acid. A fission gas recovery system, provided with cryogenic traps will permit to recover radioactive iodine and xenon. The amount and specific activity of ⁹⁹Mo obtained depend on target loading, neutron flux density and irradiation time.
The results of calculation presented below shown activity fission products and other isotopes during cooling time for period from 1 to 10 000 h [6]. The calculation was made by means ORIGEN code for:

- total mass of $^{235}$U in irradiated target - 37.6 g,
- density of thermal neutron flux - $2.0 \times 10^{14}$ n/cm$^2$s,
- irradiation time – 100 h

The waste will be generated as a gases, liquids and solids and will include material in different levels radioactive categories. Initial treatment of waste will be required at the production site prior to transfer to local facility final treatment. The treatment and elimination of gaseous waste is solved by efficient off gas system. Xenon and iodine isotopes are trapped on special materials for decay before elimination into atmosphere. Liquids containing enriched uranium will be stored till decision concerning recycling for recovery will taken. Other waste solutions, containing fission products and actinides will be mixed and adjusted to neutral pH and immobilized with cement.

**Gaseous waste**

The gaseous radioactive waste consists of tritium and noble gases: krypton and xenon. The activity of $^3$H is practical constant during cooling time and it is equal ca $7 \times 10^8$ Bq. Variation of total activity of noble gases and xenon isotopes is presented on Fig. 3.4. After 20 hours of cooling time the base component of noble gases activity is xenon - especially isotope $^{133}$Xe ($T_{1/2}=5.27$ d). During the next cooling period covering the time of few weeks total activity of noble gases depends of isotope $^{85}$Kr ($T_{1/2}=10.76$ y). Its activity is ca $2 \times 10^{10}$ Bq. Xenon and krypton isotopes could be eliminated during freezing out process and after this procedure will be deposited for the next cooling time as a liquid waste. Amount of condensed xenon will be belong 30 mg and krypton ca 1.5 mg.

**Sediment**

The activity of isotopes deposited in sediment during the first stage chemical treatment and after 10 h of cooling is $5 \times 10^{15}$ Bq and decrease 2 order of magnitude after 3 months (Fig.3.5). Among isotopes in sediment dominate after few months of cooling activity of $^{90}$Y, $^{91}$Y, $^{95}$Nb, $^{106}$Ru, $^{106}$Rh, $^{144}$Ce, $^{144}$Pr, $^{147}$Pm as well as isotopes of plutonium: $^{239}$Pu and $^{241}$Pu.

**Filtrate**

The activity of filtrate is on the same level as activity of sediment. The base component is activity of iodine which will come to solution. After few month of cooling activity of $^{90}$Sr and $^{137}$Cs will.
4.0. CONCLUSIONS

The accomplished in the presented work an analysis of the physical and technical conditions proved that the MARIA reactor can be used for the Mo-99 production by means of the fission method of the U-235 contained in the plate’s fuel. The MARIA research reactor as a one of the potential facilities which can irradiate the uranium targets.

5.0. REFERENCES


