Forschungszentrum Jülich

Zentralabteilung Technologie

Conceptual Design of a Cold Methane Moderator System for the European Spallation Source (ESS)

Heidrun Barnert-Wiemer

Jül-3967

Conceptual Design of a Cold Methane Moderator System for the European Spallation Source (ESS)

Heidrun Barnert-Wiemer

Berichte des Forschungszentrums Jülich ; 3967
ISSN 0944-2952
Zentralabteilung Technologie Jül-3967

Zu beziehen durch: Forschungszentrum Jülich GmbH · Zentralbibliothek
52425 Jülich · Bundesrepublik Deutschland
☎ 02461/61-5220 · Telefax: 02461/61-6103 · e-mail: zb-publikation@fz-juelich.de

Conceptual Design of a Cold Methane Moderator System for the European Spallation Source (ESS)

Heidrun Barnert-Wiemer

Forschungszentrum Jülich GmbH
Zentralabteilung Technologie

ABSTRACT

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 GmbH is also concerned with the moderators, particular attention being given to the development of cold methane moderators.

This report discusses the technical feasibility of solid methane moderators. Methods to tailor the neutron output by adding absorption materials (decouplers or poisons) are not considered here, neither are composite moderators.

Based on the given target-moderator-reflector assembly of the ESS project a concept for the ESS cold methane moderators has been developed and is being examined at the Forschungszentrum Jülich.
According to this moderator concept the moderator is a fixed bed of small spheres, which makes moderator container filling homogeneous and reproducible. Since spheres form a defined packed bed, cooling of the moderator bed by H_2 is reliable. The process of filling the moderator container and of removing the pellets is batchwise to ensure complete removal of the pellets, so that no spent methane pellets accumulate in the system. For removal of the moderator spheres the fixed bed in the moderator container is fluidized with subsequent hydraulic transport of the pellets.
The spent methane pellets are separated from the transport fluid and the methane is released over the stack or purified and reused. Depending on the kind and amount of the radioactive isotopes present these may have to be separated and stored.

Table of Contents

1. Introduction

Spallation neutron sources are being built to satisfy the needs of condensed matter science requiring intense pulses of neutrons. Accelerated protons strike a target and fast neutrons are produced by spallation in the target. They are slowed down by an assemly of moderators and reflectors.

The only existing continuous spallation source SINQ is in operation at the Paul Scherrer Institut in Switzerland / BAUER 97 /. The presently operating pulsed spallation sources are listed in Table1 /CARPENTER 90a /.
In the planned European Spallation Source (ESS) the design calls for a proton beam energy of 1.3 GeV and a mercury target . The reflector material will be lead which is cooled by water. Of the four moderators two are planned to be water at ambient temperature and two will be cold (~20 K) ones / ESS 96/.

Cold neutron moderators have come to be an essential ingredient for any new neutron souce to be built in the future and they have been retrofitted to older neutron sources if at all feasible. Research reactors are operated with cold hydrogen (protium 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, protium and especially deuterium are less suited since they lead to pulse broadening due to inadequate slowing down properties.

The long-term goal of the present moderator concept and test phase 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.

2. Cold Moderator Materials

The function of moderators is to reduce the energy (or temperature) of neutrons to levels that are interesting for different instruments. A great advantage being that these neutrons can be transported through guidelines to the experiment stations. Different kinds of moderators allow for thermal, subthermal, or epithermal neutrons. Cold moderators, also called cold sources, slow the neutrons down from the MeV into the meV region and thus provide a flux of very low energy neutrons (< 10 meV) at an effective temperature of below 40 K.

When a neutron loses energy, the wavelenght is increased and the energy distribution is narrowed. Neutrons being uncharged do not directly interact with orbital electrons but lose their energy primarily by interaction and elastic collisions with atomic nuclei. Protons are most efficient for slowing down fast neutrons because their mass is the same as that of a neutron and because their low energy neutron scattering cross section is larger than that of any other practical species.The maximum energy E_t that can be transferred is given by

$$E_t = 4\,M / (M + 1)^2$$

For a hydrogen atom, M = 1, and therefore all the neutron energy may by transferred to the atom. For a carbon atom, M = 12 and the fraction of energy transferred is 0.28.
It follows from the above that hydrogenous materials are preferred for moderating neutrons / EVANS 95 /.

Many research reactors are outfitted with cold (~ 20 K) neutron sources, whose moderator material is either H_2 or D_2 or mixtures of both. Deuterium has the advantage of a good scattering quality combined with low absorption. But because of the mean free path of the neutrons of > 100 mm in D_2 bigger moderator volumes than with hydrogen are necessary. In hydrogen collisions are frequent (mean free path < 20 mm) but absorption of neutrons is higher by a factor of

~ 300. Activation in hydrogen is lower than in deuterium because tritium is formed only from the ~ 140 ppm deuterium normally present in hydrogen.
Deuterium cold sources are not in use for short-pulsed spallation sources, because the required moderator volume of several liters leads to pulse broadening.

The use of hydrogen as moderator poses a further problem : the ortho-para conversion. At room temperature only one fourth of the H_2 molecules is of the para type, but with lower temperatures the equilibrium moves to higher concentrations of para – H_2 . At 25 K more then 98 % of the molecules are para-hydrogen. With each conversion of an ortho- to a para- molecule 1.47 meV are released (at 25 K). Below a neutron energy of 100 meV the mean neutron scattering cross-section depends strongly on the ratio of ortho and para H_2 as can be seen in Fig. 1 / AXMANN 84 /, where scattering cross sections are are given for p-H_2 and a mixture of 50% p-H_2 and o-H_2.

Based on the amount of protons in a material, potential cold moderator materials are listed in Table 2 / CARPENTER 90a /. Carpenter goes on to discuss the merits of the candidate materials : „Water (ice), polyethylene, and metal hydrides in and of themselves are not effective cryogenic moderator materials because they have too few low frequency modes to provide efficiently for the final stages of thermalization, although their proton densities are quite high. Liquid hydrogen is quite good in this regard, but the large (15 meV) rotational level spacing makes for large differences in the scattering between the low temperature para form and the normal ortho-para mixture; its low proton density leads to broad pulses of short wavelength neutrons. Liquid and solid methane are very attractive because of their good low temperature thermalizing power, and have high proton densities as well. Ethane and propane are similarly attractive, with even lower rotational level spacings than methane, but have fewer rotational modes per proton than methane. Mellitine and mesitylene have freely rotating methyl groups but even fewer low frequency modes per proton and lower proton densities.“

In the course of moderator studies NH_3 has also been considered / FLUHARTY 69, GRAHAM 72 /. The proton density is higher than that of methane by about 14%, so pulse widths should be shorter compared to a methane moderator of the same dimensions. In measurements at 77 K NH_3 was found to be inferior to CH_4, presumably because of neutron capture by N-14. This problem might be solved by substituting N-15 for N-14. Substituted ammonia is commercially available.

3. Methane as Moderator

Early on the advantages of methane over hydrogen have been recognized. The proton density (protons/ $Å^3$) for H_2 is 0.042 at 20 K and for CH_4 the value is 0.079 respectively / CARPENTER 90 /. These numbers show that methane represents a dense source of hydrogen. Additionally, because of rotational freedom of the molecules, methane will effectively exchange energy with even low-energy neutrons. It has been postulated though, that in solid methane due to lattice bonds only part of the molecules are available for rotational modes.

Experimental data / KIYANAGI 90, CARPENTER 90 / show the advantages of solid methane over hydrogen concerning the intensity and the pulse shape of cold neutrons leaving the moderators (Fig. 2 and 3).
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 in lattice defects (Wigner effect). The energy must be specifically removed at regular intervals. Otherwise at a certain level the energy will be set free in a spontaneous recombination reaction whose nature is not yet understood / CARPENTER 87 /. Due to the rather weak bonds in the methane molecule it can be assumed that the contribution of the lattice relaxation (Wigner effect) is small compared to the energy of the radical reaction.

- Radiation leads to polymerization reactions in methane forming wax-like alkane homologues, which deteriorate the moderator properties and also deposit in the moderator system. In the end the alkanes will be reduced to carbon and hydrogen because of the ongoing abstraction of hydrogen / PYNN 90 /. It is doubtful whether alkanes can be removed from the moderator system by rinsing with organic solvents.

- The moderators are exposed to a high radiation field and radioactive isotopes are formed from the atoms of the methane or methane hydrate. This must be taken into account in treating the spent methane, because it may not be allowed to release the radioactive isotopes into the environment. No reliable data are yet available on the kind and amount of the isotopes to be expected.

- The heat conductivity of solid methane is low (< 4 mW / cm K below 32 K). This leads to a temperature gradient in the solid methane / CARPENTER 87 /. Due to the high heat generation in ESS moderators (~ 7.5 kW in each moderator), solid methane can not be used as a compact block because the heat is not conducted fast enough to the surface to be removed by the coolant. This would lead to melting and finally evaporation of the methane.

- Material data for solid methane are not complete. It is feared that - even at 20 K - methane pellets might be too soft to withstand transport and handling. An alternative is seen in using methane hydrate.

Solid methane moderators are in operation at Argonne National Laboratory (IPNS-facility)/ USA, at the Neutron Science Laboratory KENS (KEK-facility)/ Japan, and at the pulsed reactor IBR-2 at the Joint Institute for Nuclear Research in Dubna / Russia. They are all solid slabs of methane. This is possible, because the energy input is low (Table 1). Why this design can not be used for ESS will be discussed in chapter 5.

3.1 Radical formation and chemical reactions

At the Rutherford Appleton Laboratory / UK a spallation source (ISIS) is in operation, where, besides other moderators, a 100 K liquid methane moderator is in use / BROOME 97 /. Serious operational problems are associated with radiation damage to methane:

- Hydrogen production. This leads to temperature and density variations which are of concern to the users.
- Formation of high molecular weight hydrocarbons $–(CH_2)_n$--, which leads to blockage in the system.

The mechanism for the production of hydrogen gas and „wax“ is considered to be

$$\text{Energy} + CH_4 \rightarrow C^*H_3 + H^*$$

Followed by

$$H^* + C^*H_3 \rightarrow H_2 + C^*H_2$$

Which is the mechanism for production of H_2 gas. The C^*H_2 then recombine to form „wax“ (CH_2)$_n$. H_2 production by recombination of H^* radicals is suppressed by energy considerations

$$H^* + H^* \rightarrow H_2 + 104 \text{ kcal}$$

Recombination of C^*H_3 with H^* to form CH_4 has low probability as does the reaction

$$C^* H_3 + C^* H_3 \rightarrow C_2H_6$$

It is assumed that the build-up of „wax“ can be limited by adding a chemical with which the H^* radical will react in preference to the C^*H_3 radical. A molecule with an active double bond such as propane (C_3H_8) might be suitable.

The volume of hydrogen production is related to the dose in Gy (Joules/kg) by the „G-Value“ (µmoles/Joule). For ISIS conditions (liquid methane, 100 K, beam current 200 µA) per 100 cm^3 of methane this will give 19 cm^3 / h for a dose rate of 0.03 MGy/h. The corresponding production rate of „wax“ is 0.09 g/h.

At the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory / USA a solid methane moderator is in operation at temperatures as low as 7 K. The otherwise effective performance was marred by roughly periodic temperature excursions during or shortly after continuous operation / CARPENTER 87 /. After temperature excursions, H_2 was seen in the cover gas above the moderator.
It is discussed whether aluminium or methane could be responsible for the observed effects. Radiation energy accumulates in the aluminium as Frenkel (vacancy – interstitial) pairs. But it is concluded that the damage energy is not enough to explain the measured temperature excursions. Thus methane must be involved.
It is assumed that fast neutrons and the ensuing damage cascade in irradiated methane produce monoatomic hydrogen, CH3 radicals, and other reactive species, some of which accumulate in interstices of the solid CH4. In a test run the contents of the methane moderator at IPNS were evaporated after 1 week of irradiation. Gas chromatography analysis indicated about 5.4 mol% H_2 and 2.3 mol% C_2H_6, with smaller concentrations of other hydrocarbons, the bulk being CH_4. A crude calculation based on the fraction of hydrogen evolved and an estimation of n-p collisions comes to the conclusion that the reactions

$$H + H \rightarrow H_2$$

$$CH_3 + CH_3 \rightarrow C_2H_6$$

are the main contributors to the temperature excursions. Reasoning from the close relationship between the energy equivalent of the radiolysis products and the calorimetrically observed energy release, it is asserted that released energy is stored as chemical energy in H and CH_3 , although energy stored in lattice defects or distortions remains a possibility whose magnitude could not be assessed.

It is postulated / SHABALIN 97 / that „as radical recombination reactions (RRR) are exothermic (the energy release in the

$$CH_3 + CH_3 \rightarrow C_2H_6$$

reaction is 336 kJ / mol), the presence of CH_3 radicals in irradiated methane is the cause of the stored energy, similar to Wigner energy in irradiated graphite". Shabalin considers this reaction to be a „thermal explosion". Whereas Beljakov et al. / BELJAKOV 93 / postulate that the „fast RRR has a nature other than thermal explosion, maybe, of a chain reaction", because the activation energy should be ~700 K for this case, but was found to be only ~ 290 K. The authors found the production of CH_3 radicals to be one third as much as the H_2 production rate.

