XA04C1708

Use of the FRJ-2 for Molybdenum Production

by

J. Wolters, G. Hansen, G. Stollwerk

Research Center Jülich, Germany

Central Research Reactors and Nuclear Operations Division (ZFK)

D-52425 Jülich

Telefon: 49 2461 61 3011

Telefax: 49 2461 61 3841

E-mail: J.P.Wolters@fz-juelich.de

Abstract: On the basis of an order from the Belgian Institut National des Radioéléments, the FRJ-2 research reactor (23 MW) will be used for the production of Mo-99 starting this autumn. The targets are MTR fuel plates with a U-235 content of up to 4.5 g formed into small tubes (outer/inner diameter: 22/19.5 mm; length: 160 mm). The irradiation conditions are that the targets should produce an average power of about 21 kW and that they should be loaded and unloaded during full power operation. It was not easy to fulfill these conditions with a tank-type research reactor, particularly due to cooling and shielding problems during irradiation and handling. The targets will be irradiated in three rigs, four per rig simultaneously. The rigs are cooled by a separate main cooling circuit. For safety reasons and for cooling of the rig during loading and unloading, each rig is equipped with an emergency cooling circuit to be installed in the gallery of the reactor block. At the end of irradiation, the targets loaded on a carrier one on top of the other are first lifted into a decay position in the shielding plug of the reactor tank. After 10 h, they are loaded into the fuel handling flask and are carried to the storage block in the reactor hall. During transport, the targets are cooled by forced circulation of air. It has been proven by an experiment that the targets will not exceed 500 °C if all active cooling fails. Thus, a melting of the targets during handling is impossible. In the storage block, the target carrier with the targets is loaded into a transport tube of about 40 mm wall thickness. The high heat capacity of the transport tube ensures that under normal conditions the targets do not exceed a temperature of 100 °C during transport in the uncooled fuel transport flask from the reactor hall to a water pool in the handling bay. Here, the targets are unloaded from the carrier and loaded into the customer's container. Due to the safety implications the project might have for the existing reactor, the supervisory authority required a licensing of the project according to the German Atomic Energy Act. This license was granted within 8 months at the beginning of 1998.

1. Irradiation Conditions

On the basis of an order from the Belgian Institut National des Radioéléments, the FRJ-2 research reactor will be used for the production of Mo-99. This opens a new chapter in the history of the 35-year-old reactor, which until now has only been used as a research instrument.

The irradiation conditions were set by the customer, who will provide the targets and arrange for the transport of the irradiated targets to the hot cells in Belgian, where they will be processed.

The targets to be irradiated are MTR fuel plates formed into small tubes (outer/inner diameter: 22/19.5 mm; length: 160 mm). They contain up to 4.5 g of U-235 in the form of HEU and should produce an average power of about 21 kW in order to obtain an average Mo-99 content of 840 Ci after 150 h of irradiation.
The targets must be back at the customers site 36 h after irradiation. Since there is a direct highway link between the two sites, the actual road transport of the containers with the irradiated targets will take not more than 3 hours.

2. Irradiation Rig and Target Carrier

There are two positions in the FRJ-2 reactor which provide an adequate neutron flux at a reactor power of 20 MW, the preferred power instead of the allowed power of 23 MW. But these positions are very small in diameter (50 mm), so that there is little space for the installation of a rig which fulfils all requirements with respect to the cooling of the targets during all steps of the irradiation and with respect to the handling of the targets during full power operation. The rigs are cooled by light water, which has to be separated from the heavy water of the reactor by two physical barriers. The coolant connections are at the top of the rig so that internal water guidance down to the targets and back to the top is needed. The solution is a rig with a central guide tube which is open at the bottom and which is accommodated in two concentric thimbles. The gap between the two thimbles is filled with helium and will be monitored for moisture. The central guide tube houses the so-called target carrier with the targets.

The target carrier is designed to carry four targets one on top of the other as is shown in Fig. 1. It consists of a central rod on which sleeves the length of the targets are positioned like beads on a string. The sleeves hold the targets. They are equipped at the bottom with six ribs symmetrically distributed around the periphery. The ribs are stepped in order to center the targets on the sleeves and the sleeves in the central guide tube of the rig. Thus, they ensure that the annular gap between the targets and the guide tube on the one side and the sleeves and the targets on the other side will have a nominal width of 2 mm.

