INDUSTRIAL AND COMMERCIAL APPLICATIONS OF FRM II

H. GERSTENBERG, A. KASTENMÜLLER, X. LI
Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), Technische Universität München,
Garching,
Germany
heiko.gerstenberg@frm2.tum.de

1. INTRODUCTION

The Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II) is the youngest and most powerful German research reactor. It is located on the campus of the Technische Universität München (TUM) in Garching close to Munich. The research reactor FRM II was designed and built in cooperation with AREVA (formerly Siemens Framatome). Its nuclear commissioning was successfully performed in 2004. In March 2005, the operating license for FRM II was issued by the Bavarian State authorities and since May 2005 it has been operated under the responsibility of TUM without major undesired interruptions.

The by far most important application of FRM II is the provision of thermal and cold neutrons for basic research by means of neutron beam tube experiments. Nonetheless, however, it was an important task already from the very beginning of the reactor project, making the reactor available not only to scientific users but also to interested parties dealing with technical, medical or industrial applications. Consequently, right from the beginning of routine operation of FRM II, cooperation with industrial partners mainly based on but not restricted to irradiation services was implemented.

2. REACTOR DESIGN AND OPERATIONAL MODE

FRM II is a heavy water moderated but light water cooled research reactor exhibiting a thermal power of 20 MW. Its core is in the form of a single very compact fuel element containing about 8 kg of highly enriched uranium (93% in U-235) at the start of the reactor cycle. The fuel assembly is embedded in a moderator tank with a volume of approximately 11 m³ of heavy water.

The fuel in use is U_3Si_2 dispersed in an Al matrix and covered by an AlFeNi cladding. Each fuel element is made up of 113 individual fuel plates bent to an involute shape in order to provide a cooling slit between two plates of a constant width of 2.3 mm. The fuel density within each fuel plate is 3 g/cm³ in the inner region and 1.5 g/cm³ in the outer region, i.e., the region adjacent to the moderator tank, of the plate. This disparity in fuel density was necessary in order to avoid an unacceptable peak in power density due to the high flux density of thermal neutrons being reflected from the moderator tank.

Typically FRM II is run in cycles of 60 full power days to be operated without interruption. This period corresponds to a total produced energy of 1200 MW·d, which is the maximum licensed burn up per fuel assembly. The four cycles forming the standard yearly operational regime are separated by maintenance periods used for changes of the fuel assembly and the majority of the more than 1800 periodic inspections to be carried out per year according to the operational license. It is noteworthy that many of the inspections are to be carried out under the supervision of external experts.

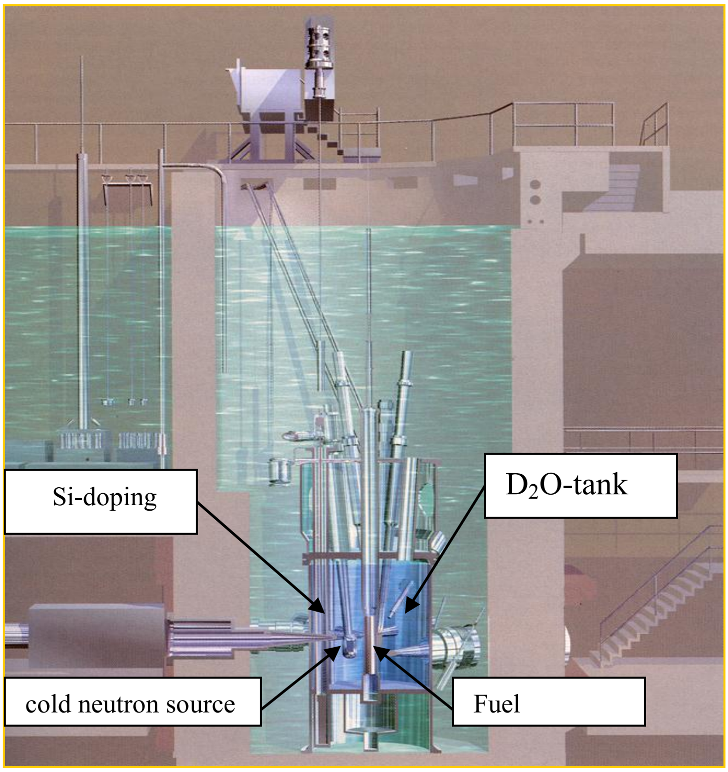

Fig. 1. Conceptual design of the FRM II moderator tank with some key components indicated.

