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Development of a Cold Methane Pellet Moderator for ESS

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Introduction

As part of the work for the target station of the planned European Spallation Source (ESS) the Central Department of Technology at the Forschungszentrum Jülich is also concerned with the moderators, particular attention being given to the development of cold moderators. Whereas in the USA the interest of users is focused on moderators operated at a temperature of 100 K, users in Europe are mainly interested in 20 K moderators.

Research reactors are operated with cold hydrogen or deuterium moderators whose advantage is a time-independent high flux of cold neutrons involving low radiation damage. For short-pulsed spallation sources like the ESS, H_2 and D_2 are less suited since they lead to pulse broadening due to inadequate slowing down properties.

The long-term goal of the concept and test phase only just beginning is the design of an advanced cold 20 K moderator for ESS, which is better by at least a factor of 2 than conventional H_2 moderators with respect to neutron intensity and which shows better slowing down properties in the thermal and epithermal range. The most favourable moderator material for neutrons is considered to be methane, in the liquid form for 100 K moderators and in solid form for 20 K moderators.

Methane as a Moderator

The clearly favourable neutronic properties of methane as a moderator, however, are faced with disadvantages in technical application:

- Methane is radiolysed under irradiation. During this process, energy is stored in the radicals formed and must be specifically removed at regular intervals. This can be achieved by temperature increase /1/, but every time this process has to be carried out measuring time for the user is interrupted.

- Radioactive isotopes are formed from the carbon atoms of methane (or other organic substances) due to spallation. This must be taken into account in treating the spent methane. Emission may not be possible. Known isotopes wirth half-lifes over 1 min are C-11 with a half-life of 20.3 min, Be-7 with 53.4 d, H-3 with 12.3 a and Be-10 with 1.6×10^6 a /2/.

- Neutron radiation leads to polymerization reactions in methane forming wax-like alkanes which deteriorate the moderator properties and also deposit in the moderator system. In the ISIS moderator system operated with liquid methane, wax-like alkanes occur which clog the system and cannot be completely removed by the use of solvents. Experiments are under way to slow down the formation rate of wax-like alkanes by adding further substances (e.g. propane) /3/.

- The heat conductivity of solid methane is low (< 1 mW / cm K). Due to the high heat generation in ESS moderators (~ 7.5 kW), solid methane can not be used as a compact block because the heat can not be removed fast enough, which would lead to evaporation /4/.

The problems described above have led to the following considerations:

- In order to prevent the distribution of wax-like polymers in the moderator and piping system, CH_4 is used as a solid, which is removed without melting so that system contamination is prevented. Designs /5, 6/ that use sheets of solid methane evaporate the methane to empty the moderator container. This may lead to residue in the moderator system.

- In order to transport solid methane in the moderator system, it has been proposed to use this in the form of small pellets /4/.

- In order to ensure adequate cooling of the solid methane the small pellets should be spherical so that a defined fixed bed is formed in the moderator container through which coolant flows in a reproducible way. Designs that feature cooled metal rods in sheets of solid methane have also be published /5, 6/.

- Operating conditions must be established to allow regular warming and recooling of the solid methane to remove the energy stored.

- The problem limiting the undisturbed measuring time when using solid methane is the energy build-up, whose removal leads to frequent down-times, rather than polymer formation, which gradually deteriorates the moderator efficiency. Additions to the methane, as they are proposed for ISIS, can reduce the problem if they diminish radical formation or lead to a faster radical recombination at operating temperature thus lengthening the time between two necessary energy removal phases.

Conceptual Design for a Methane Moderator for ESS

On the basis of the above considerations, a concept for the ESS cold moderator is being examined at the Forschungszentrum Jülich, which is characterized by the following features:

- The moderator is a fixed bed of small spheres. This makes moderator vessel filling homogeneous and reproducible. Since spheres in contrast to pellets of other shapes form a defined packed bed, good and reliable cooling of the moderator bed by H_2 is given.

- The transport of solid moderator spheres from the radiation zone largely prevents system contamination by "wax". A certain degree of abrasion must be expected however, but nothing is known so far about the abrasion resistance of solid methane spheres at 20 K.

- Good transportability of the moderator spheres due to fluidization of the bed in the moderator vessel with subsequent pneumatic transport through the pipes is given.

- The process of filling the moderator container and removing the pellets is batchwise to simplify the loading step and ensure complete removal of the spent pellets, so that no spent methane pellets accumulate in the system.