This design had to be chosen for two reasons. First, there is not enough space in the rig for the accommodation of a shell in which the targets could be fixed and, second, a shell would have too greatly hindered heat removal from the targets during handling in air, which is necessary in this type of research reactor.

The maximum allowed power of a rig is 110 kW resulting in a maximum heat flux of the targets of 230 W/cm². This heat flux requires a coolant velocity of about 6 m/s in order to keep the surface temperature of the targets below 130 °C. A higher surface temperature was considered not to be acceptable in view of transients such as the loss of forced flow or a power pulse. Since the pressure in the rigs is imposed by the flow, the loss of flow is associated simultaneously with the loss of pressure. A downward flow along the targets was chosen for several safety reasons. It has no safety disadvantages since afterheat removal by natural convection is not possible anyhow due to the high friction of the rig.

In total, three rigs will be installed, one in a position with a somewhat lower neutron flux for redundancy. All rigs will be cooled by one main cooling circuit, the main components of which will be installed in the basement of the reactor. But for safety reasons and for cooling of the rigs during loading and unloading, each rig is equipped with an emergency cooling circuit to be installed in the gallery of the reactor block. These separate circuits use the N-16 decay tanks as water reservoirs and heat sinks. Redundant emergency cooling pumps circulate the water through the rigs on demand if the main circuit fails.

2. Loading and Unloading of the Targets during Full Power Operation

At the end of irradiation, the target carrier attached to a thin pulling rod by a bayonet holder is first lifted into a decay position in the shielding plug of the reactor tank. For this purpose, the pulling rod penetrates the shielding plug of the rig so that it can be seized by the lifting machine. The pulling rod consists of two parts bolted together and after the carrier has reached the decay position, the upper part of the rod is unscrewed and removed whereas the lower part is kept in position by a clamping device at the top of the inner rig plug. This plug together with the carrier and the targets forms the removable unit.

After a decay time of 10 h, the removable unit can be unloaded from the rig. The four targets produce a maximum thermal power of 180 W at this time. First, the rig is disconnected from the main cooling circuit and depressurized. Cooling of the rig is taken over by the emergency cooling circuit. Then, the inner plug of the rig can be unlocked. Unloading as well as loading of the removable unit is done with the fuel handling flask. However, since the rig is open after having unloaded the unit, a separate shielding device equipped with a shielding gate is needed to shield the radiation from the reactor after removal of the flask with the unloaded unit. This device must first be placed over the rig position on top of the reactor plate before the flask can be positioned. After having opened the gate of the flask and of the shielding device, the removable unit is lifted into the flask. During the lifting procedure, the targets are practically uncooled so that the energy produced must be stored in the targets and the carrier. This is acceptable due to the short time of about 2 minutes. In the flask, the targets are cooled by forced circulation of air. Although the average air velocity is very low, the cooling is adequate as a test with an electrically heated simulator has shown. A maximum target temperature of 190 °C is expected.

The removable unit is transported within the closed flask from the top of the reactor to the storage block of the fuel elements. There, it is inserted with the flask into a transport tube of about 40 mm wall thickness. At first, the target carrier is in a position where the air gap between the targets and the transport tube is 2.5 mm. Later, when the flask has been removed the carrier is lowered into a position with a gap of 1mm only in order to obtain a better heat coupling of the targets to the tube. The ribs of the carrier, which exceed the size of the targets by 2 mm must run in grooves in this part. Therefore, lowering is done by hand using the pulling rod, which is lengthened again for this purpose. After lowering, the target carrier is submerged in the water of the storage block.

The unit is loaded into the rig in the opposite sequence. The unit with fresh targets is loaded into the fuel handling flask from the transport tube, transported to the top of the reactor and lowered into the rig after the flask has been positioned and the two gates (of the flask and of the shielding device) have been opened. Subsequently, the flask and the shielding device are removed, the rig plug is locked and the rig reconnected to the main cooling circuit. Conditions are now fulfilled for lowering the carrier into the irradiation position, which is carried out by the lifting machine.

3. Dry Transport of the Targets to the Water Pool in the Handling Bay

The removable unit with the targets is transported from the storage block in the reactor hall to the water pool in the handling bay with the fuel transport flask. This flask is identical with the fuel handling flask except that there is no cooling facility. The lack of a cooling facility led to the selection of the transport tube which provides such a high heat capacity that under normal conditions the targets do not exceed a temperature of 100 °C during transport.

