COMMERCIAL APPLICATIONS AT FRM II BASED ON NEUTRON IRRADIATIONS

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ABSTRACT

Due to its design as a heavy water moderated reactor with a very compact core FRM II, Germany's most modern and most powerful research reactor, offers excellent conditions for basic research using beam tubes. On the other hand it is equipped with various irradiation facilities to be used mainly for industrial purposes. From the very beginning of reactor operation a dedicated department had been implemented in order to provide a neutron irradiation service to interested parties on a commercial basis.

As of today the most widely used application is Si doping. The semiautomatic doping facility accepts ingots with diameters between 125 mm and 200 mm and a maximum height of 500 mm. The irradiation channel is located deep in the heavy water tank and exhibits a ratio of thermal/fast neutron flux density of > 1000. This value allows the doping of Si to a target resistivity as high as 1100 Ωcm within the tight limits regarding accuracy and homogeneity specified by the customer. Typically the throughput of Si doped in FRM II sums up to about 15 t/year.

Another topic of growing importance is the use of FRM II aiming the production of radioisotopes mainly for the radiopharmaceutical industry. The maybe most challenging example is the production of Lu-177 n.c.a. based on the irradiation of Yb_2O_3 to a high fluence of thermal neutrons of typically 1.5E20 cm^{-2}. The Lu-177 activity delivered to the customer is in the range of 750 GBq. With respect to further processing it turned out to be a highly advantageous to have the laboratories of ITG, the company extracting the Lu-177 from the freshly irradiated Yb_2O_3 on site FRM II.

Further irradiation facilities are available at FRM II in order to allow the activation of samples for analytical purposes or to irradiate samples for geochronological investigations using the fission track technique. Finally a project on the future installation of a facility dedicated to the irradiation of U-targets for the production of Mo-99 is in progress.

It is noteworthy that all of the irradiation facilities at FRM II have been certified according to the ISO 9001:2008 standard.

1. Introduction

Since nearly 10 years the Technische Universität München is operating the FRM II, Germany's most modern research reactor on its campus in Garching about 20 km north of Munich. Doubtlessly the mission of FRM II is to provide intense neutron beams for basic research and to offer excellent working conditions for a national and international community of scientific users. Nonetheless, it was generally agreed since the very early days of the project that FRM II will be made available to interested parties from the industry on a commercial basis. It turned out that many of those activities are based on neutron irradiation. Consequently a dedicated department was established right from the start of rector operation. As of today approximately 10 employees are operating the irradiation facilities of FRM II in 2 shifts during the working week. They are carrying out approximately 1000 irradiations per year mainly for the purpose of Si-doping, isotope production, activation of samples for analytical purposes but also for further topics.

2. Characteristics of the research reactor FRM II

FRM II is a heavy water moderated, light water cooled research reactor exhibiting a thermal power of 20 MW. The moderator tank is filled with roundabout 11 m³ of heavy water and is located within a 700 m³ light water pool. The reactor's main design feature is a single cylindrical fuel assembly, the so-called compact core, containing approximately 8.1 kg of highly enriched (92% U-235) uranium in form of U_3Si_2. Under standard conditions the reactor is operated in cycles of 60 days in a row. Typically the numbers of cycles per year is 4.

