System for Continuous Chemical and Isotopic Purification of Hydrogen for the MuCap Experiment

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Why we need ultra pure hydrogen?

- Heavier elements

Negative muons preferentially transfer from ***μp*** atoms to heavier elements with high rates. Once a muon has transferred to a μN_Z atom, nuclear muon capture proceeds more than 100 times faster than on a ***μp*** atom.

Thus, even tiny amounts of impurities in the hydrogen gas distort the observed ***μp*** lifetime spectrum.

Consequently, muon transfer and subsequent capture must be suppressed by keeping the gas contaminants below a level of ***10 ppb***.

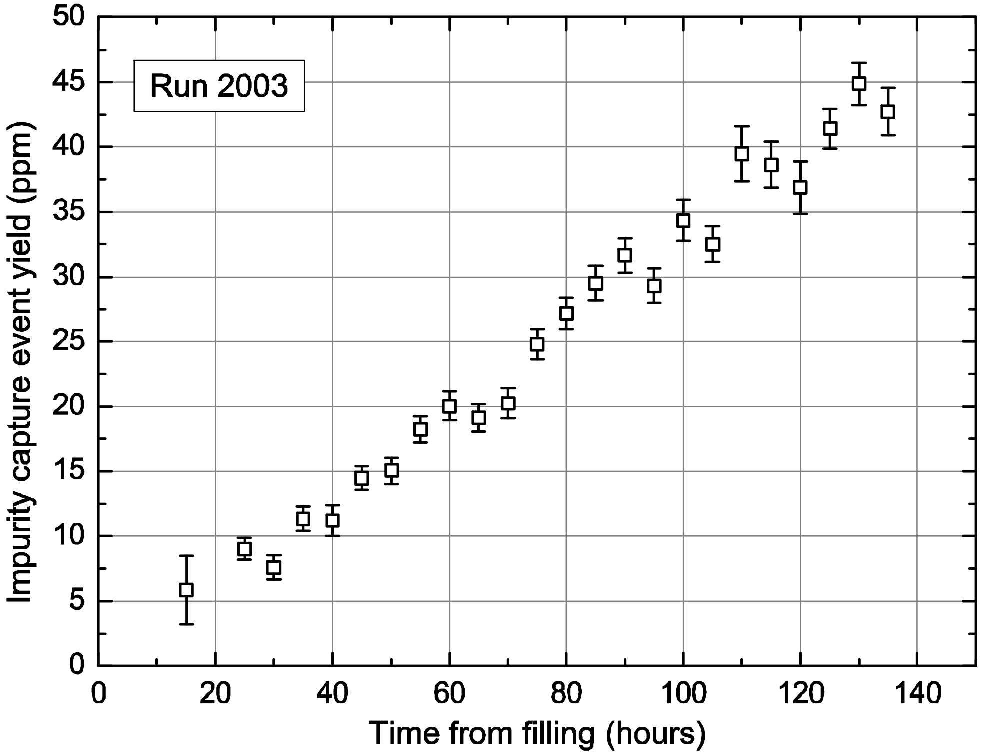
- Deuterium

Isotopically pure hydrogen is required, since muons transfer to deuterium, where they pose a systematic problem for the MuCap experiment.

Due to a ***Ramsauer-Townsend*** minimum in the ***μd + p*** scattering cross-section, ***μd*** atoms can diffuse over macroscopic distances and can either escape from the stopping volume in the TPC in a time-dependent way and can even reach the chamber materials, where muons are quickly captured.

Why we need continuous purification?

Impurity capture events measured **before purification system installation**, shows the impurity capture event yield increasing in the hours after filling the hydrogen vessel through the palladium filter (better than 1 ppb).



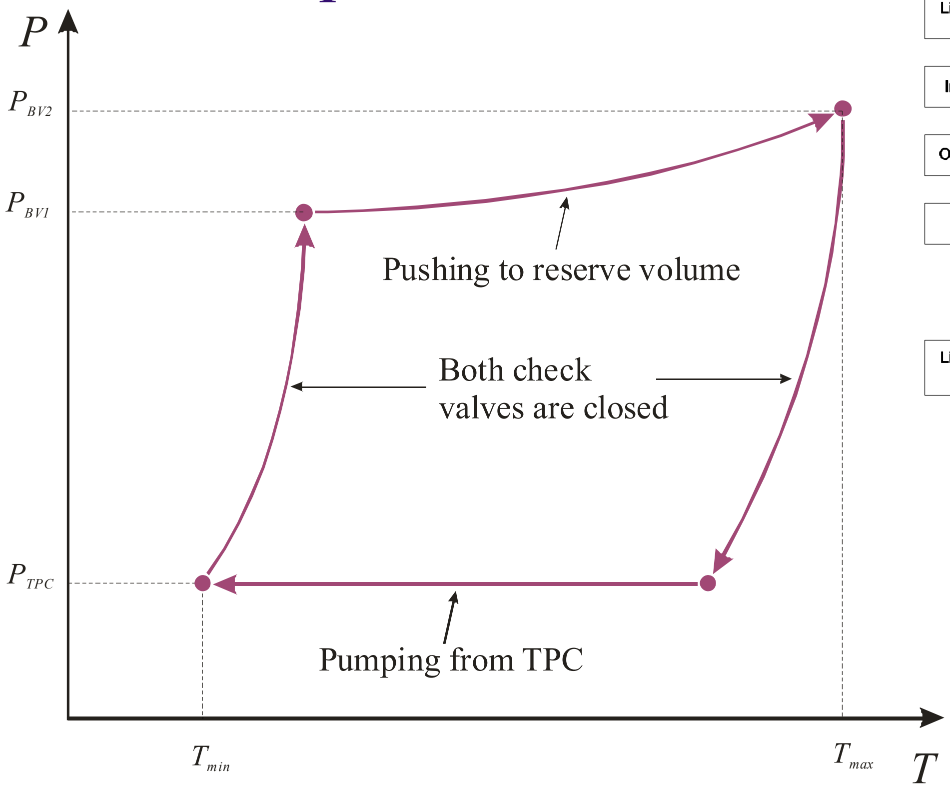
Reasons for Circulation system?

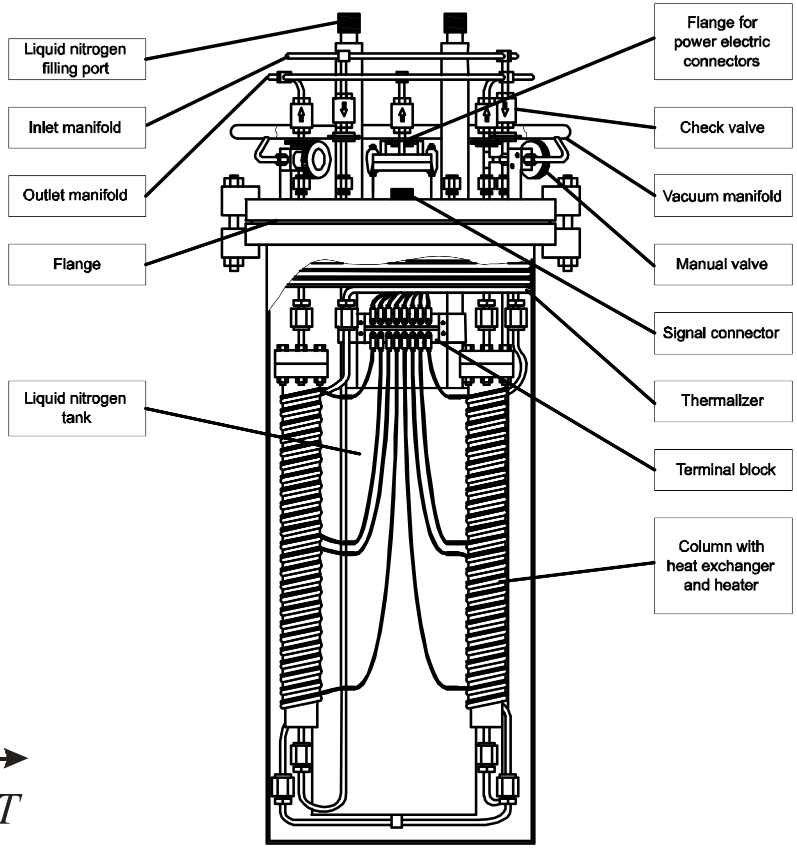
- constant flux is necessary for the permanent gas purification.
  about 3 l/min
- price of pure hydrogen is essential.
  about 1000 Eur/m^3
- stable pressure in TPC is important.
  10 bar +/- 0.1% (10 mbar)

Type of compressor? Normal or Cryo-?