- The pellets are warmed up regularly to release the stored energy.

- The spent methane pellets are separated from the transport fluid and the methane is either purified and reused or released over the stack.Depending on the kind and amount of the radioactive isotopes present retentive measures may have to be considered.

The development of the concept for the cold source was based on the given target-moderator-reflector assembly of the ESS project. Therefore, considerations on a horizontal discharge of the pellets by a screw conveyor /7/ or swinging the moderator tank out of the beam path /5/ could not be included in the concept.

Fig. 1 shows the target-moderator-reflector assembly with top shielding /8/. The two cold moderators are arranged above and below the target. The cranked coolant pipes routed over several meters through the shielding and reflector have an inside diameter of 25 mm.

The currently valid planning documents provide for hydrogen moderators of approx. 1 l volume. When methane is used as moderator instead of hydrogen, the same amount of hydrogen has to be transported as coolant and the methane pellets must additionally be transported through the coolant pipes to the moderator container. In the chosen target-moderator-reflector assembly, additional space requirements for operating the moderator (like the screw conveyor for pellet discharge or revolving the moderator container out of ist operating position) would lead to a loss of reflector material and reduce the number of beam tubes, so that concepts requiring additional space /5, 7/ are not pursued at the Forschungszentrum Jülich.

Fig. 2 schematically shows the concept for the use of solid methane pellets. CH_4 is solidified at about 20 K and then extruded. In this way, cylindrical pellets are produced which are shaped into spheres in a spheronizer. They fall into a storage vessel from where they are proportioned into the moderator vessel via a star feeder and a stream of hydrogen.

After a given time, which is determined by the extent of the polymerization processes, they are discharged by reversing the hydrogen coolant flow and subsequently separated from the hydrogen. The methane can be purified and fed back to the solidification stage.

Alternatively, the spent methane could be emitted. In this case, the emission limits for radioactive isotopes produced are to be observed.

Not included in Fig. 2 is the configuration for the H_2 circuit during a pellet warming cycle for energy removal. It is being contemplated to temporarily send a warmer hydrogen flow, whose temperature must, of course, be below the melting point of methane, through the fixed bed of pellets so that the pellets are warmed to approx. 50 K. The process has to be optimized for rapidity, because during the warming/cooling cycle no measurments are possible.

Specific Tasks for the Development of the ESS Methane Moderator

The concept for the cold moderators contains many unsolved problems which need to be tackled step by step.The activities planned for the next three years will set in at several points:

1. Production of CH_4 pellets
   - from pure methane
   - from methane hydrate
   - from adsorber materials such as zeolites or porous substances by loading with methane

For the production of solid methane pellets it is helpful to look at fusion research were solid deuterium pellets are injected into the plasma. Extrusion cryostats are used to produce these pellets. Under changed temperature conditions these devices can also be used to produce methane pellets instead of deuterium pellets. Fig. 3 shows the cryostat adopted from fusion research. Methane is condensed in the given volume in the vertically arranged cryostat. The solid condensate is then slightly warmed and ejected downwards through a nozzle by a piston. A cutting device serves to cut pellets of the desired length.
The facility is under construction. It will serve to test and, if necessary, optimize the fabrication principle as well as to produce pellets for the COSY tests (see below). For use in ESS these cylindrical pellets would have to be rounded in a further step.

Methane hydrate is regarded as an alternative to solid methane. Gas hydrates are ice-like crystalline structures of a water lattice with cavities which contain guest gases.They are bonded by van der Vaals forces under low temperature and moderate pressures. The guest molecule is necessary to support the cavity. In the natural environment methane is the most common guest molecule /9/. It is being contemplated to have this compound produced at a university institute and test it in the COSY experiments (see below).

Furthermore, it is being considered to use methane adsorbed on porous bodies such as zeolites or porous polymer spheres. This should substantially simplify the production process and handling of the moderator pellets.

Irrespective of the neutronic properties of the different moderator pellets mentioned above, their mechanical properties are to be evaluated since the pellets are exposed to considerable mechanical loads during transport in the moderator system. Another task is to evaluate the behaviour in the radiation field with respect to energy build-up, polymerization processes and the formation of radioactive isotopes.

2. Testing the transport phenomena of spheres in the moderator vessel and pipe systems at room temperature under similarity conditions.

