U-TARGET IRRADIATION AT FRM II AIMING THE PRODUCTION OF MO-99 – A FEASIBILITY STUDY

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ABSTRACT

Following the shortage in radioisotope availability the Technische Unversität München and the Belgian Institut National des Radioéléments conducted a common study on the suitability of the FRM II reactor for the generation of Mo-99 as a fission product. A suitable irradiation channel was determined and neutronic calculations resulted in sufficiently high neutron flux densities to make FRM II a promising candidate for Mo-99 production. In addition the feasibility study provides thermohydraulic calculations as input for the design and integration of the additional cooling circuit into the existing heat removal systems of FRM II. The required in-house processes for a regular uranium target irradiation programme have been defined and necessary upgrades identified. Finally the required investment cost was estimated and a possible time schedule was given.

1. Introduction

Tc-99m is by far the most widely used radioisotope in nuclear medicine. Its low gamma energy and its short half life of only 6 h make it an ideal probe for diagnostic imaging of many human organs.

The most common way for the production of Tc-99m requires the irradiation of uranium targets leading to the generation of Mo-99 the mother isotope of Tc-99m which itself has a relatively short half life of 66 h too. Consequently neither Mo-99 nor Tc-99m can be stockpiled for weeks or longer. On the other hand at present only three European research reactors are equipped with facilities allowing the irradiation of uranium targets for the production of the medical isotopes mentioned above. In addition, all of these reactors are in operation since several decades and turned out to become vulnerable against malfunction. This situation led to a serious shortage for medical isotopes in 2008 and consequently a growing public awareness for the reliability of their supply [1].

Already before the shortfall of Mo-99/Tc-99m the Belgian Institut National des Radioéléments (IRE) and the Technische Universität München (TUM) agreed to cooperate on a feasibility study dealing with the possible use of TUM´s research reactor FRM II for the irradiation of IRE’s uranium targets. The study was completed in mid 2009 and provided promising results.

2. General Boundary Conditions

The Forschungsneutronenquelle Heinz-Maier-Leibnitz (FRM II) is a 20 MW heavy water moderated, light water cooled research reactor being operated since 2005 by the TUM on its campus in Garching close to Munich. The basic design feature of FRM II is the single cylindrical compact fuel assembly forming the entire reactor core. Typically FRM II is run in cycles of 60 operational days in a row and for up to four cycles per year. Although FRM II has by its design clearly been optimised for basic research by means of beam tube experiments it claims to be a multi purpose reactor offering already presently several irradiation facilities [2, 3]. All of their irradiation channels are located within the heavy water moderator tank but hermetically separated from the heavy water itself by means of a water tight flange. In addition they are all accessible from the working area around the reactor pool for loading and unloading during reactor operation. These features had been transferred as an imperative condition for the target irradiation facility under investigation.

The feasibility study was a cooperative effort between TUM and IRE. Consequently the study was a based exclusively on IRE's target design, i.e. tubular targets containing 4.0 g of highly enriched uranium (HEU ≤ 92.2% in U-235) with a total height of 160 mm and an outer diameter of 22 mm.

3. Design of the Future Irradiation Facility

3.1 Irradiation Channels

In the first step after the launch of the study various spare positions within the heavy water moderator tank of FRM II were examined with respect to their suitability for being allocated to the irradiation of uranium targets. The most promising candidate is a vertical thimble being located in a distance centre to centre of 450 mm from the reactor core. It was originally foreseen to be used in a fast rabbit irradiation facility which, however, had never been built. Unfortunately the existing thimble reaches only down to the mid plane of the reactor core. In order to use the entire height of the fuel assembly and to irradiate as many targets as possible in parallel it will be replaced by a new, longer thimble going down to the bottom of the reactor core. In addition the new thimble will be manufactured from zircalloy instead of AlMg3 in order extend its lifetime considerably.