The cylindrical (h = ø ≈ 2.5 m) heavy water moderator tank of FRM II houses most of the major experimental installations. The most important ones for the use in the framework of basic research are the cold and the hot neutron source as well as the thimbles of the beam tubes. On the other hand, however, all irradiation positions of the various irradiation devices are located within the moderator tank and consequently provide a very well thermalized neutron spectrum characterized by high Φ_{th}/Φ_f ratios. Only the so-called converter facility, an installation of two fuel plates used for the production of fast neutrons for cancer therapy, is located at the outer edge of the moderator tank.

3. IRRADIATION FACILITIES

3.1. Mechanical irradiation facility JBE70

For very short term irradiations requiring a thermal neutron fluence of not more than 5×10^{16} cm^{-2}, FRM II is equipped with a mechanical irradiation device. Before irradiation the sample is packed into a watertight welded Al capsule, which allows a maximum sample volume of 60 cm^3, and is fixed to a nylon ribbon. The irradiation channel is a light water filled vertical thimble within the moderator tank that is accessible for the loading of irradiation samples through its open top. It is located in a distance of 1000 mm from the fuel element (center to center).

In order to load the Al capsule into the irradiation channel, it is lowered into the irradiation channel by simply unwinding the nylon ribbon from a coil.

The standard vertical irradiation position of the mechanical irradiation device is the center plane of the fuel element. The corresponding neutron flux densities were determined by means of activation of AlAu (1‰) wires and pure Ni foils to be $\Phi_{th}=1.1\times10^{13}$ $cm^{-2}\cdot s^{-1}$ and $\Phi_f=1.5\times10^{10}$ $cm^{-2}\cdot s^{-1}$. It is noteworthy that it is very easy to define additional irradiation positions with even lower neutron flux densities by choosing a higher vertical irradiation position within the irradiation channel.

The mechanical irradiation device attracts a lot of interest from geological institutes from all over the world. Due to the comparatively low neutron flux density and its low flux gradient of only few percent over a typical sample length of 60 mm, it offers an ideal experimental situation for the fission track dating technique of mainly apatite or zircon minerals. An overview of this technique can be found in [1].

3.2. Rabbit systems

FRM II is equipped with a conventional gas driven rabbit system which is mainly used for the activation of samples for neutron activation analysis and a water driven system which is suited for long term irradiations up to, in principle, the cycle length of 60 days or in other words a thermal neutron fluence of $>5\times10^{20}$ cm^{-2}.

The gas driven system RPA (abbreviation of the German word **R**ohr**p**ost**a**nlage) offers in total six independent irradiation channels with different neutron flux density parameters. The differences are due to the fact that the depths of the irradiation channels within the moderator tank are graded. For these entire irradiation channels, neutron flux densities and neutron temperatures were determined via the irradiation of flux monitors according to the Høgdahl or Westcott convention [2]. The results are shown in Table 1.

TABLE 1. NEUTRON FLUX DENSITY PARAMETERS FOR THE PNEUMATIC RABBIT SYSTEM RPA

Irradiation position	Φ_{th} ($cm^{-2}s^{-1}$)	Φ_{epi} ($cm^{-2}s^{-1}$)	Φ_f ($cm^{-2}s^{-1}$)	T_n (°C)	Φ_{th} / Φ_f
RPA-1	3.57E13	6.70E09	2.02E09	23.0	17673
RPA-2	1.52E13	3.18E09	4.11E08	30.3	36983
RPA-3	4.84E12	7.61E08	7.17E07	21.2	67503
RPA-4	7.26E13	2.34E10	5.56E10	23.8	1306
RPA-5	3.88E13	1.14E10	5.87E09	27.4	6610
RPA-6	7.12E12	1.24E09	1.51E08	27.6	47152

The RPA uses carbon dioxide as a process medium in order to avoid the undesired production of ^{41}Ar that would be generated during the irradiation of air. The irradiation capsules are made from polyethylene with an inner diameter of Ø=15 mm and a length of 100 mm. They are labelled individually as a precaution against mix up and have a weight of approximately 10 g. The maximum sample load is again 10 g but typically neutron activation samples weigh well below 1 g.

