

# DEUTERIUM REMOVAL UNIT FOR THE MUCAP EXPERIMENT

I. Alekseev<sup>1</sup>, T.I. Banks<sup>2</sup>, O. Fedorchenko<sup>1</sup>, V. Ganzha<sup>1</sup>, P. Kravtsov<sup>1</sup>,  
P. Kammel<sup>3</sup>, C. Petitjean<sup>4</sup>, V. Trofimov<sup>1</sup>, A. Vasilyev<sup>1</sup> and M. Vznuzdaev<sup>1</sup>

## 1. Introduction

A cryogenic unit for highly efficient isotopic separation of hydrogen has been designed and built at the Petersburg Nuclear Physics Institute (PNPI), Russia. The unit's main task is to produce isotopically pure hydrogen of deuterium concentration below 1 ppm, for use in the MuCap experiment conducted at the Paul Scherrer Institute (PSI), Switzerland. The goal of MuCap is to measure the rate of the basic electroweak process of nuclear muon capture by the proton from the hyperfine singlet ground state of muonic hydrogen. MuCap aims to achieve a 1% measurement of the capture rate, which will in turn provide the most precise (~7%) estimate to date for the weak nucleonic charged-current pseudoscalar form factor.

MuCap is a high-precision measurement that requires ultra-pure and deuterium-depleted hydrogen. The issue of chemical purity was solved by introducing a circulating hydrogen purification system, which is described in detail elsewhere [2]. To meet the isotopic purity requirements, the experiment first attempted using deuterium-depleted gas supplied by an outside vendor [3]. However, the deuterium content was still too high to be completely satisfactory, strongly affecting the interpretation of the first experimental results. It was therefore decided to specially design and build a device for isotopic purification of hydrogen. The principles of the device's design and the results of its operation are described in this report.

## 2. Principles

### Selection of the isotopic purification method

The most common method for separating hydrogen isotopes is to distill water and then perform electrolytic decomposition, followed by hydrogen isolation with a palladium filter. This method was inappropriate for our situation, for several reasons. First, it simply could not achieve the desired level of isotopic purity, as the best commercial hydrogen samples available contained more than 1 ppm of HD. Second, it was desirable to integrate a new isotopic separation unit into the existing hydrogen recirculation system, in order to ensure that the gas was continually cleaned of any possible deuterium contamination. The only method suitable for an in situ isotopic purification of hydrogen is cryogenic distillation.

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<sup>1</sup> Petersburg Nuclear Physics Institute (PNPI), 188350, Gatchina/Russia

<sup>2</sup> University of California, Berkeley, CA 94720-7300, USA.

<sup>3</sup> University of Illinois at Urbana-Champaign, UIUC, IL 61801, USA

<sup>4</sup> Paul Scherrer Institut (PSI), CH-5232 Villigen PSI/Switzerland

## Principles of cryogenic distillation and main column requirements

The principles of distillation (i.e. rectification) are well-known. Distillation under cryogenic conditions conforms to the same rules as high-temperature rectification, which is prevalent in the chemical industry. The method exploits the difference in the saturation vapor pressure of mixed species above the surface of the liquid mixture. It can be regarded as a multi-step fractional distillation that uses a column filled with a special facility for increasing the phase contact surface, where the facility can be either a set of perforated plates or packing, the latter being more suitable for a column with a relatively small inner diameter. A condenser located in the top of the column converts the vapor mixture to liquid form; this returning liquid is called “reflux.” The vapor can also be partially siphoned from the top of the column as a purified product of the process. The reflux drains down along the column and moistens the packing. The amount of liquid suspended on the packing is the column holdup. The lower end of the column is equipped with a reboiler. The separated mixture boils in the reboiler to form the vapor, which rises upward along the column and interacts with the counterflow of draining reflux. The liquid is saturated with the high-boiling component while the gas is saturated with the low-boiling component. In our case, the mixture is of regular hydrogen (“protium”, or  $H_2$ ) and “deuterohydrogen” (HD). Deuterium ( $D_2$ ) molecules are very rare in the source gas and can be ignored.

The column was designed to separate an initial mixture injected into the device through a feed port, with pure  $H_2$  (whose HD content is at least 30 times lower than that of the feeding gas) being the distillation product. The HD component has to be accumulated in the reboiler with the possibility to discharge it.

In order to find the optimal operating mode for the column, a number of studies and verifications were carried out, including investigations of the pressure drop along the column, assessment of the holdup, and the cleaning performance at different modes.

## HETP concept

The separation factor  $\alpha$  defines an elementary separation effect achieved at one contact of liquid and vaporized hydrogen. The ideal separation factor is a ratio of saturation vapour pressures above pure components, and depends on temperature: a higher temperature means a lower separation factor. By passing through the separation column, liquid and vapour are brought into repeated contact, thereby multiplying the elementary separation effect. The downflowing liquid hydrogen is thus steadily enriched in its high-boiling component (in our case, HD) while the rising vapour is steadily depleted.

In accordance with the concept of the Equilibrium Theoretical Plate [3], the Height Equivalent to a Theoretical Plate (HETP) is the main performance characteristic of a separation column. HETP is the height of a column part left by liquid and gas flows which are in mass-transfer equilibrium (the deuterium concentration in the liquid is  $\alpha$  times higher than in the gas). A lower HETP corresponds to a higher column separation power.

The separation ratio of the column is the ratio of relative concentrations of the component of interest at the bottom and top of the column:

$$SR = \frac{X_{Bottom} / (1 - X_{Bottom})}{X_{Top} / (1 - X_{Top})}. \quad (1)$$

The separation ratio is associated with the separation factor by the Fenske [4] equation:

$$SR = \alpha^N, \quad (2)$$

where  $N$  = the number of theoretical plates.

### The separation factor for hydrogen components

In addition to its isotopic composition, hydrogen has spin-isomeric components [5].  $H_2$  molecules can be grouped according to their nuclear spin orientations: In the first group, the proton spins are aligned in the same direction (triplet state, orthohydrogen), while in the other group, the proton spins are aligned in opposite directions (singlet state, parahydrogen). Ordinarily, transitions between the ortho and para states is relatively rare, so  $H_2$  can be regarded as a mixture of two distinct components with rather different physical and chemical properties. The ratio between the ortho and para states is about 3:1 at standard temperature and pressure, but the para form dominates at low temperatures.

For ortho-para and  $H_2$ -HD compositions, we can treat factor  $\alpha$  as the ideal one. Thus, for ortho-para separation the factor  $\alpha$  can be shown to be

$$\alpha_{o-p} = \frac{P_{sat,para}}{P_{sat,ortho}}. \quad (3)$$

Here  $P_{sat,Para}$  and  $P_{sat,Ortho}$  are the pressures of saturated vapour above pure parahydrogen and pure orthohydrogen, respectively.

Further, for the deuterium-protium case,

$$\alpha_{HD-H_2} = \frac{P_{sat,H_2}}{P_{sat,HD}}, \quad (4)$$

where  $P_{sat,H_2}$  and  $P_{sat,HD}$  are the pressures of saturated vapour above pure  $H_2$  and pure HD. The temperature dependencies of separation factors for HD- $H_2$  and for the ortho-para hydrogen system are shown in Figure 1 and Figure 2 [6].