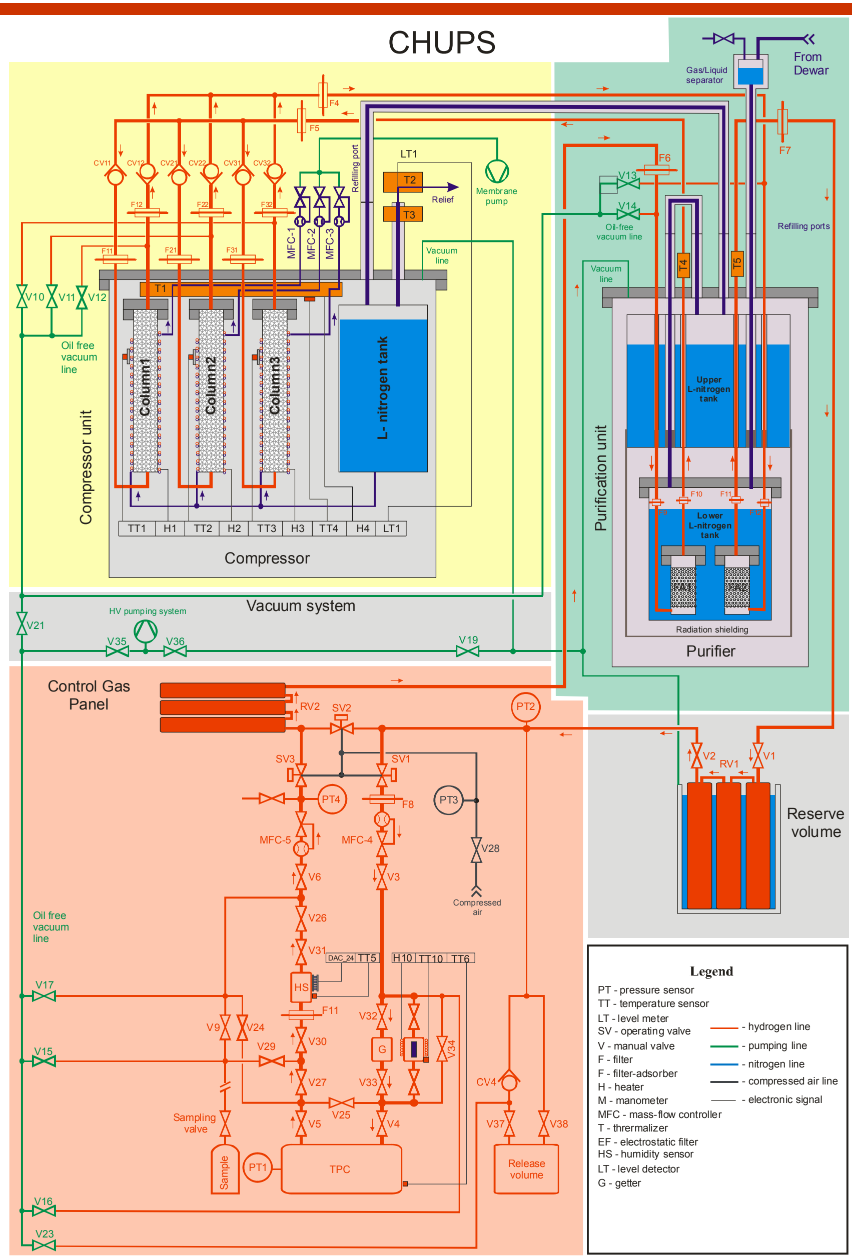
10 bar absolute pressure
3 bar differential pressure
3-4 l/min flux
ultra pure!!!

Simplified P-T diagram of a Compressor column

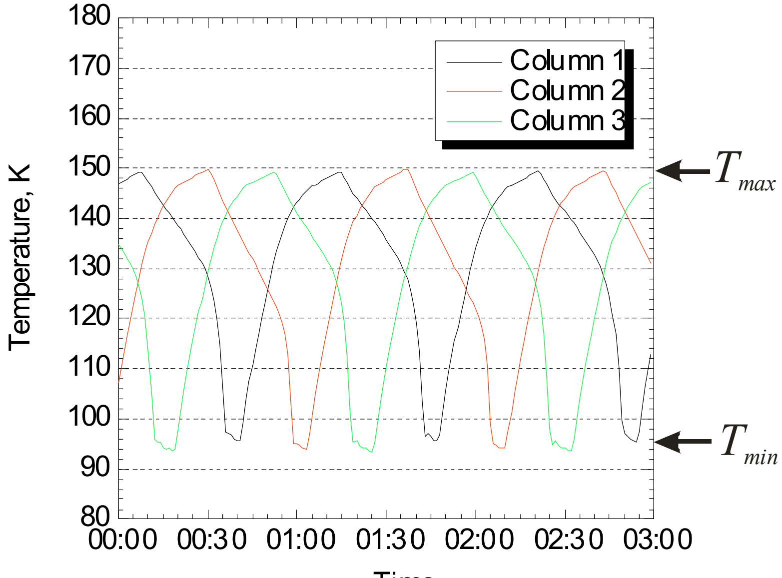


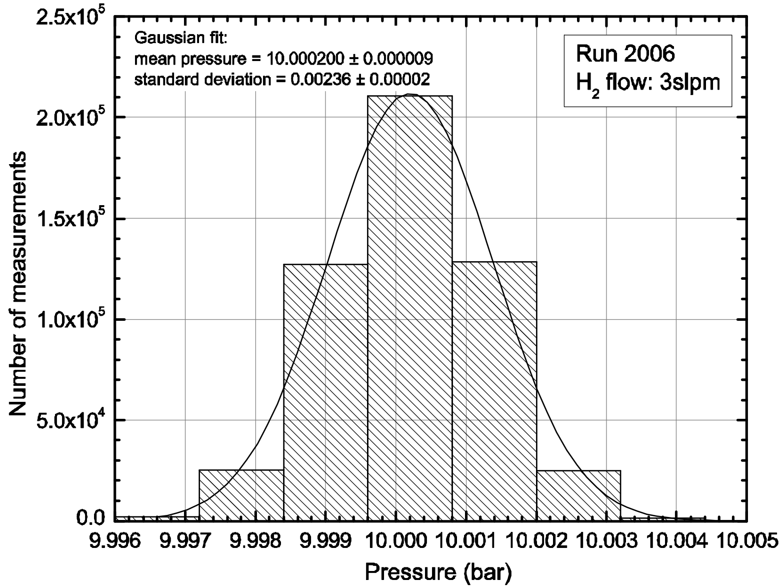


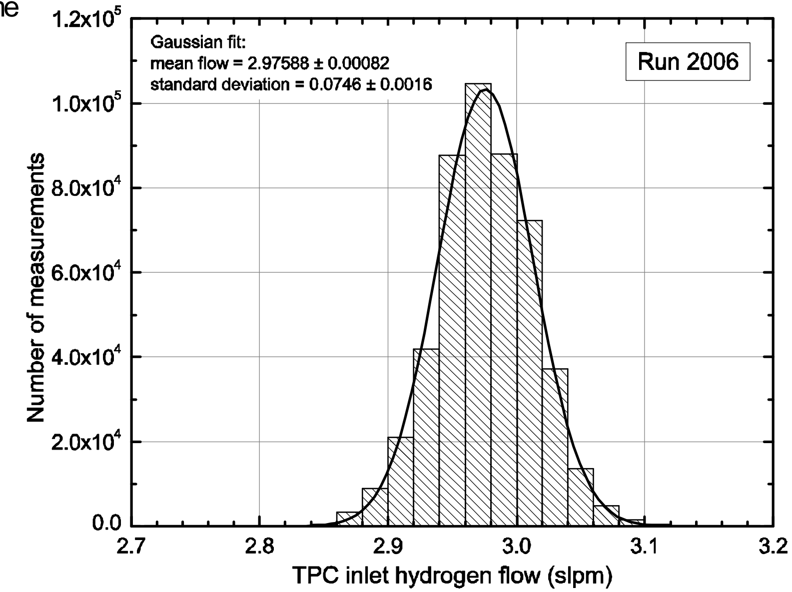
CHUPS scheme



CHUPS, Hydrogen flux 3 l/min

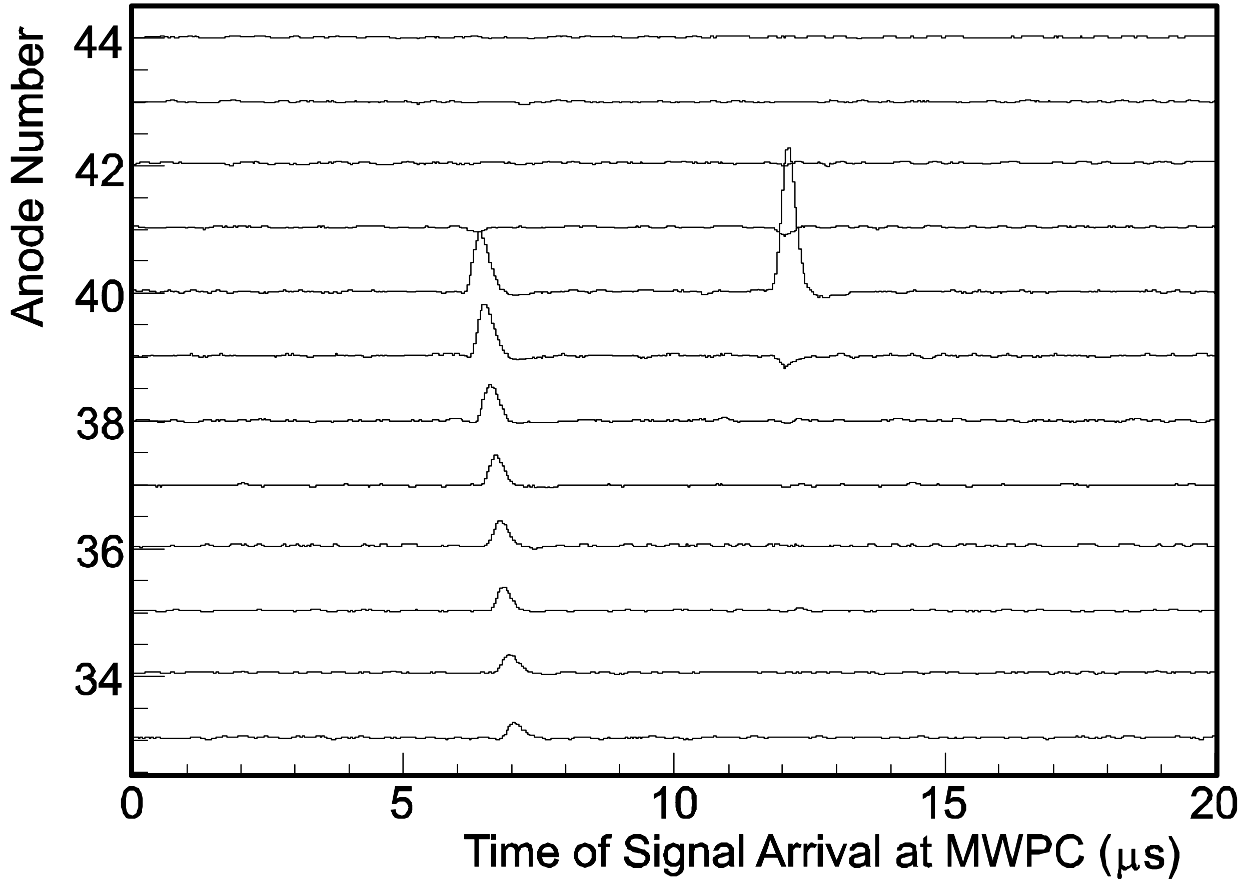






Nitrogen purification

The capture process $\mu N_Z \rightarrow N_{Z-1} + \nu$ is identified by its distinct signature in the TPC