After being loaded into the irradiation facility, the capsules are rinsed with fresh carbon dioxide again to avoid the undesired side irradiation of air, identified by the computer control and sent into the irradiation position. The stream of transport gas is generated by a ventilator without the use of any pressure vessels; thus the pressure within the system is kept at only 1.2 bars abs. The transport velocity is about 10 m/s. The applicable neutron fluence is limited by the capsule material PE to 5×10^{17} cm^{-2}. After completion of the irradiation the sample is blown out of the irradiation position and stopped in a well shielded (165 mm Pb) decay position where the induced gamma dose rate is measured. After the decay of the radioactivity to a value below 6 mSv/h in 50 cm unshielded, the sample can be unloaded optionally in a shielded (10 cm Pb) box that is equipped with manipulators for further handling, or directly into a lead shielded transport container. Alternatively the sample can be automatically handed over to an independent pneumatic dispatch for direct transport into the neighbouring institute for radiochemistry (RCM) at TUM, one of the major customers for radioactive samples.

For longer term irradiation the water driven capsule irradiation facility KBA (abbreviation of the German word Kapselbestrahlungsanlage) is used. This system exhibits 2 independent irradiation channels each of which can be loaded by up to 5 irradiation capsules simultaneously. The central component of the KBA is a disc shaped storage device, the so-called roundabout, located approximately 4 m below the surface of the reactor's storage pool. This device is designed to manage up to 38 irradiation capsules.

For long term irradiations in the KBA, the samples are contained in standard capsules made from extremely pure $AlMg_3$ which turned out to be necessary in order to allow unloading shortly after the irradiation. The available space within the capsule is a diameter of Ø=26 mm and a length of 90 mm, while the maximum sample mass is 100 g. Water sensitive samples are additionally sealed into tight quartz ampoules or welded Al tubes before being placed into the irradiation capsule. The irradiation positions are located deep in the moderator tank. They are the most intense ones in FRM II. The corresponding neutron flux parameters [2] are given in Table 2.

TABLE 2. NEUTRON FLUX PARAMETERS IN THE CAPSULE IRRADIATION FACILITY

Irradiation position	Φ_{th} ($cm^{-2}s^{-1}$)	Φ_{epi} ($cm^{-2}s^{-1}$)	Φ_f ($cm^{-2}s^{-1}$)	T_n (°C)	Φ_{th} / Φ_f
KBA 1-1	1.29E14	2.82E11	3.87E11	11.7	333
KBA 1-2	9.35E13	1.07E11	1.94E11	8.9	483
KBA 2-1	1.07E14	8.18E10	2.06E11	11.2	520
KBA 2-2	7.65E13	4.18E10	1.05E11	13.1	731

To launch an irradiation in the KBA, the $AlMg_3$ capsule is inserted into the loading tube. It sinks down into the "roundabout" which rotates the sample into a position connected to the irradiation position within the moderator tank. A pump generates a stream of pool water that transports the sample into the irradiation position. The speed of transport is only about 0.5 m/s, which is acceptable because of the long irradiation times. After completion of the irradiation the sample is returned into the "roundabout" using the same process where it is kept underwater for the necessary decay time. For unloading from the KBA, it is pumped in the first step into the position for dose rate measurement, which is located several meters below the surface of the pool. Only after verification of the conformity of the measured dose rate with the limiting value of 0.1 Sv/h (only pool water shielded to a distance of 15 cm) it is

