STATUS AND SCIENTIFIC USE OF THE TRIGA RESEARCH REACTOR AT THE UNIVERSITY OF MAINZ

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ABSTRACT

The research reactor TRIGA Mainz (FR MZ) is situated at the Johannes Gutenberg-University in Mainz, Germany. This TRIGA mark II reactor is one of the last three operating research reactors with a thermal power above 50 W in Germany. The FR MZ provides maximum thermal power of 100 kW in steady state operation mode and up to 250 MW pulse peak power at a pulse width of about 25 ms in pulsed operation mode. By now, it is under operation for 52 years and represents about 49.000 operating hours and 22.700 pulses in total.

The reactor is utilized for both education of students and maintenance of nuclear technology competence as well as a neutron source for a wide scientific spectrum.

In this context, the FR MZ is a participant of the Cluster of Excellence Precision Physics, Fundamental Interactions and Structure of Matter (PRISMA) [1]. One of the main goals of PRISMA is a high precision measurement of the neutron's lifetime, an important natural constant that is needed for some fundamental physical theories. Therefore, two ultra-cold neutron (UCN) sources have been built up at the FR MZ. These sources are used for various experiments. In the last two years, the upgrade of one of the two operating sources at the FR MZ has been completed successfully.

1. Introduction

The Institute of Nuclear Chemistry, as a part of the Johannes Gutenberg-University in Mainz, pursues a TRIGA mark II research reactor called FR MZ. It is, together with the reactor BER II of the Helmholtz Zentrum Berlin and the FRM 2 of the Technical University of Munich (TUM), one of the last research reactors in Germany with a thermal power above 50 W. In the past, Germany used to be a research site with several research reactors, which most of all are shut down or decommissioned by now [2].
The FR MZ looks back at an outstanding history of more than 50 years of operation. The former Federal Ministry for Nuclear Affairs under the leadership of Prof. Siegfried Balke promoted the installation of a research reactor at the university in Mainz. It was only when General Atomic introduced the TRIGA research reactors that a reactor located in the middle of the campus could be realised. Prof. Fritz Straßmann led the installation of the FR MZ at the university. He also invited his former research fellow and Nobel laureate Prof. Otto Hahn for the inauguration on 03.04.1967, who executed the first official reactor pulse.
Even though the FR MZ yields a relatively weak thermal power of 100 kW, it still provides not at least because of the pulsed mode manifold application possibilities. Some of these shall be exemplary presented in this paper.

2. Layout

2.1 Reactor design and irradiation facilities

The FR MZ is a standard TRIGA reactor type mark II, constructed and distributed by General Atomic. The reactor is controlled and operated by three control rods: a shim rod, an optional automated regulation rod and a pressurized air driven pulse rod. With the shim rod and the regulation rod, the reactor can be stabilized at any power level from 10 mW up to 100 kW thermal power. For a pulse, the pulse rod is expelled from the core with the help of about 5 bar pressurized air (see section 3).

For the scientific use, the reactor has several experimental facilities available. Their neutron fluxes are summarized in Table 1 [3].

Table 1: Thermal and epithermal neutron fluxes at the different irradiation positions of the FR MZ at a thermal power of 100 kW

Irradiation Position	Thermal flux 1) [$cm^{-2}s^{-1}$]	Epithermal flux 2) [$cm^{-2}s^{-1}$]
Rotary specimen rack	$0.7 \cdot 10^{12}$	$4 \cdot 10^{10}$
Rabbit systems	$1.6 \cdot 10^{12} - 1.8 \cdot 10^{12}$	$4.6 \cdot 10^{10} - 5.6 \cdot 10^{10}$
Beam tubes	$0.1 \cdot 10^{12} - 0.5 \cdot 10^{12}$	$7.6 \cdot 10^{8} - 1.6 \cdot 10^{10}$
Central thimble	$4.2 \cdot 10^{12}$	$1.4 \cdot 10^{11}$
Thermal column (hot end) 3)	$3.1 \cdot 10^{10}$	$2.1 \cdot 10^{8}$
Thermal column (cold end) 3)	$2.7 \cdot 10^{7}$	$6.8 \cdot 10^{2}$

1) $E_n \leq 0.4$ eV 2) $E_n \geq 0.4$ eV 3) Central irradiation channel

The FR MZ provides three pneumatic transfer systems ("rabbit" systems) as schematically shown in Fig. 1. These end in the outer periphery of the core and start in the laboratories of the institute. The sample can be transported from experimental places to the core and back within seconds. Thus, the rabbit systems are usually used for the production of short-lived nuclides.