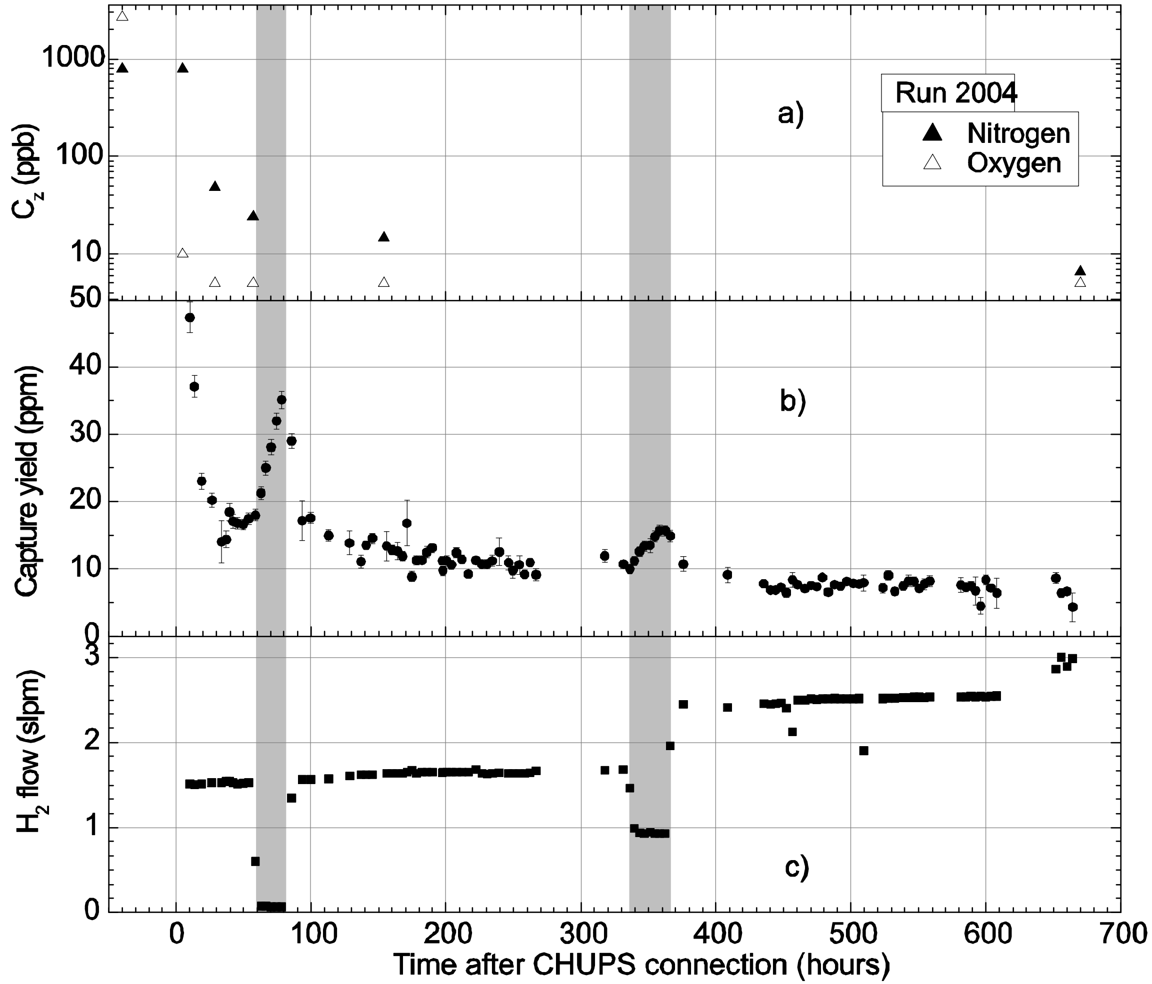


Nitrogen+Oxigen

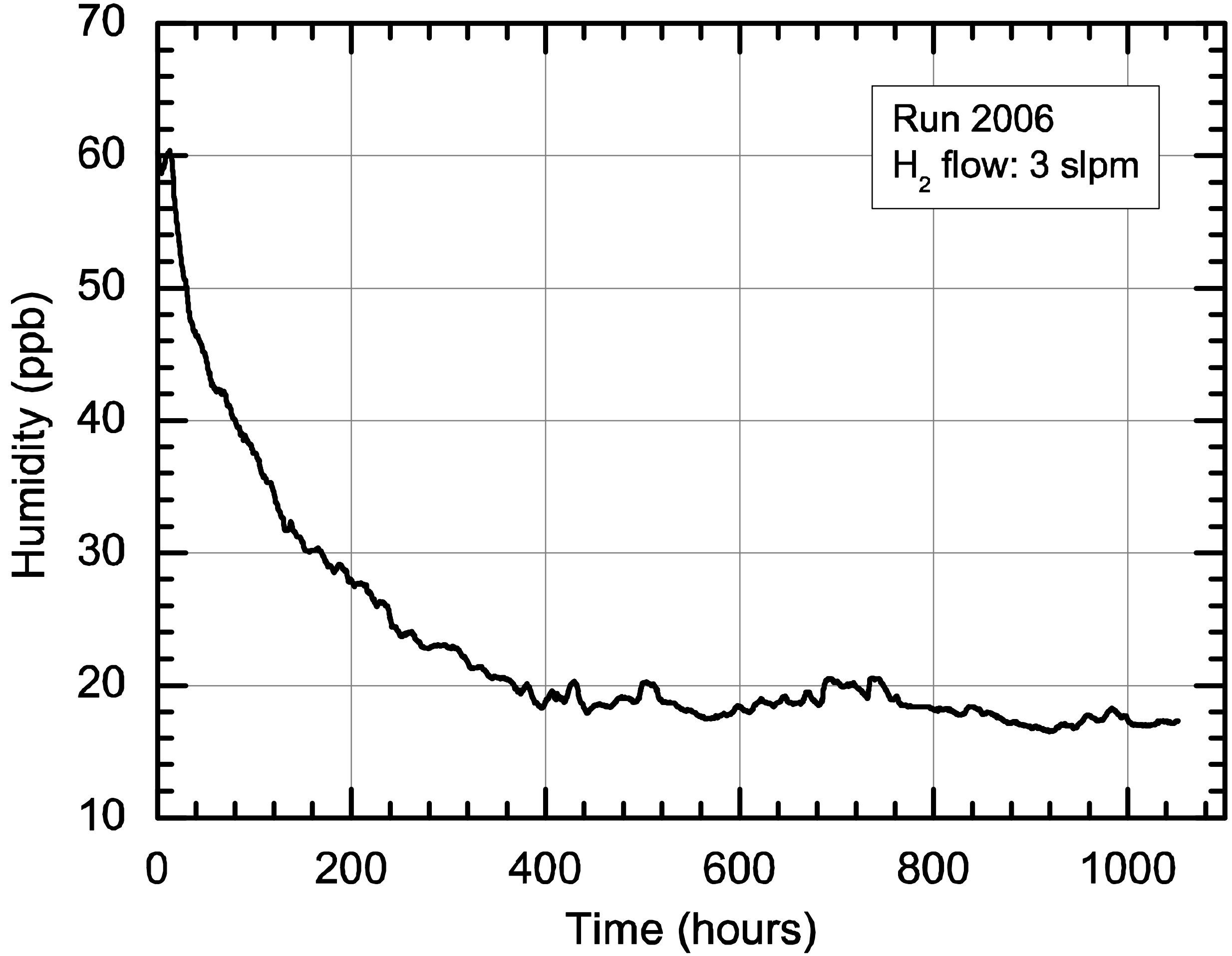
Chromatography measurements also track the purification process (-a).

Oxygen traces dropped below the apparatus' sensitivity of ***5 ppb*** within two days after starting the circulation.

Nitrogen concentrations below the apparatus' sensitivity of ***7 ppb***.