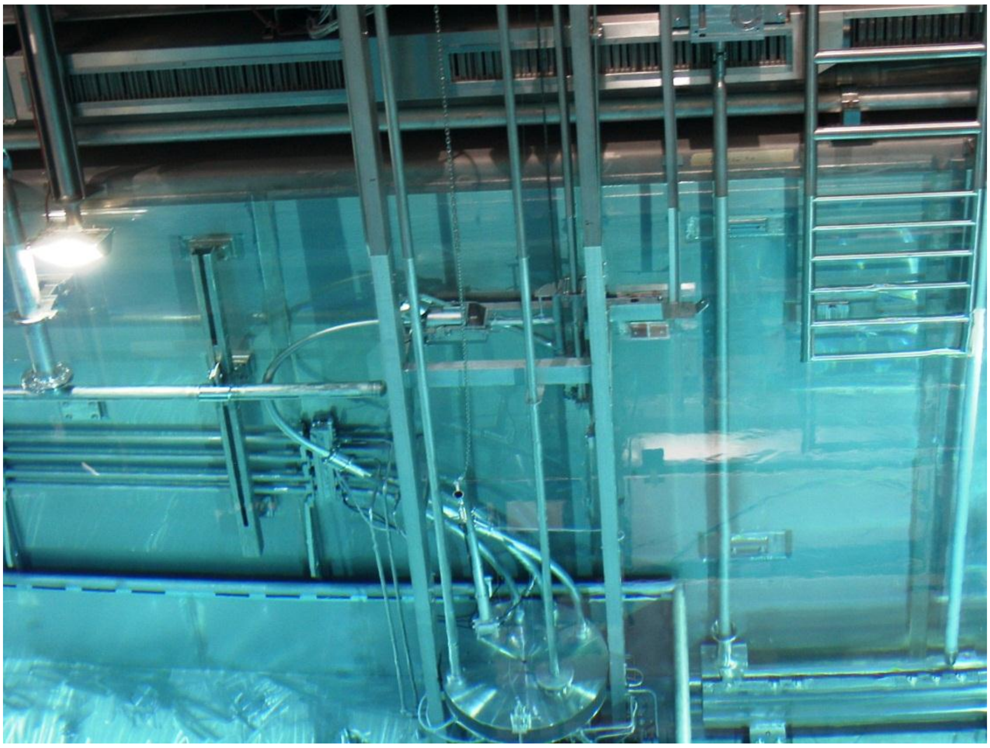

Fig. 2: Capsule irradiation facility of FRM II

transported into a Pb shielded container that waits in the bottom position of a lift 2.5 m below the pool surface. Otherwise it falls back into the “roundabout” for further decay. Optionally the sample can also be transferred into the hot cell of the FRM II.

The main application of the KBA is the production of isotopes for industrial or radiopharmaceutical applications. Major examples are:

- Production of ^{60}Co for industrial purposes up to 185 GBq per batch;
- Production of ^{177}Lu NCA (non carrier added) for the therapy of small, i.e., exhibiting a dimension in the range of few mm, neuro-endocrine tumours. This work is done in cooperation with ITG GmbH, Garching, a company for the production of radioapharmateuticals whose laboratories are located in the immediate vicinity of FRM II on the reactor site. The ^{177}Lu activity is labelled with dotatate and applied to patients in several steps of typically 7.4 GBq each;
- Production of seeds containing ^{125}I for the therapy of prostate cancer. This work is done in cooperation with Eckert & Ziegler IBT Bebig GmbH, Berlin;
- Production of tracers for, e.g., leak testing or the investigation of flow parameters in the chemical industry; and
- Irradiation of high purity Si for trace element detection by neutron activation analysis.

3.3. Silicon doping

The P-doping of Si by means of the neutron transmutation effect (NTD) is based on the nuclear reaction

$$^{30}Si(n,\gamma)^{31}Si \xrightarrow{\beta^-} {}^{31}P \quad (1)$$

The motivation for doping Si in a nuclear reactor is the high homogeneity of the doping profile, which is hardly achievable with other doping techniques. Therefore NTD Silicon is mainly used in power consuming electronic components.