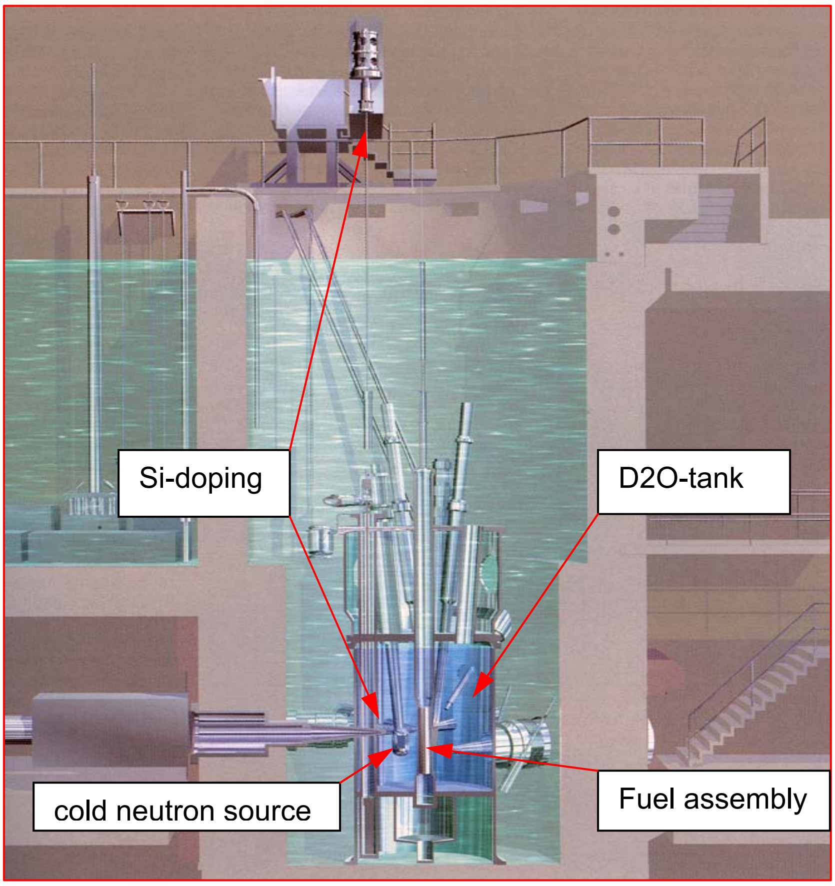


Fig 1: FRM II reactor pool and moderator tank (dark blue) with some key components

For scientific and commercial applications FRM II is equipped with 11 beam tubes allowing the operation of approximately 30 neutron scattering instruments, a cold neutron source (liquid D_2 at 24 K), a hot neutron source (irradiation heated carbon block at approximately 1900 °C), a high intensity positron source, a facility for cancer treatment by means of direct irradiation with fast neutrons and 5 irradiation facilities - one of them for gamma irradiations in spent fuel assemblies. Except for the gamma irradiation device all of the irradiation channels are located within the heavy water moderator tank and consequently provide high values of the thermal/fast neutron flux density ratio Φ_{th}/Φ_f between 300 and more than 10000.

3. Commercial applications

3.1. Silicon doping

The P doping of Si in a nuclear reactor is based on the neutron capture reaction

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

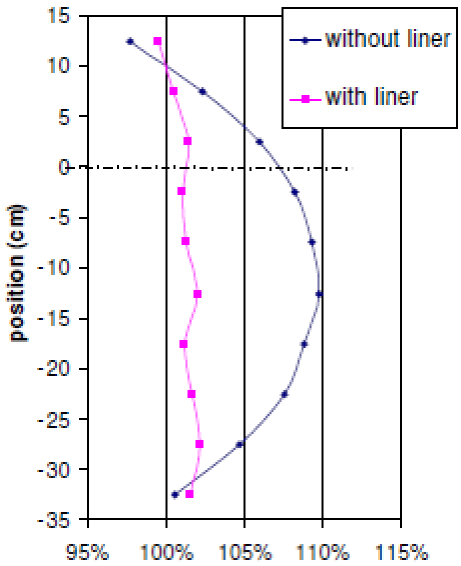
It is important to note that this doping technique takes advantage from the fact that practically no other activation reaction takes place during the irradiation of Si by thermal neutrons, that the half-life of the radioactive ^{31}Si is only 2.62 h and that extremely pure Si is available as starting material. As compared to non nuclear doping techniques neutron transmutation doping (NTD) is able to provide very accurately the required amount of dopant atoms – usually measured in terms of electrical resistivity – and a very homogeneous doping profile over the entire area of the wafer. Consequently NTD Si is mainly used for components in high power electronics. A detailed overview about Si doping can be found in [1].