A frequently used irradiation facility is the rotary specimen rack that is located around the reactor's core and embedded inside the graphite reflector. It can receive up to 80 sample capsules in 40 positions. The rotary specimen rack is used for long-term irradiations, the production of radioactive isotopes and Neutron Activation Analysis (NAA). Unlike the rabbit systems, the samples for the rotary specimen rack must be inserted by hand. Another irradiation facility with manual sample loading is the central thimble. It can receive only one sample and is located in the centre of the core. For this it provides the highest thermal neutron flux of about $4{,}2 \cdot 10^{12} cm^{-2} s^{-1}$.

The thermal column, a further irradiation unit, is a large volume filled with graphite bars and sealed with a moveable concrete door. It is located in the biological shield of the reactor (see Fig. 2). It can be opened only if the reactor was not operating for about 30 minutes. This is necessary concerning the high radiation level. The graphite blocks behind the concrete door are 20 cm x 20 cm x 120 cm in size and can be removed so that big samples can be irradiated. The thermal column can also be used for online detector measurements, guiding the required cabling through a small aperture inside the concrete gate.

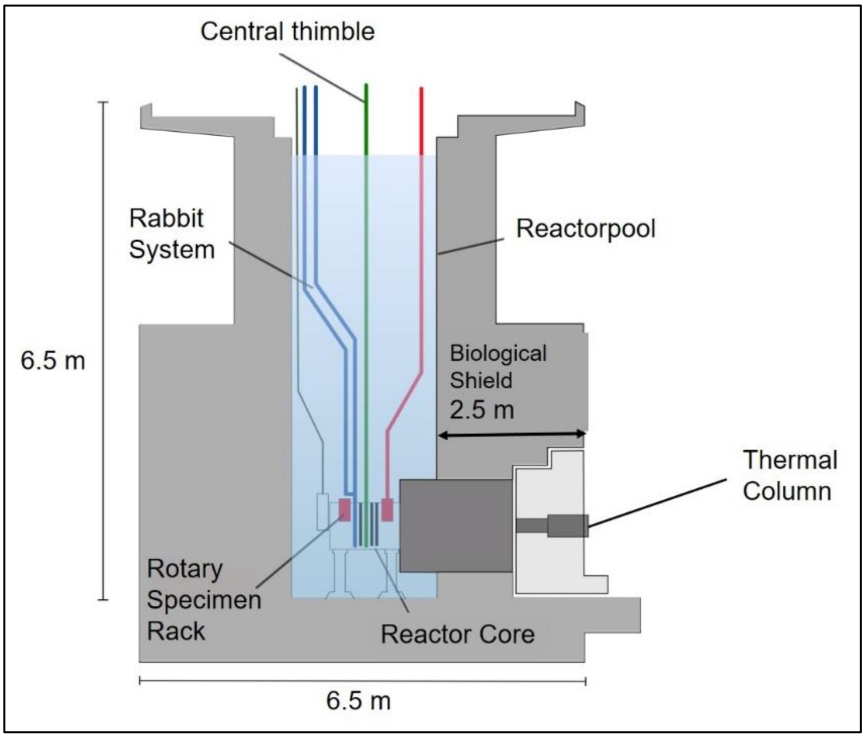

Fig. 1: Simplified drawing showing the longitudinal section of the reactor. The experimental stations are shown in this picture: in green the central thimble, in red the rotary specimen rack and in blue the pneumatically operated "rabbit" systems.

Furthermore, the FR MZ is equipped with four beam tubes that are for the use as caves for experiments close to the reactor core. All four beam tubes penetrate the biological shield and have the same diameter of 150 mm, but their orientation to the core varies. The beam tubes A and B have a radial configuration and end both in front of the reflector (see Fig. 2**Error! Reference source not found.**). Beam tube C is pointing tangentially to the reactor core. This configuration reduces the gamma flux compared to the other beam tubes. Beam tube D has a radial orientation as the beam tubes A and B, but it pierces the graphite reflector and ends close to the core, resulting in a high thermal neutron flux (see Table 1).