Like for the mechanical irradiation facility presented earlier, the Si doping facility is housed in a vertical, light water filled thimble at a distance of 1000 mm (center to center) from the reactor core The main design requirement for its layout was to allow the simultaneous irradiation of a stack of Si ingots with a diameter of up 200 mm and a total stack length of 500 mm. In addition, in spite of the fact that due to the very limited space and the high demand for experimental installations within the moderator tank FRM II could only be equipped with a single Si doping channel, the doping facility needed to be available also to ingots of lower diameter. This requirement is fulfilled by the use of standard irradiation containers suitable for being loaded with Ø=200 mm ingots. If needed, however, the inner diameter of the containers may be made narrower by ring-shaped Al spacers for the irradiation of smaller ingots. Up to now the demand of the semiconductor industry has concentrated on the processing of ingots with Ø=125 mm and Ø=150 mm, but interest in the irradiation of Ø=200 mm ingots is growing, however.

The basic condition to be met for a successful doping of Si in a research reactor is to provide an irradiation position with a very homogeneous neutron flux density profile. Already during the commissioning phase of FRM II the vertical neutron flux density profile in the irradiation position was determined using a simplified irradiation rig to be manipulated via the crane of the reactor hall. For this purpose two Si ingots (Ø=150 mm) were equipped with a total of 15 AlAu (2‰) monitor wires and irradiated. In order to increase the radial homogeneity of doping, the ingots were rotated during irradiation with a frequency of ≈ 5 turns/min. The result was a maximum inhomogeneity of the flux profile of 12% along the cylinder axis; in contrast the radial inhomogeneity was below 3 %. On the basis of these data the profile of a Ni absorber layer, the so-called liner, for the smoothening of the neutron flux density along the axis of the Si ingots was calculated using MCNP methods. For the final Si doping facility, this absorber layer was embedded into the Al tube surrounding the irradiation position by means of an injection moulding technique. The experimental setup for the flux density measurement and the resulting reduction of the inhomogeneity along the central axis of the Si ingot under irradiation to about 2% are shown in Figure 3.

The absolute thermal neutron flux density in the Si ingot was determined to be $\Phi_{th}=1.7\times10^{13}$ $cm^{-2}s^{-1}$, and the ratio of thermal and fast neutron flux density was $\Phi_{th}/\Phi_f \approx 1700$. A very important and desired consequence of this high ratio is that the irradiation does not produce many extended defect clusters that can hardly be annealed. Therefore the target resistivity for Si to be irradiated in FRM II can be as high as 1075 Ω·cm.

In addition to the use of a Ni liner and the rotation of the Si during irradiation, also the vertical shift of the neutron flux density with increasing burn up of the fuel element has to be anticipated. For this purpose the entire irradiation facility can be lifted by up to 150 mm with respect to the vertical position of the fuel element. On the other hand the change of the form of the neutron flux density during the reactor cycle turned out to be of minor importance.