Already during the design phase of FRM II it was decided that the reactor needed to be equipped with a Si doping facility suitable for the irradiation of ingots up to a maximum diameter of 200 mm and a maximum stack height of 500 mm. Due to the fact that only one irradiation channel is available for Si doping ring-shaped Al spacers are introduced into the irradiation basket in order to allow the doping of ingots with smaller diameter. Since the demand for NTD Si is concentrating more and more on weakly doped (high resistivity) material a vertical thimble in a distance of 1 m (centre to centre) from the fuel assembly was chosen to house the facility. The irradiation channel is a light water filled thimble located deep within the heavy water moderator tank. It allows easy access from the top and exhibits a thermal neutron flux density of 1.7E13 $cm^{-2}s^{-1}$ combined with a ratio $\Phi_{th}/\Phi_f \approx 1700$. In order to provide the required axial homogeneity of the doping profile the outer Al tube of the irradiation insert was equipped with a suitably shaped Ni layer – the so called liner - which due to the higher neutron absorption cross section of Ni as compared to Al smoothens the neutron flux density along the vertical axis of the ingots. The detailed shape of the Ni liner was determined by means of MCNP calculations [2] and verified by the irradiation of Si ingots which had been equipped with plenty of Al:Au (2‰) neutron flux monitors. The effect of the Ni liner is to reduce the axial inhomogeneity of the neutron flux density to less than 3 % (see fig. 2). In addition, of course, the irradiation basket containing the Si ingots is rotated upon irradiation around its central axis by 5-7 turns/min in order to limit the radial inhomogeneity of the doping process and finally the entire irradiation channel is allowed to be moved vertically by up to 150 mm in order to follow the burn-up based shift of the neutron flux density distribution during the 60 days reactor cycle.

Since 2007 the semiautomatic Si doping facility at FRM II which had been developed in cooperation between the Technische Universität München and the Hans Wälischmiller GmbH, Markdorf, Germany, is operated routinely in 2 shifts on week days. Due to its extremely well thermalized neutron spectrum it is particularly suited for the production of high resistivity NTD Si. The corresponding short irradiation times allow a throughput of approximately 15 tons/year. Fixed contracts with leading Si producers from Europe and Asia guarantee the operation of the Si doping facility at least for the medium term future.

Like all of the irradiation facilities at FRM II the Si doping facility is certified according to the ISO 9001:2008 standard.

A more detailed description of the Si doping facility has been published in [3].



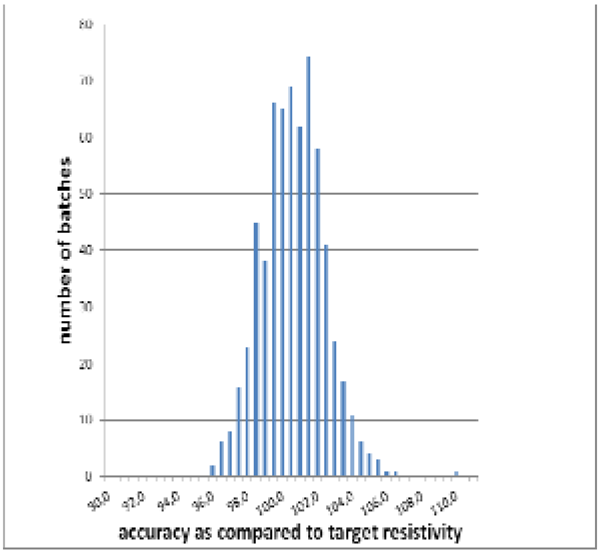


Fig.: 2 Relative neutron flux density along the vertical axis of a Si ingot and accuracy of the measured resistivity as compared to the target resistivity (based on the results of a major customer in 2012)

3.2 Isotope production

At FRM II the production of isotopes for industrial and radiopharmaceutical purposes is mainly done using the so called capsule irradiation facility. Easily spoken this facility is a water driven rabbit system suitable for the exposure of several grams of sample material to a considerable thermal neutron flux density of >1E14 $cm^{-2}s^{-1}$ for irradiation times of days to weeks. The facility provides 2 independent irradiation channels. Each of them is able to accept a stack of up to 3 capsules at the same time. The neutron flux density in the outer positions, however, falls considerably below 1E14 $cm^{-2}s^{-1}$.