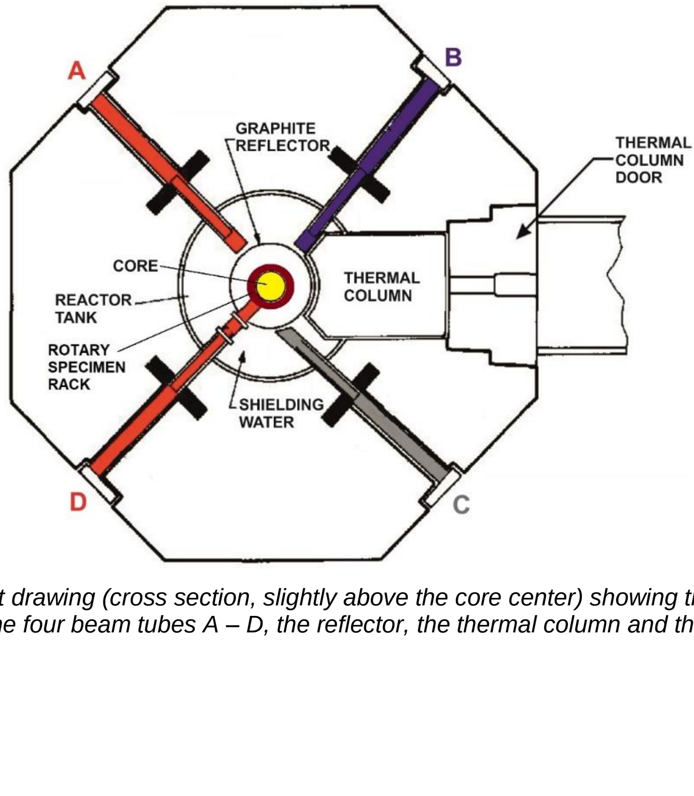

Fig. 2: Simplified cut drawing (cross section, slightly above the core center) showing the rotary specimen rack, the four beam tubes A – D, the reflector, the thermal column and the core

2.3 Fuel

The TRIGA reactor employs special fuel-moderator elements. They contain uranium, enriched in U-235 (< 20%, LEU fuel). The uranium is embedded in a ZrH matrix and contributes to only 8% of weight of the fuel elements. Currently the FR MZ is equipped with 76 fuel elements. The U-235 burnup is about 4 g per year resulting in about 200 g burnup over the last five decades. Thus, the FR MZ actually has a life-time core. However, in order to overcome the slow decrease of the reactivity over time, it is necessary to replace a graphite placeholders in the core with fresh fuel elements every three to five years.

The particular about these fuel-moderator elements is the behaviour when the fuel temperature increases. The hydrogen in the ZrH lattice has the ability to moderate the fast neutrons down to an energy about 0,14 eV. With rising fuel temperature, the moderating ability decreases and therefore the ability of transferring energy from the matrix to the neutrons increases. Therefore, the energy spectrum of the neutrons shifts to a higher average energy. Thus, the fission chain collapse and the reactor power declines rapidly within a few thousandths of a second, faster than any engineered device can operate [4, 5]. The described effect is the reason for the prompt negative temperature coefficient of the reactor and the characteristic that allows to operate the reactor in pulsed mode.

3. Operation modes

The reactor operates in two different modes. On the one hand there is the steady state operation mode and on the other hand the pulse mode. In steady state mode, the FR MZ is operated for minutes up to hours between 10 mW and 100 kW thermal power. A predominant part of irradiations takes place at 100 kW thermal power.

Additionally, the reactor can be operated in pulse mode. In order to pulse, the reactor power needs to be stabilized at 50 W thermal power. Once the reactor reaches this power level, the pneumatically driven transient rod (pulse rod) is expelled from the core with pressurised air within in a few 10 ms.

