

# THE CRYOSYSTEM OF THE MUNICH ACCELERATOR FOR FISSION FRAGMENTS (MAFF)

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## Abstract

The Munich Accelerator for Fission Fragments (MAFF) will be a facility to produce high-intensity beams of radioactive ions from neutron-induced fission at the new research reactor FRM-II in Garching. While the fission fragments will be produced in a through-going beam tube near the reactor fuel element, a cryopump will be installed near the fission source designed to localize volatile radioactivity close to its origin. The layout of the MAFF facility will be outlined and a characterization of its expected radioactive inventory will be given, thus defining the specifications for the cryosystem.

## 1 MAFF: THE MUNICH ACCELERATOR FOR FISSION FRAGMENTS

High-intensity radioactive ion beams are key prerequisites of current experimental nuclear physics. Fission induced by thermal neutrons provides is capable of producing the highest yields of neutron rich isotopes due to the large cross section for  $^{235}\text{U}$  fission ( $\sigma = 580 \text{ b}$ ). Therefore one of the main experimental projects (MAFF: Munich Accelerator for Fission Fragments) at the new high-flux research reactor FRM-II in Garching aims at the production of intense neutron-rich fission fragment beams [1].

### 1.1 Overview of the MAFF facility

Figure 1 gives an overview of the planned facility. On the left hand side it shows the reactor hall with the reactor vessel and the fuel element in the centre. In red the beamline of MAFF (SR-6) is indicated, which is the only evacuated and through-going beamline at the FRM-II.

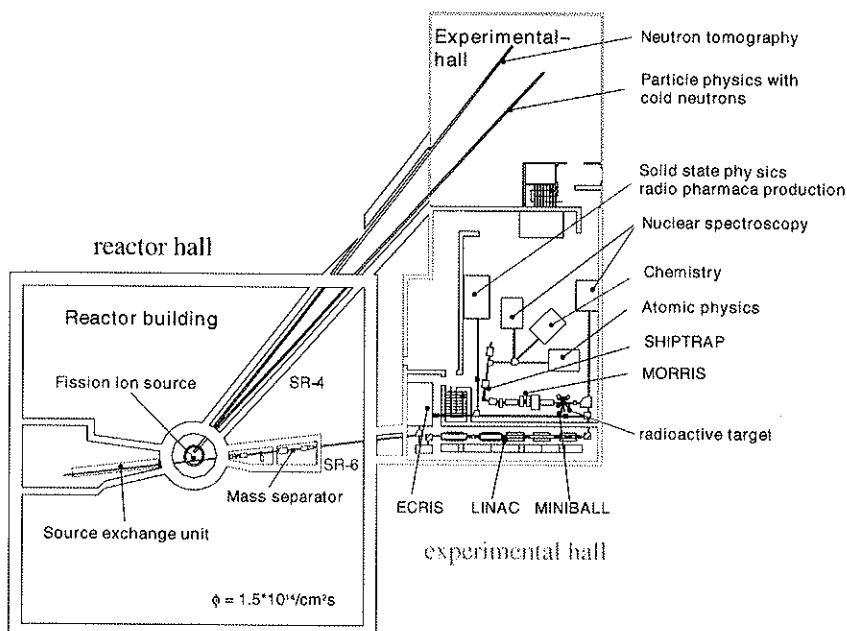


Fig. 1: Layout of the MAFF facility at the research reactor FRM-II in Garching.

The  $^{235}\text{U}$  fission source will be placed close to the fuel element in a neutron flux of about  $10^{14}$  neutrons/sec  $\text{cm}^2$ . The fission products will be extracted and after mass-separation and emittance improvement (cooling) they will be guided to an experimental hall, which will house the accelerator and the experimental installations.

Figure 2 shows the inner part of MAFF within the reactor building. In the center the concrete-shielded reactor vessel with openings for the various beam lines is visible. The beamline for MAFF (SR-6) is divided into two sections (SR-6A, SR-6B) to either side of the reactor vessel. This reflects the fact that mounting and exchanging the fission source will be performed from one side (SR-6B), while the extraction of the beam will take place from the other side, followed by a first stage of mass-separation.

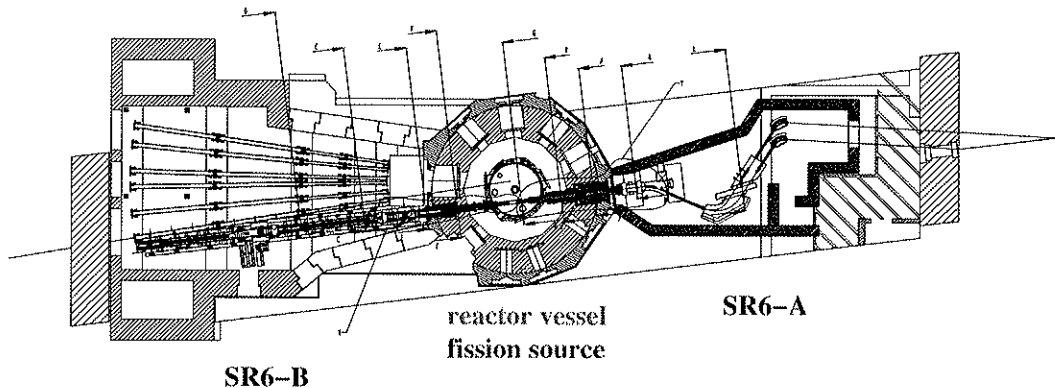


Fig. 2: Horizontal cut through the reactor hall of the FRM-II, showing the central part with the reactor vessel and the through-going beam tube of the MAFF project (SR6-A and SR6-B).