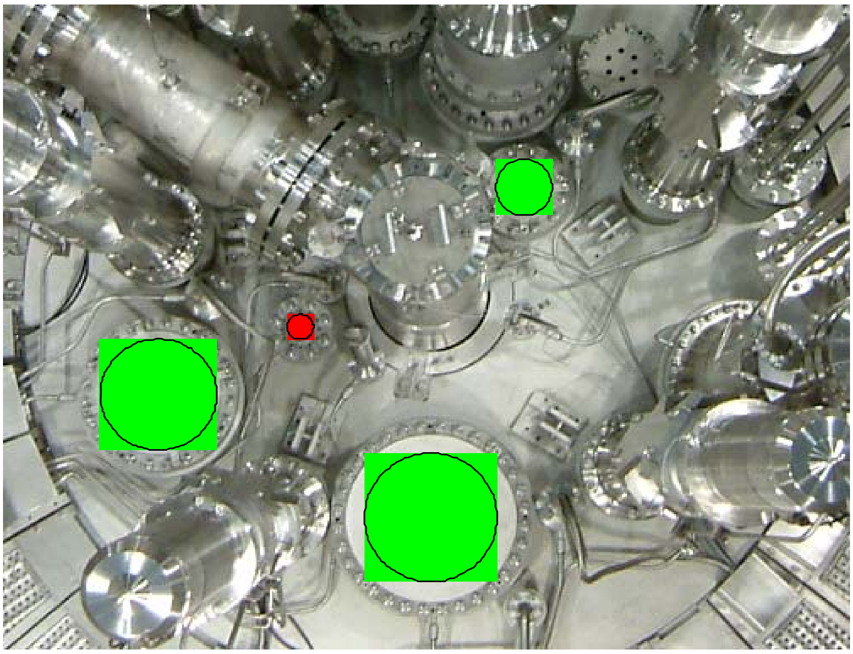

Fig. 1:
Top view to FRM II moderator tank. The future uranium target irradiation position is indicated in red, rejected spare positions are indicated in green.

The thimble will be housing three independent irradiation channels, i.e. three tubes containing up to five uranium targets each and a corresponding tube for each of them in order to close the cooling water circuit. The open space within the thimble will be He-filled and the humidity of the He gas will be monitored. This feature will control the leak tightness of the thimble and of its installations as well.

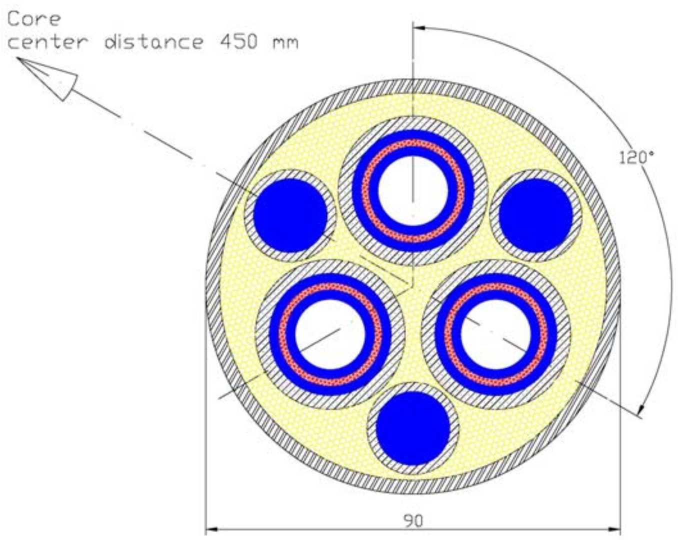

Fig. 2:
Arrangement of targets (red circles), target tubes and cooling water supply tubes within the thimble.

3.2 Neutronic Calculations

A major parameter for the suitability of the irradiation position is doubtlessly the neutron flux density. In the present study it has been examined using the 3d-transport code MCNP [4]. The input model took into account the single fuel assembly arrangement including all technical and experimental installations in the moderator tank and of course the design of the thimble containing the HEU targets described above.

The values of the flux density and their geometrical profiles along the entire irradiation channels (cross sections formed by target, tube and light cooling water) were calculated to be approximately 2.2E14 $cm^{-2}s^{-1}$ in the average with a maximum value in the mid plane of the core close to 3E14 $cm^{-2}s^{-1}$. It is to be remarked that the use of zircalloy for the thimble and, more important, the use of He instead of light water in the open space of the thimble resulted in a gain in neutron flux density. The corresponding heat load resulting from the fission processes in the irradiated targets was estimated to be equal to 430 kW provided that all 15 irradiation positions are taken.

The Mo-99 activity being produced during irradiation is estimated to sum up to 17 kCi right after a typical 6 days irradiation with the maximum number of targets, namely 15, exposed.

In addition to the neutron flux density and power production the neutronic calculations showed that the penalty caused by the target irradiation facility to neighbouring beam tube experiments is marginal, i.e. in the range of about 1%. The same applies to the influence of the targets to the reactivity of the reactor core. It was calculated to be 0.28% for fresh targets and will be even lower after the amount of Xe-135 in the targets will be in the equilibrium. Finally also the transient caused by a sudden drop of a stack of 5 targets due to a technical interference was looked at. The response curves for several sink velocities were calculated. In order to get a realistic scenario for that kind of incident a mock-up of the irradiation facility is foreseen to be built and used with various designs for target stacks, in particular for the spacers between the individual targets, and different coolant velocities.