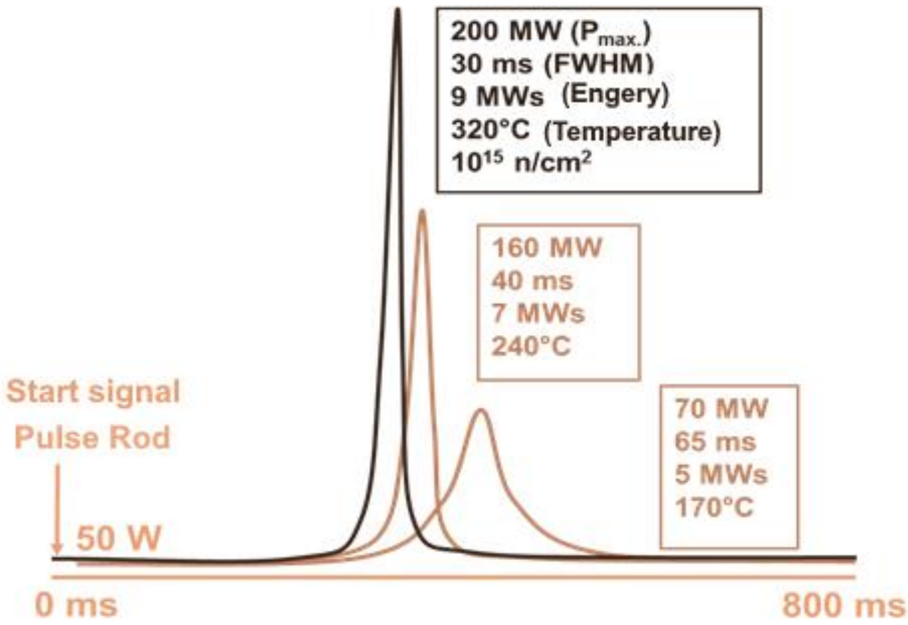

Fig. 3: Time structure of a 2 $, 1.75 $ and 1.5 $ neutron pulse at the FR MZ with exemplary values for the maximum thermal power, the full width half maximum (FWHM), the pulse energy and the fuel temperature.

The reactor reaches prompt criticality and the fuel elements start to warm up. After a few 10 ms to 100 ms the negative temperature coefficient of the fuel element ZrH matrix takes effect and the moderation capability decreases rapidly. The chain reaction collapses and the reactor's power level decreases immediately. In this operation mode, the peak reactor power is up to 250 MW, depending on the excess reactivity that is contributed in the core. The excess reactivity is typically between 1.25 $ minimum up to 2.0 $ maximum and is controlled by adjusting the expelling distance of the pulse rod. Fig. 4 presents the time structure of different pulses with an excess reactivity of 2.0 $, 1.75 $ and 1.5 $.

4. Utilisation

The embedment of the FR MZ into a nuclear chemistry institute and its inherent safety of a TRIGA type reactor allows for a wide range of applications. The examples in this chapter can only give an incomplete overlook of the applications and experiments that benefit from the FR MZ.

4.1 Utilisation Capacities

The FR MZ is available for experiments and teaching for about 200 days per year. The participation at the PRISMA cluster of excellence raised the request for ultra-cold neutrons to evaluate the free neutron’s lifetime (see Section 4.3). To fulfil these requirements, two new operators and a deputy radiation protection officer strengthened the reactors staff, in order to introduce a regular two-shift operation at the FR MZ. During the two-shift operation, which is provided for 12 weeks per year, the reactor is available for experiments for 16 hours on