Prior to loading the transport tube with the loaded unit into the transport flask, the flask is filled with helium in order to improve the heat conduction across the gap between the targets and the transport tube. After having been loaded, the flask is transported on a vehicle through the truck air lock into the handling bay. There, the flask is taken up by the crane and put on a shielding cone on the water pool. Then the transport tube is lowered into the central hole of the cone and the target carrier is thus immersed in water. The whole transport process from picking up of the transport tube in the reactor hall to setting it up in the shielding cone takes no longer than half an hour.

After having removed the flask and lengthened the pulling rod, the carrier is lowered into a receiving fixture below the shielding cone. The carrier is disconnected from the pulling rod and the receiving fixture swung around horizontally so that there is free access to the carrier. It is taken up by a manipulator and set up on a table at a depth of 3 m. There, the targets are removed from the carrier and loaded into the customer's container. Loading fresh targets onto the carrier and transport back to the storage block of the reactor is performed in the opposite sequence. All manipulations in the water pool are observed via a submerged video camera.

4. Incidents to be Considered

Incidents to be considered are related to the irradiation process and to the handling procedures. The countermeasures are mainly directed towards protecting the targets from becoming overheated.

4.1 Incidents during Irradiation

The main cooling incident during irradiation is the loss of flow. It will in general be detected by 3 flow rate gauges at each rig causing a reactor trip, the disconnection of the corresponding rig from the main circuit and the demand of the emergency pumps when 2 of the gauges measure a flow of 80 % or less of the nominal flow rate. At nominal flow rate, the safety factor against flow instability and burnout does not drop below 2. This is also ensured by monitoring the temperature (2oo3) and the pressure (1oo1) at the outlet of each rig. The temperature causes a reactor trip but no disconnection of the rigs from the main circuit. The pressure releases a warning only, which requires operator actions.

The main cooling circuit is equipped with two main cooling pumps which are both normally operated. But for availability reasons, the three rigs may also be operated with one pump only. In this case, the frequency of the operating pump is monitored additionally in order to obbtain a reactor trip as quickly as possible if the operating pump should also fail.

For the detection of a target failure, the activity level at the outlet of the decay tank of each rig is monitored by 3 activity measuring instruments. They would respond if the activity level increased by two orders of magnitude and would scram the reactor and disconnect the corresponding rig from the main circuit in order to retain the radioactive substances released in the internal circuit. The reactor trip should avoid a propagation of the target failure. An analysis has shown that in the case of a burnout of an area of 1 cm^2 about 14 % of the fission products released would escape into the main cooling circuit.

Small leaks in the cooling circuits are detected by leak detectors and require operator actions. Larger leaks are only assumed in the main cooling circuit. They cause the automatic trip of the reactor and of the main cooling pumps by the level gauges (2003) of the storage tank. The emergency cooling procedure is demanded by the flow rate gauges of each rig. A guillotine fracture of a pipe in the main cooling circuit is excluded by design.

A number of interlocks prevent human errors during loading and unloading of the removable units. For instance, a special tool is needed to unlock the inner plug of a rig. This tool is released after 10 h of decay and after the rig has been disconnected from the main circuit.

Of course, the targets must not be damaged in all cases of design basis incidents of the reactor. There is only one reactor incident which affects the targets, namely a reactivity transient. The allowed reactivity addition to the reactor is a jump of 0.6 % $\Delta k/k$ increasing the reactor power by 100 % before the rapid shutdown system shuts the reactor down. The corresponding transient analysis with the Canadian CATHENA code has shown that the maximum target temperature reaches 186 °C within 200 ms and then decreases below 100 °C just as quickly as is had previously increased.

4.2 Incidents during Handling of the Irradiated Targets

The countermeasures in the case of incidents during handling are directed towards avoiding a target temperature of higher than 400 °C in the reactor hall and 300 °C in the handling bay. The problem in choosing the allowable temperature was that no quantitative figures were available on the temperature-dependent release of fission products in this temperature range. Therefore, we will perform an experiment during the start-up of production to demonstrate that up to 300 °C practically no fission products are released from the targets. This is necessary since no fission product release is allowed in the handling bay due to the lack of control of radioactive effluents.