Verification of the column's separating power was complicated by the absence of a fast, on-line deuterium concentration analysis, and also by the fact that the residual deuterium content of the depleted product can lie below the detection limit of any available analysis methods. This compelled us to find another approach: namely, the use of gas chromatography (described below) for ortho-para hydrogen analysis. Gas chromatography proved to be a relatively fast and simple way of evaluating column performance during testing.

The lower  $\alpha$  (compare Figure 1 and Figure 2) makes it possible to measure the concentrations for ortho and para modifications in probes taken from each control point of the column (condenser, reboiler, and feeding line). Using the Fenske equation and the data from the chromatographic analysis, it is easy to determine the HETP for the column. Knowledge of HETP enables calculation of concentration profiles along the column, using a mathematical model.

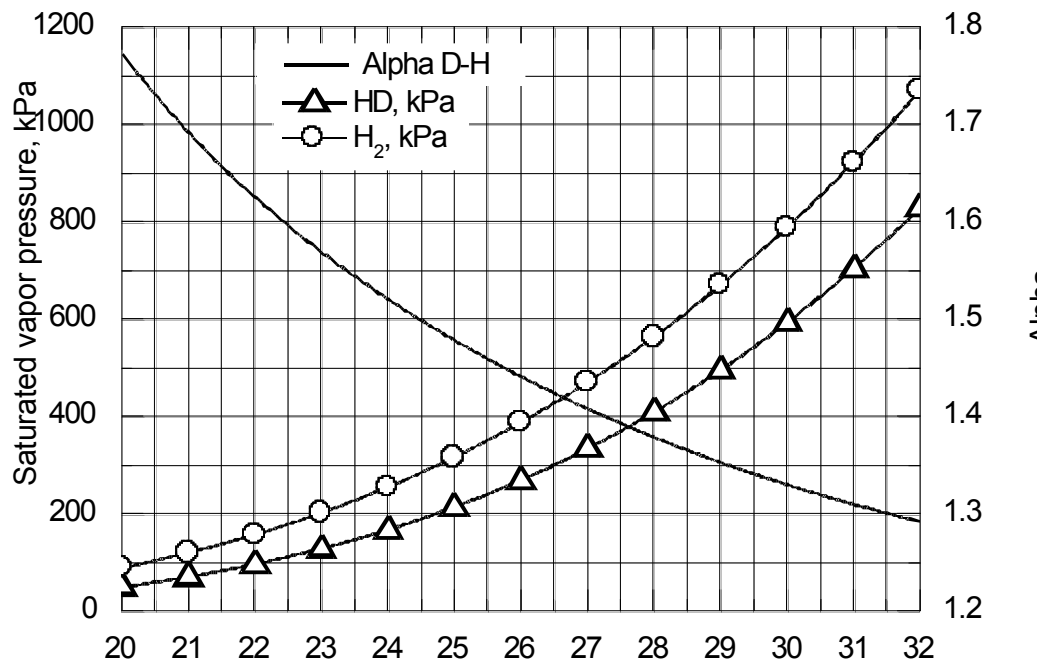


Figure 1. Temperature dependencies of saturated vapor pressures and separation factor for  $H_2$ -HD system

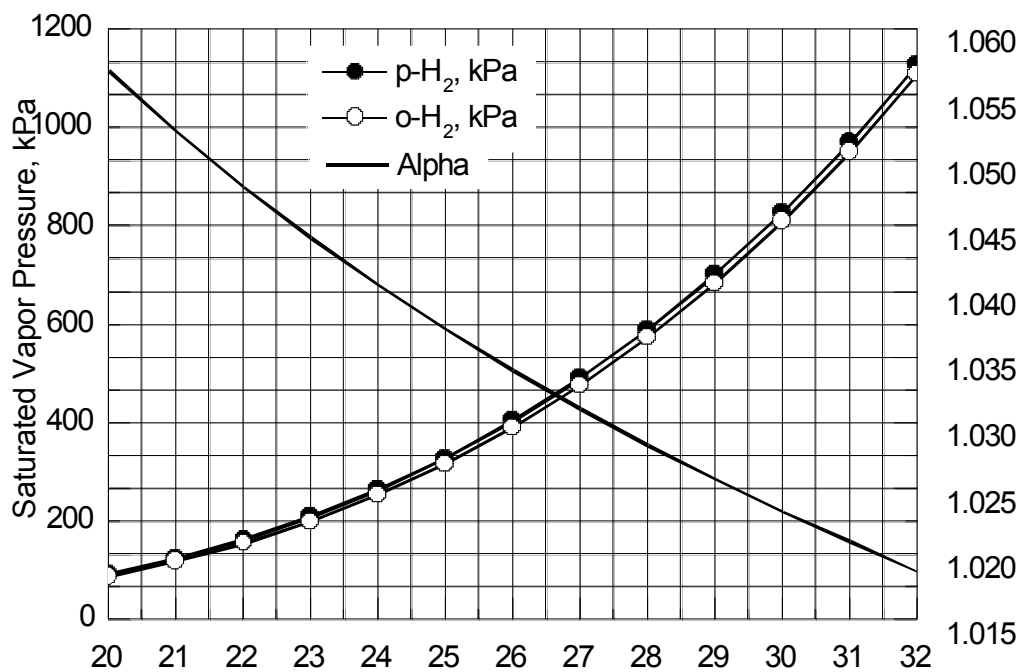


Figure 2. Temperature dependencies of saturated vapor pressures and separation factor for ortho-para hydrogen system

### 3. Design

#### Layout

A simplified layout of the deuterium separation unit (DRU) is presented in Figure 3. Historically, the DRU has been part of a chemical purification system [2] gas scheme, and has a through sequence of enumeration for the units and devices. The separation column of 2.2 cm inner diameter and 155 cm overall packing height is the main part of the unit. It consists of two sections connected by flange coupling. The column is encased in a vacuum jacket containing upper and lower attachments (adaptors) that support various hardware and vacuum terminals. The jacket and adaptors comprise a common vacuum volume. The volume is fixed on an adjustable tripod which provides precise vertical positioning of the column. All working parts of the device are placed inside the vacuum volume.