A particular problem is the transport of moderator pellets in the moderator system. They must not be broken when charged into the moderator vessel, because only a-bed of spheres can guarantee an even cooling without hotspots, and they should not be destroyed during discharge, so that they can be easily separated from the coolant and to prevent fine particles from depositing in the system. The pellets must be quantitatively discharged so that no spent material accumulates in the moderator container, and the process should be carried out in a very short time so that the beam time is not significantly shortened. If the residence times of liquid methane in the ISIS moderator /10/ are used for comparison, it must be assumed that the methane in the ESS moderators may have to be replaced once a day.
The first transport tests will be carried out under similarity conditions at room temperature. A Plexiglas apparatus that allows visual inspection was set up.

3. Design and construction of the moderator test facility at JESSICA on the COSY cooler synchrotron of the Forschungszentrum Jülich.

COSY is a 2.4 GeV proton synchrotron at the Forschungszentrum Jülich. Initial experiments have shown that it is possible to completely extract the beam in one circulation /11/. This enables the setup of a moderator test facility where the performance of various moderators (solid methane pellets, methane hydrates, zeolites loaded with methane) can be measured under realistic ESS target geometries in the JESSICA facility (JESSICA = Jülich Experimental Spallation-Target Setup in COSY Area). Fig. 4 schematically shows the procedure: moderator pellets are externally produced in the laboratory (here as an example the production of solid methane pellets in the extrusion cryostat), transferred into the experimental hall of COSY and filled into the moderator vessel in a glovebox under inert gas. The pellets are kept in liquid hydrogen in the moderator vessel. A circulation cooling is not necessary because only single pulses are measured so that the heat generation is low. The moderator vessel is cooled by a cold head.

Summary

Work for moderator design concentrates on the development of a concept for a ESS cold moderator system assuming that spherical moderator pellets will be available for the process, irrespective of the type of moderator ultimately selected. The choice of a moderator results from neutronic and technical aspects, to enable the safe and functional handling of the moderator pellets and, at the same time, satisfy the neutronic requirements.