A more detailed insight into the innermost part of the reactor with the concrete-shielded light-water reactor vessel and the heavy-water moderator tank is given by Fig. 3.

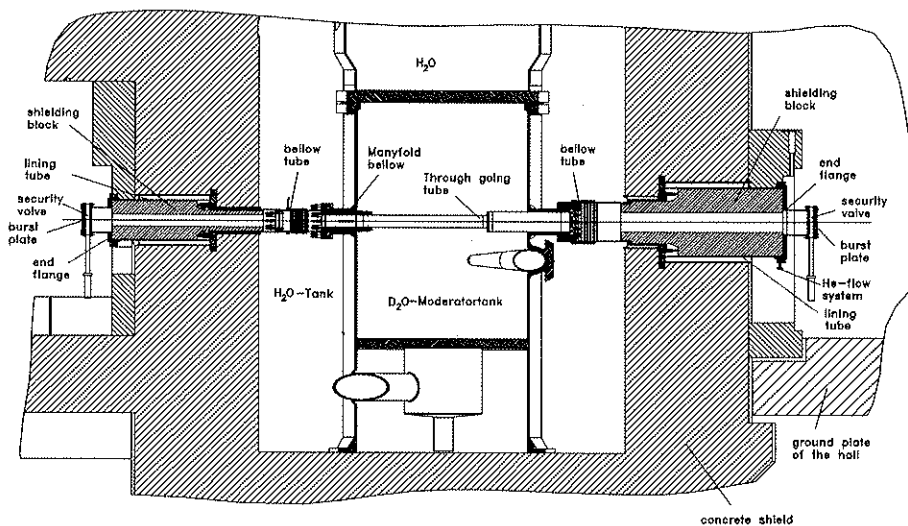


Fig. 3: Horizontal cut through the inner part of the through-going beam tube SR6 at the FRM-II.

While most components of MAFF are still in the planning and approval stage, the through-going beam tube is already installed in the reactor. In contrast to all other beam tubes at the FRM-II, which are made from Aluminum, the MAFF beam tube beam tube is made from Zirkaloy, a material that is resistant against embrittlement by neutron radiation. Therefore the lifetime of the MAFF beam tube is projected to be the lifetime of the reactor, while all other beam tubes have to be replaced every five years due to neutron damages. This would not have been possible for the MAFF beam tube, which has to

be considered as highly contaminated with long-lived fission fragment radioactivity. Another advantage of the Zirkaloy material is its high melting temperature of almost 2000°C, which is favourable for the operation of the  $^{235}\text{U}$  fission source which will be heated to about 2400°C.

Figure 4 summarizes in a schematical way the principle of the MAFF facility. The  $^{235}\text{U}$  fission source will be mounted on a trolley for mounting and exchanging after each reactor cycle of 52 days. The source itself consists of up to 1 g of  $^{235}\text{U}$ , dispersed in a porous graphite matrix surrounded by a Rhenium cylindre of about 8 cm length and 2 cm diameter, operating at a HV potential of 30 kV. The fission products will leave the hot source (about 2400°C) via diffusion through a small central hole. Thus the fission source has to be considered a potentially open source. Fission fragments will be surface-ionized on the hot Rhenium metal and will be extracted by electrostatic lenses and further transported for mass-separation and emittance improvement by beam cooling. In addition Fig. 4 shows a cryopump installed close to the fission source, which be needed since a significant fraction of the Uranium fission fragments will be volatile species. It is foreseen to freeze out this part of the unwanted radioactivity on the cold panels of the cryopump, where they will be localized and migration through the beamline system can be prevented.

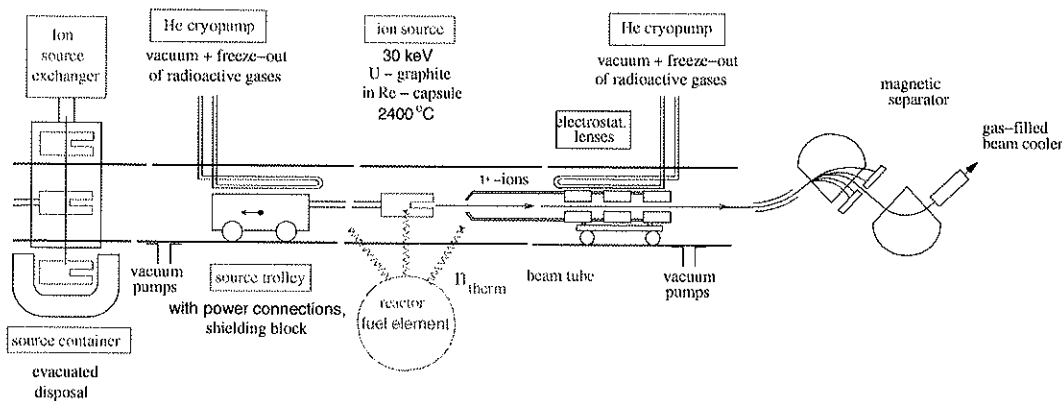


Fig. 4: Schematical view of the layout of the MAFF facility to produce neutron-rich ion beams from n-induced  $^{235}\text{U}$  fission products.