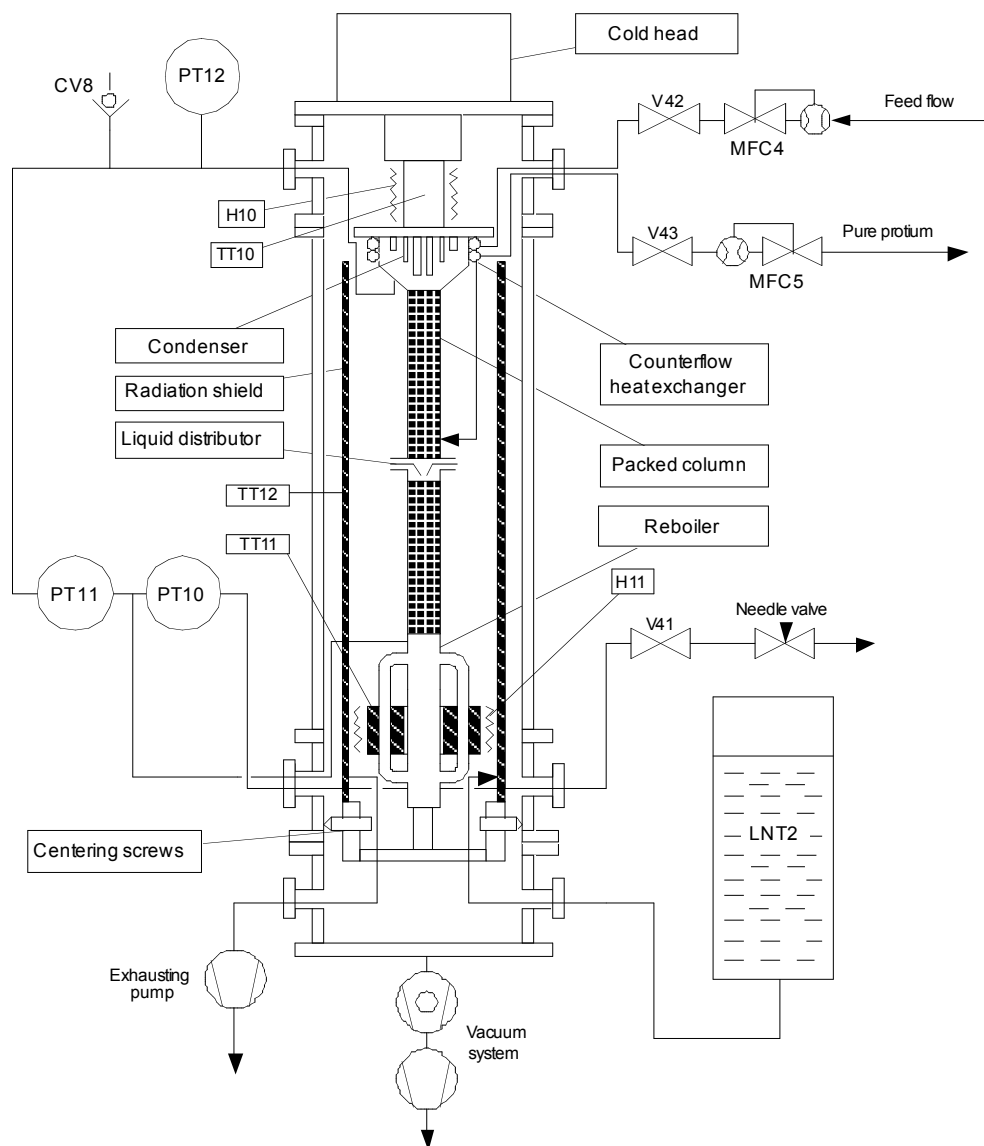


Figure 3. Simplified scheme of the DRU

The column has a condenser and a reboiler connected to its upper and lower part, respectively. A COOLPOWER 140T (Oerlikon Leybold Vacuum GmbH<sup>5</sup>) cryogenerator with a maximal cooling power of 20 W at 20 K is used for the cold operations.

A control system provides the necessary algorithms for column operation in all modes. Four modes were tested during our experiments: (1) “feedthrough” with purging of the deuterium-enriched mixture, (2) “feedthrough” without purging, (3) continuous circulation, and (4) so-called Rayleigh depletion. The differences between these modes are considered below. In all modes, the system operates two mass-flow controllers (MFC4 and MFC5 in the scheme), two heaters (H10 and H11), and reads signals from the temperature and pressure sensors.

### Condenser

The condenser is a closed, conical, bimetallic volume (Figure 4) whose upper part is made of copper plate. The inner part of this plate is processed in the form of lamellas to develop the heat transfer surface area. The vertical orientation of the lamellas facilitates the downward motion of the condensed hydrogen drops. The conical part has a flange for connecting to the column that is welded to the copper plate by electron-beam. The condenser has two connections for conducting product hydrogen and its pressure to an upper point of the differential manometer PT11.

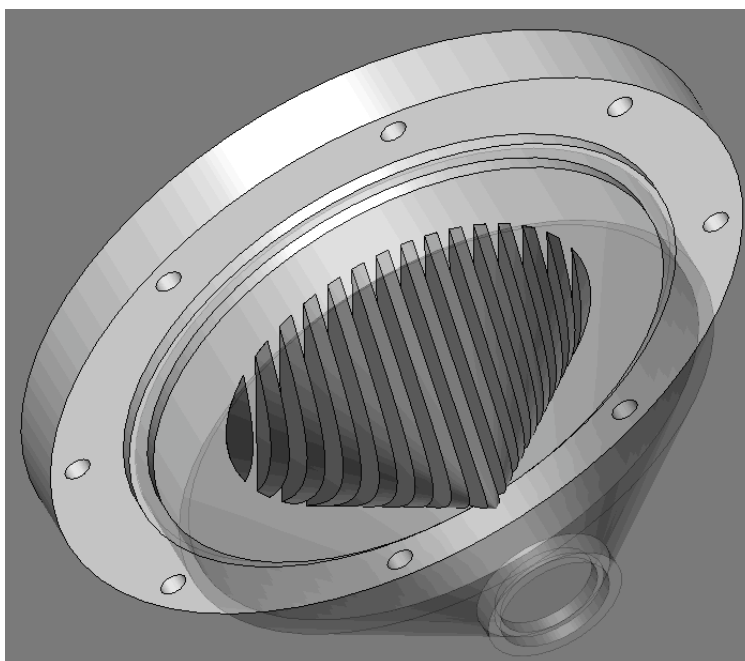


Figure 4. Condenser

The cold head tightly contacts with the outer surface of the copper plate through a thin indium foil to provide better heat flow. The cooled part of the condenser is mounted in the upper adaptor and therefore in the common vacuum insulation of the column. A counter-flow heat exchanger is mounted around the condenser. The heat exchanger first cools the feed flux using the product flux. The feed flux is further cooled and ultimately liquefied by

<sup>5</sup> <http://www.oerlikon.com/vacuumsystems/>

the cooling power of the cryogenerator. The cooled feed flux is delivered to the center of the column just above the liquid distributor.

### Reboiler

The reboiler is used to evaporate the separating mixture. Its rather complicated arrangement (Figure 5) is needed to provide steady and controllable boiling [7]. This unit mainly consists of a central tube and a massive copper muff equipped with an electric heater. Upper and lower collectors are welded to the central tube and connected with the muff by four lateral tubes (from above) and the manifold (from below). The connection between the steel parts and the copper body of the muff is provided by electron-beam welding.

Several longitudinal apertures are drilled in the muff around the large central hole. An electric heater is coiled around its body and a PT100 temperature sensor (TT11) is fixed on it. The sense of its structure is shown on the scheme of fluxes (Figure 6).

The central tube keeps most of the liquid hydrogen collected in the bottom of the column. This hydrogen forms the steady lift of the liquid which can be precisely measured by the differential manometer (upper and lower arms of the manometer are connected to the lower part of the column and to the lower collector of the reboiler, respectively).