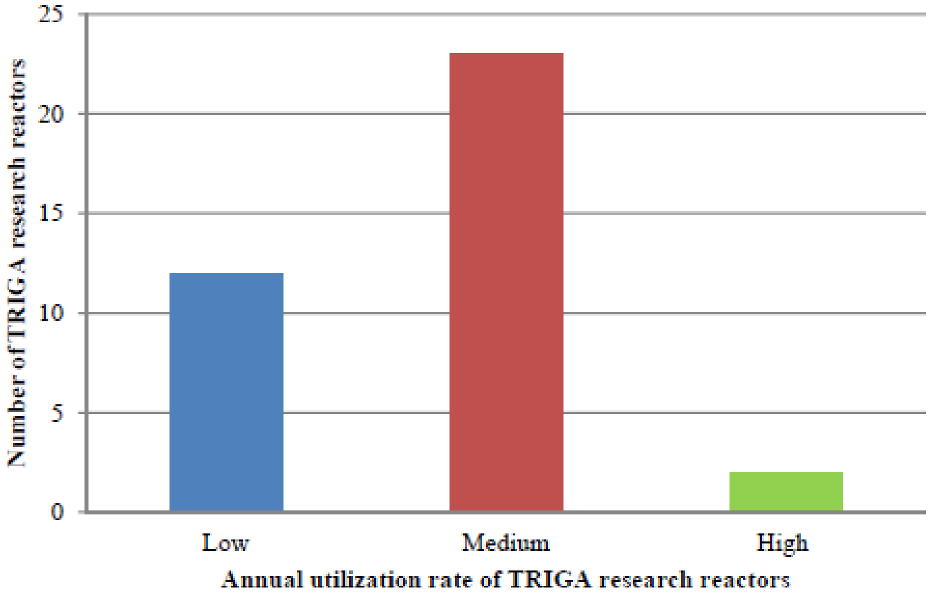

Fig. 4: Evaluation of the annual utilization rate of TRIGA research reactors around the world. The reactors have been categorized in three levels: low utilization rate with less than 4 effective weeks in operation (blue bar); medium utilization rate with more than 4 effective weeks but less than 20 weeks (red); high utilization rate with more than 20 effective weeks per year (green). Picture taken from [6]

Mondays to Thursdays (and 10 hours on Fridays) instead of 6 hours on ordinary beam times. Due to intensive requests of the FR MZ operation by internal and university-external users, the reactor is often in parallel utilisation for two or even up to three applications at the same time. According to the statistics of the IAEA, the FR MZ is one of two TRIGA research reactors in the world with more than 20 effective weeks of utilization (see Fig. 4) [6].

4.2 Applied Science

Neutron Activation Analysis (NAA) and tracer production for various applications in research and industry are high-frequented applications of the FR MZ. NAA is a versatile method for various analytical problems due to its simplicity, multi-element capacity and sensitivity. Nowadays the most used kind of NAA at the FR MZ is the instrumental neutron activation analysis (INAA). It is performed without any chemical separation steps. Nevertheless, there are laboratories available in the institute where any chemical procedures either prior to or after the neutron irradiations can be performed (RNAA). Delayed neutron activation analysis (DNAA) is a special version of NAA and uses the counting of beta-delayed neutrons emitted from very neutron-rich fission products as obtained by the irradiation of fissile material.
First highlight in the history of the FR MZ, in collaboration with the Max-Planck-Institute of Chemistry in Mainz, was the INAA of lunar rock samples from the Apollo 11 mission only a few months after the landing on the moon [7]. More than 45 years later, there are still interesting questions that are examined with this method.

Another field of applied science at the FR MZ is the simulation of accelerated aging for detector and electronic materials under the influence of neutron irradiation. It has been shown, that a probe inside the rabbit system is irradiated by an equivalent 1 MeV neutron flux of 5.15 (15) (75) $cm^{-2}s^{-1}$ [8]. This makes the FR MZ a perfect test facility to test the effect of long-term radiation on, e.g. detectors or electronics, provided to be used in highly radiative areas such as the ATLAS-detector at CERN. In these cases, the materials cannot easily be replaced in case of damages, due to the high dose rate in the experiment's environment. Therefore, it is necessary to know that the chosen material can resist long-term irradiation.

4.3 Fundamental Research

The FR MZ provides the special ability to produce neutron pulses which can be used for the research of fundamental questions in physics and chemistry. For an overview, three experiments shall be presented here.

4.3.1 The TRIGA-SPEC collaboration

The TRIGA-SPEC collaboration consists of a common beamline and the two experiments TRIGA-TRAP and TRIGA-LASER that are the prototypes for the future FAIR experiments MATS and LaSpec [9]. The common beamline parts are a surface ionization source that is connected to beam port B, a mass separator magnet and a radiofrequency quadrupole cooler and buncher (RFQCB). The ion source ionizes neutron-rich atoms produced via fission of a uranium or trans uranium target placed inside the reactor beam tube and an isotope-selective ion beam is focussed into the RFQCB. There, ions are stored, cooled and finally sent to the experiments.