## 1.2 Isotope yields at MAFF

In order to demonstrate the potential of neutron-induced fission to produce high yields of neutron-rich isotopes, Fig. 5 displays a calculation of the isotopic yields after the MAFF mass separator, based on an initial fission rate of  $10^{14}$  fission/s. The resulting distribution reflects the Uranium fission mass distribution with intensities for individual isotopes of more than  $10^{11}$  ions per second in the maxima of the fission mass distribution. The red arrows mark the gaseous species among the fission products, consisting of the noble gases Krypton and Xenon and of the halogenides Bromium and Iodine.

Table ?? gives a list of some typical beam intensities expected for MAFF compared to the respective intensities presently achieved at radioactive ion beam facilities like ISOLDE (CERN) or SPIRAL (GANIL) or expected for the planned Rare Isotope Accelerator in the US.

It is obvious that along with such high radioactive ion beam intensities large quantities of unwanted radioactivity have to be controlled. The MAFF cryosystem is designed to localize the dominant fraction of volatile and non-ionized radioactivity close to its origin at the fission source.

## 2 RADIOACTIVE INVENTORY AT MAFF

Figure 6 contains an attempt to characterize the distribution of the radioactivity produced in the fission ion source along the MAFF beam line system. The diagram shows the distribution of the radioactive  $^{131}\text{I}$

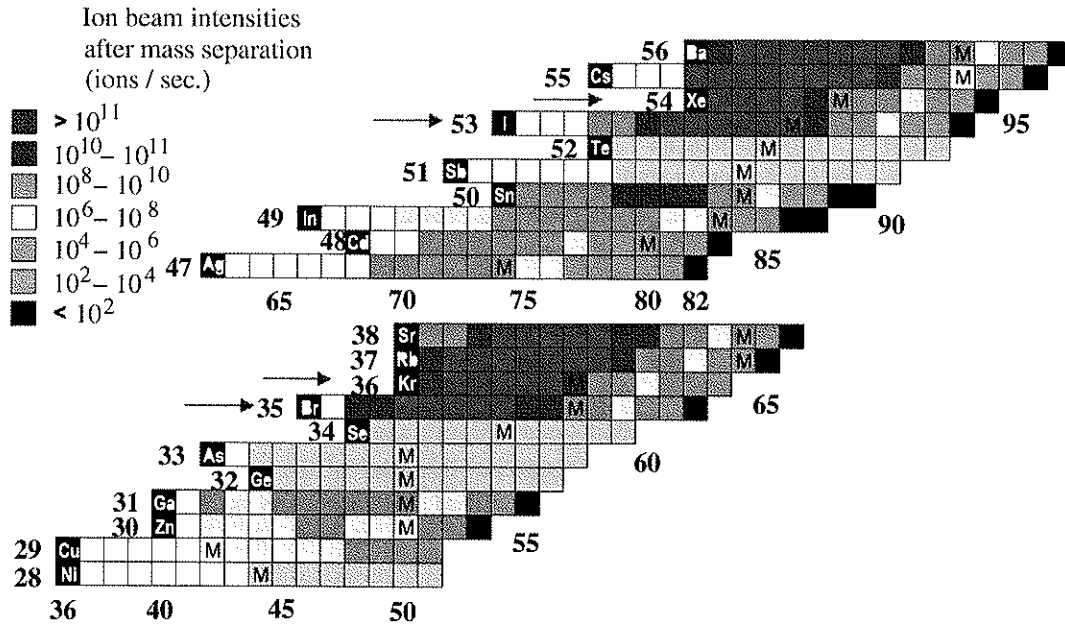


Fig. 5: Expected ion beam yields behind the MAFF mass separator based on a fission rate of  $10^{14}$  fission/s. Isotopes where too few information was available for a reliable yield estimate are marked in grey. The black boxes indicate the most neutron rich stable isotopes. The heaviest isotopes, for which the mass was experimentally determined [2] are marked by 'M'.

dose applied in a distance of 1 m after 1 day of decay time.