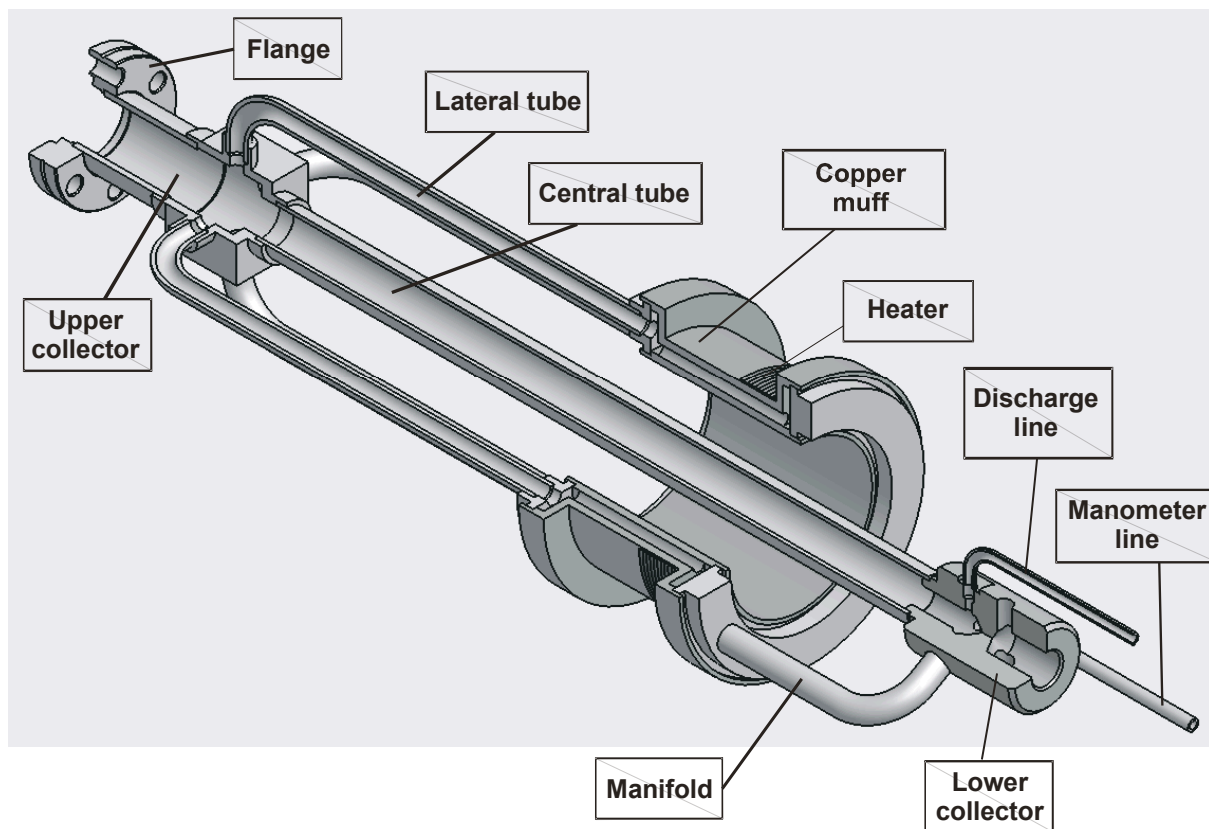


Figure 5. Sectional drawing of the reboiler

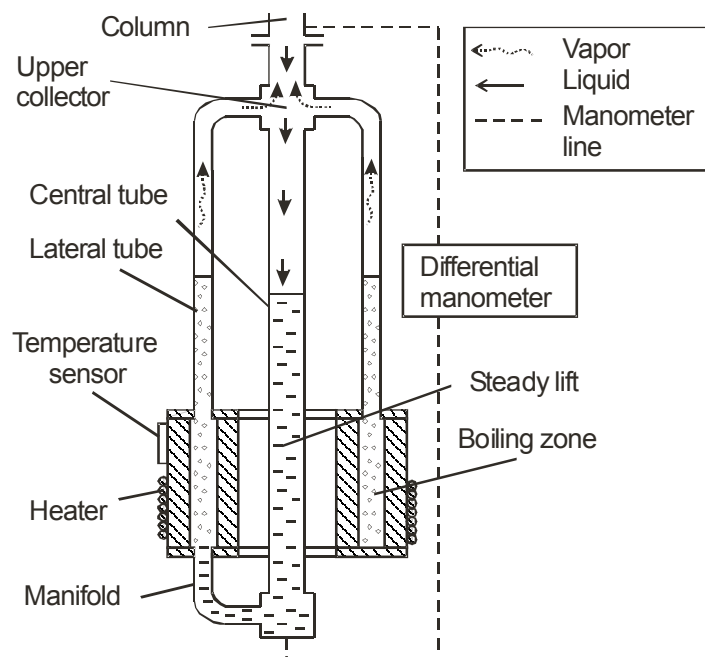


Figure 6. Scheme of fluxes in the reboiler

The boiling zone is removed to the apertures of the muff. The forming vapour moves from the muff upward through four lateral tubes to the upper collector and further to the column. The liquid is delivered to the boiling zone from the central tube through the manifold of the lower collector under the action of hydrostatic pressure of the steady lift.

### Packing

The column is filled with specially designed spiral prismatic random packing made of 0.2 mm stainless steel wire (Figure 7). The packing is intended to provide a maximal surface for phase contact. Given the  $2 \times 2$  mm size of the prismatic springs and the 560 ml volume of packing in the column, it provides the surface area of  $1.95 \text{ m}^2$ . The choice of stainless steel was dictated by the purity requirements for hydrogen. The packing surface was pickled with nitric acid to develop its roughness and improve wettability by liquid hydrogen.

This was the first test of this particular packing in a cryogenic distillation column. A similar type of packing, but of bigger size ( $3 \times 3$  mm), was tested earlier in PNPI water distillation columns of 80 mm inner diameter, and exhibited good separation characteristics ( $\text{HETP} = 2.5\text{-}3 \text{ cm}$ ) [9]. A similar packing made of stainless steel (Heli-Pak and Coil Pack) tested in a cryogenic distillation column of small diameter gave an HETP value about 5-6 cm [10]. Thus, the HETP value of 5 cm was put in the design according to a conservative approach.