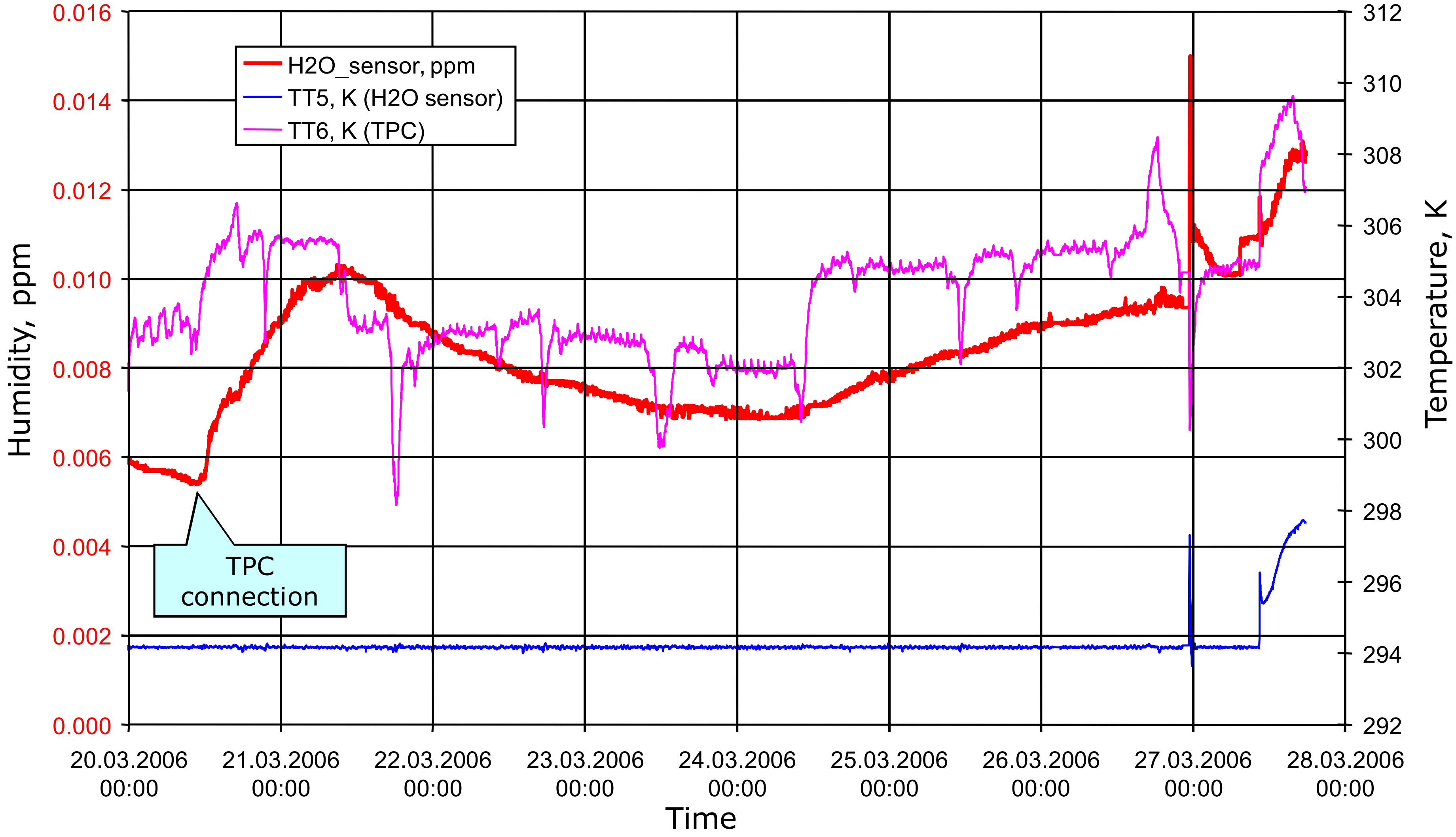


Humidity



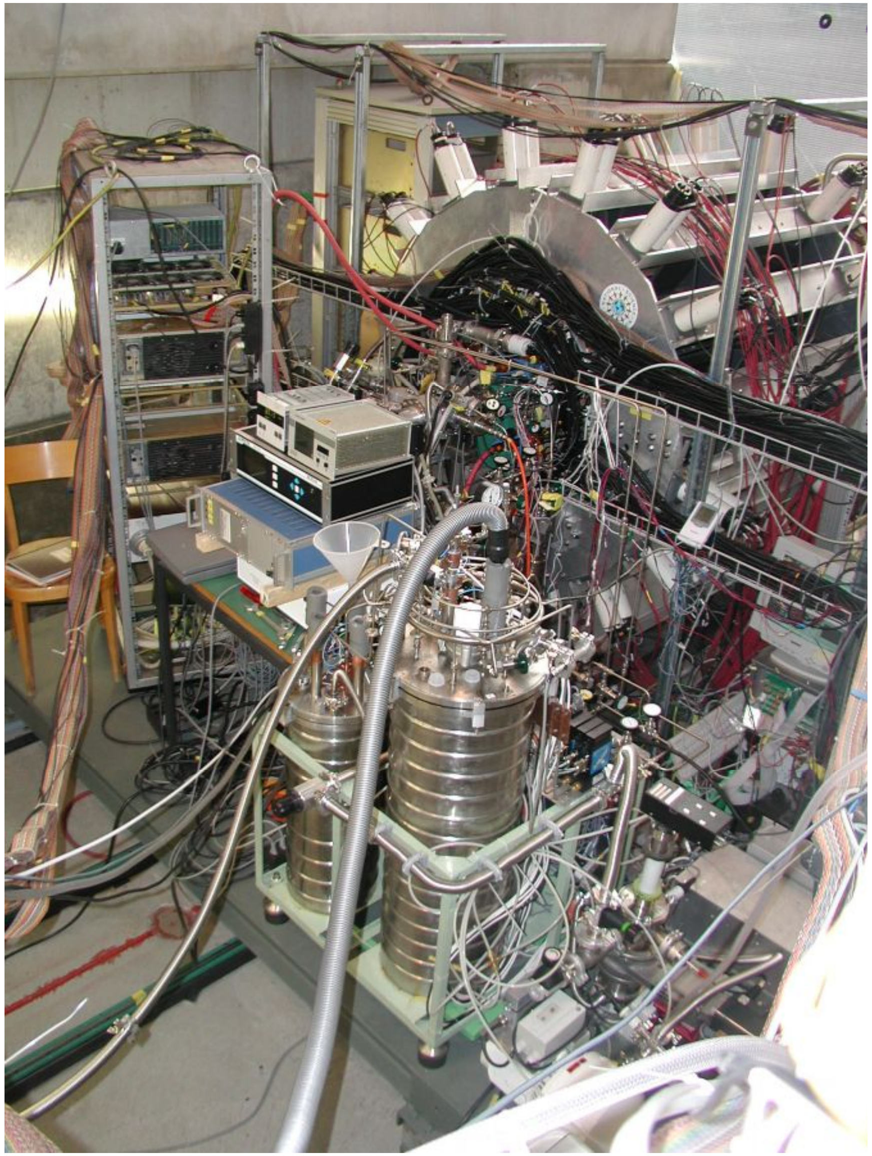
Run 2006
H2 flow: 3 slpm
Humidity (ppb)
Time (hours)
70
60
50
40
30
20
10
0
200
400
600
800
1000