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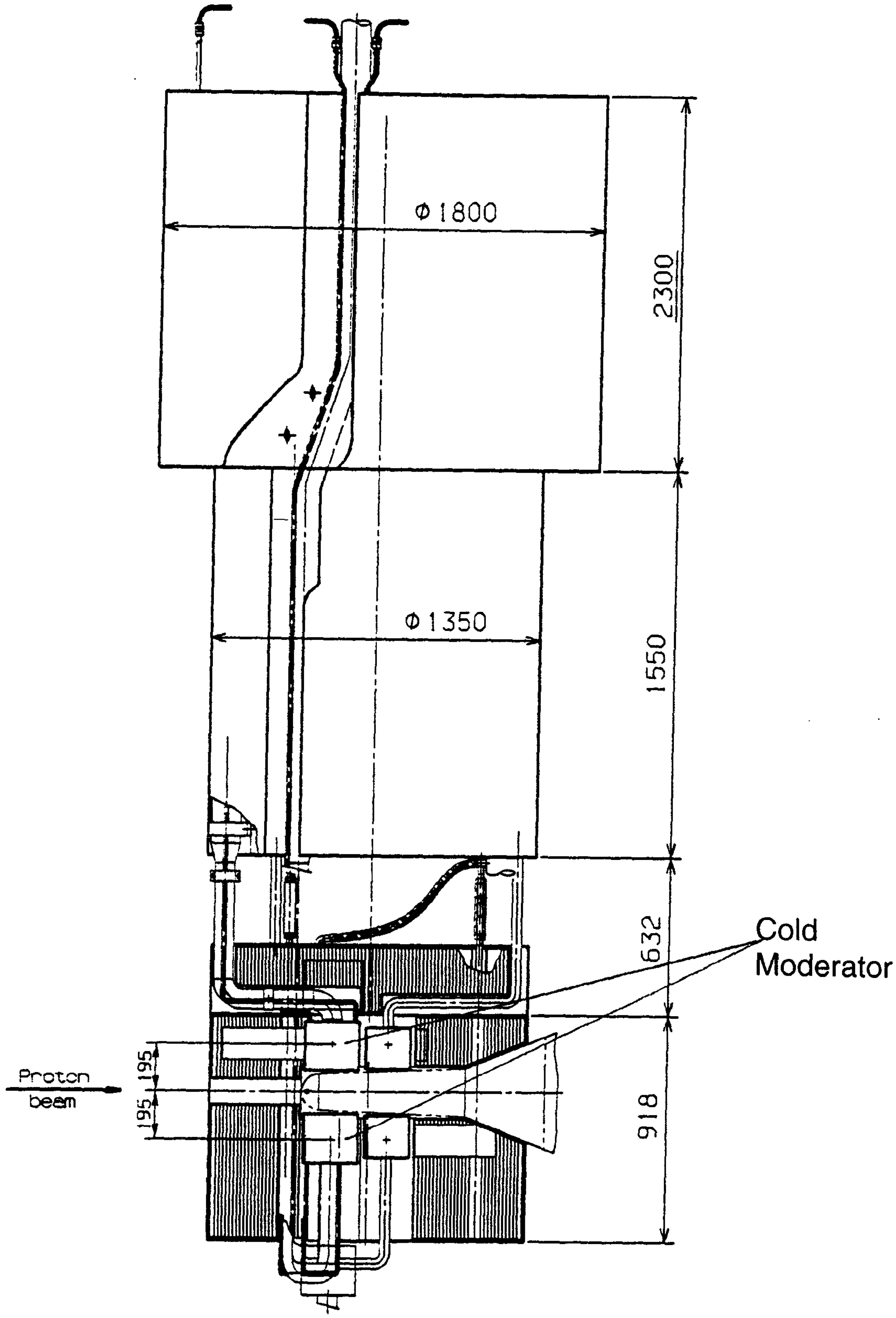
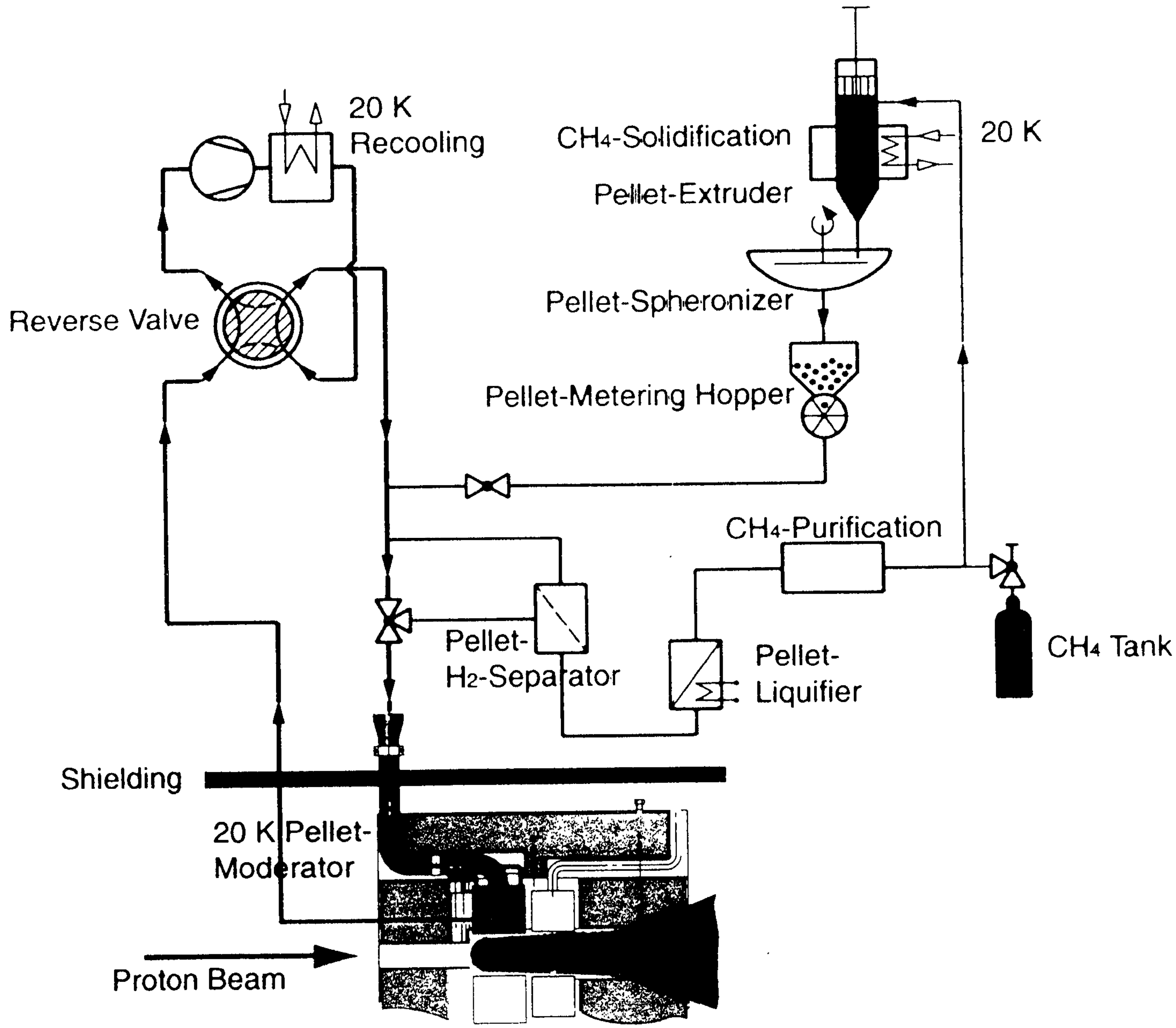