In experiments on the formation of atomic as well as molecular hydrogen via interaction of galactic cosmic-ray particles with extraterrestrial organic ices Kaiser et al. / KAISER 97 / irradiated methane ices between 10 and 50 K with 9.0 MeV α-particles and 7.3 MeV protons. It was found that more than 99% of the energy is transferred via inelastic interactions to the electronic system to form electronically excited CH_4 molecules decomposing to a CH_3--H radical pair. Two H atoms recombine in a diffusion limited step to H_2. Further, secondary dissociation of CH_3 to H and CH_2 contributes to the H production. Fourier transform infrared spectroscopy (FTIR) and quadrupole mass spectroscopy (QMS) analyses indicate that if these energy-loss processes accumulate up to 6 ± 3% H atoms in the CH4 target, more than 90% of the methane ice is released in an explosive ejection into the gas phase.
In experiments where oxygen was present these explosions were avoided

/ KAISER 95 /, because molecular oxygen serves as radical scavenger by reacting with hydrogen to form O_2H followed by conversion to H_2O_2 and H_2O according to the equations

$$O_2 + H \rightarrow O_2H$$

$$O_2H + H \rightarrow H_2O_2$$

$$H_2O_2 \xrightarrow{\alpha} 2HO$$

$$HO + H \rightarrow H_2O$$

CH_4 / O_2 frosts formed brown to yellow residues comprising up to 5% of the deposited carbon, whereas pure methane targets never yielded any residues after heating the irradiated frosts to 393 K. FTIR spectra of irradiated samples varied widely in the IR absoption region of 3800 – 2800 cm^{-1}, i.e. stretching vibrations of hydroxyl groups (-OH) in alcohols, carbonyl acids, and water in spite of identical experimental parameters. It was found that this was due to experimental errors in preparing CH_4 / O_2 mixtures , i.e. an absolute error of 0.5% oxygen increased the yields of selected carbonyl groups up to 1000%.

In a first set of experiments solid methane was irradiated at 77 K in a closed cavity with MeV protons and helium ions from the CV28 compact cyclotron of the Forschungszentrum Jülich / ROESSLER 99 /. The formation of polycarbon molecules up to coronene was followed off line by gas and liguid chromatograpy. In a second set of experiments at 15 K in a 10^{-6} mbar vacuum of a cryostat the product formation was followed off line by infrared spectroscopy in transmission at 15 K and by NMR and SEM of the carbonacious residues after evaporation. At a dose of 17 eV per carbon atom by 17MeV helium ions up to 2% of the methane were converted into a dark matter consisting of hydrocarbons with H/C- ratios ≤ 1 (PAHs and a-C:Hs) and amorphous carbon.
A third set of experiments was carried out at 10 K in a 10^{-9} mbar vacuum of the CSR chamber at the Forschungszentrum Jülich. The product formation was

followed in situ by mass spectrometry of the molecules emitted from the surface and by FTIR spectroscopy in reflectance. The results of the previous experiments were corroborated and in addition vigorous explosions were observed at doses of approximately 145 eV per carbon atom by 13 MeV helium ions. The dose corresponds to the radiolytic formation of 6% H_2 . By analysis of the residues it could be shown, that the explosions were due to the recombination of H atoms to molecular hydrogen. At 10 K the migration of H atoms in solid methane is relatively limited and recombination seems only to be possible if their concentration is high enough that they can interact without far reaching diffusion.

It can be seen from the above that the chemical mechanisms involved with the use of methane in a radiation field are not yet understood. From the fact that explosive reactions of radicals do not happen at the liquid methane moderator at ISIS, it can be judged that it is a problem of solid methane moderators, where the recombination of species is hindered by the low diffusion rate, so that a concentration of radicals builds up that leads to an explosion at a concentration of ~6 % hydrogen. The temperature to which the solid needs to be warmed regularly - before an explosive concentration is reached – has not yet been determined with certainty, but is believed to be around 60 K. The IPNS methane moderators, which are operated at 28 K are melted to 90 K every two to three days to anneal out the stored energy and remove the accumulated hydrogen / IPNS 01 /.

Also unknown are the radioactive isotopes that will have to be dealt with when methane is used as moderator. Unpublished data indicate that tritium, C-11, Be-7, and Be-10 might be produced in amounts that shall prohibit the release of the spent methane into the environment.

3.2 Material data of solid methane

The data base for solid methane is small. A number of publications, especially those from the Kharkow Institute of Low Temperature Physics, are available only in russian.

3.2.1 Thermodynamic properties

Under equilibrium vapor pressure solid methane can exist in two phases depending on temperature : phase I at 20.49 K $\leq T \leq$ 90.68 K and phase II at $T \leq$20.49 K. Both have fcc-structures , phase I with 4 molecules per unit cell and phase II with 32 molecules per unit cell / BOLSHUTKIN 71, PROKVATILOV 82 /. The phase transition temperature has been determined to be 20.49 ± 0.01 K and the enthalpy of transition to be 10.6 cal/mol / COLWELL 63 /. The difference in molar volume at phase transition is ΔV = 0.11 ± 0.01 cm^3/mol / KRUPSKII 74 /. The heat capacity between 2.3 and 90 K is given in / COLWELL 63 /.

Thermal conductivity has been measured by / MANZHELII 68 / between 22 and 80 K and by /GERRITSEN 42 / between 19.9 and 20.6 K.

Molar volume, density, and thermal expansion between 10 and 90 K can be found in / PROKVATILOV 83 /.

Saturated vapor pressures over solid methane between 51 and 90 K are given in / MALKOV 85 /. The handbook also contains a diagram where vapor pressures of additional technical gases are given.

3.2.2 Mechanical properties

Stress-strain curves were measured between 4.2 and 77 K by / BOLSHUTKIN 68 /. The data were used to estimate the mechanical properties characterising strength : ultimate strength, deforming stress, relative elongation.

The hardness was studied by using a conical indenter / BOLSHUTKIN 65 /at temperatures between 4.2 K and 77 K. Measurements at 64 K and 77 K showed methane to be plastic and the indenter creaping steadily and rapidly. This effect decreases with falling temperature.
In / TOWLE 67 / the temperature dependence of of the shear strength is given. The diagram shows the shear strength in bar versus the temperature normalised with the melting temperature.

The velocities of ultrasonic sound, both longitudinal and transverse, have been measured at different frequencies and used to calculate compressibilities / TARASENKO 75 /.

3.3 Explosion limits of gaseous methane and hydrogen

The whole moderator system contains methane and hydrogen in various states of aggregation and most of the time both are also mixed. Both gases are flammable. In case of a leak and consequent mixture with air explosions are possible if a source of ignition is present. Conditions for the propagation of flame in mixtures of gases are discussed at length in / COWARD 52 / and / BARTKNECHT 93 /.
Fig. 4 and 5 show the limits of flammability for H_2 and CH_4 in air.
The limits of flammabilty for mixtures of H_2 and CH_4 in air have been calculated according to / BARTKNECHT 93 / and are given in Fig. 6.

4. Target - Moderator - Reflector Assembly for ESS

Fig. 7 shows the target-moderator-reflector assembly with top shielding / ESS 96 /. The mercury target is positioned horizontally within the watercooled lead reflector. In the reflector voids are left for four moderators and the beam extraction tubes.The two cold moderators are arranged above and below the target in front position. The cranked coolant pipes to and from the cold moderators are routed over several meters through the shielding and reflector. They have an inside diameter of 25 mm and are designed in triple containment mode like all hydrogen containing equipment.

The currently valid planning documents provide for 20 K hydrogen moderators of approx. 1 l volume. In the chosen target-moderator-reflector assembly, additional space requirements for operating advanced moderators would lead to a loss of reflector material and reduce the number of beam tubes. For this reason a methane moderator system is proposed (chapter 6) that fits into the space given by the present design for hydrogen moderators. Concepts requiring additional space / LUCAS 95, JESSEN 98 / have so far not actively been pursued at the Forschungszentrum Jülich. When methane pellets are used as moderator instead of hydrogen, the same amount of hydrogen still has to be transported as coolant for the pellets and the methane pellets must additionally be transported through the coolant pipes to the moderator container. This will put a severe mechanical strain on the pellets and it will have to be tested whether they can withstand this strain. It was the fear that methane is too soft, that led to the ideas of using methane hydrate, which is harder than methane, or methane condensed in zeolites or highly porous plastic spheres.

5. Design requirements for methane moderator systems

A cold source system includes the necessary hardware to maintain the defined moderator operating conditions, which enable a cold moderator to slow thermal

neutrons and produce an energy spectrum and wavelength distribution in a range defined as cold or very cold.
To this end the moderator has to be thermally and chemically stable at operating temperature under high radiation conditions.

The requirements and the consecutive problems described above have led to the following general 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 / JESSEN 98, BELIAKOV 97 / that use sheets of solid methane evaporate the methane to empty the moderator container. This leads 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 / WILLIAMSON 88 /.

- 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. If pellets form badly cooled clusters these can melt. It has been calculated that spheres of approx. 20 mm diameter will melt at a volumetric heat generation of 10^6 W / m^3 / LUCAS 95 /.

- Designs that feature cooled metal rods in sheets of solid methane have also be published / JESSEN 98, BELIAKOV 97 /. In these designs methane is removed from the moderator container by evaporation.

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

- Undisturbed measuring time is limited by the energy build-up in the methane, whose removal by heating with consequent recooling leads to frequent down-times, whereas polymer formation deteriorates the moderator efficiency gradually. Additions to the methane can reduce these problems 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.

Based upon the above said, spheres of 2 – 2.5 mm diameter were choosen for the ESS concept (Chapter 6). These spheres should facilitate pneumatic transport / SIEGEL 91 / while the temperature distribution in the spheres during irradiation is nearly uniform, which cannot be upheld if particles are too big, due to the low heat conductivity of methane.

5.1 Pellet production and transport concepts

For the production of solid methane pellets it is helpful to look at fusion research were solid deuterium pellets are injected into the plasma / JET 96 /. 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. 8 shows the extrusion cryostat adopted from fusion research. The vacuum jacket is removed. This extruder had been used for making deuterium pellets before it was replaced. Data of this extruder have been lost. It was outfitted at the Forschungszentrum Jülich with LN_2 cooling. Methane is condensed in the given volume (diameter 4mm, length 30 mm) 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 / BARNERT-WIEMER 98 /.
This extruder has been fitted with a LN_2 cooled discharge unit to collect the pellets (Fig. 9). It was used to test the fabrication principle. The pellets were 4mm in diameter and 25 mm long and were transparent. But with one pellet every ten

minutes the throughput of this unit was too small to produce pellets for the JESSICA experiment (Chapter 7). Therefore the further development was stopped after the feasibility had been proven. A new helium cooled extruder has been designed / STELZER 01 /. For use in JESSICA pellets can be cylindrical but for use in ESS these cylindrical pellets would have to be rounded in a further step. A description of of the whole moderator system is given in chapter 6.

Tests to remove spherical pellets from a transparent model of a moderator container were made at room temperature with glass spheres of 3 mm diameter and water as fluid / BARNERT-WIEMER 99 /. Distributor plates were tested with the goal to the optimise the system for fast and quantitative removal of pellets. In parallel a study was made to define similarity conditions for further experiments / TEICHMANN 99 /. Archimedes and Reynolds numbers were calculated for the methane – hydrogen system. Similarity conditions were established for a system of water at 60°C and lead spheres. The lead spheres should be 1.5 times as large as the methane spheres. To adapt the geometrical dimensions of the model container according to the bigger spheres the hydraulic diameter of the model container has to be 2.2 times bigger than the original and the water throughput thus will have to be 7.7 times as large as the hydrogen flow.

A different approach, which is also derived from fusion research, is followed by Cryogenic Applications F, Inc. / FOSTER 00 /. A test apparatus produced 3mm hexagonal cylindrical pellets. A 100mm x 100mm stainless steel honeycomb was soldered to a copper plate, which was cooled by a refrigerator. The temperature was controlled by resistance heaters. The pellets were formed by condensing methane gas. The honeycomb was mounted on top of the pellet growing chamber so that the pellets dropped down, when the honeycomb was slightly heated. The pellets were grown in 28 min. They were wellformed and transparent, but sticky at 70 K and no method was found to separate the pellet clump.
Plans are under way to build a pelletizer capable of producing 1.5 l of pellets per hour. It is to be outfitted with subcooler which shall cool the pellets from the formation temperature near the triple point to below 20 K und liquid helium cooled

storage vessel. A prototype moderator cell, similar to SNS design, is also planned, where the pellets entrained in the coolant are deposited in the cell. To remove the pellets they are melted.

A system complete with fabrication, transport and removal of the pellets was suggested in / LUCAS 95 /. Fig. 10 shows the schematic of the system : formation of pellets from liquid methane, continuous transport of the solidified droplets with liquid hydrogen into the moderator container and continuous removal of the pellets from the container by an auger. Outside the high radiation field the methane pellets are melted, the liquid is purified, cooled, and returned to the pelletizer.