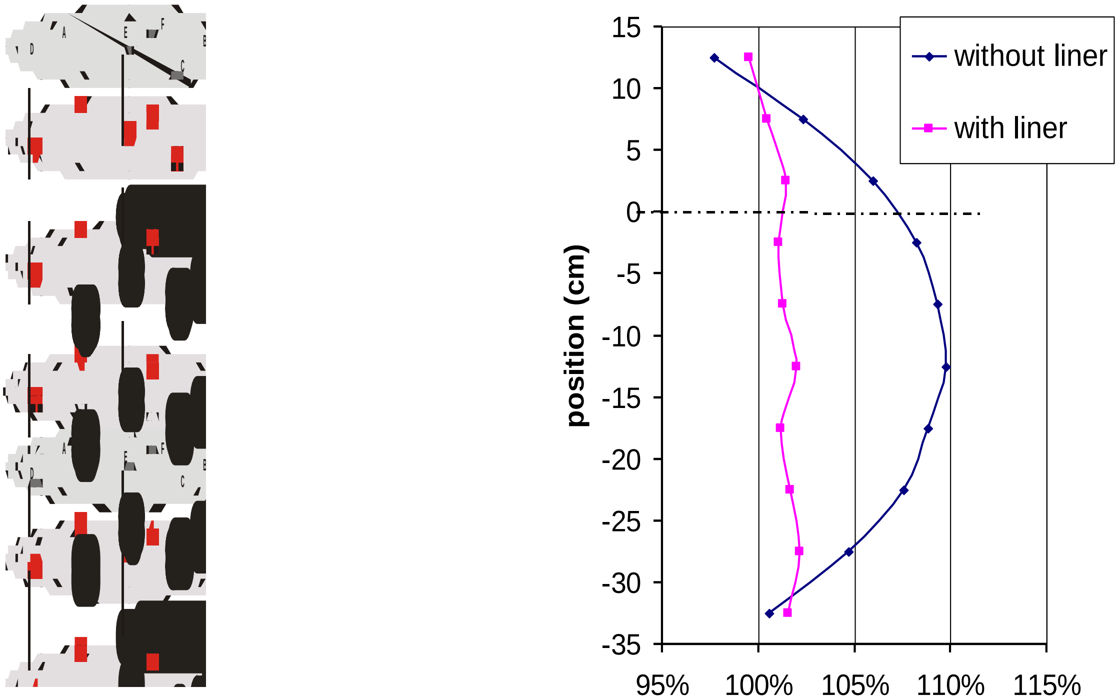

Fig. 3. Si ingot equipped with Al:Au(2‰) neutron flux monitors and measured neutron flux density profile with and without Ni-liner.

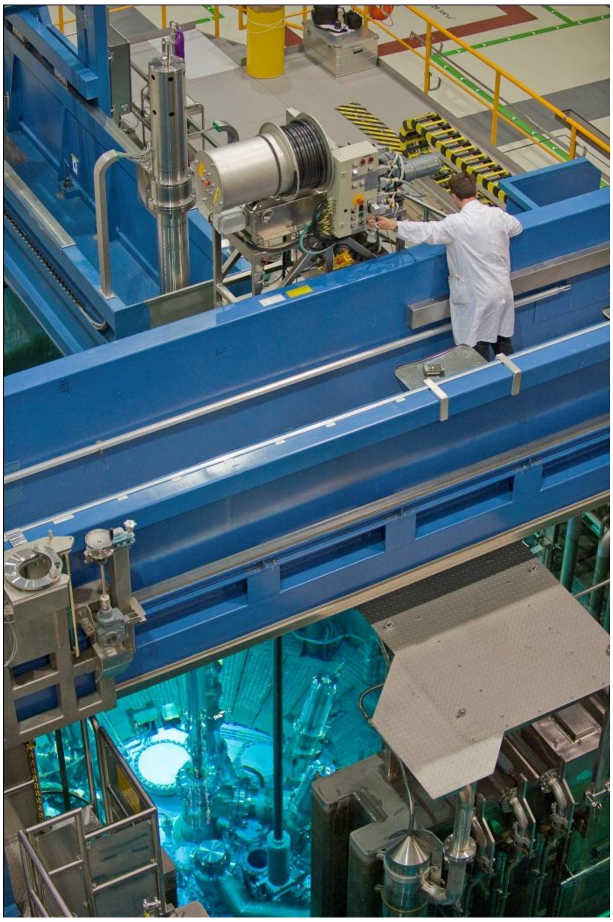

Fig. 4. Si doping facility of FRM II in operation.

The semi-automatic final Si doping facility was taken into operation in January 2007. All major parameters like the transport route of the Si within the reactor pool, the rotation frequency of the Si during irradiation, the irradiation time and many others are controlled by a computer system. After only 48 hours of decay time the Si is unloaded from the pool and can be released from the regulations for radioactive material after cleaning in an ultrasonic bath.

Already in the first year of operation an amount of almost 4 t of Si was irradiated in FRM II for various customers from Europe and Asia. Due to the increasing demand in 2008, a 2-shift working regime was established and the throughput of NTD Si from FRM II increased to approximately 15 t in 2009. More details on NTD at FRM II can be found in [3].

4. CANCER TREATMENT BY IRRADIATION WITH FAST NEUTRONS

Already before the availability of FRM II, considerable experience regarding tumour therapy with fast neutrons was gained at TUM from the treatment of more than 700 patients which had been carried out between 1985 and 2000 at the former FRM reactor. Therefore it was an obvious decision to equip FRM II also with a medical irradiation device using fast neutrons. The patient irradiations at FRM II started in 2007 under the guidance and medical responsibility of Klinikum Rechts der Isar, which is also part of the TUM.