For the reactor hall as well as for the handling bay, dropping the flask has to be considered since the cranes are not designed in accordance with the latest KTA-rule. For such an event in the handling bay, it could be shown that the temperatures of the targets would not exceed the limit of 300 °C. But in the reactor hall, the temperature of 400 °C would be exceeded by about 50 °C. Thus, it was assumed pessimistically that a fraction of 1E-4 of the noble gases and 1E-5 of the iodine escapes from the targets into the reactor hall. The reactor hall will be closed and kept at subatmospheric pressure by the so-called incident filter system. This reduces the iodine discharge into the environment to less than 1 ‰ of the daily allowed value. The same is true of the discharge of noble gases.

5. Time Schedule

Nearly three years will have passed since the customer's first inquiry when production starts this autumn.

The first year was used for the development of the concept and for the clarification of fundamental questions such as the adequate production yield in the positions in question and sufficient cooling of the targets during handling in air. Both these questions were clarified by experiments. This phase lasted such a long time because most of the FZJ experts on the design of irradiation rigs have now retired so that there was a lack of know-how.

When the project was discussed for the first time on the FZJ internal safety committee in February 1997, the supervisory authority as a guest on the committee decided that the project as a whole should be licensed according to the German Atomic Energy Act due to the safety implications the project might have for the reactor itself. The application was made in the first half of June and the safety document submitted at the end of July. TÜV, the technical expert called upon by the authority, issued its draft report at the end of October and the final version in mid February 1998. The license was granted at the beginning of March 1998, after 8 months, which is very quick for German circumstances.

During the licensing procedure, the detailed design was elaborated and the call for tenders started. In parallel, the specifications of the components were discussed with TÜV experts in order to get their approval before the documents could be officially submitted. On this basis, the components and semi-finished products with a long supply time have already been ordered.

The rig, the circuits and the control boards will mainly be installed in the long shutdown period of the reactor scheduled for September and the first half of October. It will be followed by the cold start-up tests, which include the demonstration of all handling procedures in the presence of a TÜV expert. Provided there is a satisfactory outcome of all the tests, the production of molybdenum can be started in the second half of October.