Also connected with fusion reactor work hydrogenic pellets between 0.5 and 10 mm in diameter had been made with droplet generators. The method was tested for making methane spheres in experiments / LUCAS 98 / where liquid methane drops were formed by spraying the methane through a nozzle. Surface tension causes the drops to form spheres. The liquid methane droplets fell down 8 m in hydrogen gas, loosing heat by evaporation, and dropped into a liquid helium cooled product container which contained a pool of liquid hydrogen. The droplets fell apart when hitting the container and it was not cleared up whether they had frozen during the fall or were still liquid. The experiments were not continued.

The use of an auger to transport the pellets out of the moderator container has been tested at room temperature / BAUER 01 /. Polyethylene spheres of 3 mm diameter have been removed from a transparent model of a moderator container by means of a screw conveyor that was positioned at the bottom of the container.

6. Concept for a cold methane moderator system for ESS

In the ESS feasibility study / ESS 96 / the reference design of the target-moderator-reflector-assembly has been described (Chapter 4, Fig. 7). The reference cold moderator material is supercritical hydrogen. A more detailed description of this moderator systems is given in / STENDAL 96 /.

It is assumed that the cold methane moderator development will be parallel to the ESS development, and that ESS shall be started up with cold hydrogen moderators in place. The methane moderators would then have to be retrofitted. For this reason the concept for the moderator system developed at the Forschungszentrum Jülich is designed to fit into the reference target-moderator-reflector-assembly.

On the basis of the above considerations and constraints, a concept for the ESS cold moderator system is being examined at the Forschungszentrum Jülich / BARNERT-WIEMER 98 /. In Fig. 11 this process is shown : cylindrical methane pellets are formed with an extrusion cryostat followed by spheronizing of the small cylinders. The spheres should have a diameter of 2 to 2.5 mm. From a metering device the pellets are flushed with hydrogen into the moderator container. Independent of the finally chosen moderator material, the moderator has to be 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.
For transport of the pellets out of the moderator container the hydrogen flow is reversed and the moderator spheres are fluidized with subsequent pneumatic transport. 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.
The process of filling the moderator container and removing the pellets is batchwise to simplify the loading step and ensure complete removal of the pellets, so that no spent methane pellets accumulate in the system.
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.

As was already detailed above, the philosophy behind the development of the described advanced moderator system for ESS is, that such a system should fit into the target-moderator-reflector assembly designed for a hydrogen moderator with only minimal adjustments. The process of filling the moderator container with spheres – which is independent of the moderator material that will finally be chosen – needs to be developed. But for the removal of the methane from the container other methods are conceivable. A simple – and reliable - method might be to evaporate the methane. This will lead invariably to incrustation of the container and tube walls with polymers and entrains a frequent replacement of the moderator container and part of the piping. The whole target-moderator-reflector assembly would have be redesigned in such a manner that the moderator system can be replaced regularly. At present it is planned to replace the moderator system every two to three years.

7. The JESSICA experiment

At the Forschungszentrum Jülich the experiment JESSICA / TIETZE-JAENSCH 99 / has been installed at the synchrotron COSY(Cooler Synchroton). JESSICA stands for Jülich Experimental Spallation Target Set-up In COSY Area and is designed for prototyping experiments on the target - moderator - reflector system of the European Spallation Source (ESS). Because of its low proton beam intensity ($2 \cdot 10^9$ protons per pulse, 100 – 300 µs pulse length, 10 s repetition rate) COSY is particularly suitable for studying the neutron performance of advanced moderators, whereas radiolysis, high activation levels and background are negligible. Furthermore, only moderate shielding is necessary, which means that the whole prototyping experiment is much easier to accomplish without any lack of scientific quality of the results. Not only can the proton beam energy be tuned from 0.8 - 2.5 GeV at COSY, the negligible activation rate enables easy modifications to the geometry, construction details and materials involved. On the other hand, using a realistic 1:1 mock-up model of the anticipated ESS target yields reliable data for the technical optimization process (Fig. 12). All this makes JESSICA ideally suited for direct measurements of the neutronic performance of target - moderator - reflector systems.

A full-size liquid Hg target has been placed in the short-pulsed proton beam of COSY to produce a low intensity pulsed neutron beam. A number of ambient temperature and cold moderators are being investigated at various positions relative to the target which is surrounded by a lead reflector (Fig.13). The moderators are located in wing geometry , i.e. the neutron beam tubes do not view the target through the moderator directly. This way the fast neutron background is reduced considerably. Furthermore, the moderator surfaces are oriented perpendicular to the neutron beam axes in order to conserve the time structure of the neutron pulse. The aim is to validate the very complex simulation methods of particle interaction and transport by experiments to optimize the technical layout of an ESS-type target-moderator-reflector system. This includes investigations of advanced cold moderators and development of scattering kernel data. With the JESSICA experiment the neutronic performance shall be measured by means of neutron time of flight and scattering techniques. All results of the JESSICA experiment are important data to validate the simulation models and code systems - as CALOR, HERMES, LCS, NMTC - which are used to optimize the layout of high power spallation target systems / FILGES 01 /.

7.1 Goals of the moderator experiments in JESSICA

For the design of cold moderators the moderator tests at JESSICA are of great importance. The moderator experiments can be divided in two goals :

- Measurement of the neutron spectra and time structure of the respective neutron pulses of candidate cold moderator materials relative to the conventionally used hydrogen.

Because from a neutronic standpoint methane (CH_4) is considered the best moderator, the candidate materials to be tested at 16 K are all methane based:

- solid methane in the shape of small pellets (Methane has a melting point of 90.7 K at 1 bar)
- methane hydrate in pellet form (Gas hydrates are ice-like crystalline structures of a water lattice with cavities which contain guest gases, e.g. methane. Gas hydrates belong to a special family of inclusion compounds called clathrates. Methane hydrate is solid at 198 K and 1 bar.)
- methane adsorbed on zeolites (Zeolites are crystalline alumosilicates that are commercially fabricated with high porosities up to 40 %. Handling of this material is expected to be much simpler than the other two materials.)

Methane or methane-containing pellets shall be surrounded by liquid hydrogen.

The reference materials to be tested at 16 K also are liquid hydrogen, solid water ice, and solid methane as a block.

- Influence of the moderator geometry on the neutron pulse time structure.

A moderator container with two convex surfaces has been designed for the ESS cold hydrogen moderator / ESS 96 / with the goal to improve the time structure of neutron pulses. This effect is to be tested in JESSICA by varying the viewing angle of a moderator container with flat sides and the same dimensions as they are described for the cold moderator tests. These data can be gained with water as moderator at room temperature.

7.2 Experimental set-up of the JESSICA cold moderator system

To test the various moderator materials a cold moderator system has been built, complete with methane pellet production and transport / BARNERT-WIEMER 98 and 99 /.

Fig. 14 shows a schematic of the moderator system for the JESSICA experiment.The moderator container has a cross section of 50x150 mm and a height of 135 mm (inside dimensions). It is made from $AlMg_3$. The moderator container is surrounded by two vacuum jackets and two radiation shields. The one in the inner jacket is connected to the 80 K stage of the coldhead. Both jackets are bolted to a flange, which is welded to the motor housing of the cold head. Through this flange all ducts and electrical cords for the operation of the moderator container are threaded. The outer vacuum jacket is opened when pellets are filled into the container (s. below). The moderator vessel is cooled by a cold head type RGD 1245 by Leybold Vakuum GmbH in Köln / Germany, combined with a helium compressor type COOLPAK 4000 also from Leybold Vakuum. The cold head has a cooling capacity of 45 Watt at the first stage (80 K) and 12 Watt at the second stage at 20 K. The minimum temperature is ~ 10 K. Since the temperature of the cold head can not be regulated the second stage has an electrical heating so that moderator temperatures can be controlled. Experiments with hydrogen are designed to be operated between 14.01 K (melting point of hydrogen) and 20.28 K (boiling point of hydrogen). Experiments with methane up to the boiling point of 109.15 K are also feasable.

The moderator container and the cold head are mounted on a trolley, together with vacuum pumps and mass spectrometers. It is moved on rails which run between the glovebox and the target (Fig. 15).

When both vacuum jackets are in place and functional the trolley is positioned under the target such that the moderator container together with the cold head can be lifted into its exact position under the target. After the moderator container has been cooled down to the experimental temperature the condensation of hydrogen from the hydrogen storage container can begin. The hydrogen is precooled in coils around the first cold head stage (80 K). During the duration of the experiment the moderator container is connected to to the H_2 storage container for pressure compensation if the moderator temperature varies. As an additional safety measure a duct, outfitted with a rupure disc, leads to safety container where all gases can be stored in case of a pressure excursion in the moderator system.

When pellets are to be tested in JESSICA the process is much more complicated. Fig. 16 describes schematically the process of making methane pellets, transporting them to the COSY building, and filling them into the moderator container : Methane pellets are made in an extrusion cryostat , discharged into a collecting vessel and transferred into to a transport vessel under a helium blanket in a glovebox. After transportation to the COSY building the transport vessel is transferred to another helium-filled glovebox . The evacuated moderator container, which has been mounted to the cold head before, is flanged to the bottom of the glovebox. The container is cooled down to the lowest achievable temperature before the outer vacuum jacket is removed. The inner vacuum mantle stays intact. The stopper on top of the container is removed and the methane pellets, which are at liquid helium temperature are filled into the container. The stopper is put back on, then the outer vacuum jacket, and the container is briefly evacuated to remove most of the helium which has entered the container during the filling process. The moderator container on its cold head is decoupled from the glovebox, lowered into the trolley and moved under the target, where it is lifted into the experimental position. After this has been finished and the temperature adjusted hydrogen is condensed into the container as described above.

As of the end of the year 2001 the function of the target has been verified with a ambient water moderator in place, neutronic measuring techniques are being optimized, and the moderator system components are being tested.

8. Areas defined for research

For the above described moderator system – in fact for every advanced moderator concept - not a single componenent can be taken off the shelf. So work has been started on several subtasks of the complete moderator system.

8.1 Moderator materials

From a neutronic standpoint methane is considered the best. But for a working system a material can not be considered from this standpoint alone but chemical behaviour and engineering constraints need to be weighed. Because the few available data on methane indicate that it might be too soft for extensive handling, other methane containing materials have been considered.
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 ice lattice alone is unstable and the guest molecule is necessary to support the cavity / SLOAN 90 /. It is planned to test the neutronic properties in JESSICA. (Chapter 7)
It has also been discussed to adsorb methane on zeolites, which are available in qualities up to 40% pore volume. Their neutronic qualities are unknown. Most zeolites are not abrasion resistant at room temperature. All abrasion products will be transported with the hydrogen coolant to the refrigerator where they need to be removed to avoid blocking of the refrigerator. Similar constraints also hold for condensation of methane in hollow or porous spheres of plastic.
The advantage in both cases is the perfect spherical form.

8.2 The making of spherical pellets

It has been mentioned before, that spherical pellets are a must to ensure a uniform packing and reliable cooling to avoid hotspots, which, depending on their size, can lead to melting of solid methane. A spherical shape can be gained by either making non-spherical pellets, like cubes or zylinders, and shape them into spheres in a spheronizing prozess, or making spheres directly.

The process under development at present is of the first kind : extruder plus spheronizer . The spheronizing process might be done by pressure compaction (molding, tableting) at a temperature where solid methane is soft or by grinding of hard methane pellets between rotating plates.

Two techniques to make spherical pellets directly should be considered : prilling and fluidized bed processes. The prilling process consists of spraying droplets of liquid into the top of a tower and letting them solidify while dropping. The process has been tried with unsatisfactory results before / LUCAS 98 /, but an optimization should be tried before giving up the idea for good.

In a fluidized bed process particle enlargement occurs when layers of methane freeze on seed kernels. This would probably be a batch process, because methane would freeze on all cold surfaces and would have to be evaporated after each cycle.

8.3 Design of moderator container and pellet transport

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 for easy separation from the coolant and to prevent fine particles from depositing in the system. Hydraulic transport processes make good grinders, so special attention has to be given to attrition of pellets and and wall materials. The hydrogen coolant shall be contaminated by coarse (from broken pellets) and fine (from attrition) particles.These accumulated particles do

not only put a high load on the filters upstream of the refrigerators but the particles in the hydrogen may be detrimental to other parts as well, like valves, metering and measuring devices.
Therefore a supercritical hydrogen test loop is being considered / BARNERT-WIEMER 01 /. It is designed such that components of ESS size can be testet (Fig 17). Two test rigs are considered, one for bigger components like a moderator container and one for smaller devices like valves. The fluid is supercritical hydrogen at 20 to 30 K, cooled by a refrigerator, which has to remove the heat input from the environment and the pumps only. No heating to simulate radiation is being considered at present. Even without this feature the experimental set-up is going to be costly due to refrigeration at low temperatures and safety measures like a design of ducts and components in triple containment mode.

9. Summary

The European Spallation Source (ESS) is designed for an average proton beam power of 5 MW at a repetion rate of 50 Hz with proton pulse lengths on the target of 1 µs or less. This makes ESS 30 times as powerful as the presently biggest spallation facility ISIS. From these data it is recognizable that practically every component has to be redesigned. This holds true for advanced cold moderators on methane basis as well.
This report describes the concept for a cold (20 K) methane moderator system, which fits into the target-moderator-reflector assembly of the ESS reference design. The moderator is used in the shape of small pellets which are cooled by supercritical hydrogen. The pellets have to be spherical to ensure defined and reliable cooling. Solid methane pellets are made by an extrusion process followed by a spheronizing step. The diameter of the pellets is 2 – 2.5 mm. A batch of approx. 1 liter is transported with the hydrogen into the moderator container where the pellets form a fixed bed through which the hydrogen coolant flows. To remove

the pellets from the container, the flow is reversed, the pellet bed is fluidized and the pellets are removed by pneumatic transport. They are separated from the hydrogen and the methane is either purified and reused or rejected and realeased to the environment – if the amount of radioactive isotopes present in the spent methane allows it.