Moisture measurements in CHUPS with TPC

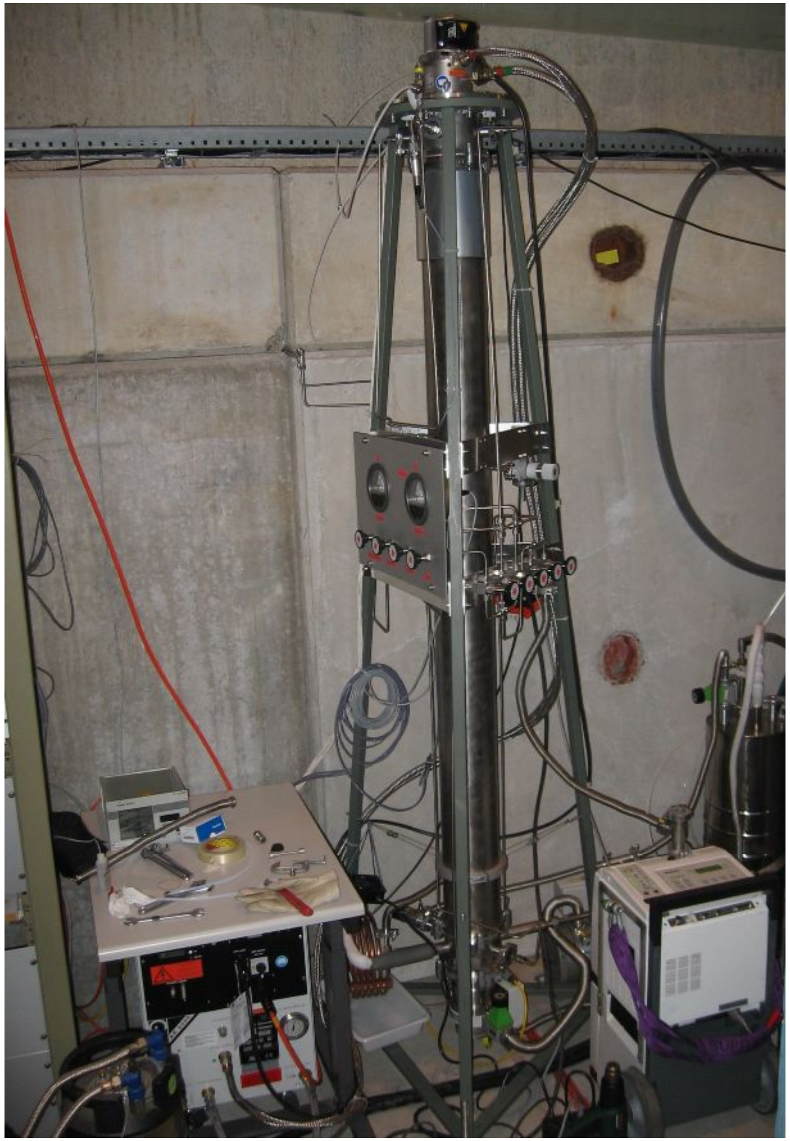


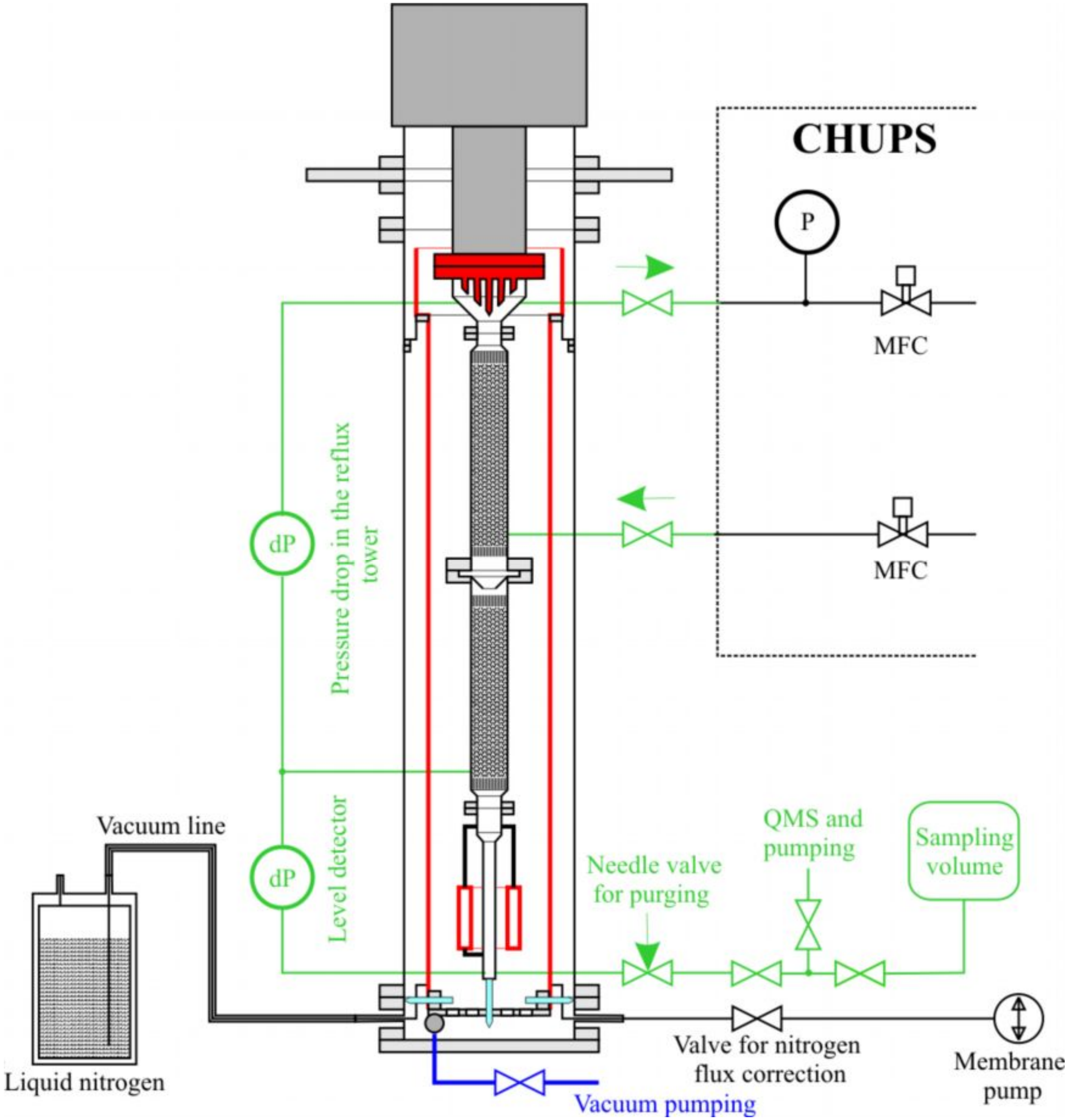
CHUPS results

N_2 less than 5-7 ppb
O_2 less than 5 ppb
H_2O about 20 ppb

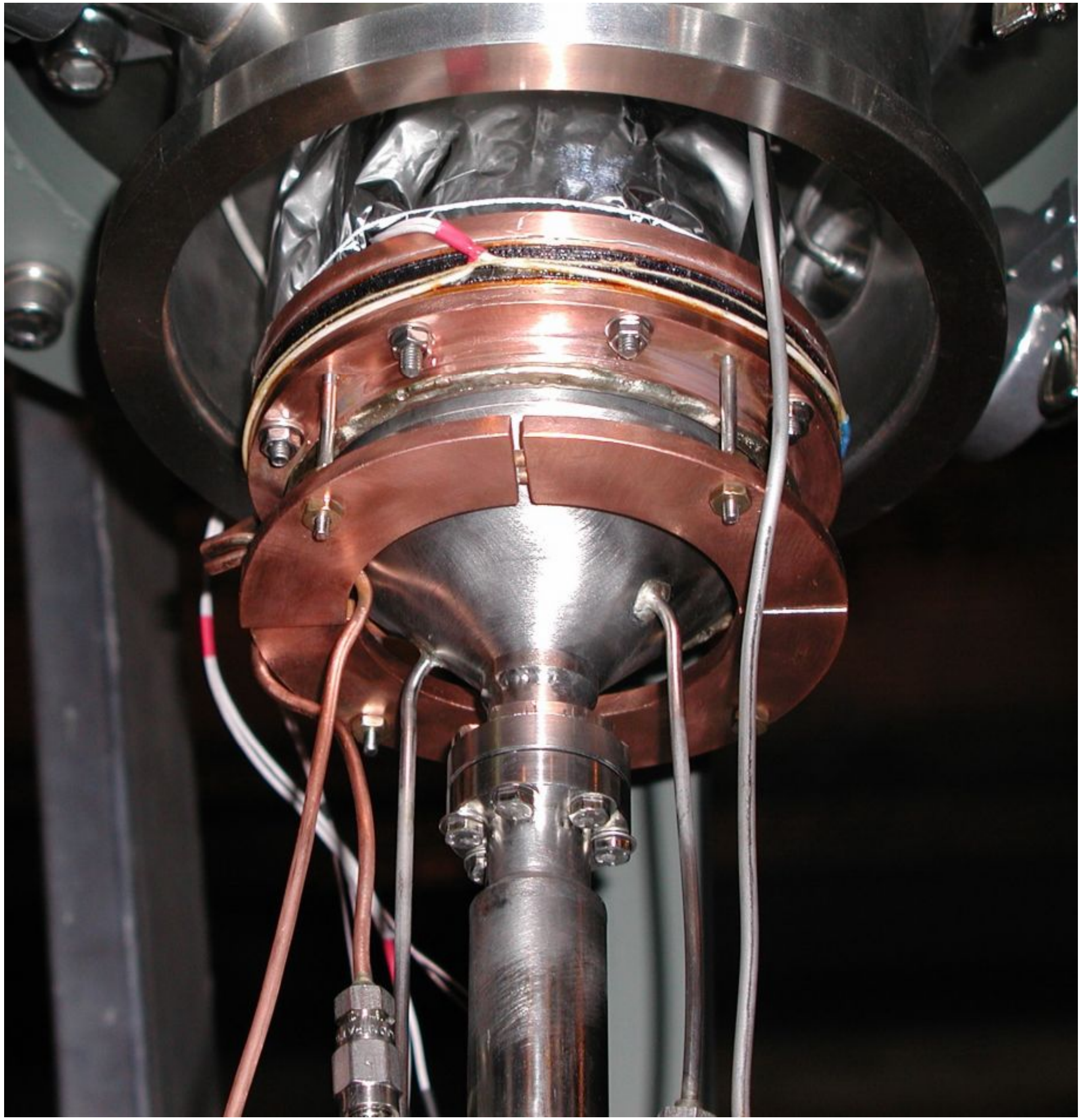


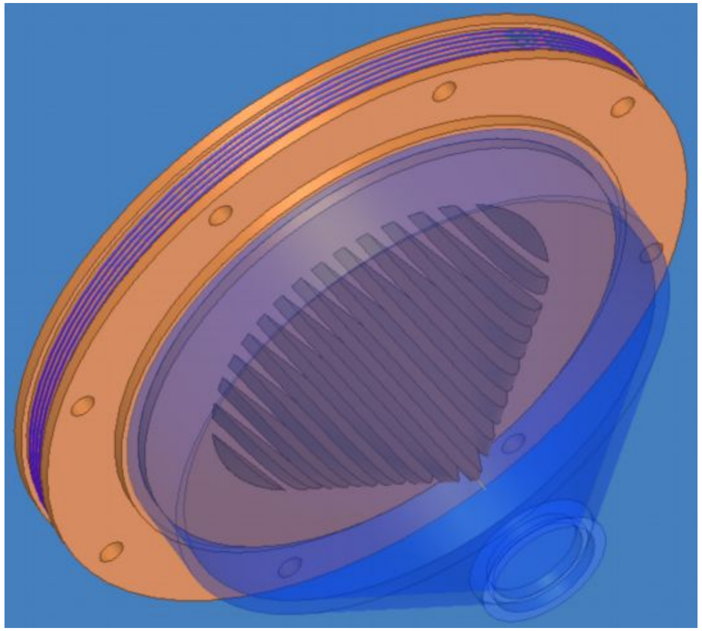
General layout of the cryogenic column

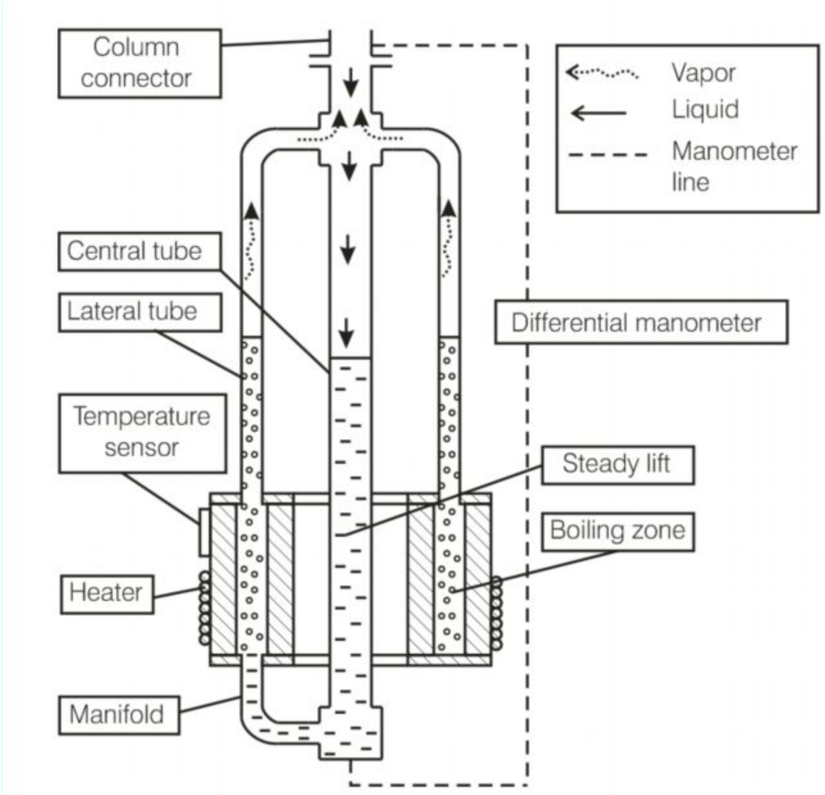


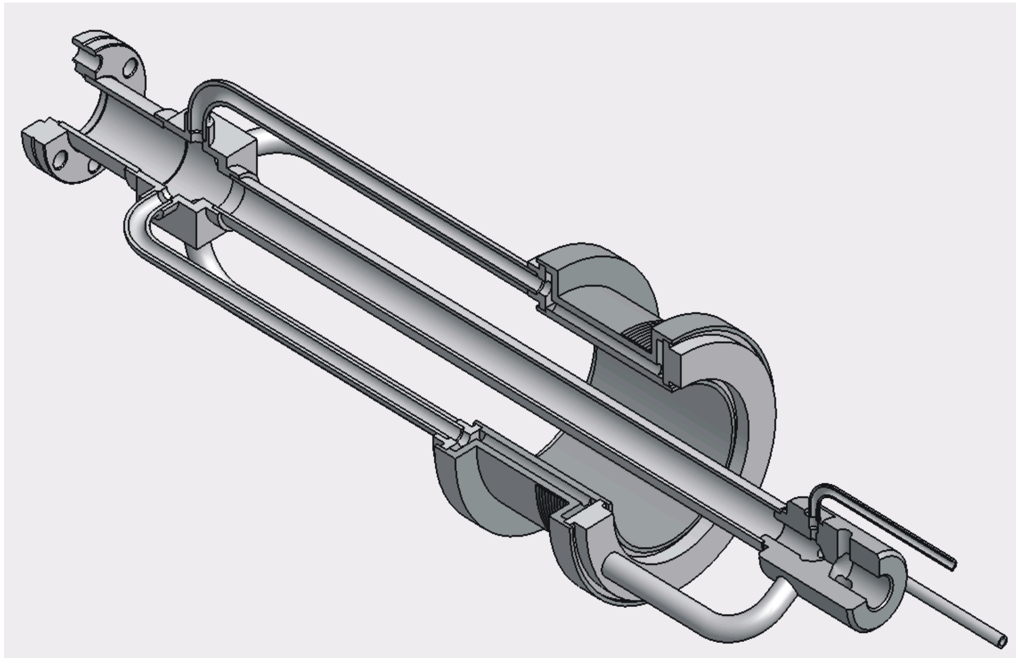


Condenser