Fig. 1 : Schematic of the ESS Target-Moderator-Reflector-Assembly

Fig. 2 : Concept of the 20 K Methane Pellet Moderator System



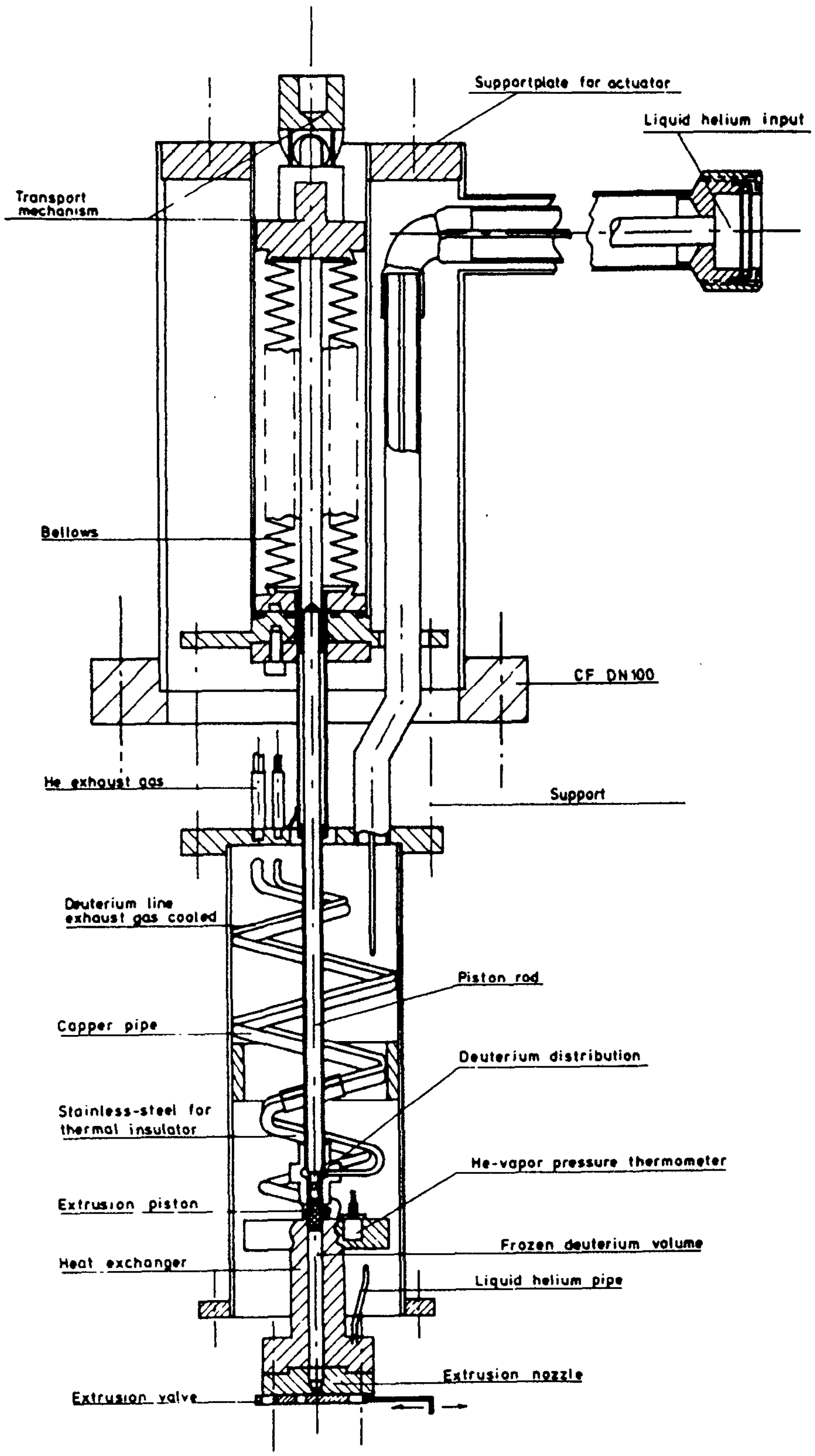