Work is under way to make pellets and test them under ESS conditions in the JESSICA experiment.

10. References

/ AXMANN 84/ : A. Axmann und K. Gobrecht, Die Wahl des kalten Moderators für die Kalte Neutronenquelle des BER II, HMI-B 479 (1984)

/ BARNERT-WIEMER 98/ : Proc. of the 14 th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS) Utica,Illinois/USA

/ BARNERT-WIEMER 99 / : 4th Meeting of the International Collaboration on Advanced Cold Moderators (ACOM IV), Forschungszentrum Jülich, Jülich / Germany , Feb. 25 – 26, 1999

/ BARNERT-WIEMER 01/ : H. Barnert-Wiemer, H. Stelzer, Cold moderator work for ESS at the Forschungszentrum Jülich, Workshop on moderator concepts and optimisation for spallation neutron sources,12.-14. 3.2001 Hahn-Meitner-Institut, Berlin / Germany

/ BARTKNECHT 93 / : W. Bartknecht, Explosionsschutz – Grundlagen und Anwendung, Springer Verlag 1993

/ BAUER 97 / : G.S. Bauer, Pulsed Neutron Source Cold Moderators – Concepts, Design, and Engineering, Proc. of the International Workshop on Cold Moderators for Pulsed Neutron Sources, Argonne National Laboratory / Argonne, Illinois / USA Sept. 29 – Oct. 2, 1997,

/ BAUER 01 / : G.S. Bauer, H. Spitzer, T. Hofmann, Status of the development efforts towards a pelletized moderator at PSI, Workshop on moderator concepts and optimisation for spallation neutron sources,12.-14. 3.2001 Hahn-Meitner-Institut, Berlin / Germany

/ BELJAKOV 93/ : A.A. Beljakov, V.G. Ermilov, V.L. Lomidze, V.V. Melichov, E.P. Shabalin, First Experience of Cold Moderator Operation and Solid Methane Irradiation at the IBR-2 Pulsed Reactor, Proc. of the 12th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS), 24-28 May 1993, Abingdon Oxfordshire / UK

/ BELIAKOV 97 / : A.A. Beljakov, I.T. Tretiakov, E.P. Shabalin, V.V. Golikov, V.I. Luschikov, Solid Methane Cold Moderator for the IBR-2 Ractor,
Proc. of the International Workshop on Cold Moderators for Pulsed Neutron Sources, Argonne National Laboratory / Argonne, Illinois / USA Sept. 29 – Oct. 2, 1997,

/ BOLSHUTKIN 71 / : D.N Bolshutkin, V.M. Gasan, A.I. Prokhvatilov, Zhurn. Strukt. Khimii 12 (1971) 734

/ BOLSHUTKIN 68 / : D.N Bolshutkin, A.V. Leonteva, A.I. Prakhvatilov, Y.E. Stetsenko, Fiz. Kondens. Sostayan, Inst. f. Low Temp. Phys. and Engineer., Kharkov / USSR „ (1968) 136

/ BROOME 97 / : T. A. Broome, The ISIS Moderators, Proc. of the International Workshop on Cold Moderators for Pulsed Neutron Sources, Argonne National Laboratory / Argonne, Illinois / USA Sept. 29 – Oct. 2, 1997,

/ CARPENTER 87/ : J.M. Carpenter, Nature 330 (1987), p.358 - 360

/ CARPENTER 90/ : J.M. Carpenter et al. , Proc. of the 11 th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS), Tsukuba / Japan 1990

/ CARPENTER 90a / : J.M. Carpenter, Cold Moderators for Pulsed Neutron Sources, Proc. of the International Workshop on Cold Neutron Sources, Los Alamos, NM / USA, March 5 – 8, 1990

/ COLWELL 63 / : J:H: Colwell, E.K. Gill, J.A. Morrison, J. Chem. Phys. 39 (1963)635

/ COWARD 52 / : H.F. Coward, G.W. Jones, Limits of flammability of gases and vapors, Bureau of Mines, Bulletin 503, United States Government Printing Office, Washington,1952

/ ESS 96/ : The European Spallation Source Study, ESS-96-53-M (1996)

/ EVANS 95 / : D. Evans, Cryogenics 35 (1995) p.763 –766

/ FILGES 01 / : D. Filges, The performance of moderators for the ESS short and long pulse target stations, Workshop on moderator concepts and optimisation for spallation neutron sources,12.-14. 3.2001 Hahn-Meitner-Institut, Berlin / Germany

/ FOSTER 00 / : C.A. Foster, J.M. Carpenter, An Advanced Cold Moderator Using Solid Methane Pellets, 15 th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS), November 6 – 9, 2000, Tsukuba / Japan

/ FLUHARTY 69 / : R.G. Fluharty, F.B. Simpson, G.J. Russell, J.H. Menzel, Nuclear Science and Engineering, 35, 45 – 69 (1969)

/ GRAHAM 72 / : K.F. Graham, J.M. Carpenter, Nuclear Science and Engineering, 49, 418 – 437 (1972)

/ IPNS 01 / : Intense Pulsed Neutron Source (IPNS) Progress Report 1096 – 20001, Argonne National Laboratory,Illinois/USA 2001

/ JESSEN 98 / : N.C. Jessen, K. Stendal, Solid Methane Moderators for the ESS-Project, Third Meeting of the International Collaboration on Advanced Cold

Moderators (ACOM III), Paul Scherrer Institut (PSI)Villigen / Schweiz, 24./25. 2. 1998

/ JET 96 / : JET Joint Undertaking Progress Report 1995, EUR 16873-EN-C 1996

/ KAISER 95 / : R.I. Kaiser, A. Gabrysch, K. Roessler, Rev. Sci. Instrum. 66 (4), April 1995

/ KAISER 97 / : R.I. Kaiser, G. Eich, A. Gabrysch, K. Roessler, The Astrophysical Journal, 484:487-498, 1997 July 20

/ KIYANAGI 90 / : Y. Kiyanagi et al., Proc. of the 11 th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS), Tsukuba / Japan 1990

/ LUCAS 95 / : A.T. Lucas, G.S. Bauer, C.D. Sulfredge, A pelletized solid methane moderator for a medium-to-high power neutron source, Proc. of the 13 th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS), Oct.11 – 14, 1995, Paul Scherrer Institute, Villigen/Switzerland

/ LUCAS 98 / : A.T. Lucas, First attempts to generate methane pellets at ORNL, Discussion meeting on an International Collaboration on Advanced Cold Moderators (AcoM), Paul Scherrer Institut, Villigen/Schweiz,24.-25. Feb. 1998

/MALKOV 85 / : M.P. Malkov, I.B. Danilov, A.G. Zeldovich, A.B. Fradkov, Physical and Engineering Fundamentals Of Cryogenics, Handbook (in russian), Energoatomizdat, Moskov (1985)

/ MANZHELII 68 / : V.G. Manzhelii, I.N. Krupskii, Fiz. Tverd. Tela. 10 (1968) 281

/ PROKHVATILOV 82 / : A.I. Prokhvatilov, A.P. Isakina, I.N. Krupskii, Fiz. Nizk. Temp. 8 (1982) 635

/ PROKHVATILOV 83 / : A.I. Prokhvatilov, A.P. Isakina, Fiz. Nizk. Temp. 9 (1983) 419

/ PYNN 90 / : R. Pynn , Cold Moderators at Pulsed Spallation Sources: A Personal View, Proc. of the International Workshop on Cold Neutron Sources, Los Alamos, NM / USA, March 5 – 8, 1990

/ ROESSLER 99 / : K. Roessler, Chemistry of solid methane under MeV ion radiation, 4th Meeting of the International Collaboration on Advanced Cold Moderators (ACOM IV), Forschungszentrum Jülich, Jülich / Germany , Feb. 25 – 26, 1999

/ SHABALIN 97/ : E.P. Shabalin, Proc. of the International Workshop on Cold Moderators for Pulsed Neutron Sources, Argonne National Laboratory / Argonne, Illinois / USA Sept. 29 – Oct. 2, 1997

/ SLOAN 90 / : E.D. Sloan, Clathrate hydrates of natural gases, Marcel Bekker Inc. New york 1990

/ STELZER 01 /: H. Stelzer, N. Bayer, H. Barnert-Wiemer, The JESSICA Moderator System, 3rd JESSICA Workshop, Berlin, Germany March 12-15, 2001, ESS 114-01-T, Mai 2001

/ STENDAL 96 / : K. Stendal, Description of The Moderator Systems for the ESS Project, RISØ- R-908(EN), 1996

/ TARASENKO 75 / : L.M. Tarasebnko, Fiz. Nizk. Temp. 1(1975) 688

/ TEICHMANN 99 / : R. Teichmann, Ähnliche Moderatorströmungen, not published

/ TIETZE-JAENSCH 99 / : H. Tietze-Jaensch et al., JESSICA, the ESS-target/reflector prototype and advanced cold moderator test facility at Jülich, Deutsche Neutronentagung, Berlin / Germany, 25. – 27. 5. 1999

/ TOWLE 67 / : L.C. Towle, Appl. Phys. Lett. 10 (1967) 317

/ WILLIAMSON 88 / : K.D. Williamson, A.T. Lucas, A Combined H_2 / CH_4 Cold Moderator for a Short Pulsed Neutron Source, Proc. of the 10th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS), Los Alamos / USA, 3.- 7. Oct. 1988

11. Tables

Source	Location	Target	Proton Current, µA	Proton Energy, MeV
IPNS	Argonne	235-enriched U	16.	450.
ISIS	Rutherford	238-U, Ta	110.	750.
KENS	KEK (Tsukuba)	238-U	10.	500.
LANSCE	Los Alamos	Tungsten	70.	800.

Table 1 : Operating pulsed spallation neutron sources / CARPENTER 90a /

Material	Temperature, K	Proton Density, protons/Å3	Melting and Boiling (1 atm) Temperatures, K
H_2O (for reference)	293	0.067	273, 373
$(CH_2)_n$, 0.94 gm/cm^3 (ref)	293	0.081	-------
TiH_2 (for reference)	293	0.095	-------
H_2	20	0.042	14, 21
CH_4	109	0.070	90, 110
CH_4	10	0.079	90, 110
C_2H_6	165	0.068	90, 184
C_3H_8	228	0.064	83, 231
$C_{12}H_{18}$, Mellitine	293	0.047	439, 538
C_9H_{12}, Mesitylene	293	0.039	228, 438