The source will typically produce a fission rate of  $10^{14}$  fission events per second, which is equivalent to an activity of about  $4 \cdot 10^{16}$  Bq (10 kCi). During the reactor cycle of 52 days, when the source is heated by the fission process and the fission products diffuse out of the Rhenium container, the source has to be considered to be an open source. It can be estimated that in a distance of 1 m from the fission source a total dose of  $3 \cdot 10^8 \mu\text{Sv}/\text{hour}$  resulting from non-volatile fission products will be produced. Only about 2% of the fission products will leave the source as ions, while about 30% of the fission products will be produced as volatile species. The dominant part of the unwanted ionized activity will be deposited onto the well-shielded slits of the mass pre-separator. Due to their long lifetimes the main part of the activity remaining on those slits after some days of decay time after the end of a reactor cycle will originate from mass 95 (Zirkonium) and mass 140 (Barium). In order to avoid a buildup of long-lived activity from those isotopes in the experimental part of the MAFF facility it is foreseen to avoid an extraction and acceleration of beams from those isotopes. The activity deposited onto the slits of the subsequent

Isotope	Prod. rate ( $\text{s}^{-1}$ )	half-life (s)	release (%)	ioniz. (%)	MAFF ( $\text{s}^{-1}$ )	SPIRAL-II ( $\text{s}^{-1}$ )	ISOLDE ( $\text{s}^{-1}$ )	RIA ( $\text{s}^{-1}$ )
$^{78}\text{Zn}$	$2.3 \cdot 10^9$	1.47	36	10	$8 \cdot 10^7$	$1.3 \cdot 10^9$	$1 \cdot 10^6$ SC	$8 \cdot 10^7$
$^{91}\text{Kr}$	$3.3 \cdot 10^{12}$	8.6	89	15	$4 \cdot 10^{11}$		$2 \cdot 10^9$	$7 \cdot 10^{11}$
$^{94}\text{Kr}$	$1.1 \cdot 10^{11}$	0.2	16	15	$3 \cdot 10^9$		$4 \cdot 10^6$	$5 \cdot 10^{10}$
$^{97}\text{Rb}$	$3 \cdot 10^{10}$	0.17	10	80	$2 \cdot 10^9$		$3 \cdot 10^8$	$1 \cdot 10^{10}$
$^{132}\text{Sn}$	$7 \cdot 10^{11}$	39.7	89	10	$6 \cdot 10^{10}$	$6.6 \cdot 10^9$	$8 \cdot 10^7$ SC	$1 \cdot 10^{10}$
$^{131}\text{Xe}$	$5.2 \cdot 10^{11}$	1.24	7	25	$1 \cdot 10^{10}$	$7.8 \cdot 10^9$	$2 \cdot 10^8$	$1 \cdot 10^{10}$
$^{144}\text{Xe}$	$9.3 \cdot 10^9$	1.15	7	25	$2 \cdot 10^8$		$5 \cdot 10^6$	$1 \cdot 10^9$
$^{144}\text{Cs}$	$4.3 \cdot 10^{11}$	1.0	47	80	$2 \cdot 10^{11}$		$3 \cdot 10^{10}$	$4 \cdot 10^{11}$

Table 1: Comparison of selected MAFF beam intensities with other radioactive ion beam facilities

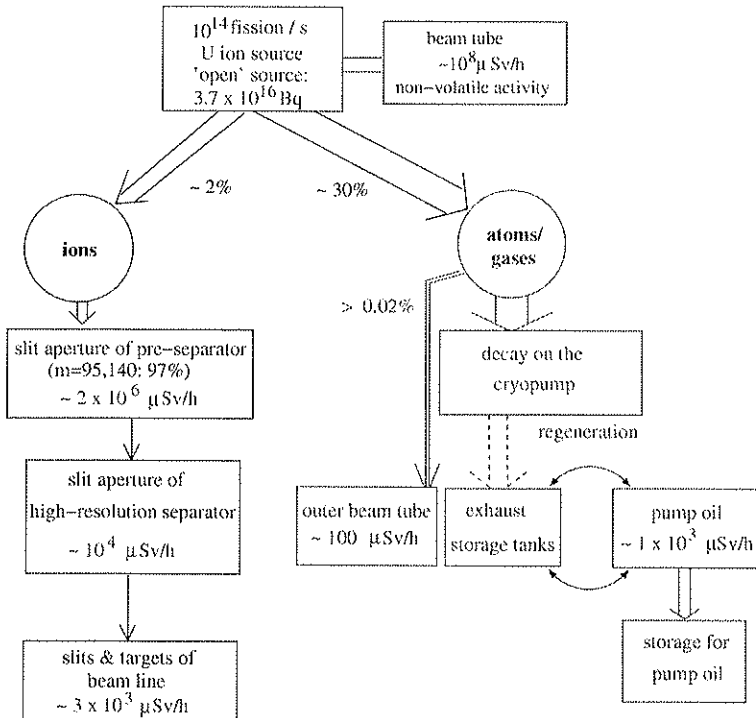


Fig. 6: Schematic view of the radioactivity distribution along the MAFF system (analog to Ref. [3]).

high-resolution separator and also finally in the experimental area will already be reduced by two or three orders of magnitude.

Radioactive gases amount to about 30% of the produced radioactivity. The cryopump of the MAFF beam tube is designed to localize the dominant part of this volatile component onto its cryopanel, so that only a fraction with a lower limit of 0.02% will escape to the outer beamline or will be deposited in the pump oil or the exhaust storage tanks of the vacuum system.