3.3 Cooling and Thermohydraulics

As mentioned above the power released by the targets during irradiation was estimated to be approximately 430 kW. Adequate cooling is foreseen to be guaranteed by a system of four pumps. Thus, during irradiation the pump capacity is 4 × 50%; with respect to the after heat removal of the irradiated targets it is increased to 4 × 100%. Two pumps must be able to provide sufficient cooling capacity for the targets but three of them are in operation all the time; the fourth one is only a spare which is taken into operation only in case of a failure occurring in one of the others. This concept has been chosen in order

- To continue reactor operation and target irradiation in case of a failure occurring in a single pump and
- to reliably avoid an emergency shut down of the reactor itself due to a cooling failure within the uranium target irradiation facility.

The proposed cooling circuit is designed to be as far as possible independent from other reactor installations. It will take the water from the reactor pool at a rate of about 6.5 kgs/s and feed it back into the pool at its outlet. The heat removal from the coolant circuit is foreseen to take place in two heat exchangers: one of them supplies 95 kW for the heating of the reactor pool's warm water surface, which is heated electrically so far, whereas the excess power produced in the targets is transferred into the reactor's secondary cooling circuit.

Besides the reliable removal of the heat produced within the targets during irradiation the coolant circuit is designed to cover the following safety aspects:

- The target temperature must not exceed 400 °C according to the specification of the manufacturer.
- The pipelines have to withstand the water pressure created by the pumps. A pressure stage of 16 bars for the piping was established to be sufficient.
- The coolant flow has to be guided in bottom → top direction in the irradiation channel and it has to be strong enough to lift the target stack against its weight force in order to reliably avoid an uncontrolled drop of targets into the irradiation position even in case of a defect of the handling tools.

3.4 In House Handling Aspects

In addition to the irradiation technique the handling steps in the periphery – in particular the preparation of freshly irradiated targets for the shipment on public roads – had to be looked at.

Calculations show that after completion of the irradiation the targets have to stay in a water cooled position for a total of about 12 hours - only the first few hours, however, under forced cooling - before they are allowed to be handled in air. After expiry of this decay time the targets are foreseen to be transferred into the hot cell of FRM II which offers direct access from the reactor pool through a hatch in the floor. In the hot cell the targets will be removed from the support which had held them during irradiation and inserted into the transport containers provided by the customer. From the radiation protection point of view the dry packaging procedure is regarded to be considerably advantageous as compared to under water loading in the reactor pool. In addition this procedure will reduce the time consumption for packaging and radioprotection control procedures. Finally the containers need to be brought to the truck lock at the ground floor of FRM II, where the shock absorbers will be mounted.
Several shortcomings have been identified in the analysis of the in house handling of the transport containers. In particular the goods lift between the 4th floor housing the reactor pool

and the ground floor has to be upgraded and additional crane capacity has to be provided. On the other hand it has to be noted that the connection of FRM II to the European highway grid is excellent with the nearest exit only 1 km apart from the reactor.

4. Cost Estimation and Time Schedule

The total cost of the design, construction and installation of the uranium target irradiation facility was estimated to be 5.4 M€. This number includes additional personal, supervision by external experts as required according to German regulations and the upgrade of the periphery. Although the local and the federal government as well as the industrial suppliers of medical isotopes recommend unanimously the presented project it is still suffering from a lack of funding. Since, however, an extraordinary long maintenance period at the end of 2010 which is connected with the drainage of the heavy water moderator tank offers the rare chance to change the thimble in the foreseen irradiation position, TUM nonetheless undertakes at that time to supply the reactor with the necessary zircalloy thimble on its own expense. On the other hand, TUM will hardly be able to finance the entire project from the regular operational budget.

Finally TUM will have to apply at the Bavarian authorities for an extension of the operational license, which so far covers the use of enriched uranium only in form of fuel assemblies and so-called converter plates, i.e. fuel plates for cancer treatment by irradiation with fast neutrons [5]. For this purpose a safety report needs to be written in the first step.

Provided the funding issues will be solved in due time it is foreseen to build the mock-up for the testing of components in summer 2010. With respect to the installations in the reactor pool it has to be noted that access is only possible in the maintenance periods, since in spite of the importance of the project the scientific use of FRM II must not be obstructed by its realization. In summary a realistic estimation of the necessary timeframe predicts the begin of the irradiation service for Mo-99 production at the end of 2013/begin of 2014.

5. References

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