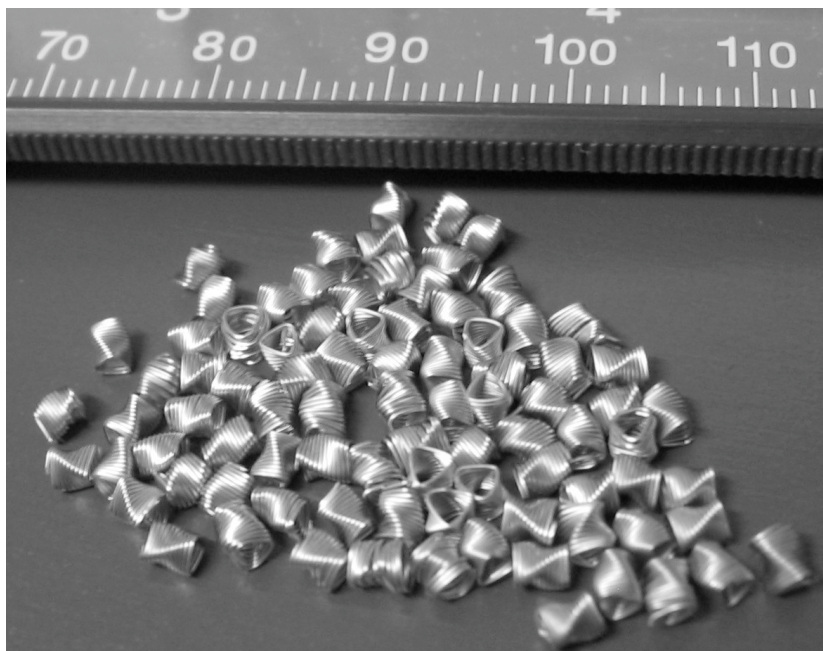


Figure 7. Packing

### **Operating modes**

As mentioned earlier, there are several possible DRU operating modes for producing deuterium-depleted hydrogen. Four modes were tested during the experiments: “feedthrough” with purging of the deuterium-enriched mixture, “feedthrough” without purging, continuous circulation, and so-called Rayleigh depletion.

During Rayleigh depletion the column was fully primed with liquefied hydrogen. After the priming, the hydrogen was released through the product line. Natural hydrogen was used only for preliminary tests. The accumulation of required product was carried out from pre-depleted protium.

In the “feedthrough” modes the mass-flow controllers were used to operate the feed (MFC5) and the product (MFC4) flows. The rate of the purging flow was adjusted with a manual needle valve using the bubble flow meter.

## **4. Analysis**

### **Chromatographic analysis of ortho-para composition**

A method for quickly determining the deuterium content in deuterium-depleted hydrogen was not available during the experiment. It was essential, however, to be able to rapidly estimate column performance so that adjustments could be made. A chromatographic analysis of ortho-para isomers of hydrogen was proposed as a method of indirect estimation of the column’s separating power.

A chromatographic device equipped with a  $\text{Al}_2\text{O}_3$ -filled chromatographic column immersed in liquid nitrogen was used for the analysis. The low temperature is essential for separating the isomers, which have temperature-dependent differences in adsorption affinity. Neon was used as the carrier gas, instead of the more conventional helium, in order to

provide a greater difference in thermal conductivity between the separated species and the carrier gas.

Measurements of the concentration of ortho and para isomers in the top and bottom regions of the column make it possible to estimate its separation performance, and to calculate the concentration profile for deuterium. An example of a chromatogram for samples taken from the top and bottom part of the column is shown in Figure 8.

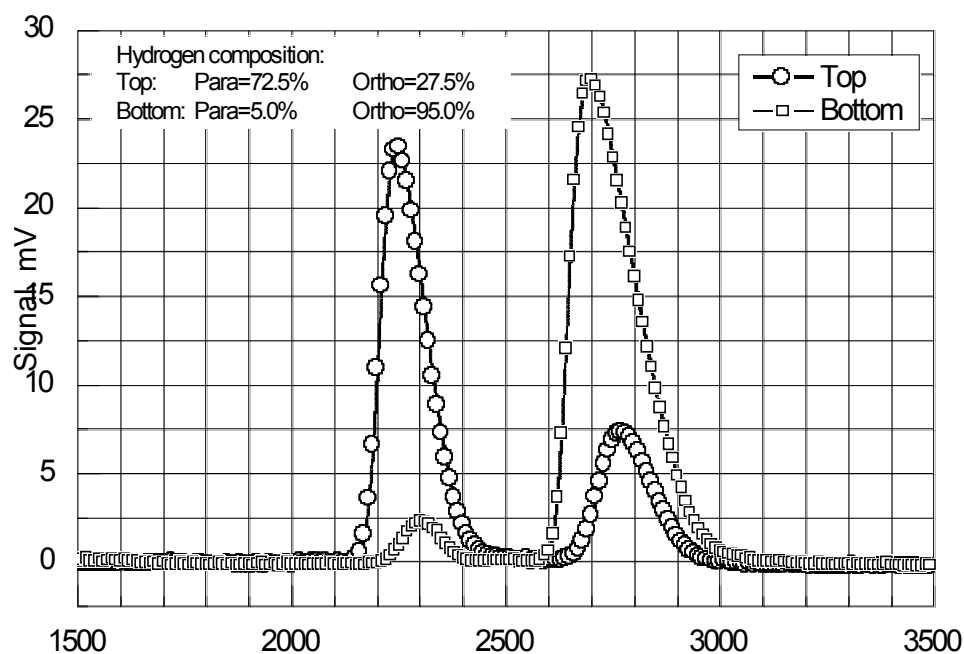


Figure 8. Chromatogram of separated ortho- and parahydrogen. (Column pressure: 1.2 bar; Reboiler power: 10 W; Operating mode: total reflux)

A specially prepared synthetic mixture of  $H_2$ , HD, and  $D_2$  was prepared for the column test. The mixture was prepared by diluting natural hydrogen with pure deuterium and subsequently passing the mixture through the catalytic column under heating conditions. The column is filled with palladium catalyst immersed in granulated silica gel. The mixture thus obtained is analyzed by the same chromatographic device as used for ortho-para composition measurements, albeit with minor readjusting of the regime [11]. The chromatogram of the converted mixture is shown in Figure 9.

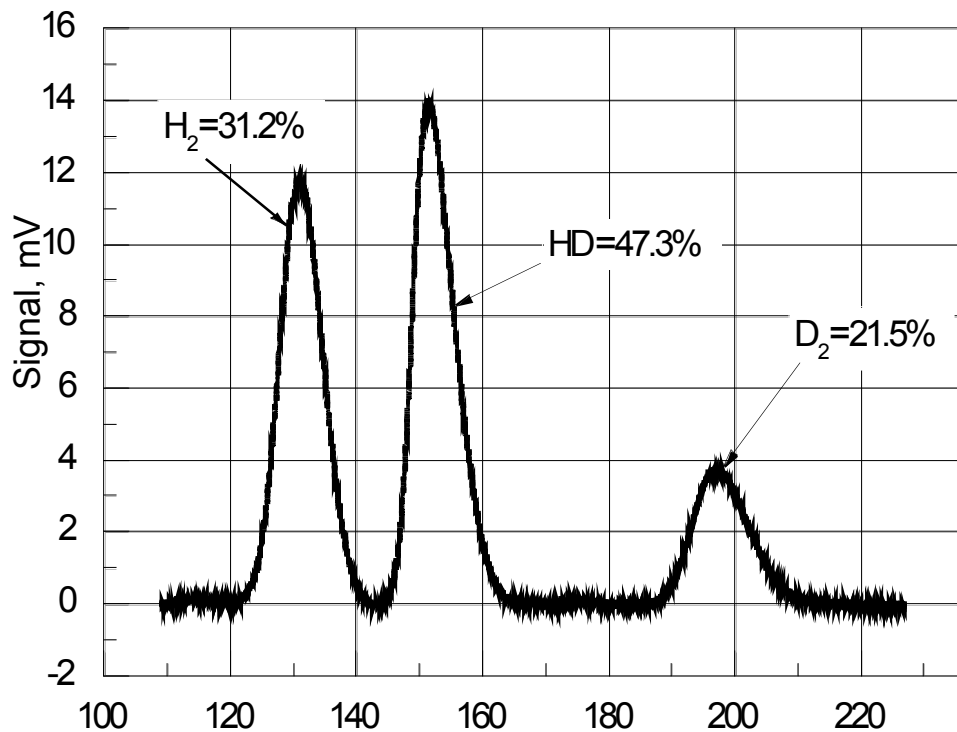


Figure 9. Chromatogram of synthetic mixture of hydrogen isotopes

#### Analysis at large tandem accelerator

Direct measurements of low deuterium concentrations in samples were carried out by a large tandem accelerator group at the Institute of Particle Physics, ETH-Zurich, Switzerland. Measurements of the probe samples were performed using a new 200 kV accelerator designed for isotope analysis in Zurich [12]. Its unique ion source provides extremely low backgrounds (e.g. hydrogen ions from walls, etc.).