Volatile Uranium fission products typically exhibit short-lived  $\beta$  decay half-lives in the order of minutes with only a few exceptions of longer-lived activity. It is the purpose of the cryopump to freeze out gaseous activity in order to localize it onto the cold surfaces until the  $\beta$  decay will transform the dominant fraction of volatile activity into non-volatile species.

It should be noted that the cryogenic concept to confine the volatile activity on the cold surfaces of cryopanel close to the fission source is continued by the beamline vacuum system, where also cryopumps and not turbomolecular pumps provide the pumping capacity. Thus additional capabilities to retain volatile species exist throughout the beam transport system, especially acting as a backup in case of a failure of the main cryo system.

The main part of volatile activity that will escape permanent localization will come from  $^{85}\text{Kr}$ , which has the longest half-life of all gaseous fission products ( $t_{1/2} = 10.76$  years). However, with a fission yield of only about  $10^{-6}$  the resulting  $10^8$   $^{85}\text{Kr}$  nuclei per second will accumulate within one reactor cycle of 52 days (being stored for that period in the exhaust storage tanks of the vacuum pumping system) to about  $4.5 \cdot 10^{14}$  nuclei. Taking into account the half-life of  $^{85}\text{Kr}$  a decay rate of  $0.7 \cdot 10^6$  Bq will originate from  $^{85}\text{Kr}$ , which is negligible in view of the licensed yearly release dose of  $10^{13}$  Bq for long-lived noble gases at the FRM-II. The concept of confining radioactivity at certain well-localized positions can be extended also to the exhaust storage tanks of the vacuum pumping system. By installing an oil-sealed circulation pump with additional filters in the storage system a large amount of activity can be deposited and retained in this filter system, thus minimizing the time until a release of the exhaust gas to the environment is acceptable.

Although the durability of the MAFF beam tube from Zirkaloy against neutron radiation damage will contain all radioactive contamination for the lifetime of the FRM-II, an estimate will be given for the longterm  $\beta$  and  $\gamma$  activity buildup within the beam transport system, assuming  $10^{14}$  fission events per second and a runtime of the cryopump of 10 years. The dominant part of the  $\gamma$  activity will originate from the decay of  $^{137}\text{Cs}$  with a half-life of 30 years, coming from the  $\beta$  decay of  $^{137}\text{Xe}$  ( $t_{1/2} = 4$  min.). After ten years of operation a  $\gamma$ -decay rate of about  $1.2 \cdot 10^{12}$  Bq can be expected.

The corresponding  $\beta$ -decay rate is of the same order of magnitude ( $1.07 \cdot 10^{12}$  Bq), mostly originating from the decay chain starting from  $^{90}\text{Kr}$  and via Rb ( $t_{1/2} = 28.5$  a) finally leading to Yttrium.

The cooling conditions needed to freeze out the radioactive gases can be clarified when looking at the vapour pressure curves of the different gaseous elements as a function of temperature, as shown in Fig. 7. For temperatures below about 20 Kelvin all relevant species including Nitrogen as the dominant component of the residual gas can be frozen out to partial pressures in the ultra-high vacuum regime. Therefore a temperature of 15 K was chosen as the operational temperature for the MAFF cryopump.

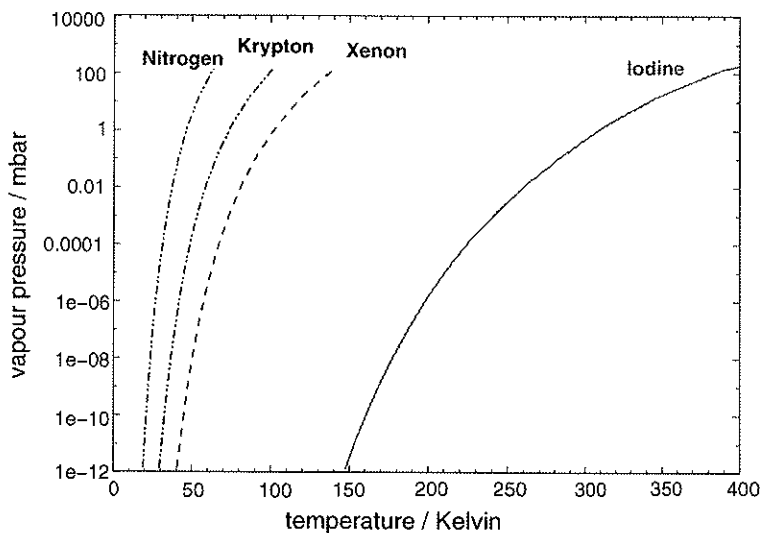


Fig. 7: Vapour pressure curves of volatile ion species.

### 3 DESIGN SPECIFICATIONS OF THE CRYOSYSTEM

In order to characterize the surrounding conditions for the operation of the cryopump the radiation fields existing along the MAFF beamline have to be calculated. Figure 8 shows the results of Monte Carlo calculations using the MCNP code [4] resulting in the neutron flux along the innermost part of the MAFF beamline SR-6. The center position marks the place of the fission source, while in addition the position of the cryopanel at each of the two sides of the beamline is indicated. The cryopanel will be located in a distance of about 2 m from the fission source, where the radiation level for neutrons is already reduced by about three orders of magnitude compared to its maximum.