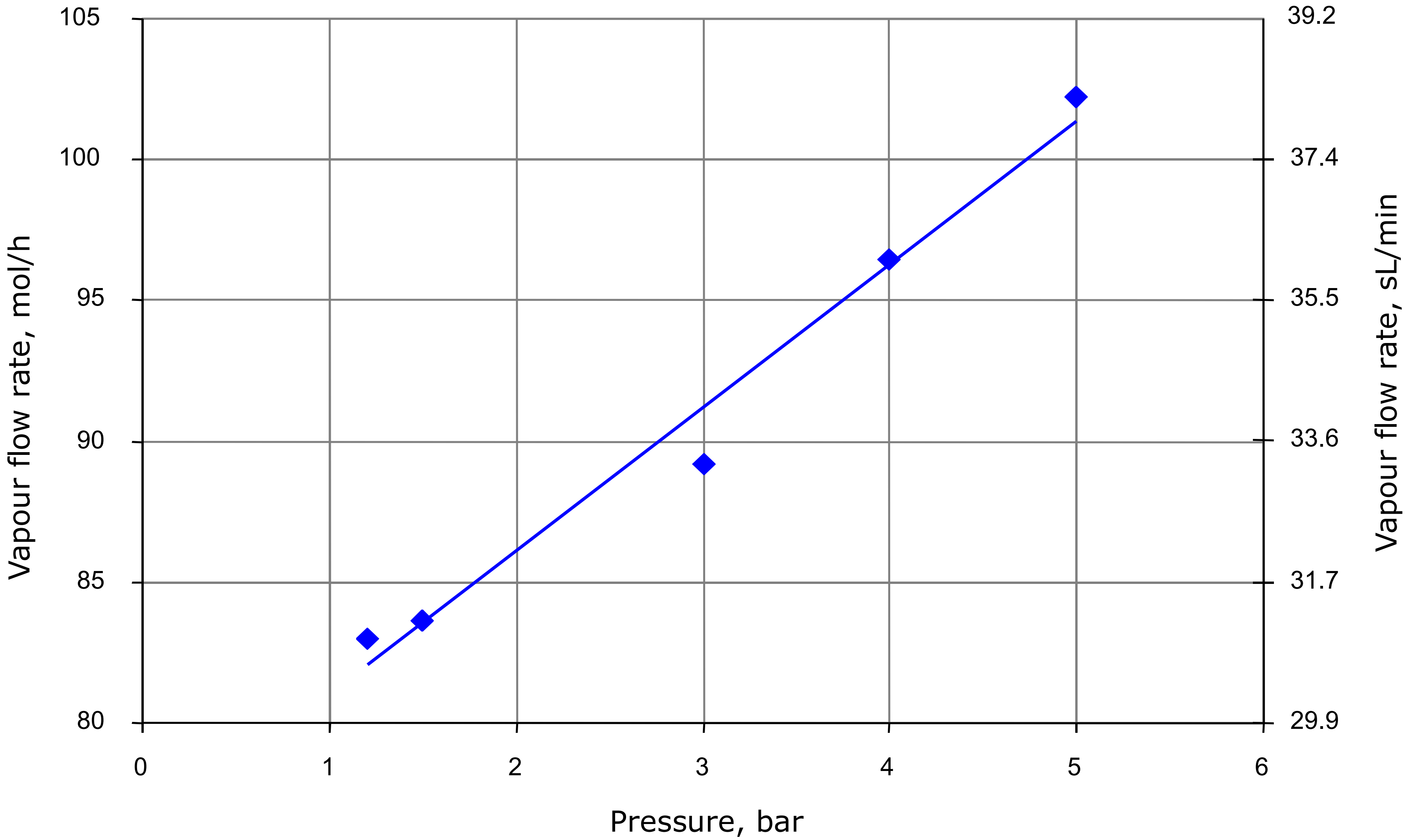


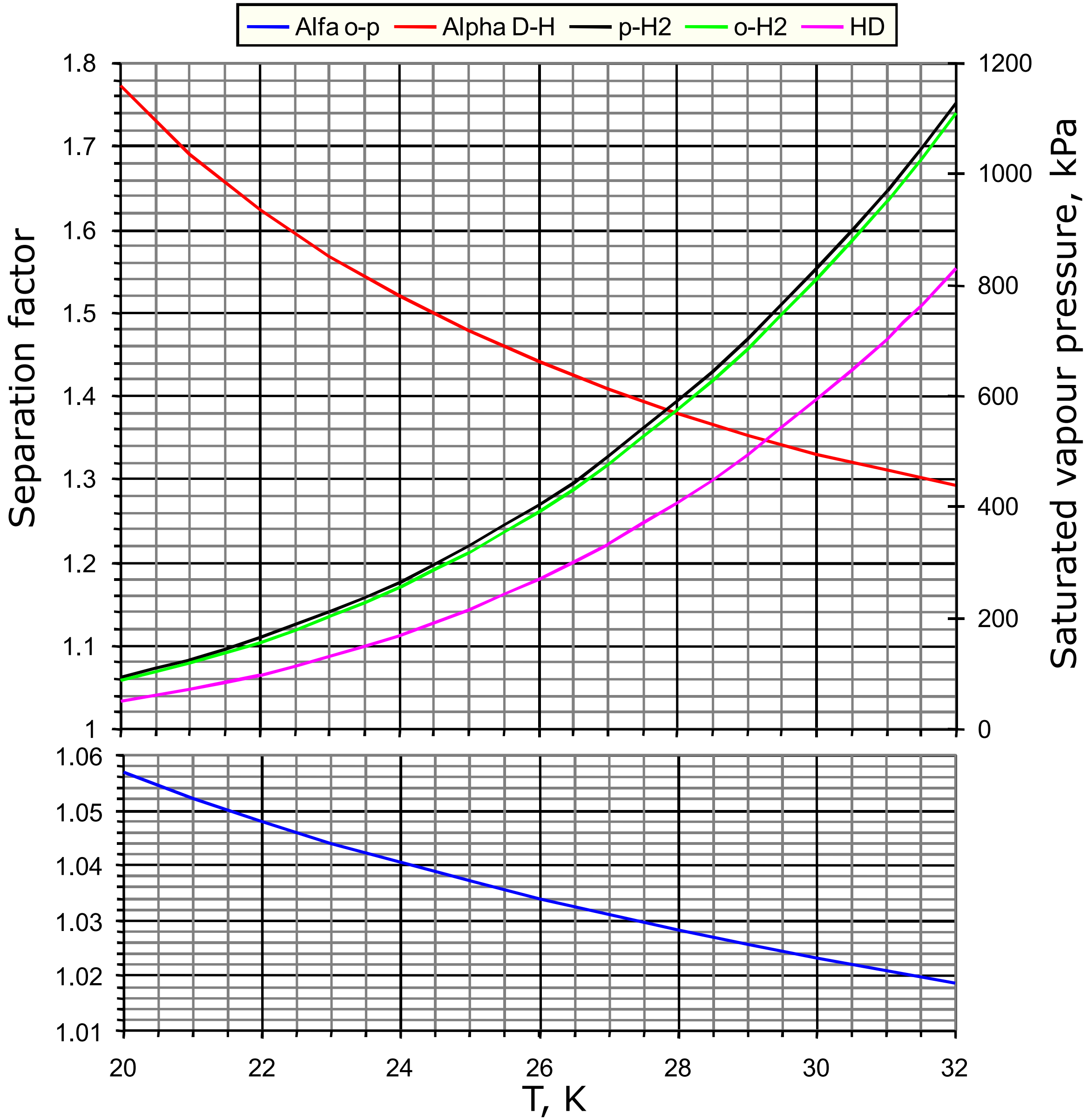


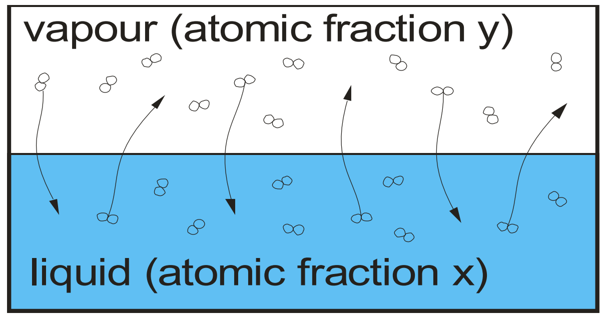

Column connector
Vapor
Liquid
Manometer line
Central tube
Lateral tube
Differential manometer
Temperature sensor
Steady lift
Boiling zone
Heater
Manifold



Vapour flow in the column at reboiler power ~20W







Temperature = const
At dynamic equilibrium the content of low-boiling component is α-times higher in liquid.