Table 2 : Pulsed source moderator materials / CARPENTER 90a /

12. Figures

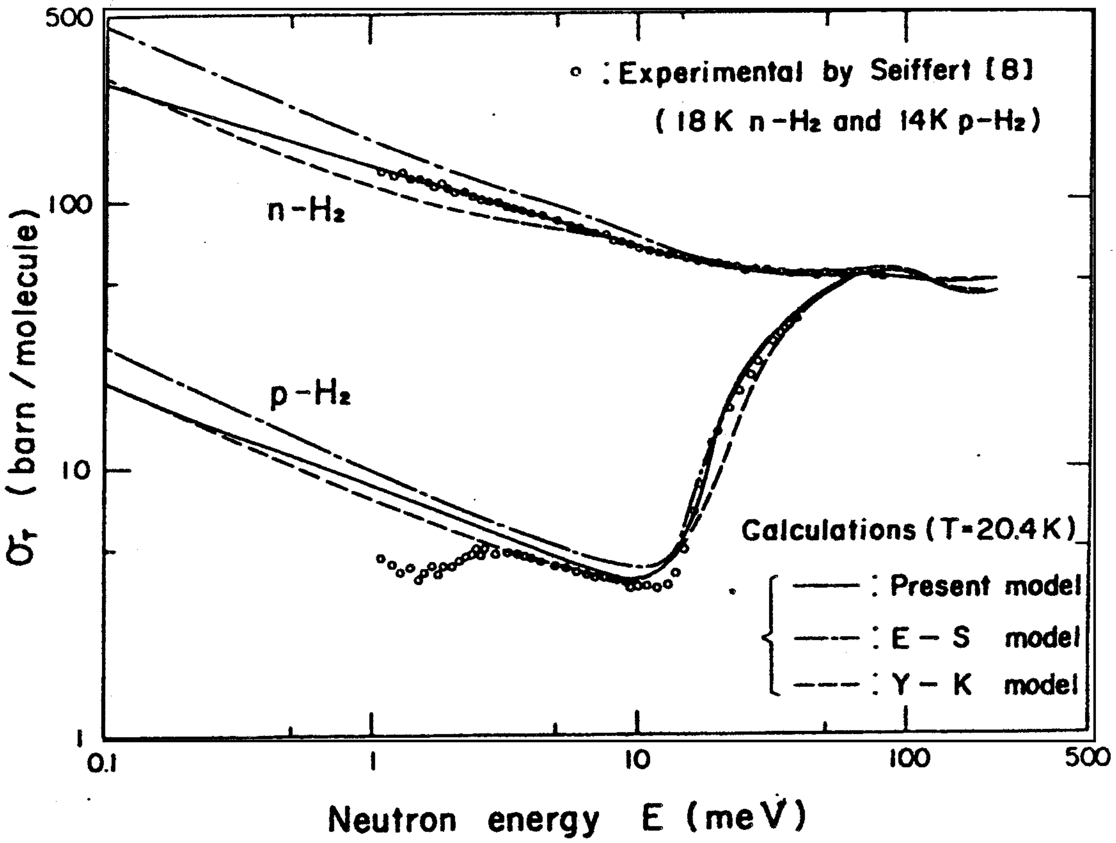


Figure 1 : Scattering cross sections of n - and p -H_2 / AXMANN 84 /

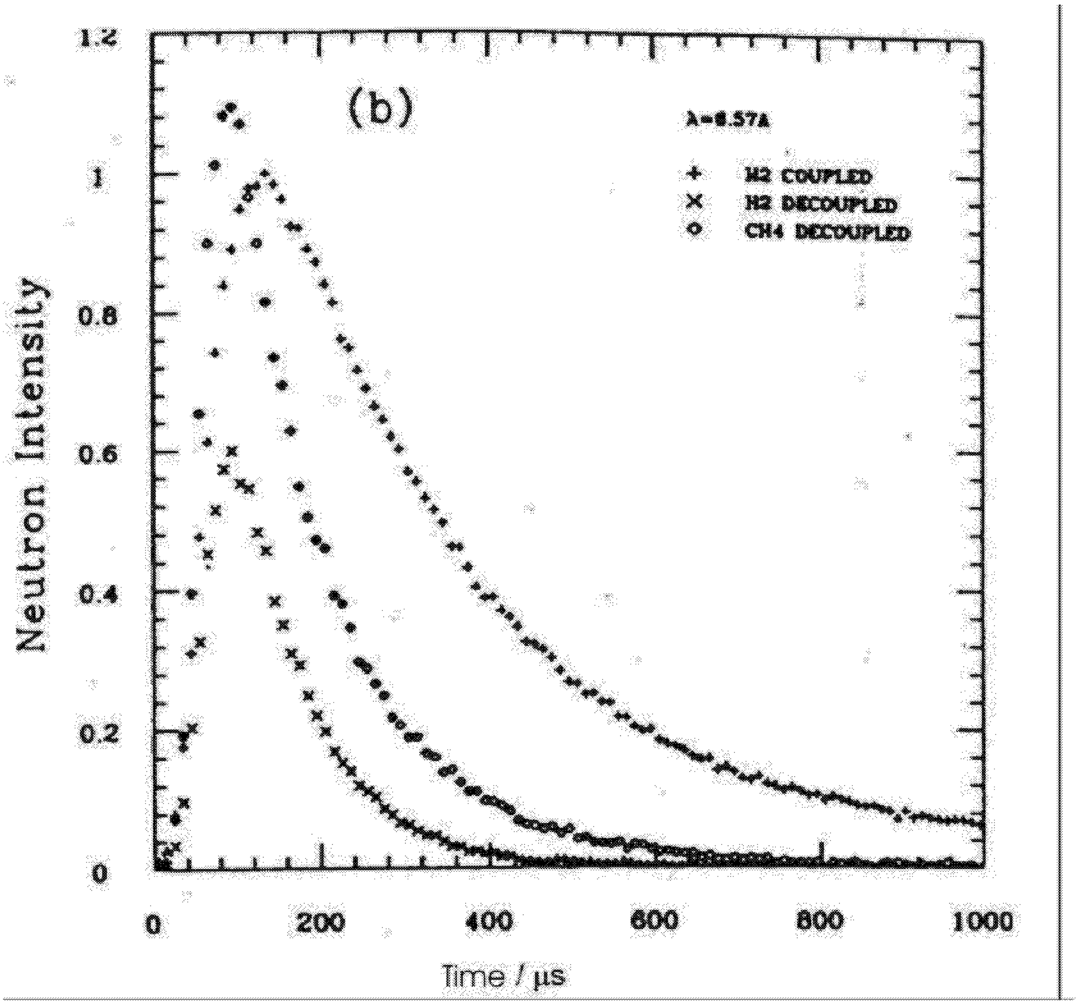


Figure 2 : Experimental puls shapes of cold neutrons (wave length 6.57 Å) for hydrogen and methane moderators

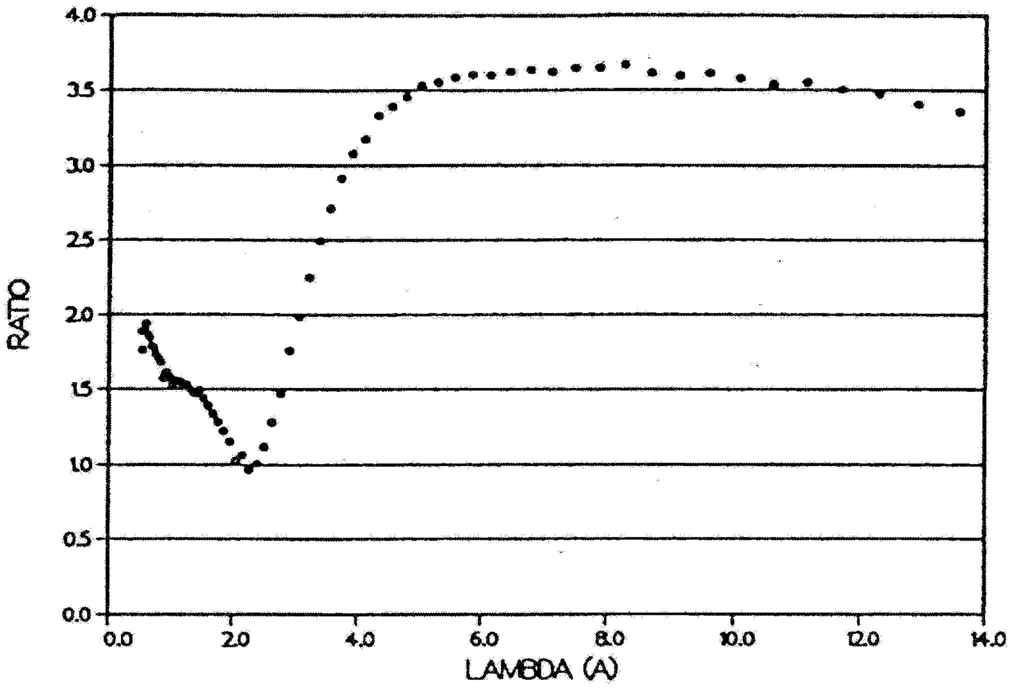


Figure 3 : Ratio of neutron intensities from a methane moderator (20 K, solid) and a hydrogen moderator (20 K, liquid) / CARPENTER 90 /

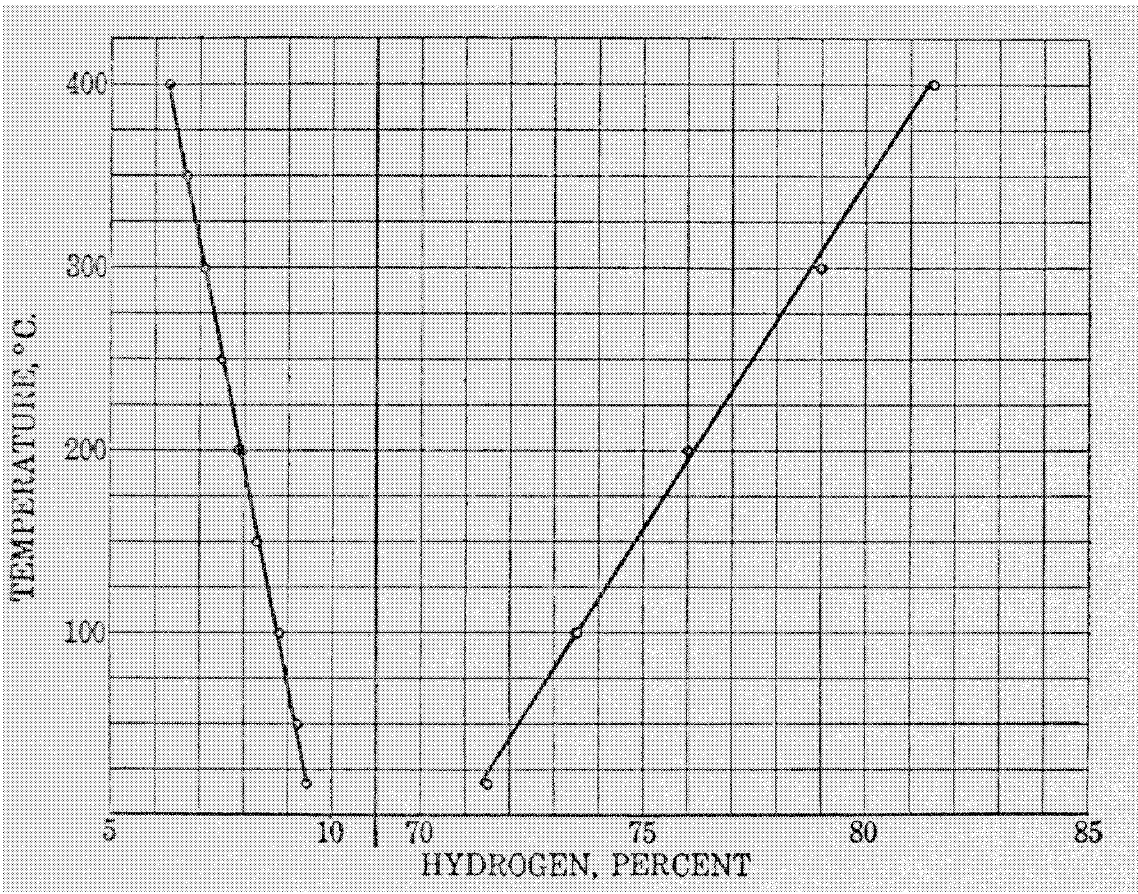


Figure 4 : Influence of temperature on limits of flammability of hydrogen in air (downward propagation of flame) / COWARD 52 /.

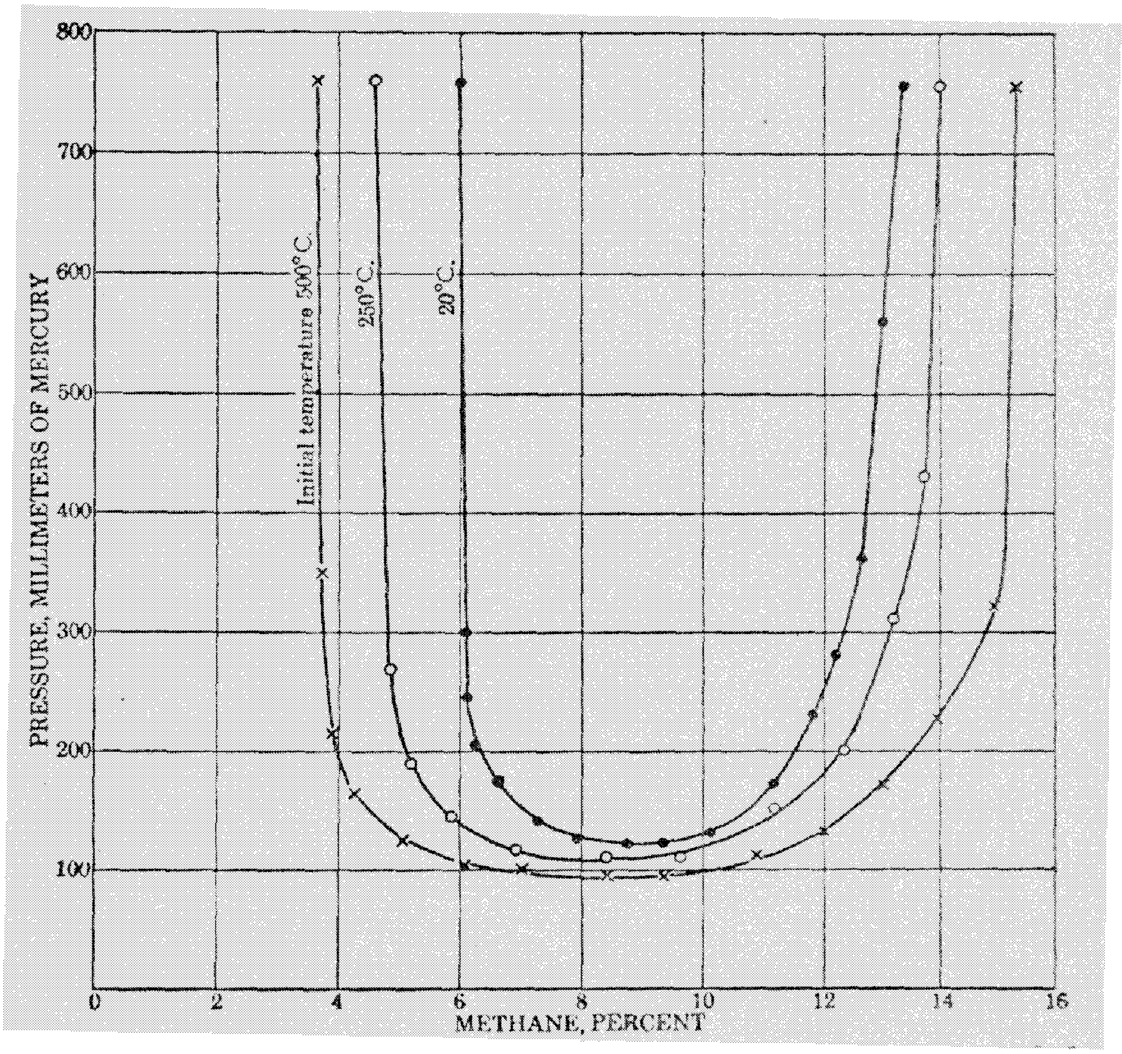


Figure 5 : Limits of flammability of methane in air (downward propagation), showing influence of pressure (below normal) and temperature / COWARD 52 /