#### 3.1 The cryopanel

The cryopanel designed to confine gaseous radioactivity near the MAFF fission source has to provide a homogeneous temperature distribution of about 15 K over a large area within a very limited space available in the MAFF beam tube. So a very compact design had to be found for the cryopanel. As a solution a double-wall tube design was chosen, where six spiral-type segments allow for a circulation of cold Helium gas. This can be seen in Fig. 9.

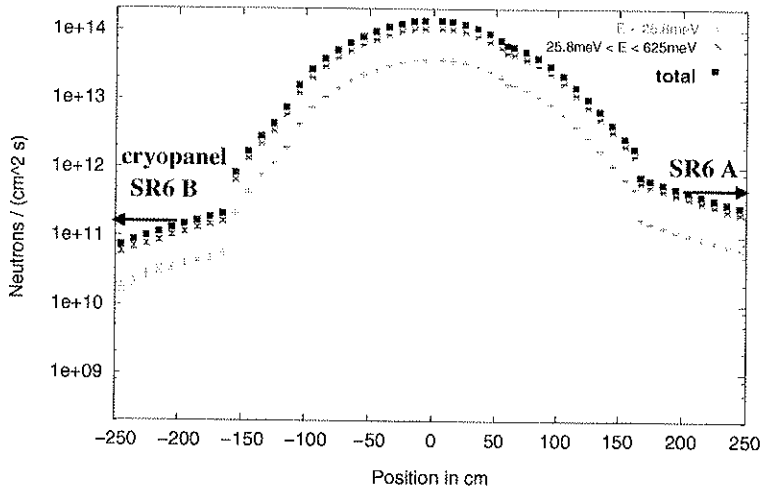


Fig. 8: Simulated neutron flux along the SR6 beamline of MAFF using the MCNP code.

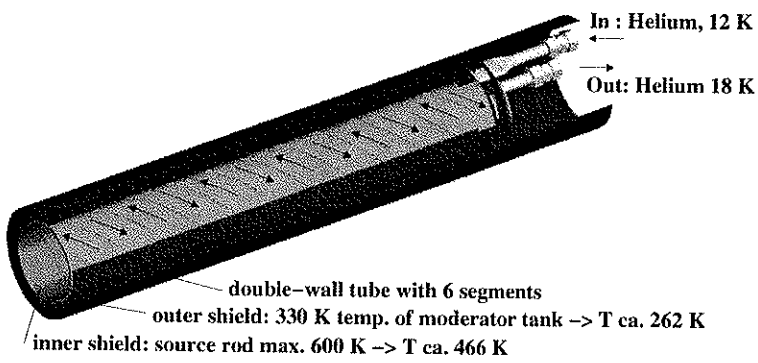


Fig. 9: Schematical view of the double-tube cryopanel designed to freeze out volatile radioactivity near the MAFF fission source.

Since the limited space does not allow for actively cooled ( $\text{N}_2$ -)shields, passive floating inner and outer shields will be installed to reduce the heat load from the outside beam tube and the inner hot Titanium rod that carries the fission source. The resulting temperatures on the shields have been estimated to be about 260 Kelvin on the outside shield and about 470 Kelvin on the inner shield. This will allow to reduce the thermal heat load on the cryopanel by a factor of two.

The cryopanel will have a length of 1 m (SR6-B) and 1.5 m (SR6-A), respectively, with an inner diameter of 157 mm, which is limited by the diameter of the fission source that needs to be moved through the cryopanel assembly. Since the cryopanel will be mounted in the sensitive inner region of the FRM-II, they have to be designed for integrity in case of all possible impacts from the inside or the outside, thus implying restrictions onto the minimum wall thickness which was chosen as 2 mm. The gap for the cold Helium gas flow between the two tubes will be 4 mm. Due to its cryogenic specifications the cryopanel will be manufactured from Al 6061T6. The assembly of the double-wall tube will be performed by shrinking the (heated) outer tube onto the (cooled) inner one.

Given the active cold pumping area of about  $2.5 \text{ m}^2$  an (ideal) pumping capacity for the heaviest volatile isotopes of Xenon can be estimated to be about  $1.4 \cdot 10^5 \text{ l/s}$ . This number can only be considered to be an upper limit, where changes of the emissivity of the cold surfaces with time have to be taken into account as well as the realistic conductance of the radioactive gases in the beam line. Since accurate calculations of those values are hard to achieve, experimental studies using a prototype setup are foreseen.

Since the pumping speed of the beamline vacuum system at the place of the cryopanel can be estimated to be about  $30 \text{ l/s}$ , an estimate of the attenuation factor for gaseous radioactivity as the ratio between this pumping speed and the above given pumping speed of the cryopump can be given, resulting in a lower limit of  $2 \cdot 10^{-4}$ . An experimental verification of this factor will be one of the central goals of the prototype testing.