In preparation for the irradiation the source material is sealed in quartz ampoules and/or cans made from pure Al (Ø = 26 mm; h = 70 mm). These containers serve as the inner barrier of the sample against contamination by reactor pool water. Their water tightness is proven in every single case experimentally before the sample is released for irradiation. Finally the packaged sample is inserted into a standard irradiation capsule made from high purity (5N) AlMg3. Due to their high purity these standard containers are reusable for up to 5-10 irradiations. Besides the irradiation facility itself FRM II provides the necessary infrastructure like storage positions and a hot cell for unloading the freshly irradiated samples from the co-irradiated capsules and their packaging according to the transportation regulations for dangerous goods.

Besides the production of Co-60 for industrial purposes as of today the highest demand for radioisotopes at FRM II is based on Lu-177, an important isotope for medical purposes. It is produced non carrier added (n.c.a.) in cooperation with Isotopes Technologies Garching (ITG) GmbH, a company whose production facilities are located on site FRM II.

The starting material for the production of Lu-177 n.c.a, is Yb-176 oxide powder. The route to Lu-177 is as follows:

$$^{176}Yb(n,\gamma)^{177}Yb \xrightarrow{\beta^-} {}^{177}Lu$$

Typically about 1 g of Yb_2O_3 is irradiated in a sealed quartz ampoule. In order to increase not only the overall output activity but also the specific activity of the product highly (>99.5%) enriched Yb-176 is used as starting material. Due to the production path via the short lived isotope Yb-176 ($T_{1/2}$ = 1.9 h) and the subsequent chemical isolation the product Lu-177 is produced non carrier added (n.c.a.), i.e it is not contained in a matrix of inactive Lu-176 after separation from the target material. In addition the undesired metastable Lu-177m ($T_{1/2}$ = 160 d) level is not populated using this production route. In the standard procedure the Yb-oxide sample is irradiated for 2 weeks in KBA in order to provide the required Lu-177 activity of about 750 GBq. In more detail the production process can be found in [4]

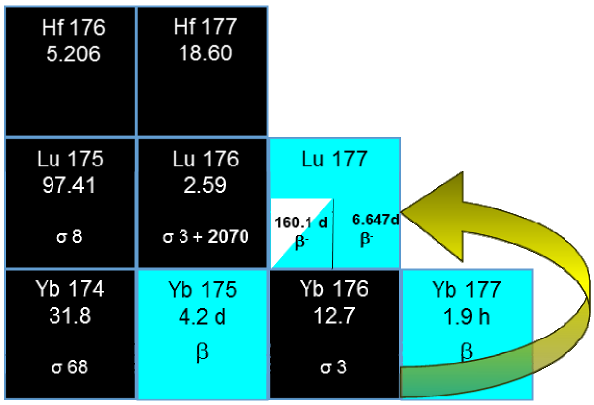


Fig.: 3: Excerpt from the chart of nuclides illustrating the production of Lu-177 n.c.a.

3.3 Future prospects

Further commercial applications are under consideration at FRM II. In particular a project team on its own is developing an irradiation facility aiming the production of Mo-99 as a fission product using the irradiation of targets from low enriched uranium (< 20% U-235). The realization of this project, however, will require an extension of the reactor’s operational license.

In contrast, the production of Ho-166 in the form of microspheres using the pneumatic rabbit system underwent already test irradiations and is hoped to enter into the routine production in the very short term future.

4. References

[1] IAEA-TECDOC-1681, International Atomic Energy Agency, Vienna, 2012

[2] A. Röhrmoser: Internal Documentation

[3] H. Gerstenberg, X. Li, I. Neuhaus: RRFM 2009, Vienna, March 22-25, 2009, Transactions, pp 50-54

[4] H. Gerstenberg, A. Draack, R. Henkelmann, M. Harfensteller: RRFM 2013, St. Petersburg, April 21-25, 2013; Transactions, pp 63-68