The Penning-trap mass spectrometer experiment TRIGA-TRAP aims to measure the mass and thereby the binding energy of short-lived nuclei with a relative precision of about $5 \cdot 10^{-9}$ with the help of the time-of-flight ion-cyclotron-resonance detection technique **[10, 11]** .

The determination of nuclear charge radii, nuclear moments and spins can be performed using the collinear laser spectroscopy experiment TRIGA-LASER. It was part of TRIGA-SPEC until 2016 when it moved to Chicago to keep on measuring nuclear properties of exotic nuclei. Finally, it will be installed at the FAIR facility.

4.3.2 Chemistry of the heaviest elements

The heaviest elements known in the periodic table are the trans-actinide elements or so-called super-heavy elements (SHE, $Z > 103$). SHE can only be produced at ion accelerator facilities. The production rate of the SHE is extremely low and varies between few atoms per hour, down to only one or two atoms per month. Due to the low production rates and the short half-lives, there are special requirements for a SHE chemistry experiment, since one performs chemical separations on a one-atom-at-a-time basis. Thus, separations must be performed as fast as possible, fully automated and the decay of the SHE must be detected with high efficiency [12].

The FR MZ gives a unique possibility to develop and test such chemistry set-ups. Short-lived lighter homologues of the SHE can be produced in the neutron induced fission of actinides and used for experiments. Systems to study the chemical properties of single atoms by means of ion-exchange chromatography, liquid-liquid-extraction and electro-deposition on various metals have been developed in Mainz [12-14]

4.3.3 Experiments with ultra-cold neutrons

A large contingent of experimental time is requested for experiments with UCN. UCN have kinetic energies below 335 neV which corresponds to a temperature below 4 mK. Neutrons at this low energy level can interact with matter and can be led to experiments and thus become storable. One motivation for experiments with UCN is the measurement of the free neutron's lifetime, an important physical constant. This constant affects many theoretical considerations such as the composition of matter in the early universe. The fast neutrons, which are produced by fission in the reactor core, are moderated down to thermal energies inside the reactor. In order to cool down these (epi-)thermal neutrons originating from