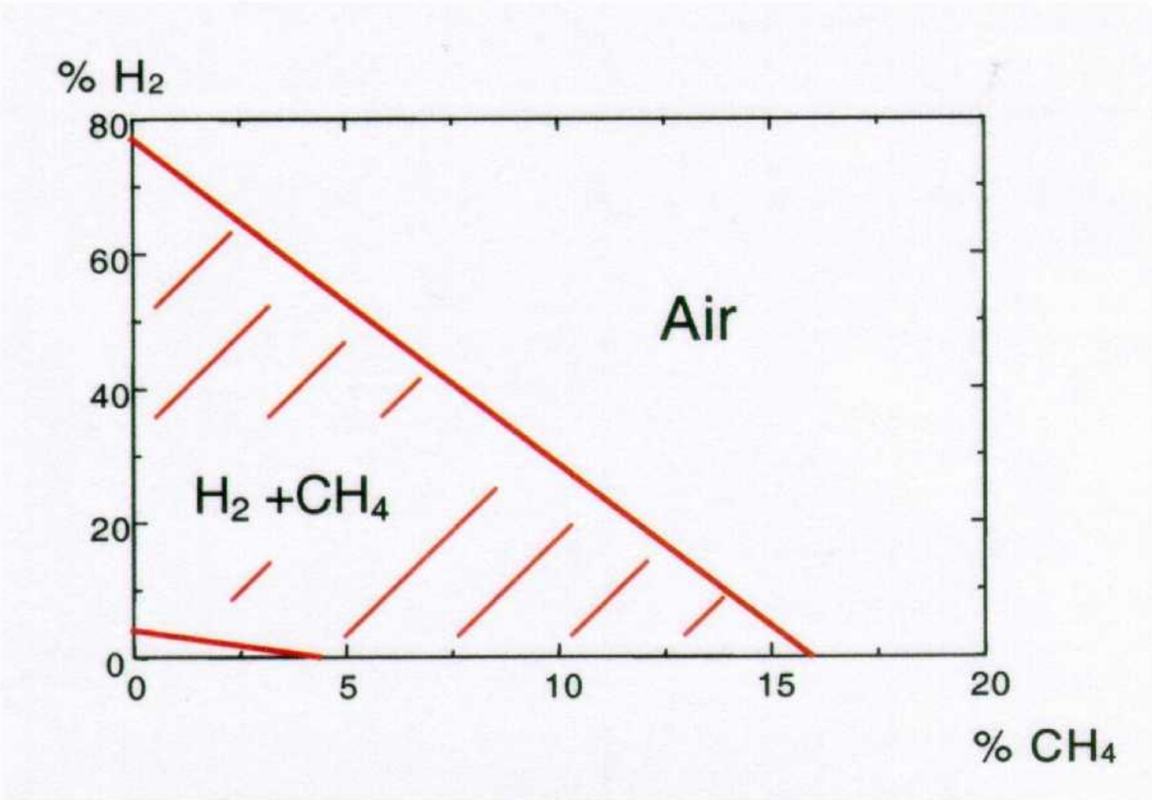
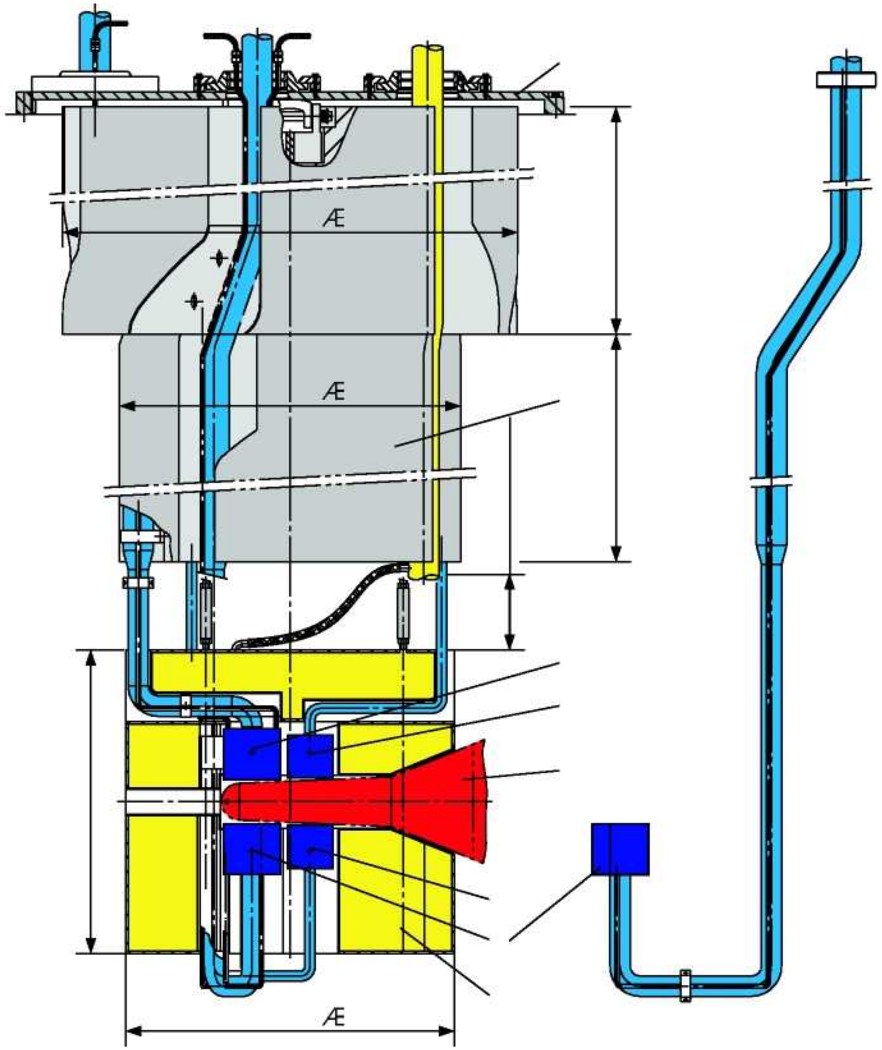


Figure 6 : Explosion limits for mixtures of H_2 and CH_4 in air



ESS Target - Moderator - Reflector Assembly

Reference Design

1 Lead Reflector
2 Target
3 Lower Cold Moderator
4 Upper Cold Moderator
5 Upper Ambient Moderator
6 Lower Ambient Moderator
7 Vertical Shielding Plug
8 Top Sealing Flange

Figure 7 : Reference design of the ESS target - moderator - reflector assembly / ESS 96 /