Fig. 3 : Extrusion Cryostat for the Production of Solid Methane Pellets

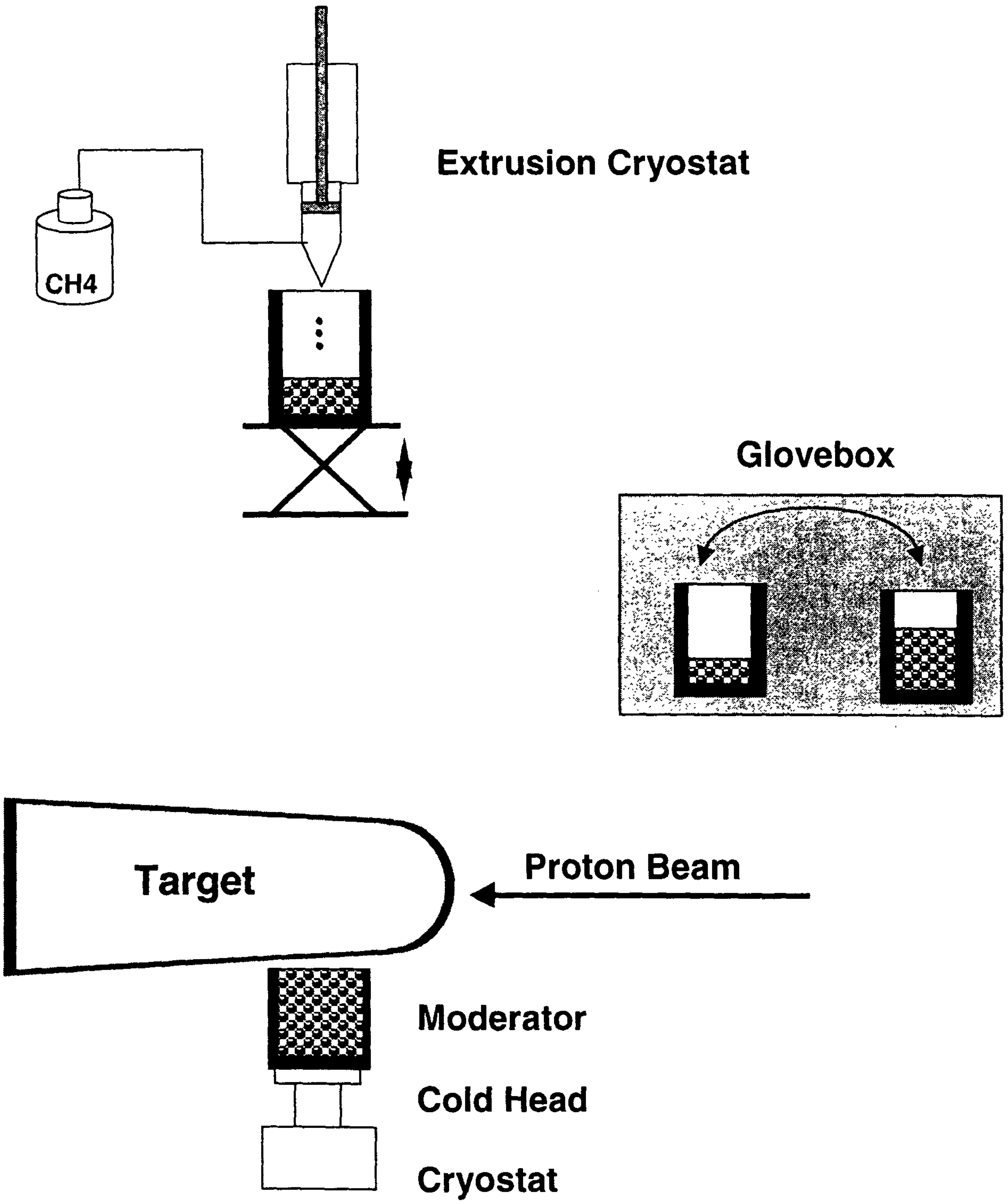


Fig. 4 : Moderator Tests at COSY