## 5. Results of tests and production

#### Pressure drop and the liquid holdup studies

For the liquid holdup measurement, a known quantity of hydrogen was batched into a column. From the observed liquid level and reboiler dimension, the quantity of hydrogen resident on the packing was calculated. It was assumed that the liquid film in the overhead condenser contributed insignificantly to the column inventory. Figure 10 shows the relation between the pressure drop across the column and the vapor flow rate of natural hydrogen at various pressures. The flooding regime was not achieved due to the relatively large (for this design) inner column diameter. The relatively low pressure drop can be explained by the moderate range of vapor flow rate. The packing holdup presented in Figure 11 had an expected high value in the full range of vapor fluxes.

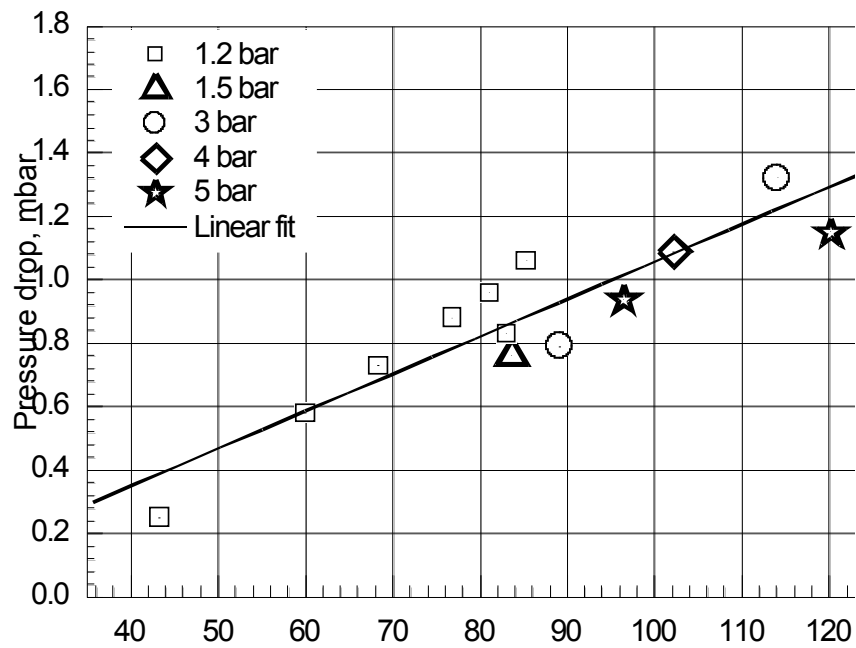


Figure 10. Pressure drop across the column

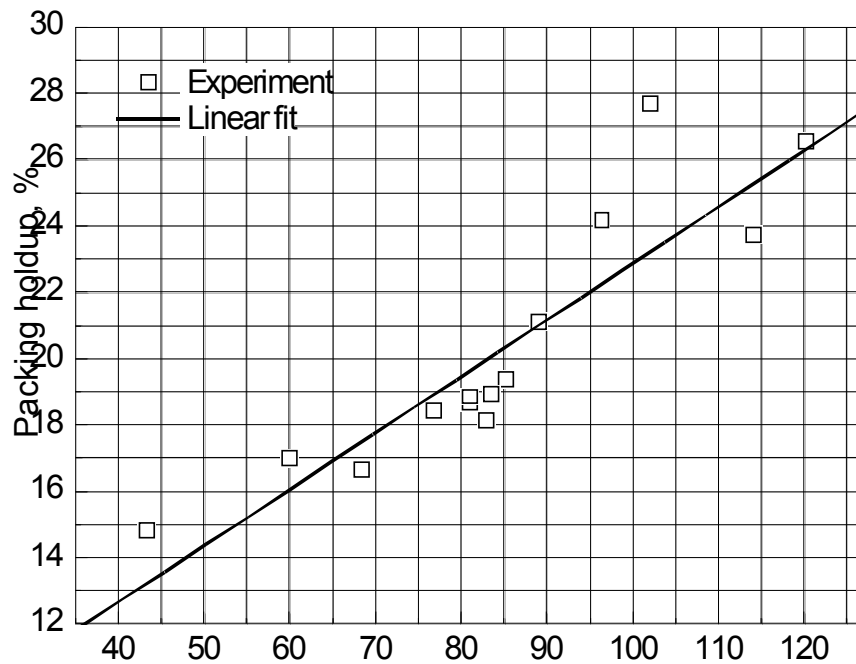


Figure 11. The holdup as a function of the flow rate

The first DRU run was carried out using natural hydrogen with  $126.9 \pm 1.9$  ppm of deuterium. The initial gas volume was 181.6 liters. The column was filled by liquid hydrogen in large excess, and after 1 hour of operation the excess gas (84 liters) was withdrawn from

the column bottom through the discharge line. This explains the low final deuterium concentration in the bottom (Table 1). Withdrawn gas was sufficiently enriched to significantly change the average concentration in the column. The negative deuterium concentration in the column's top (-1.9 ppm) indicates the precision of the measurement resolution. (The method relies on extrapolating a measurement of deuterium content from a sample of natural hydrogen, and gives low accuracy for close-to-zero concentrations.) Thus the small negative value indicates effectively zero value for the HD admixture.

Probe location	HD concentration, ppm	Orthohydrogen concentration, %
Source gas (measured)	126.9±1.9	75
Column top	-1.9	14.6
Column middle		32.2
Column bottom	56.2	85
Average concentration calculated from the mass balance	7.7	53.5

Table 1. Natural hydrogen run results

The experimental data for the ortho-para separation were fitted with the Fenske equation (Figure 12). The average HETP calculated on the basis of tests with various vapor flow rates is 2.2 cm (Figure 13), which is comparable with the best world results [12].