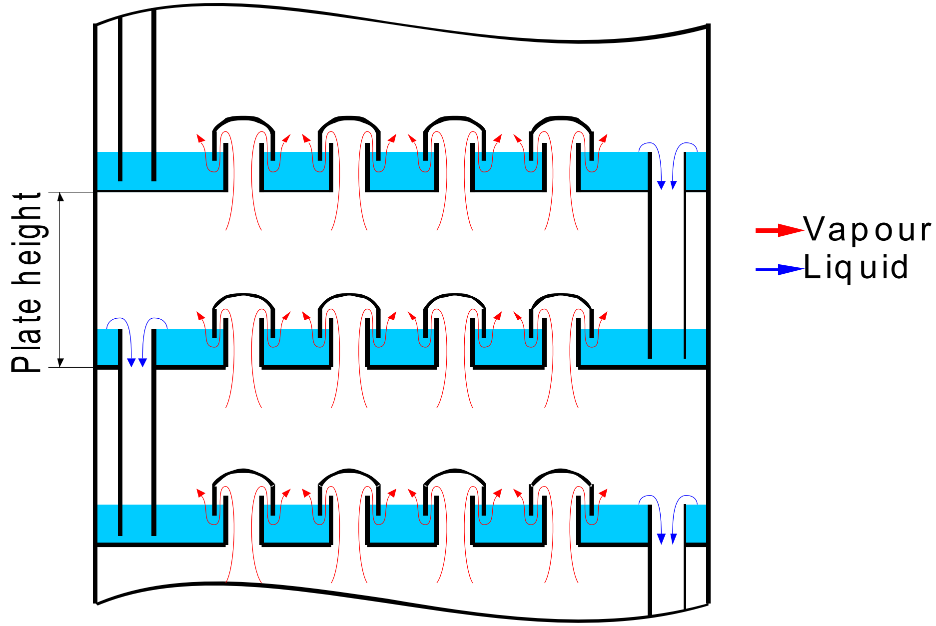
Separation factors:

$$\alpha_{o-p} = \frac{P_{Sat.Para}}{P_{Sat.Ortho}}$$

$$\alpha_{D-H} = \frac{P_{Sat.H_2}}{P_{Sat.HD}}$$

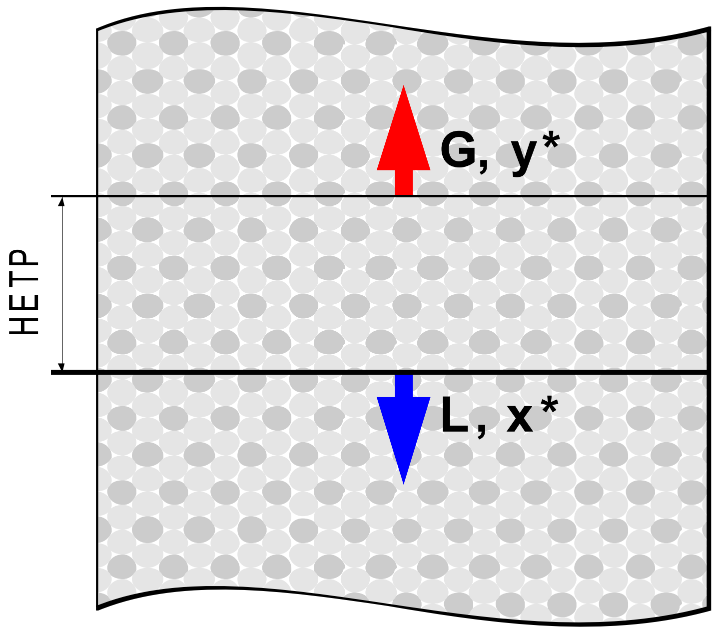
Height equivalent to a theoretical plate (HETP)

Bubble cap plate



Typical plate height ~15 cm
Efficiency = 70%

Non-regular packing



Height Equivalent to a Theoretical Plate (HETP)

Theoretical plate – a part of the column, where composition of outgoing liquid and vapour flows are in equilibrium.
(Abstract equivalent of the bubble cap plate with efficiency = 100%)

Atomic fractions:
x - in liquid
y - in vapour

Separation factor:

$$\alpha = \frac{x^*/(1-x^*)}{y^*/(1-y^*)}$$

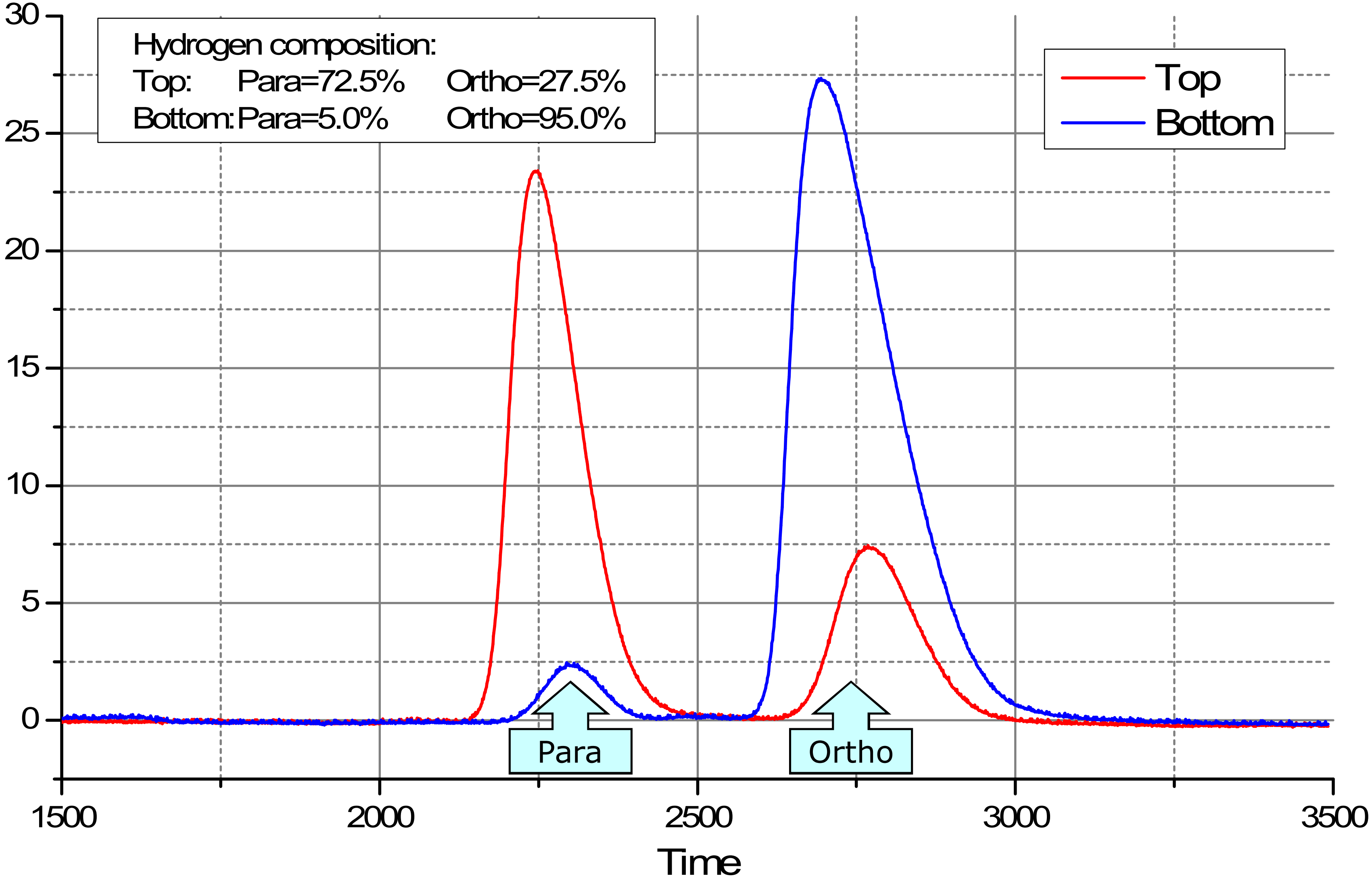
* indicates equilibrium composition

Characteristics:
- Type: spiral prismatic
- Size: 2x2x0.2 mm
- Free volume fraction: 0.82
- Specific surface: 3490 m^2/m^3
- Packed density: 1430 kg/m^3
- Material: stainless steel

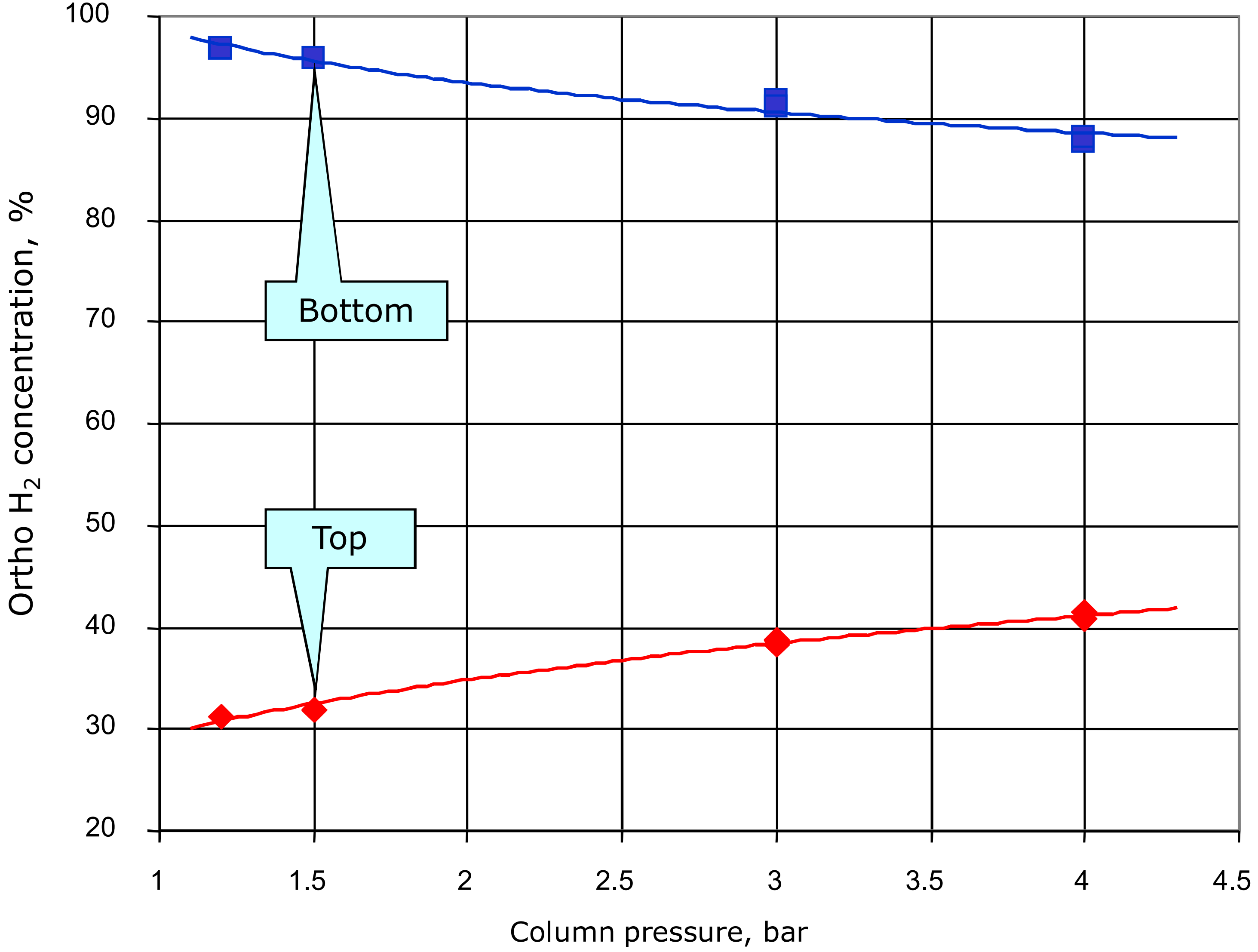
Total volume of packing in the column = 560 ml
Total packing surface in the column = 1.95 m^2