### 3.2 Cooling power requirements

The most important parameter that defines the specifications of the cryopump as the central part of the MAFF cryosystem is the amount of cooling power needed to achieve the foreseen average operational temperature of 15 Kelvin on both of the two cryopanel. In addition to the thermal heat load the nuclear  $\gamma$  heating resulting from the  $\gamma$ -radiation field along the beamline has to be considered. Figure 10 displays the result of MCNP calculations for the  $\gamma$  heating along the beamline SR-6, which amounts to only a few mW/g in about 2 m distance from the fission source.

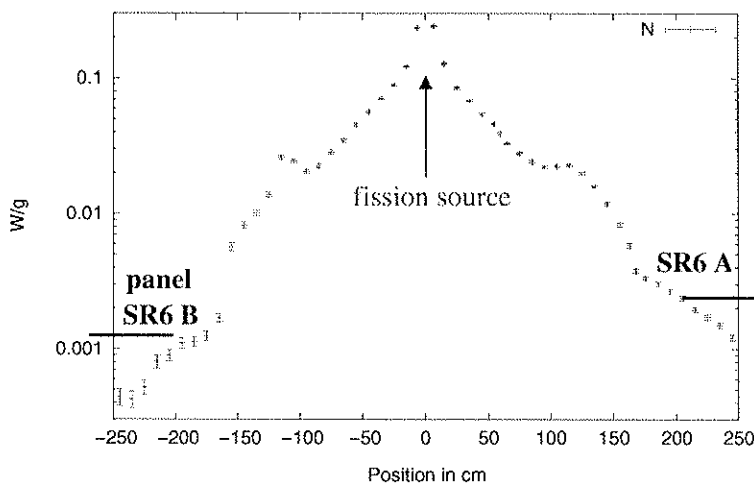


Fig. 10: MCNP calculation for the nuclear  $\gamma$  heating along the SR-6 beamline of MAFF.



Table 3.2 gives an overview of the heat loads appearing at the two cryopanel, with the dominant contribution originating from the thermal heat load, either introduced by the surrounding surface of the beam tube being at the temperature of the moderator tank (about 330 K), or by the hot Titanium rod carrying the fission source at a temperature of several hundred degrees. In comparison the contributions by nuclear heating and by heat conductance are almost negligible. In total a heat load of 700 Watt has to be compensated in order to operate the two cryopanel at an average temperature of 15 Kelvin. Including some safety margin (allowing for emissivity changes of the cryopanel surface during operation) the cooling power required for the operation of the MAFF cryosystem was finally chosen to be 1 kW at 15 K.

#### 4 LAYOUT OF THE MAFF CRYOSYSTEM

In order to operate the cryopanel as described above, a complex cryosystem has to be installed with special emphasis on the safety requirements imposed by the nuclear environment of a research reactor. A layout schematics of this cryosystem is given in Fig. 11. The design features are widely governed by the potential failure scenarios that have to be controlled under any circumstances.