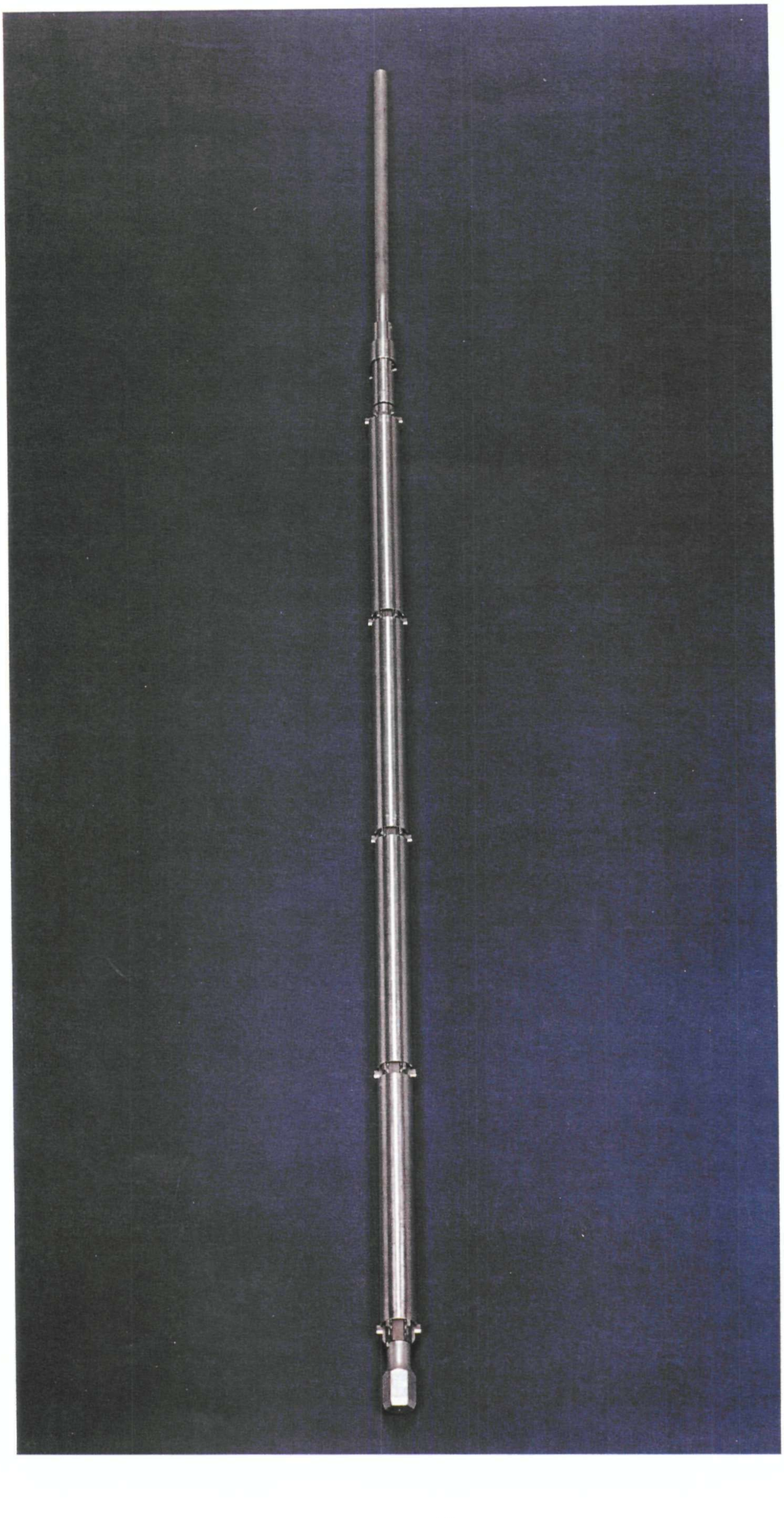


Fig. 1:
Assembled
Target
Carrier

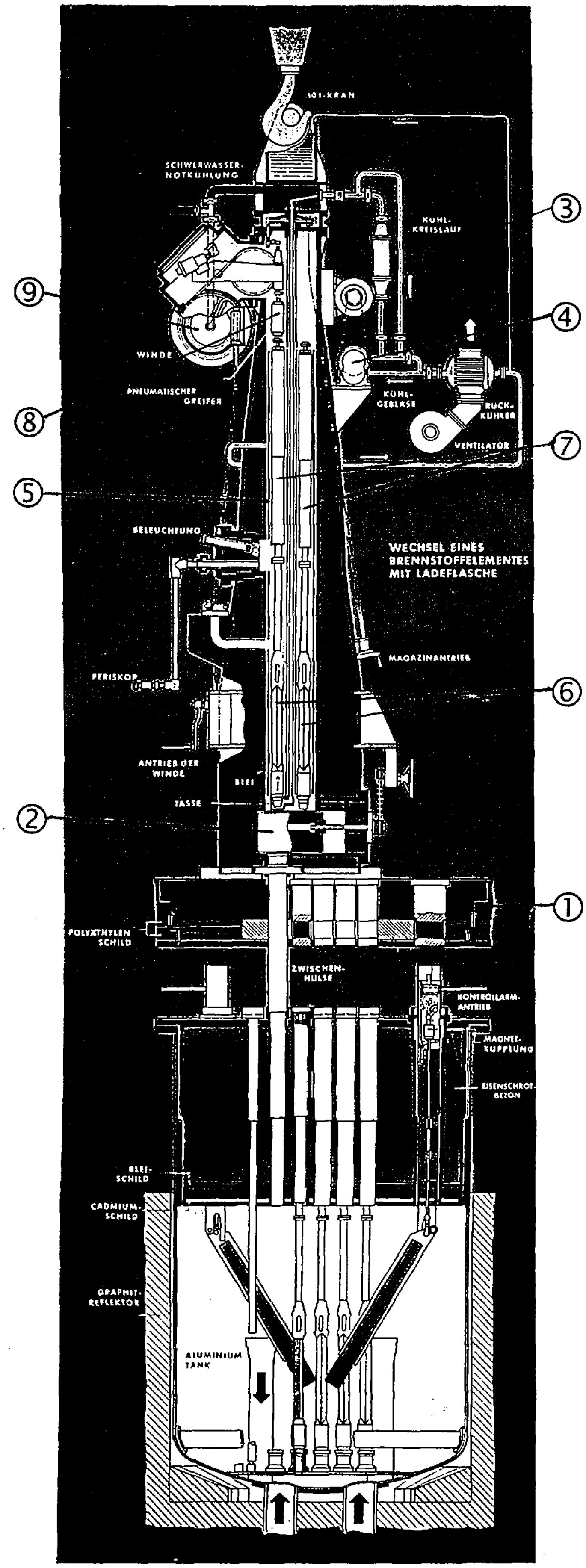
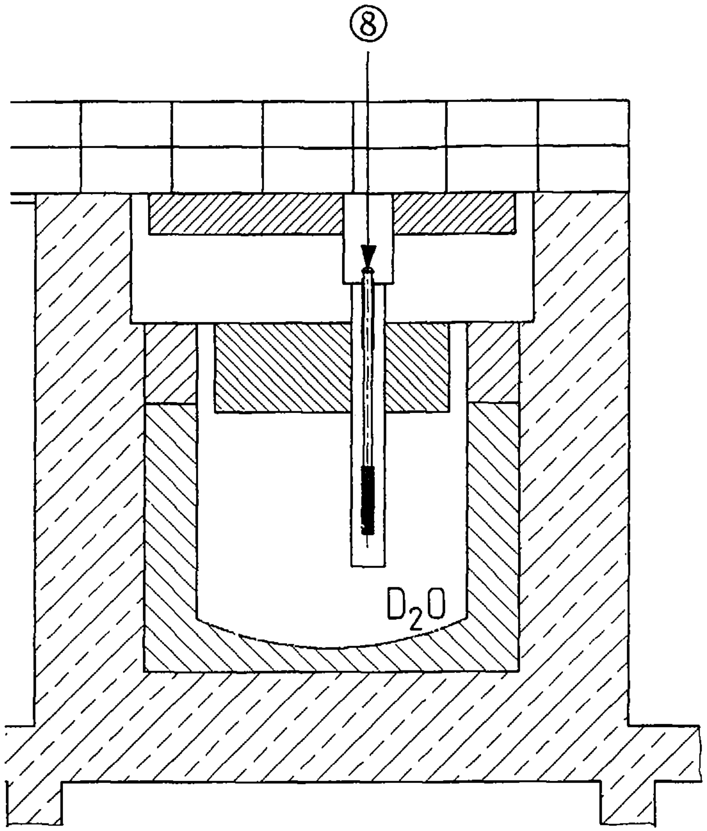
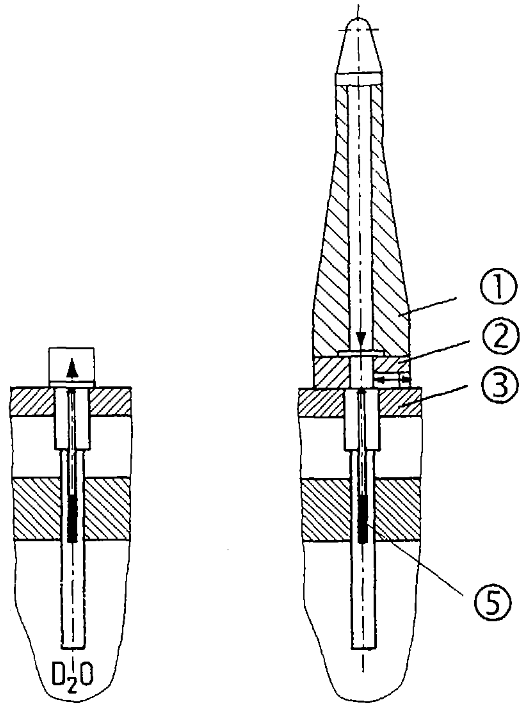


Fig. 2:
Fuel Handling Flask in Operation on Top of the Reactor

①	Reactor Top Plate
②	Flask Gate
③	Cooling Circuit
④	Blower
⑤	Magazine
⑥	Fuel Element
⑦	Shielding Plug
⑧	Pneumatic Grab
⑨	Winch





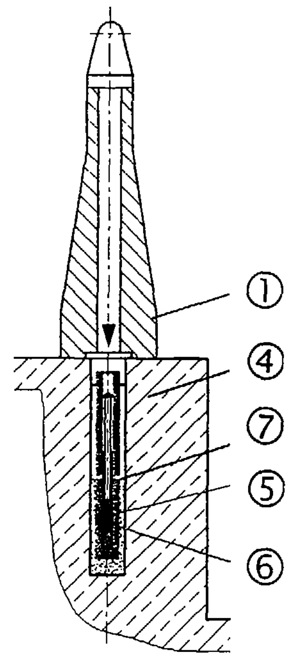


Fig. 3: Unloading the Irradiated Targets from the Reactor and Transport to the Storage Block

