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 $\mu p$ atoms to heavier elements with high rates. Once a muon has transferred to a $\mu N_z$ atom, nuclear muon capture proceeds more than 100 times faster than on a $\mu p$ atom.

  Thus, even tiny amounts of impurities in the hydrogen gas distort the observed $\mu p$ 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 $\mu d + p$ scattering cross-section, $\mu d$ 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).
Circulation Hydrogen Ultrahigh Purification System - CHUPS

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/m³
- 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

- $P_{BVI}$
- $P_{AVI}$
- $P_{TPC}$

- Pushing to reserve volume
- Both check valves are closed
- Pumping from TPC

- $T_{min}$
- $T_{max}$
CHUPS, Hydrogen flux 3 l/min

Temperature, K

Time

Temperature, K

Time

Gaussian fit:
mean pressure = 10.00020 ± 0.000009
standard deviation = 0.00036 ± 0.00002

Gaussian fit:
mean flow = 2.97588 ± 0.00062
standard deviation = 0.0746 ± 0.0016

Column 1
Column 2
Column 3

T_{\text{max}}

T_{\text{min}}

Run 2006
H_{2} flow: 3slpm

Run 2006

TPC Inlet hydrogen flow (slpm)
Nitrogen purification

The capture process $\mu N_Z \rightarrow N_{Z-1} + \nu$ is identified by its distinct signature in the TPC.
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)
A. Vasilyev 6/21/2007

Moisture measurements in CHUPS with TPC

<table>
<thead>
<tr>
<th>Time</th>
<th>Humidity, ppm</th>
<th>Temperature, K</th>
</tr>
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<tr>
<td>11/21/2006</td>
<td>0.002</td>
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<td>11/28/2006</td>
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</tbody>
</table>

H2O_sensor, ppm
TT5, K (H2O sensor)
TT6, K (TPC)

TPC connection
CHUPS results

N₂  less than  5-7 ppb
O₂  less than  5 ppb
H₂O about       20 ppb
General layout of the cryogenic column
Condenser
Reboiler
Vapour flow in the column at reboiler power ~20W

Pressure, bar

Vapour flow rate, mol/h

Vapour flow rate, sL/min

Pressure, bar

Vapour flow rate, mol/h
Saturated vapour pressures and separation factors

At dynamic equilibrium the content of low-boiling component is \( \alpha \)-times higher in liquid.

Separation factors:

\[
\alpha_{o-p} = \frac{P_{\text{Sat.Par}}} {P_{\text{Sat.Ortho}}}
\]

\[
\alpha_{D-H} = \frac{P_{\text{Sat.H}_2}} {P_{\text{Sat.HD}}}
\]
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:**

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

*indicates equilibrium composition
Packing

Characteristics:
- Type: spiral prismatic
- Size: 2x2x0.2 mm
- Free volume fraction: 0.82
- Specific surface: 3490 m²/m³
- Packed density: 1430 kg/m³
- Material: stainless steel

Total volume of packing in the column = 560 ml
Total packing surface in the column = 1.95 m²
Ortho-Para Hydrogen Chromatogram

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

Hydrogen composition:
Top: Para=72.5%  Ortho=27.5%
Bottom: Para=5.0%  Ortho=95.0%
Ortho hydrogen concentration

![Graph showing the ortho hydrogen concentration against column pressure. The graph has two lines, one for the top and one for the bottom, with the top line showing a slight increase and the bottom line showing a slight decrease as the column pressure increases.]
Separation ratio (SR)

\[ SR = \alpha^N \] - Fenske equation for total reflux mode

\[ \text{N (N - the number of theoretical plates)} \]

\[ \text{HETP= Packing height / N} \]

\[ SR = \frac{X_{\text{Bottom}}}{X_{\text{Top}}} \frac{1}{(1 - X_{\text{Bottom}})} \frac{1}{(1 - X_{\text{Top}})} \]
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.