

Internal Exposures by Tritium in a High Flux Research Reactor

Franz M. Wagner and Richard Henkelmann, Technische Universität München,
Walter Fritsch, Siemens-KWU Erlangen

The construction of the new neutron source FRM-II at Garching is in an advanced state; the commissioning is planned for the year 2001. The core of this reactor consists of one very compact element with 8.1 kg high enriched uranium (93 % U-235). The core is cooled by light water and is moderated by 10 m³ heavy water (D₂O) contained in a surrounding tank with 2.5 m diameter, see [1] and fig.1. In this way, the core is strongly undermoderated, and the maximum thermal flux density (energy 25 meV) is reached only 13 cm outside of the core with a slow radial decrease. In this way, the beam tubes can be mounted tangentially to the core so that the user avoids the direct hard neutron and gamma radiation of the core, and nevertheless gets the maximum possible thermal flux. D₂O has the best moderation-to-absorption ratio of all usable materials. According to calculations, the unperturbed maximum neutron flux density (i.e. without beamtubes) is 8E14 cm⁻²s⁻¹. Such a flux, however, generates during one day with 20 MW thermal power about 3E12 Bq tritium (T) by activation of deuterium (D). Within 2 years, an inventory of about 2E15 Bq tritium is built up as DTO in altogether 20 m³ D₂O with an average concentration of 1E14 Bq/m³.

There are also other systems where tritium is generated, the cold neutron source which contains 2 kg D₂-gas, and the primary light water circuit. Many modern applications in physics, biology, material testing etc. need neutrons with energies 1-10 meV; hence, the cold neutron source is the most important facility of the reactor, and the design of the whole reactor is optimised for it, similar to the facility of the HFR Grenoble. The build-up of tritium during the whole lifetime (about 40 years) of the cold neutron source amounts to nearly 1E15 Bq; the tritiated deuterium gas can be gathered completely in a metal hydride storage tank. The tritium concentration in the light water circuit reaches only about 10⁸ Bq/m³; this is less than in the old research reactor FRM.

The handling of large amounts of tritium is justified by the obtained degree of optimisation; in particular, the FRM-II will be the high-flux reactor with the lowest thermal power world-wide. On the other hand, however, the enhanced requirements for handling such a radioactive inventory must be met. With respect to tritium, a number of limit values have to be observed in context with transport, handling, release, leakage, incorporation, and contamination.

The working-range of the tritium concentration is given mainly by a transport condition: The mentioned amounts of activated D₂O can be transported on public roads practically only by use of 200-l-drums qualified at least as "IP-2". The international transport convention ADR, attachment A, sets an upper limit to the tritium concentration in D₂O at 4E14 Bq/m³. The use of heavy type B casks (for which the tritium activity is unrestricted) is not possible due to the big quantity of drums. Because of the quick build-up of tritium, the D₂O has to be replaced regularly by recycled D₂O which has about 3E13 Bq/m³ initial tritium concentration (other radionuclides have been extracted by ion exchange resins). *In praxi*, the average tritium concentration will be about 2 E 14 Bq/m³. Hence, the tritium concentration range of operation is rather narrow, but it still allows for sufficiently long exchange periods on the one hand, and limits the risk by still higher concentrations of D₂O, on the other hand. No advantage can be taken from virgin D₂O because, due to the fast built-up, it would not prolong the exchange period significantly (fresh D₂O can contain, by the way, more than 4E10 Bq/m³ tritium).

Radiation protection at the reactor site imposes further limit values, and necessitates a number of practical measures which are optimised with respect to the ALARA principle:

1. The maximum licensed stack release for the FRM-II is 3E10 Bq tritium during one day; this amount can be contained in 100 cm³ D₂O. As a consequence, the complete D₂O-system must be sealed in a way that it can be evacuated to about 1 mbar (in this way, the D₂O-circuit can also be dried completely in case of unavoidable changes or repairs). The exhaust air of the reactor building is controlled for the tritium concentration as well as for the rate of discharge and triggers alarms at 3E9 Bq/h and 2E10 Bq/h. In context with the licensing procedure of FRM-II, the potential radiation exposure of reference adults and infants from releases via the vent stack has been calculated [2] taking into account all possible exposure pathways according to the radiation protection regulations (defined in section 45 of the German Radiological Protection Ordinance). Using the most recent weather statistics at the Garching site, the maximum values are 11 and 13 μSv/a for adults and infants, respectively. The most important fraction of the calculated effective dose (about 67 %) is generated by Ar-41, whereas tritium contributes only about 7 %. This is due to its extremely low energetic beta radiation and absence of γ-radiation so that it can cause only internal exposure. The main tritium emissions are in the forms HTO and DTO. On this level of exposure, further measures for reduction are not reasonable.
2. According to the German IWRS-II regulations, a total dose to a radiation exposed worker during one project (e.g., exchange of D₂O) greater than 10 mSv needs a special permit by the supervising authority; with the

conversion coefficient used in the German radiation protection code, this value would be reached with $6E8$ Bq tritium uptake corresponding to about $3 \text{ cm}_3 \text{ D}_2\text{O}$ with average concentration. This example is given in order to demonstrate orders of magnitude, but no practical constraints are expected from it.