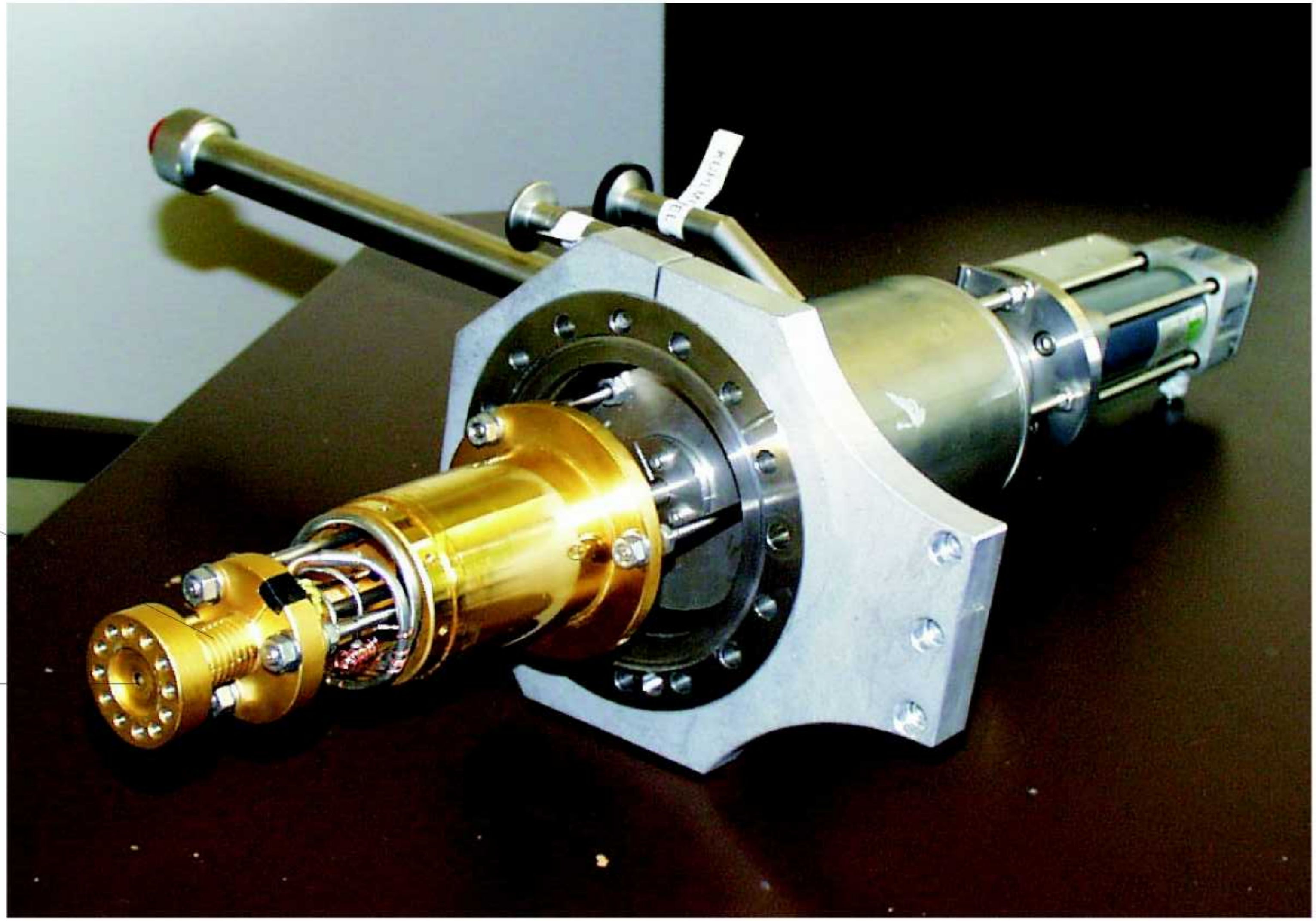


Figure 8 : Extrusion cryostat for methane pellet fabricatiom

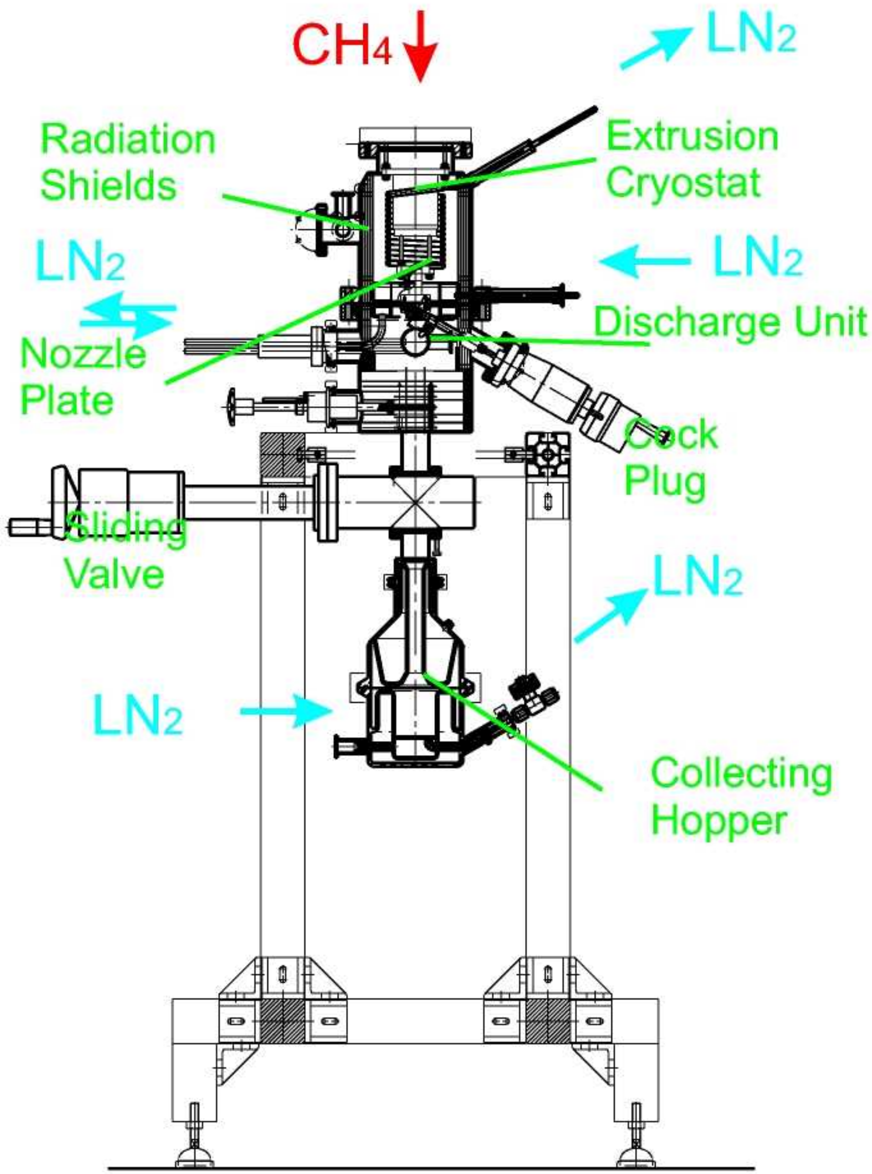


Figure 9 : Methane extruder and discharge unit

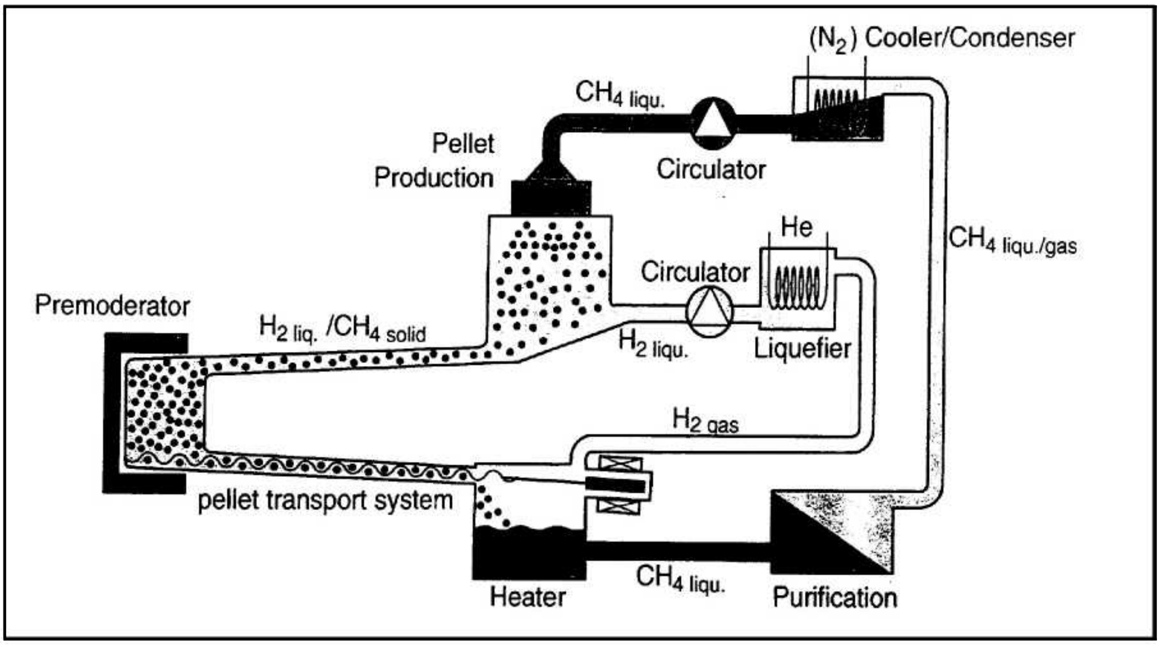


Figure 10 : Pelletized methane moderator system with pellet production outside the radiation zone and pellet removal from the moderator canister by mechanical means / LUCAS 95 /