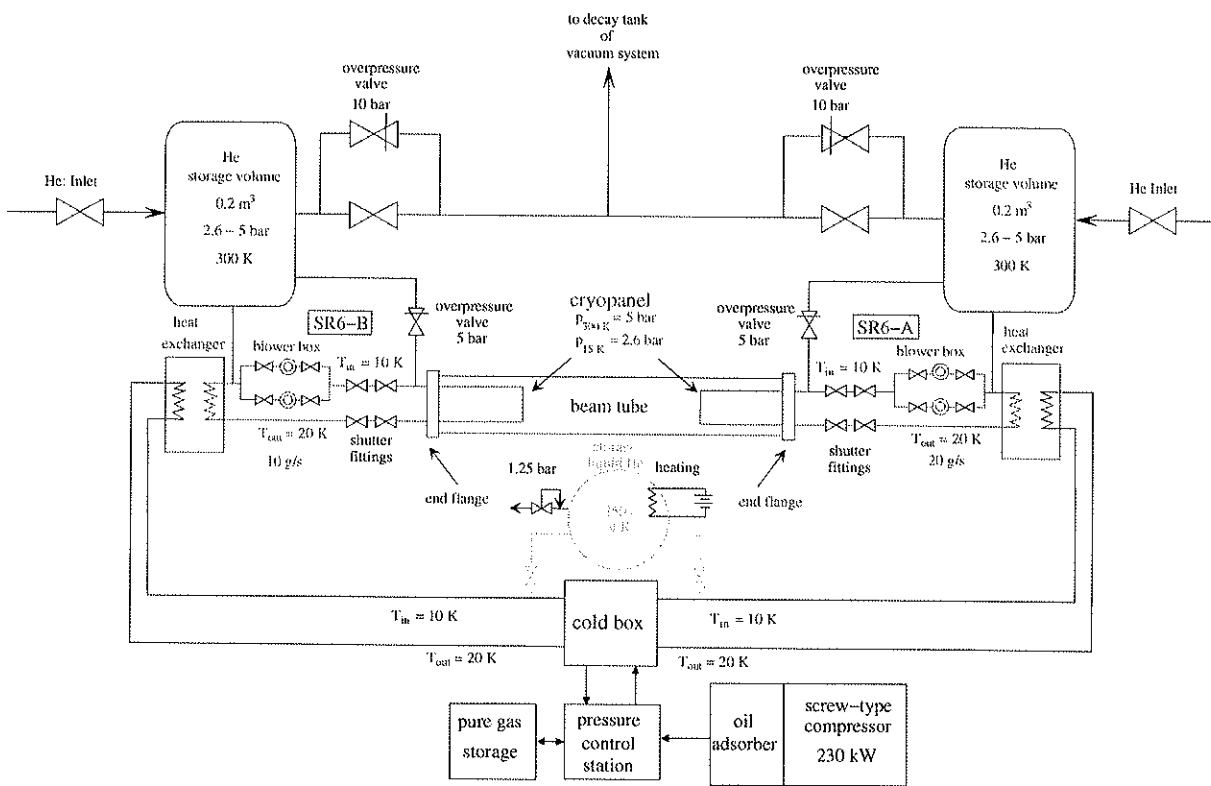


Fig. 11: Layout of the MAFF cryosystem.

Among those a complete failure of the Helium cooling system, resulting in a rapid heating-up

	SR-6A	SR-6B	Total
thermal radiation	396 W	264 W	660 W
heat conductance	21 W	14 W	35 W
nuclear heating	18 W	12 W	30 W
total	435 W	290 W	725 W

Table 2: Heat loads appearing at the MAFF cryopanel

of the cryopanel and the expansion of the cold Helium gas, can be identified as one of the major risk scenarios. Additional potential risks include a Helium leakage from the cryopanel into the evacuated beamline and any other impact from the inside or the outside like earth quakes or air plane crashes.

In order to control these incidents the general design principle is the integrity of the system in case of all of these failure scenarios, especially since part of the cryosystem (end flanges to either side of the inner part of the MAFF beamline) belongs to the second barrier of the reactor which has to keep its integrity under all circumstances to prevent water losses in the reactor vessel. This requires for example a redundant layout of valves. Since the MAFF beam tube is designed for a maximum inner overpressure of 5 bar, even in case of a complete warming-up of the Helium gas in the cryopanel the pressure must never exceed this value. The Helium gas storage tanks on either side of the beam tube compensate for pressure changes for example in case of a warming-up of the cold gas. In this case the gas would expand into the tanks without exceeding the allowed maximum inner overpressure. However, minimizing the Helium inventory in the cryopanel was one of the design goals, which was achieved by heat exchangers that decouple the secondary cooling circuit in the cryopanel from the primary circuit connected to the Cold Box of the Helium refrigerator. The flow of the Helium gas within the secondary cooling circuit is provided by blower units, which for safety reasons will be redundantly installed. Moreover, it is also favourable to minimize the amount of Helium exposed to the neutron flux of the reactor since commercially available Helium gas contains a 0.3 ppm contamination of  $^3\text{He}$  that will be activated forming Tritium by the neutron flux. For the cryosystem as displayed in Fig. 11 the resulting Tritium decay rate per reactor cycle will be about  $3.9 \cdot 10^8$  Bq (10 mCi), which is negligible compared to the approved yearly release dose of Tritium for the FRM-II ( $3 \cdot 10^{12}$  Bq).

In order to assure the localization of the volatile activity even in case of a sudden failure of the cryosystem, an emergency cooling system is foreseen using a liquid Helium storage tank. In case of a failure of the main cooling system cold gas from this reservoir will be pumped into the primary circuit replacing the He flow from the Cold Box. Thus an uninterrupted cooling for additional 30-60 minutes can be guaranteed, long enough to transform the dominant fraction of gaseous activity to non-volatile species via  $\beta$  decay. In addition also the cryopumps of the beam transport system outside the reactor vessel will be able to localize a significant amount of the released gaseous activity.

The main cooling system will be based on a commercial Helium gas refrigerator which will be designed for a gas inlet temperature of 12 Kelvin and an outlet temperature of 18 Kelvin, in order to guarantee the average operational temperature of 15 Kelvin. A screw-type compressor will compress the Helium gas to 13 bar which then will be adiabatically expanded in the cold box down to 3 bar. The whole system will be processor-controlled and maintenance-free over the period of the reactor cycle.

#### 4.1 Prototype tests

Since many aspects of the cryosystem performance can only be quantitatively characterized via experimental studies, extensive prototype tests are foreseen at the Maier-Leibnitz Laboratory in Garching. A test beamline has been set up, simulating the geometrical situation at the inner part of the MAFF beamline. A prototype of the double-wall cryopanel tube has been successfully manufactured and will be installed into the test beamline. It will be attached to an existing Helium refrigerator in order to perform a variety of tests. The pumping capacity of the cryopanel will be determined, considering also longterm tests, where we have to take into account a buildup of condensate from the residual gas on the cold surfaces, mostly from nitrogen.

The homogeneity of the temperature distribution along the cryopanel will be measured, with and without a prototype of the hot ion source installed in the centre of the beamline. Safety tests will also simulate the influence of leakages into the beamline. Finally the attenuation factor for the confinement of the volatile fission products will be measured with a highly-sensitive mass spectrometer by introducing defined amounts of tracer isotopes from Kr, Xe, Br and I.

## References

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