① Fuel Handling Flask
② Shielding Device
③ Reactor Top Plate
④ Storage Block
⑤ Target Carrier
⑥ Transport Tube
⑦ Water Level
⑧ Pulling Rod

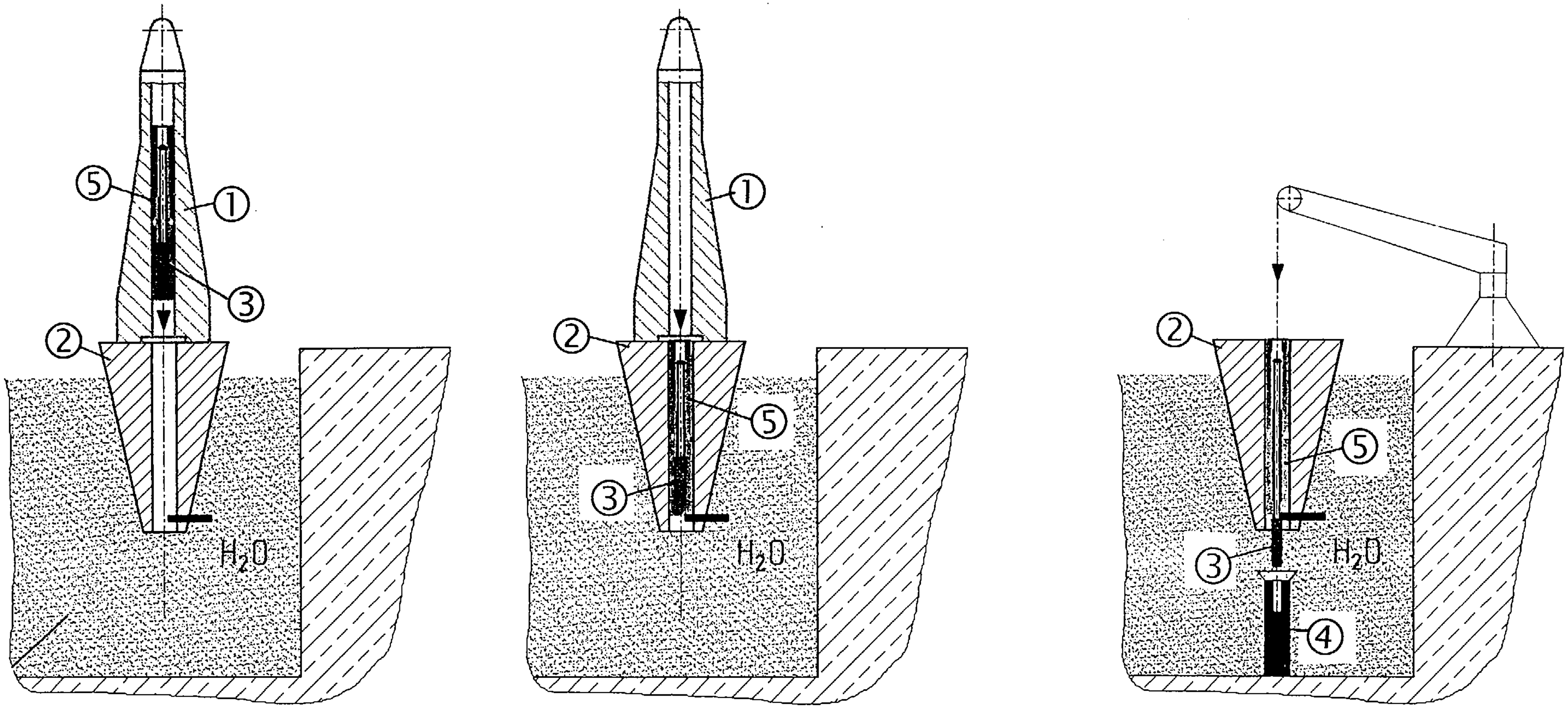


Fig. 4: Loading the Transport Tube into the Shielding Cone of the Water Pool and Unloading the Target Carrier

① Transport Flask
② Shielding Cone
③ Target Carrier
④ Receiving Fixture
⑤ Transport Tube