The biological advantage of using fast neutrons (E≈1MeV) for the therapy of malignant tumours is their high radiobiological efficiency as compared to conventional projectiles. Due to their interaction pattern with human tissue, it is most favourably applied to slowly growing and well differentiated tumours mainly of the neck and head. In addition all shallow tumour lesions like skin metastases from various cancer diseases, as well as chest wall metastases of breast cancer, are suited for neutron irradiation therapy, particularly when pre-treated by irradiation projectiles with a low linear energy transfer.

The fast neutrons are generated in the so-called converter facility, the main component of which is a pair of fuel plates containing a total of 498 g ^{235}U. The converter plates are located at the edge of the moderator tank in close vicinity to the entrance window of a neutron beam tube. The fission neutrons from these fuel plates are hardly moderated since they have to penetrate only a few mm of water and some thin Al foils before entering into the beam tube. The beam tube itself contains four shutter drums. It ends at the outer surface of the biological shielding of FRM II in the heavily shielded room for patient treatment. In order to establish the effect of the fast neutrons, the undesired gamma dose rate is decreased by a 35 mm Pb filter. In addition a B_4C filter suppresses thermal and epithermal neutrons. The beam is shaped by a multi-leaf collimator that allows matching the area of neutron radiation to the contour of the tumour up to 20–30 cm² in maximum.

The fast neutron flux density at the irradiation position is 7.1×10^{8} cm^{-2}s^{-1}; and the mean neutron energy is 1.6 MeV. The dose distribution along the depth of a water phantom was determined experimentally in order to provide the basis for working out medical treatment plans. With the combination of filters for therapeutic irradiations, 10 mm B_4C and 35 mm Pb, the ratio of neutron:photon decreases from 3.6 at the surface of the water phantom to 1.8 at a depth of 50 mm. The drastic decrease of the dose rate with increasing depth is the reason that only superficial tumours are irradiated with fast neutrons.

The converter facility and its applications are described in more detail in [4].

5. IRRADIATION FACILITY FOR THE PRODUCTION OF ^{99}MO (PROJECT)

Already before the recent crisis regarding the worldwide production of fission product ^{99}Mo, the most widely used radioisotope in nuclear medicine, TUM in cooperation with the Belgian company Institut des Radioéléments (IRE) established the suitability of FRM II to considerably contribute to the future supply of the radiopharmaceutical industry with this important isotope in a technically and economically reasonable way. A feasibility study based on the irradiation of IRE’s tubular targets containing about 4 g of highly enriched uranium was successfully completed in 2009.

In a first step a moderator tank insert, the so-called HFRP thimble, which is presently not taken by other experimental installations, was identified to be the most promising irradiation channel for the purpose under consideration. It is recognized to be suitable to house a vertical thimble foreseen to be made from zircaloy, which itself will contain three independent U-shaped irradiation loops (U-tubes) for the simultaneous irradiation of up to 15 targets. By means of MCNP calculations the mean thermal neutron flux density in the target positions was predicted to be close to 2×10^{14} $cm^{-2}s^{-1}$ resulting in a power production from the targets up to 430 kW. In a typical irradiation cycle of 6 days the produced ^{99}Mo activity is expected to add up to 17 kCi (6.3×10^{14} Bq) upon removal from the irradiation position.

The cooling circuit of the ^{99}Mo production device was designed to be independent from other reactor installations to the highest extent possible. It will be equipped with a total of four identical pumps. In standard operation, three pumps will be running. Only two of them, however, will be able to provide the necessary throughput for the cooling of the target. The fourth pump is a spare which will be taken into operation automatically only in case one of the others fails. The cooling water is taken from the FRM II reactor pool. Two more main components of the cooling circuit are the heat exchangers located within the primary cell of FRM II, which already houses the main components of the cooling circuit of the reactor itself. One of them will be used to transfer the energy from the cooling water partially into the heat supply for the warm water surface layer of the reactor pool, whereas the other will feed the balance of the heat into the secondary circuit of FRM II. A preliminary thermohydraulic layout of the irradiation device was carried out and shows that the maximum pressure in the cooling circuit will be below 14.5 bars. Consequently a pressure stage of 16 bars for the components in use will be conservative.