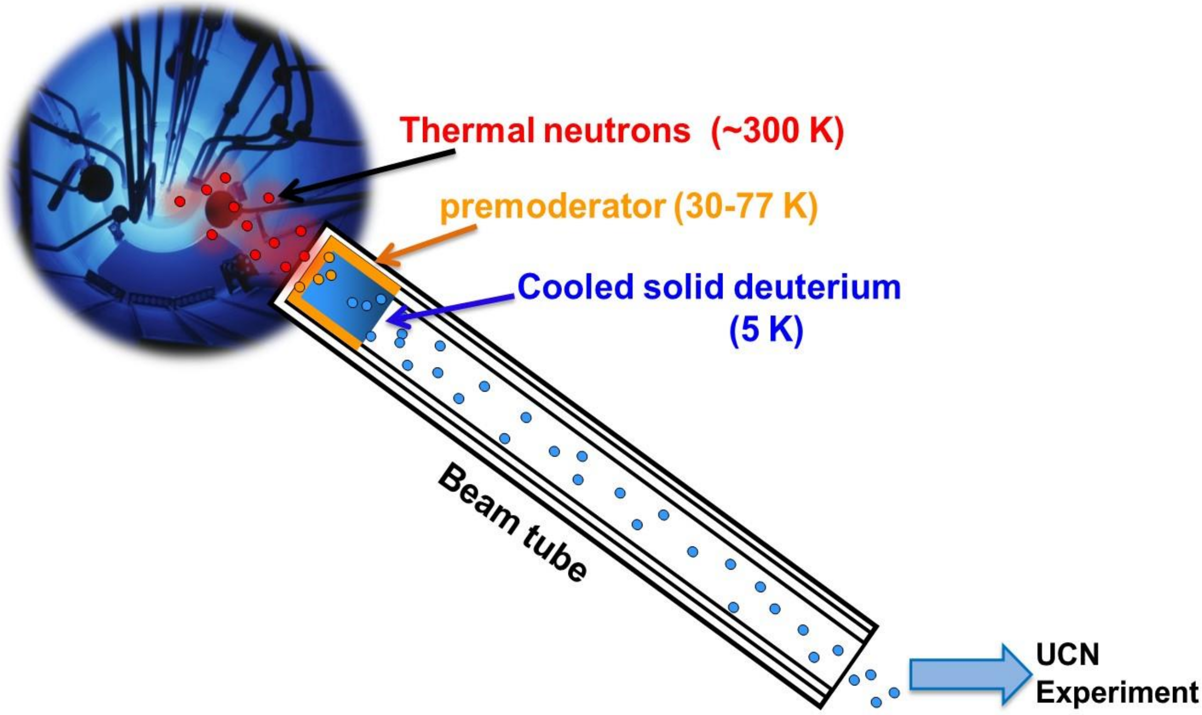

Fig. 5: Schematic presentation of the production of UCN. It shows how the thermal neutrons (red) are cooled down by a premoderator (yellow)

a reactor pulse to low energies, a special UCN source is needed (see Fig. 5) which is cooled down to around 10 – 30 K with the help of liquid helium. As a first step, the neutrons are cooled down to the µeV range by interaction with a stopper volume and the premoderator. The stopper consists of a mixture of graphite and bismuth. Around 10 mol of hydrogen is frozen out inside the source and acts as premoderator. The significant step into the ultra-cold regime is realized by an inelastic down-scattering process of the cold neutrons by mainly one phonon processes at the crystal structure of the main moderator, in this case 8 mol of solid deuterium. This super-thermal process creates ultra-cold neutrons with energy below 335 neV.

At the FR MZ two sources are pursued [1]. One source, located at beam tube C, has been developed in collaboration with the Technical University of Munich. The second source is located at beam tube D. While the UCN source at beam port C is used for continuous reactor operation, the UCN source at beam port D is used for pulsed mode operation. The source at beam port D has been upgraded in the last two years in collaboration with the Institute of Physics at the University of Mainz. The performed improvements yielded in a UCN density of 8.53 (5) cm^{-3} [15]; an increase by a factor of 3.5 compared to the former UCN source.

4.4 Training and Education

Besides the mentioned experiments above, a further important field of application is the training and education of professionals and students at the institute and the FR MZ. For this, the Institute of Nuclear Chemistry, as a part of the university, provides several options for students to train them in nuclear chemistry, nuclear physics and radiation protection.

For the traditional nuclear chemistry lab courses the required nuclides are produced in the irradiation facilities of the reactor. The aim of this course is to give a broad overview of the production, the properties and the applications of radioisotopes in chemistry, physics and life sciences. The training bases on the course Experimental Radiochemistry previously implemented by Otto Hahn at the Kaiser-Wilhelm-Institute of Physical Chemistry in Berlin. In one experiment of this 14-days course the students use the fission products of irradiated uranium to execute the same chemical experiments as the pioneers of nuclear chemistry did and which led to the discovery of the nuclear fission.
Another unique feature is the so-called reactor training, a course where the participants learn to operate the reactor under supervision. This course is provided to students and, with some variations, to the participants of the reactor operator’s school at the Paul-Scherer-Institute (PSI) in Villigen, Switzerland. Within the training, the participants are performing the daily checklists. Furthermore, they train fuel element handling, irradiation of samples and finally they have the chance to pulse the reactor by themselves.

Operating a research reactor means in the same breath to deal with radiation protection. Therefore the crew of the FR MZ also consists of a radiation protection crew with a wide ranged competence. The over many years gained experiences are passed to participants of radiation protection courses at the institute. In these courses specialized fire fighters, teachers or future radiation-protection-officers are trained for a secure handling of radioactive materials.

5. Outlook and Conclusion

After more than 50 years of operation, the FR MZ is still an intensively used university research reactor. The research community has still a benefit in this reactor, not only the research group in the field of fundamental research but also experimenters of applied sciences from inside and outside the university.
It is necessary for Germany to develop and conserve the knowledge and competences in nuclear chemistry and reactor technologies, as well as related fields such as radiation protection, to retain a voice in international committees. The student’s interest and the demand for education and training of them and other groups such as fire fighters or operators from Switzerland strengthens this assumption.
Due to the concept of a life-time-core of the FR MZ with only moderate fuel consumption in combination with a reservoir of fresh fuel elements, an ongoing operation until the end of the next decade is possible.

6. References

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