Ortho-Para Hydrogen Chromatogram

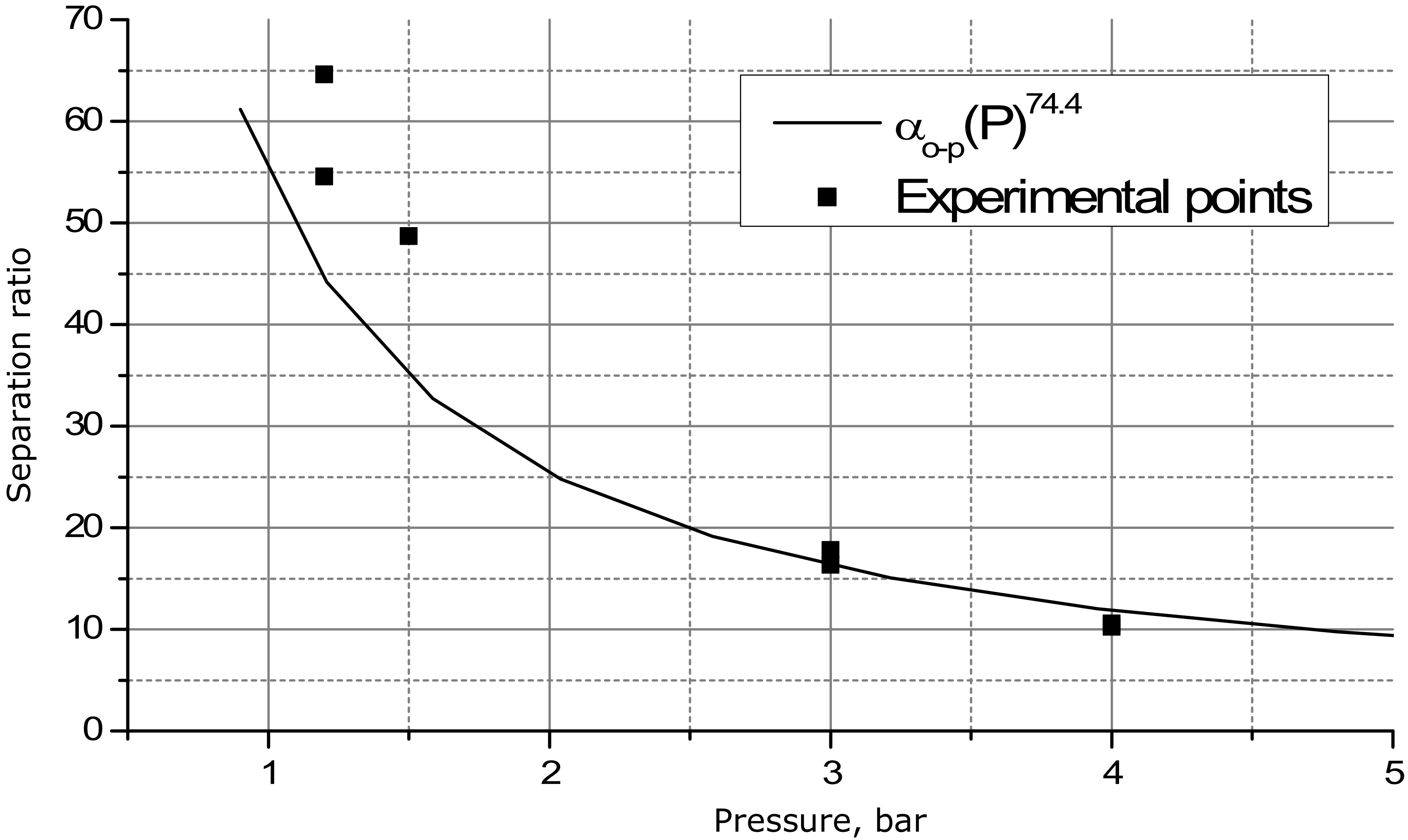
(Natural hydrogen, Column pressure = 1.2 bar; Reboiler power = 10W)



Ortho hydrogen concentration



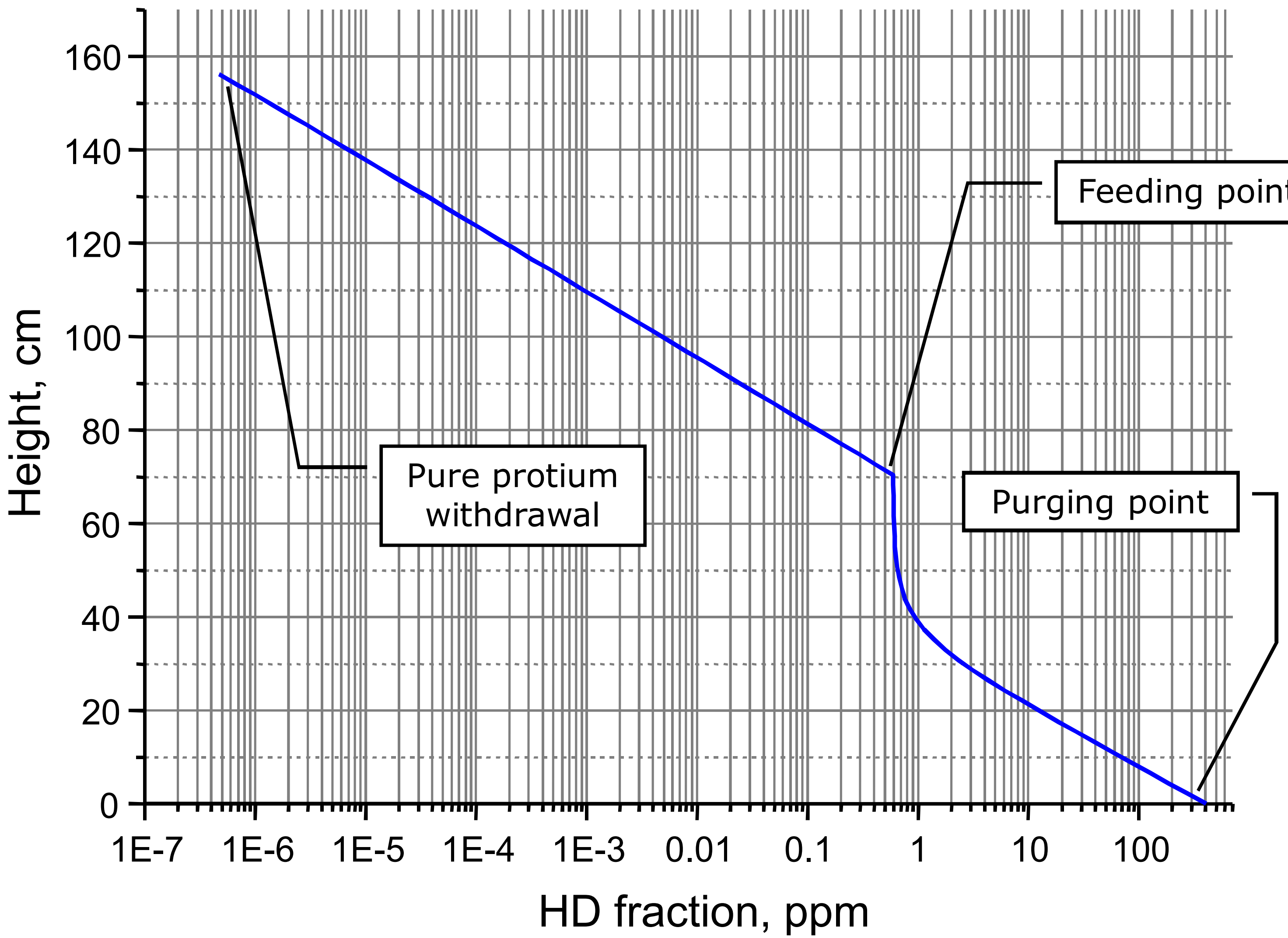
Separation ratio (SR)



$SR = \alpha^N$ - Fenske equation for total reflux mode
(N – the number of theoretical plates)
HETP=Packing height / N

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

Expected HD concentration profile



Feed flow rate = 1 L/min; Pressure = 1.5 bar; Purging flow rate = 0.015 L/min; HETP = 2.2 cm
Initial HD concentration = 6 ppm (deuterium atomic fraction = 3 ppm)

Deuterium purification results

The final measurements of the probes were performed on the new 200 kV Tandem accelerator built for isotope analysis in Zurich.
A special ion source was constructed giving extremely low backgrounds of hydrogen ions from walls, etc. The walls are continuously sputtered to keep the background low allowing measurements during 2 hours.
The existence of the zero samples from the DRU system turned out to be crucial, since the accelerator gives a different background if the ion source is not fed with hydrogen gas.
First zero sample measurement gave zero deuterium concentration at ***70 ppb*** sensitivity.
Thus, it contains less than ***70 ppb*** of deuterium.

Conclusions

- Separation column performance:
  - Height Equivalent to a Theoretical Plate (HETP) value of ***2.2 cm*** corresponds to the best medium power cryogenic column results. HETP value is almost constant in wide range of vapour flow rate.
  - Output deuterium concentration better than ***70 ppb*** does not depend on the initial concentration (natural hydrogen can be used!).
- It is possible to produce pure ortho- or para- hydrogen (99%) in the continuous mode.