Regarding the in-house handling of the targets, a general course of necessary operational steps has been fixed. The resulting time table guarantees that irradiated uranium targets can be provided to a company dealing with the extraction of ^{99}Mo within 36 h after the end of the irradiation at the very latest. In addition major technical challenges like the loading and unloading of targets into and out of the irradiation position and the loading and dispatch of the transport cask have been designed sufficiently detailed to demonstrate their general feasibility.

The possible interference of the ^{99}Mo production device with other experimental installations within the moderator tank of FRM II has been looked at and proven to be acceptable. In addition the effect of main incidents, in particular the drop of a loaded stack of 5 targets and the blockade of a cooling pump have been investigated and demonstrated to be controllable.

The feasibility study is complete via a consideration of licensing issues, a preliminary time schedule of the project and a cost estimation. Based on the results of the study the decision to build the irradiation facility for ^{99}Mo production was taken. Already in the 4th quarter of 2010 the zircaloy thimble of the future irradiation facility and its He supply will be installed, taking advantage of the rare opportunity to have access to the moderator tank when it is drained of heavy water. Consequently the future installations of the irradiation rig itself and the cooling components can be done under standard maintenance conditions of FRM II.

According to schedule the production of ^{99}Mo is foreseen to begin in early 2014.

6. COMMERCIAL APPLICATIONS OF BEAM TUBE EXPERIMENTS

The design of FRM II has clearly been optimized for neutron beam tube research. Although the huge majority of these experiments deal with problems in basic research, various technical questions are successfully tackled by means of neutron scattering techniques at FRM II.

An important example is the neutron tomography setup ANTARES operated under the responsibility of B. Schillinger. It is based on the fact that neutrons are strongly scattered by hydrogen bearing materials. By consequence neutron tomography yields a contrast not available in conventional X ray computer tomography, in particular for the investigation of biological or geological materials.

Further examples of neutron scattering experiments of industrial interest are the non-destructive measurement of stress in seriously charged components used in machines and vehicles or prompt gamma activation analysis.

For more details regarding the options of benefits of neutron scattering in technical questions I refer you to the FRM II homepage, www.frm2.tum.de, where contact with experts can be established.

7. SUMMARY

Although the Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II) is a university owned research reactor dedicated to basic science, it is able and willing to offer technical and scientific service to partners dealing with the technical application of scientific results. It has to be noted that the analytical and production techniques using neutrons as described above were recognized and adopted already shortly after commissioning of FRM II by many partners from various branches in industry. It is one of the important tasks of reactor management to cultivate and extend this partnership with all parties interested in the use of neutrons for applied science for their mutual benefit.

8. REFERENCES

[1] GALLAGHER, K., BROWN, R., JOHNSON C., Fission track analysis and its applications to geological problems, Annu. Rev. Earth Planet Sci. **26** (1998), 519–572.

[2] LIN, X., HENKELMANN, R., TÜRLER, A., GERSTENBERG, H., DE CORTE, F., Neutron flux parameters at irradiation positions in the new research reactor FRM II, Nucl. Instr. & Methods in Physics Research A **564** (2006), 641–644.

[3] GERSTENBERG, H., LI, X., NEUHAUS, I., “Silicon Doping at FRM II”, Trans. 13th Topical Meeting on Research Reactor Fuel Management (RRFM), Vienna, 2009, European Nuclear Society, Brussels (2009) 282–286.

[4] WAGNER, F.M., LOEPER-KABASAKAL B., BREITKRUETZ, H., PETRY, W., BÜCHERL, T., "Use of Fission Radiation in Life Sciences and Materials Characterization", Trans. 13th Topical Meeting on Research Reactor Fuel Management (RRFM), Vienna, 2009, European Nuclear Society, Brussels (2009), 242–246.