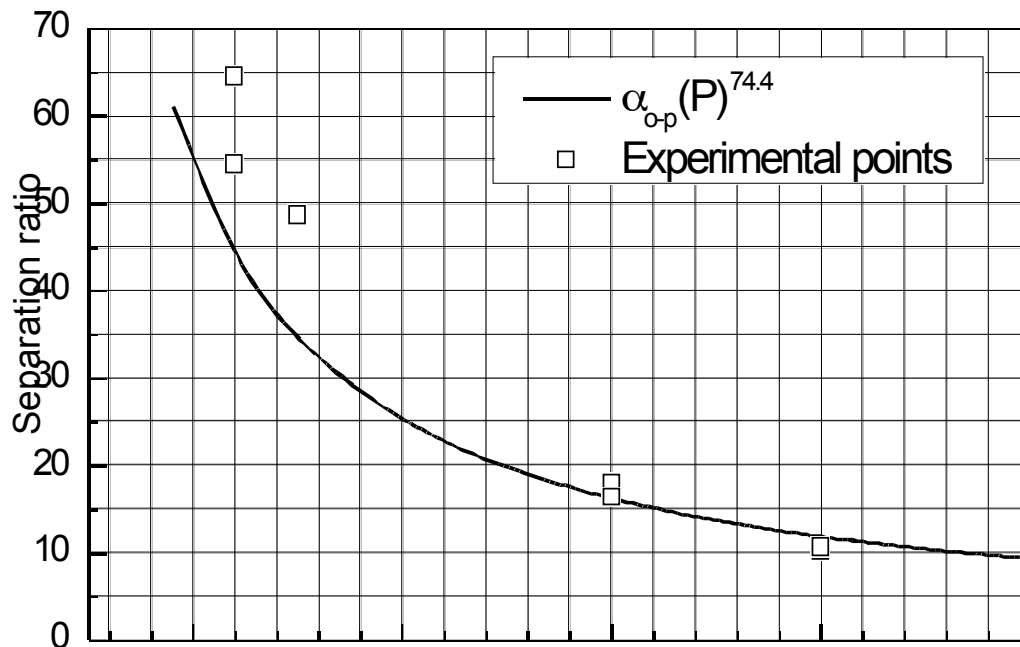


Figure 12. Fenske equation fit for total reflux mode

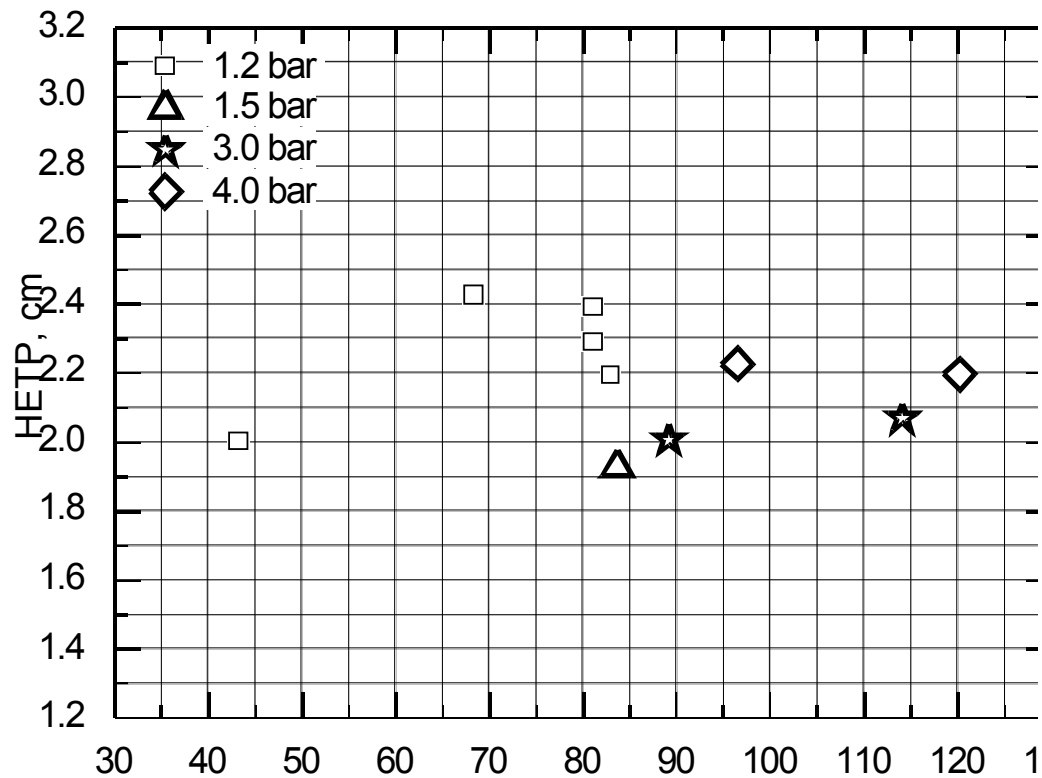


Figure 13. HETP at various flow rates

#### Natural gas separation in “feedthrough without purging” mode

During this test, a large amount of natural hydrogen (~ 6000 liters) was passed through the column to increase the average deuterium concentration and thereby obtain measurable concentrations in the middle of the column.

#### Production of deuterium-depleted hydrogen

The production procedure was carried out using all possible operating modes in order to determine which one was optimal. The results of the experiments are shown in Table 2. “Circulation” mode corresponds to the “Feedthrough without purging” regime, with the difference that the gas was continuously circulated through the column system for two days. The goal of this test was mainly to clean the circulation system of all traces of deuterium.

In addition, a number of deuterium-depleted probes were collected—so-called “zero protium” samples. The expected HD concentrations (Figure 14) were calculated using the special simulation program based upon the aforementioned mathematical model with known HETP and total packing height of the column.

Mode	“Feedthrough” no purging		“Feedthrough” with purging	Rayleigh’s depletion	Circulation
Reboiler power, W	20.5		19.0	12.0	20.0
Pressure, bar	1.5		1.5	1.5	2.0
Feed flow, l/min	0.81		0.81	1.25	0.55
Ortho-hydrogen, %	Top	67.8	69.4	–	64.8
	Middle	69.2	74.6	94.2	69.3
	Bottom	96.4	98.5	–	98.2
HD initial, ppm	$1.45 \pm 0.14$		$<1.45 \pm 0.14$	10	$<1.45$
HD in the product, expected, ppm	$< 0.01$		$< 0.01$	0.09	$< 0.01$
Amount of gas, Standard Liters	572		630	390	420
Bottom probe HD measured, ppm	$57.5 \pm 1.2$		$76.9 \pm 1.6$	–	$16.2 \pm 0.5$