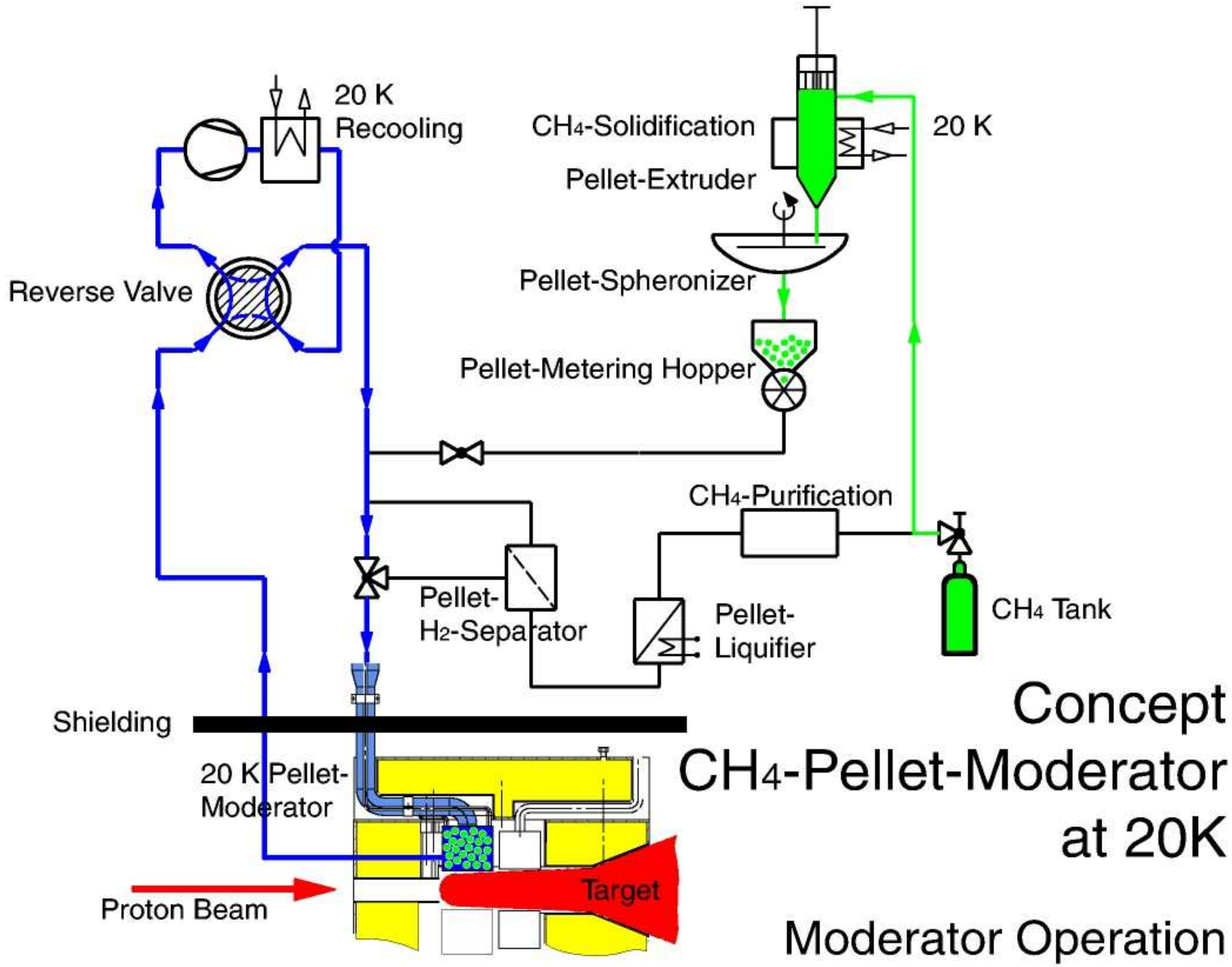


Figure 11 : Concept of the CH_4 - Pellet Moderator System for ESS

Figure 12 : Target-Moderator- Reflector Assembly for the JESSICA experiment

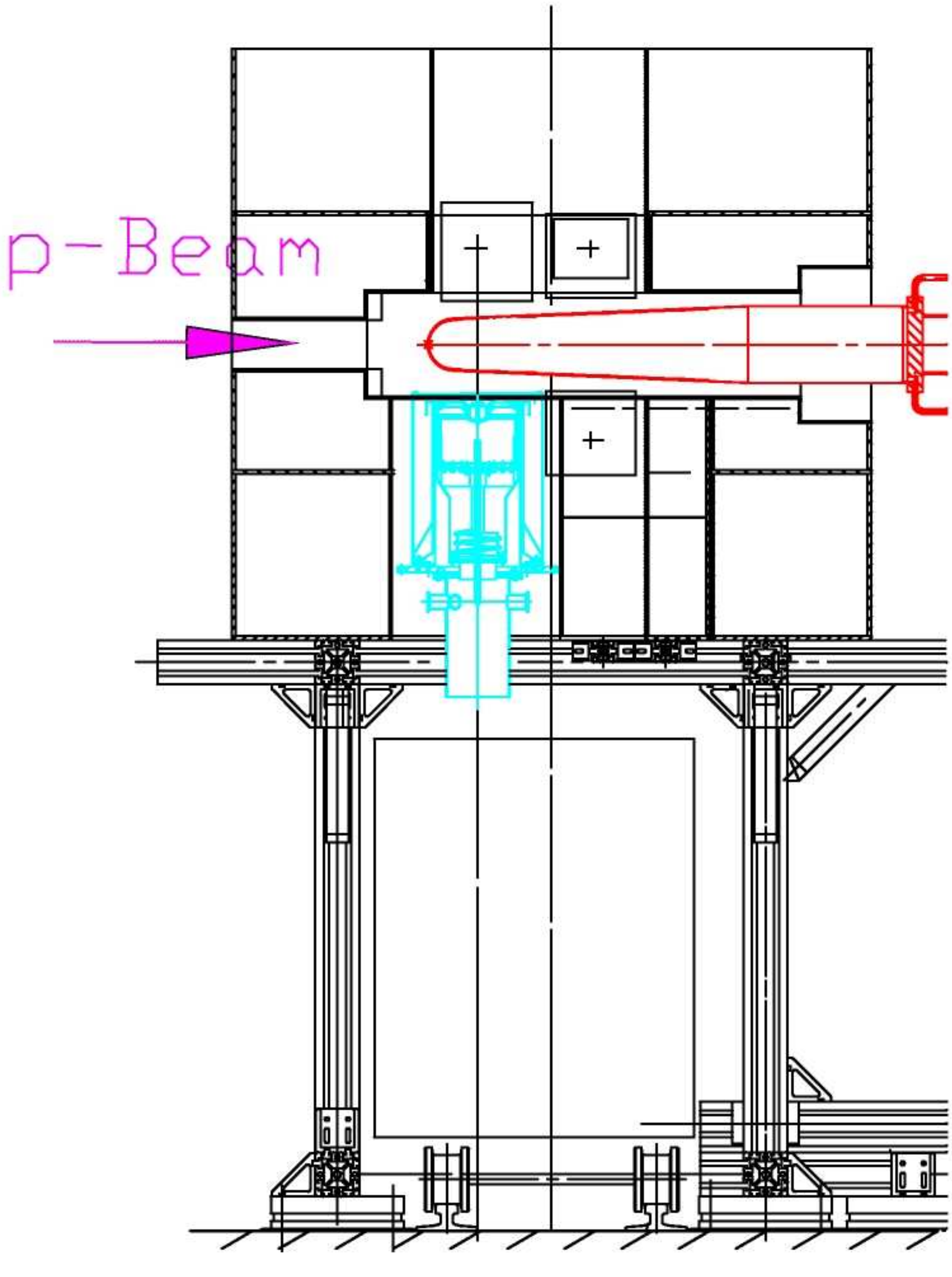


Figure 13 : The JESSICA experiment : Cold moderator in position

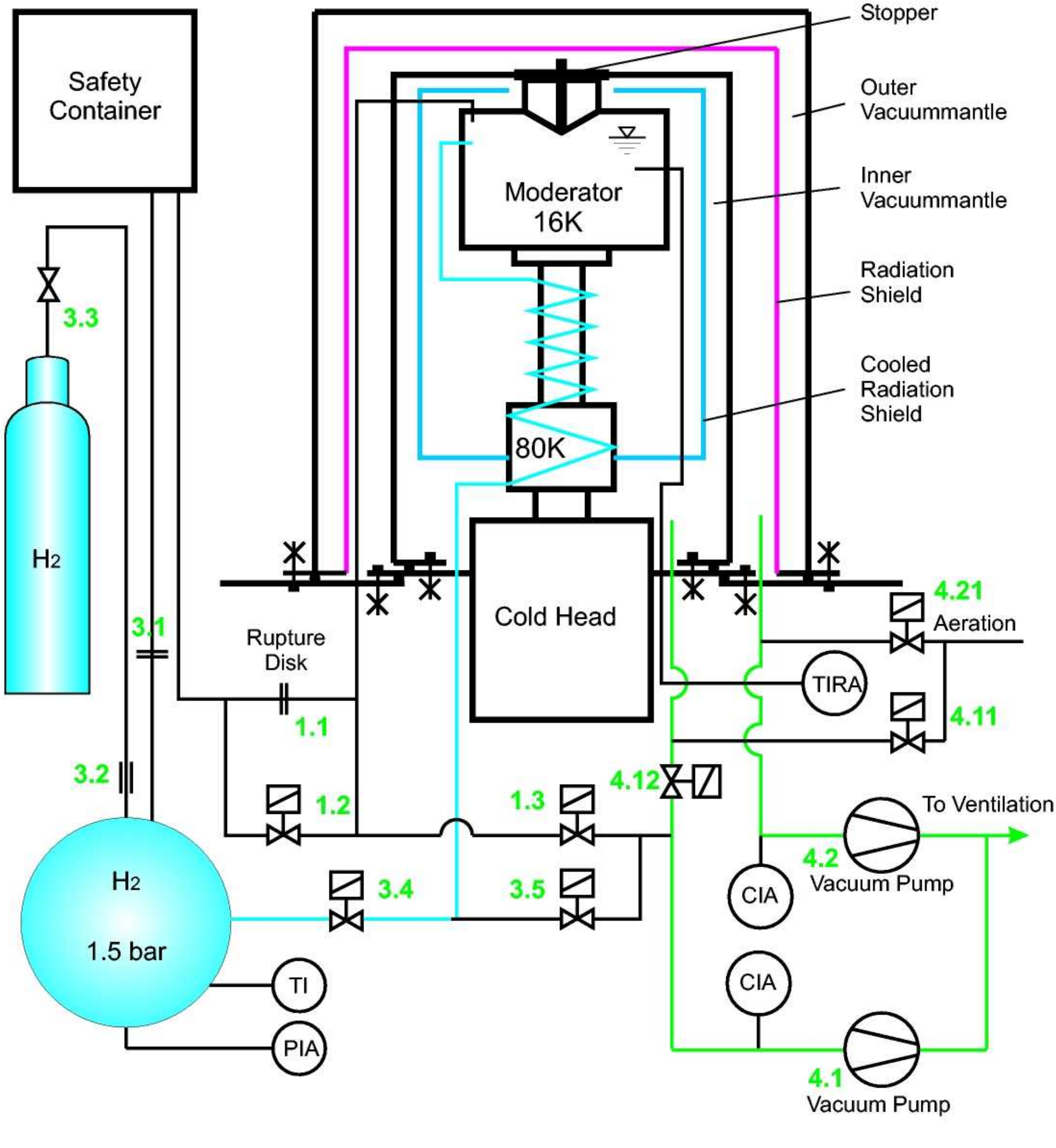


Figure 14 : Schematic of the cold pellet moderator assembly for JESSICA

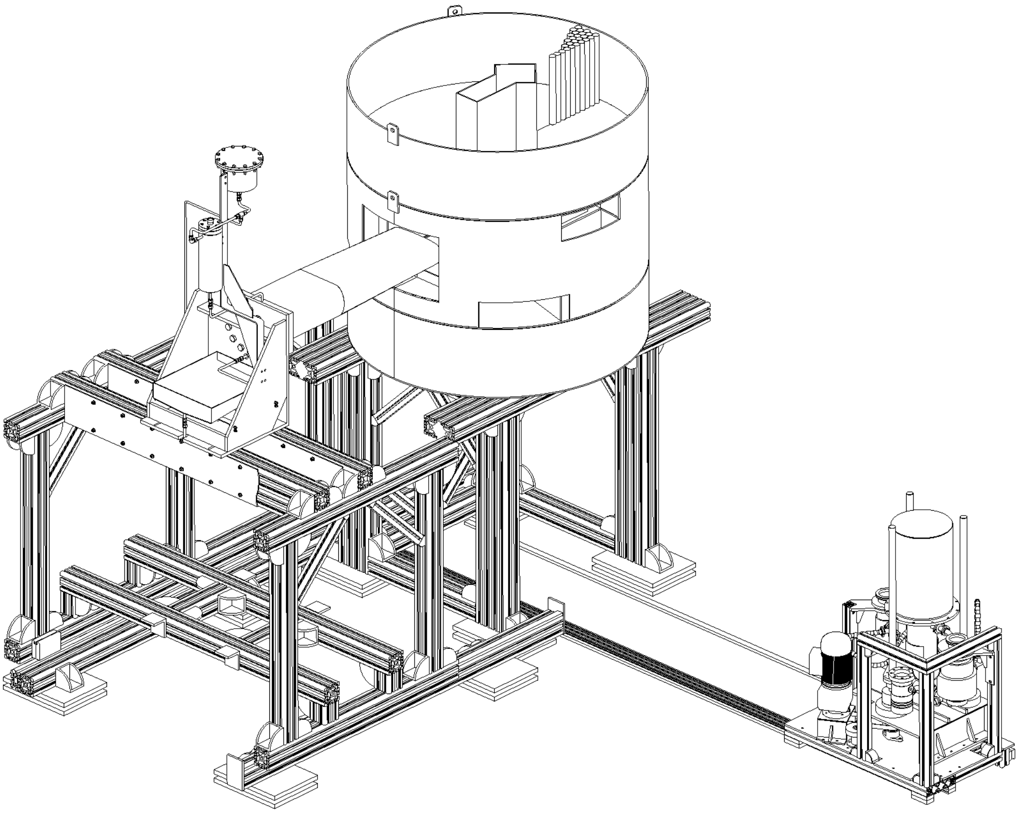


Figure 15 : JESSICA Target-Moderator-Reflector Assembly with moderator trolley outside assembly

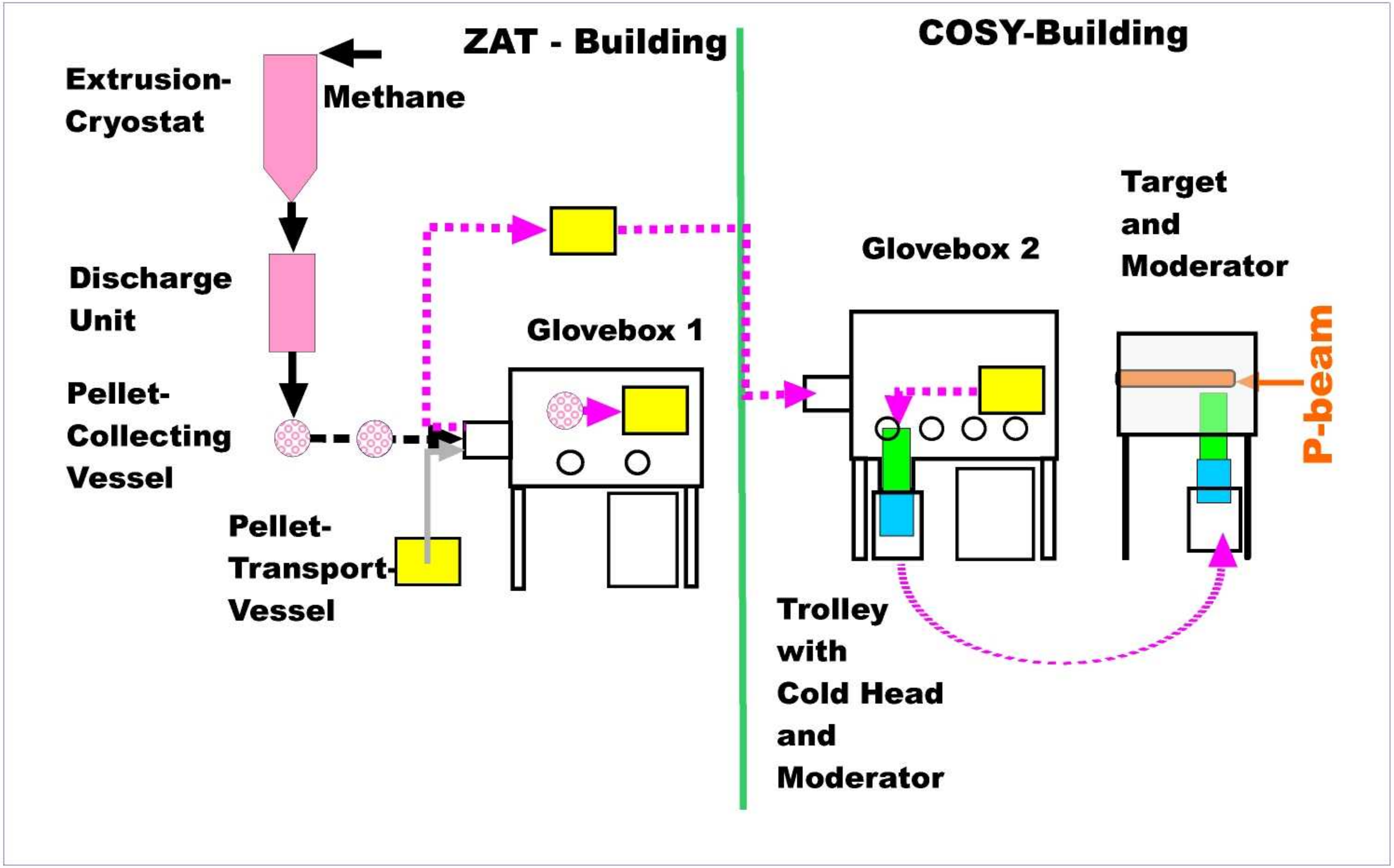


Figure 16 : Preparation and transport of methane pellets to the JESSICA experiment

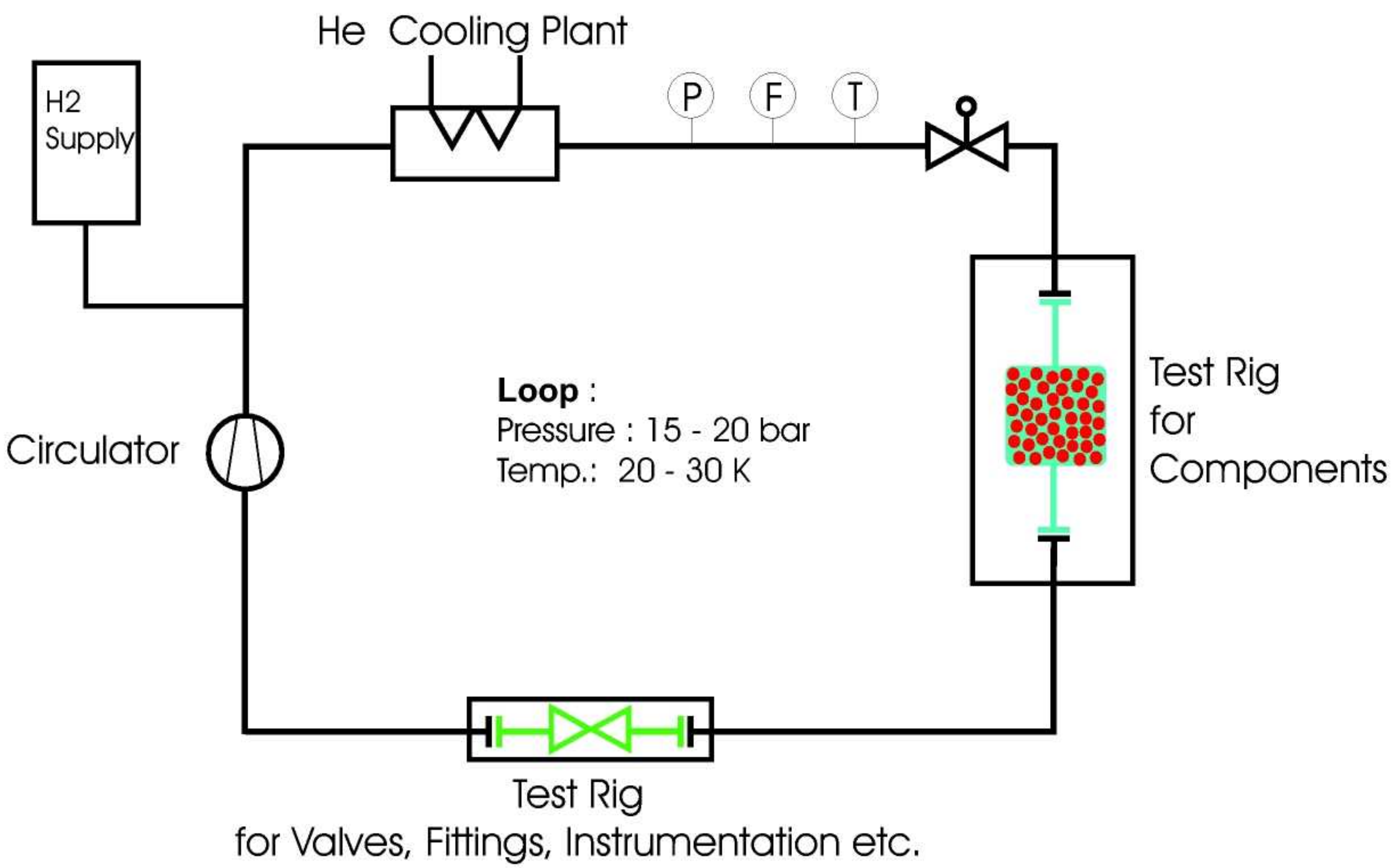


Figure 17 : Supercritical H2 loop for cold moderator system- and component development