3. The maximum permanent concentration of tritium in air is $1E6 \text{ Bq/m}_3$ assuming 1000 h working time per year and 20 mSv as European limit of the effective dose (standard 96/29EURATOM). Without permanent exchange of air, this concentration of activity would be obtained by evaporating $0.5 \text{ cm}_3 \text{ D}_2\text{O}$ in a room with 100 m_3 volume. Of course, the exchange rates are maintained at a level of at least 8 times per hour as prescribed for by the standard DIN 25425 for radionuclide laboratories so that the exposure via this pathway is excluded. Additionally to the plant exhaust air monitor, tritium monitors are installed in the local exhaust air system of the D_2O zone which comprises a personnel lock, cooling, storage, waste water, pumps for the purification, filters, handling, and transfer rooms for the ion exchange resins. The exhaust air from the cold neutron source has an own monitored pathway to the top of the stack (because of the release as gas, D_2/DT); the D_2O service laboratory has a mobile room monitor. The alarms of the local exhaust air monitors are triggered at $5E5 \text{ Bq/m}_3$ and $5E6 \text{ Bq/m}_3$ whereas the plant exhaust air (about $30000 \text{ m}_3/\text{h}$) triggers the alarms already at $1E5$ and $5E5 \text{ Bq/m}_3$. If the higher value is reached, the shift supervisor can perform a ventilation isolation of the contaminated section.
4. In context with the mentioned European dose limit 20 mSv/a , the take-up of radionuclides has to be controlled; therefore, the staff has to deliver urine samples within 4 working days after handling D_2O -samples. The potentially most delicate routine work is the exchange of the resins: First, the container with the mixed-bed filter is isolated from the moderation circuit; after about one week waiting-time, the resin is rather free from mobile heavy water. Then, the compartments of the fittings are evacuated, and the container which is shielded by 10 cm lead is disconnected. Subsequently it is moved on rails into the adjacent room where the resin is vacuum-dried during one week. The $\text{D}_2\text{O}/\text{DTO}$ emerging in the outlet of the pump is frozen out at $-40 \text{ }^\circ\text{C}$ and can be recycled. The container is then sealed and transported from the basement (1^{st} level) into the hot cell on the 5^{th} level (see Fig.1), where it can be opened. The basket with the resin is transferred into a waste drum, and the original container is furnished with fresh resin. The waste drum is enclosed in another shielded transport vessel, e.g. type "MOSAIK", and transferred to the waste storage room on the 1^{st} level. There, the drum is taken out by help of remote control, and the MOSAIK vessel becomes free for the next transport. Waste-conditioning is not foreseen at the FRM-II.
5. The practical limiting value for a surface contamination with tritium is $5E4 \text{ Bq}$ on 100 cm_2 inside controlled areas, and 1 % of this value outside. This means that already a small leakage in the D_2O system and droplets on a sheath will lead to a violation of limits. Therefore, samples of the D_2O -circuit are taken in a glove box by help of special fittings avoiding any drip water. The D_2O -vessel has to be transported in a cask to the exhauster in the service laboratory on the same floor. In this laboratory, the air is monitored by a tritium detector with a threshold at $1E3 \text{ Bq/m}_3$ which would show a quickly evaporating droplet. A residue contamination can be detected by a portable monitor with an open chamber. Waste water from the exhauster is collected in a vessel which has an own venting.

The handling procedures have been described in great detail by the general contractor SIEMENS-KWU, and are continuously subject to an examination by experts of TÜV and the authorities.

The light water circuit, in contrast to the D_2O -systems, causes no radiological problems. The temperature level at the surface of the open reactor pool, however, is generally above $40 \text{ }^\circ\text{C}$, so that up to 900 m_3 primary water evaporate per year. Therefore, the tritium release from this circuit will amount to about $1E11 \text{ Bq/a}$, i.e., 3 % of the licensed tritium release.

Concerning the tritium uptake of personnel working in the immediate neighbourhood of the open pool, there is experience at the old research reactor FRM: The mean level of HTO in urine samples of operators was 250 Bq/l ; the corresponding radiation exposure is about $4 \text{ } \mu\text{Sv/a}$. In context with these measurements, a side effect has been observed which demonstrates how readily HTO is incorporated, but also the tritium levels met in everyday life [3]: A watch with a luminous display may contain up to $2.5E8 \text{ Bq}$ tritium; a typical exhalation rate is 5 % per year. After 11 days, such a watch increased the concentration of tritium in the urine of its wearer from 30 to 750 Bq/l – i.e. three times higher than during work in the reactor hall of the FRM.

References

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Figure 1:

View from south into the FRM-II-building (ground area 40x40 m). The room on top is the reactor hall. In the centre, the reactor pool is shown with the D₂O-tank and the rising primary water outlet, control rods on top, and some beam tubes. The dark structure contains on its upper left also the storage pool and the adjacent primary cell with primary pumps and heat exchangers. The light structure above is the hot cell consisting of a pre-cell and the main cell. The dark room on the 2nd floor below the primary cell and the storage pool is the neutron guide channel (left); the horizontal slits in the western outer wall connect it to the neutron guide hall. At the right side, an insight into the experimental hall is given. The basement (1st floor) comprises the D₂O-systems, a service laboratory, shielded storage rooms for radioactive waste, ventilation, waste water and other systems.

Beitrag zu:

3rd European ALARA Network Workshop – Managing Internal Exposure, BfS Neuherberg, 15.-18.11.99

06/02/01 11:12