Table 2. Results of cleaning

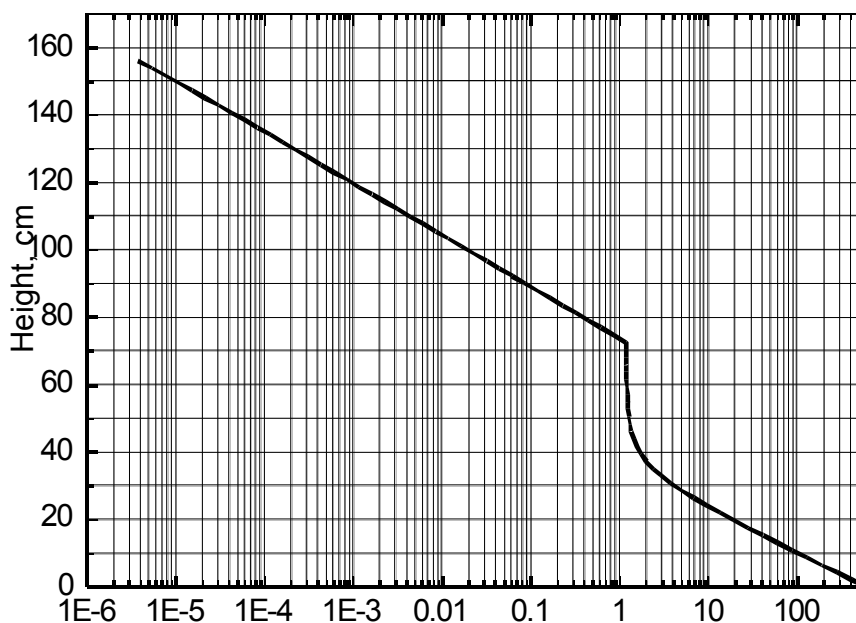


Figure 14. Expected concentration profile for HD. Feed flow = 1 l/min, pressure = 1.5 bar, purging flow = 0.015 l/min, HETP = 2.2 cm, initial deuterium atomic fraction = 3 ppm.

The column's deuterium concentration profile, as calculated from the ortho and para hydrogen separation, yields a very low value at the top of the column. This low concentration could not be confirmed by online direct measurement because of insufficient sensitivity in the mass spectroscopy.

### **Direct deuterium measurements by the accelerator method**

The direct analysis of product samples using accelerator mass spectrometry at the Institute of Particle Physics, ETH-Zurich, Switzerland [12], indicated an absence of signal at 6 ppb sensitivity, which means at least better than 6 ppb. This is an order of magnitude better than the technical requirements of the MuCap experiment, and corresponds to the most isotopically pure protium in the world.

### **Measurements of deuterium concentration in depleted hydrogen**

As mentioned earlier, even the accelerator method provides insufficient sensitivity for direct deuterium measurements at extremely low concentrations. Therefore, it was proposed to use the DRU to concentrate the deuterium from a large amount of depleted hydrogen. The HD admixture is measured in the enriched "stillage bottoms," which is hydrogen collected in the reboiler. The final result was obtained by dividing the measured value by the ratio of the concentrated "stillage bottoms" and the full volume of hydrogen.

The concentration was provided by cleaning the full hydrogen amount in "feedthrough without purging" mode. The full volume of the gas was precisely measured by the previously calibrated mass-flow controller MFC4. The calibration was provided by discharge of hydrogen from the vessel with the well-known volume under control of pressure by a precise manometer.

The entire amount of deuterium-depleted hydrogen available (846.7 liter in total) was passed through the column. As a result, 80% of deuterium was concentrated in approximately 15 ml of liquid hydrogen in the reboiler. The coefficient of concentration was calculated on a basis of the mathematical model.

### **Direct isotopic measurements via gas chromatography**

The gas chromatographic device used in the present work permitted direct measurements of large (1% and higher) concentration of components ( $H_2$ , HD or  $D_2$ ) without modifying the measuring apparatus. The mixture for this test was specially prepared by diluting pure  $D_2$  with natural hydrogen, by passing the mixture through a column filled with palladium catalyst immersed in silica gel. The equilibrated composition of the mixture after the conversion had the composition: 31.2%  $H_2$ , 47.3% HD, rest  $D_2$  (Figure 15).

The test of the  $H_2$  – HD –  $D_2$  mixture was done at higher pressure (5 bar) to reduce the separation factor. On the basis of an assumed  $N$  and observed composition of hydrogen isotope, the fraction profile of hydrogen isotopes in the column can be calculated.

Finally, the value number of theoretical plates is obtained to give the best-fit values of calculated compositions with the hydrogen isotope compositions in the top, middle and bottom of the column. The HETP calculated in this test was 1.95 cm, which conforms with the previously obtained value.



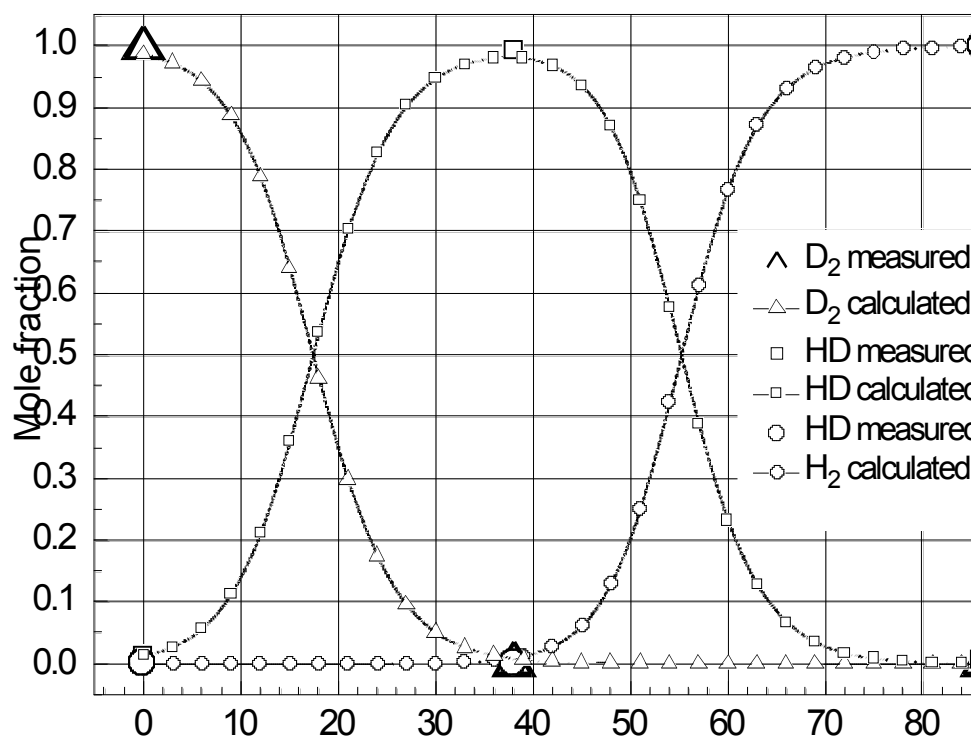


Figure 15. Isotope composition along the column

## 6. Conclusions

The height equivalent to a theoretical plate (HETP) for the column is 2.2 cm, on the basis of our measurements. It is one of the best results yet for columns of low and medium cryogenic power. A new packing design was developed and successfully tested.

The column's isotopic separation power exceeds the demands of the MuCap experiment. The output deuterium concentration is lower than 6 ppb, and this value is nearly independent of the initial concentration. The output of the system in the “feedthrough” modes, which are most appropriate for production, is 500 l/day. A sufficient amount of deuterium-depleted hydrogen was obtained for use in MuCap experimental runs in 2006 and 2007.

In addition to deuterium-depleted hydrogen, the setup can produce pure ortho-hydrogen, which is useful for some experimental physics applications.

A method for measuring low deuterium content in hydrogen (1 ppm and less) was developed and used to measure the deuterium admixture for the 2006 MuCap run.

## 7. References

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