CRYOGENIC SYSTEM FOR CONTINUOUS ULTRAHIGH HYDROGEN PURIFICATION IN CIRCULATION MODE

A. Vasilyev[1], B. Bezymyannykh[1], V. Ganzha[1], P.Kravtsov[1], P. Kammel[2], G. Shapkin[1], V. Trofimov[1], M.Vznuzdaev[1] and M.Poltavtsev[1]

1. Introduction

Hydrogen of high purity (0.1ppm impurities level and better) is used in various branches of science and technology. An instance of high purity hydrogen use is the MuCAP experiment [1]. The goal of this experiment is the precision measurements of muon capture (μ-capture) on a proton. This is achieved by comparing the lifetimes of positive and negative muons in hydrogen to determine λ – the rate of μ-capture, with 1% precision. The experiment requires pure protium (hydrogen with deuterium concentration of less than 1ppm) for the active target which is a Time Projection Chamber (TPC) operated with 10 bar of ultra-pure protium.

Extremely high purity of the gas in the detector is one of the main requirements of the experiment. To carry out the measurements at the specified precision, the total level of all contaminants (water, nitrogen, oxygen etc.) has to be lower than 0.01 ppm.

Hydrogen preparation by commercial purification units, such as palladium filters, could give a good initial level of purity. However, during the experiment, various impurities build up in the chamber volume from outgasing from the walls and electronic components and by diffusive leaks. The accumulation rate of contaminants does not allow to keep the required purity during the whole experimental run (up to 2 month). Thus, a continuous purification of the protium in the TPC is necessary. Very high price of the protium defines the closed, circulation type of the cleaning system. Contaminated gas from the TPC has to be cleaned in purification block and returned to the detector. This scheme can support high purity over long periods. However, additional difficulties, such as pressure stabilization inside the TPC, could be caused by the continuous gas flow. During the operation, maximum instability of the TPC internal pressure is 0.5 %.

CHUPS (Circulation Hydrogen Ultrahigh Purification System) is designed to solve these two tasks: providing of predefined hydrogen purity and stable pressure in the TPC detector volume.

2. Principles of operation

A schematic diagram of gas lines and control devices of CHUPS is shown in the Figure 1.

[1] Saint-Petersburg Nuclear Physics Institute, PNPI, 188300, Orlova Rosha, Gatchina, Russia; e-mail: vassilie@pnpi.spb.ru; http://lkst.pnpi.nw.ru.
[2] University of Illinois at Urbana-Champaign, UIIC, IL 61801, USA; e-mail: kammel@npl.uiuc.edu

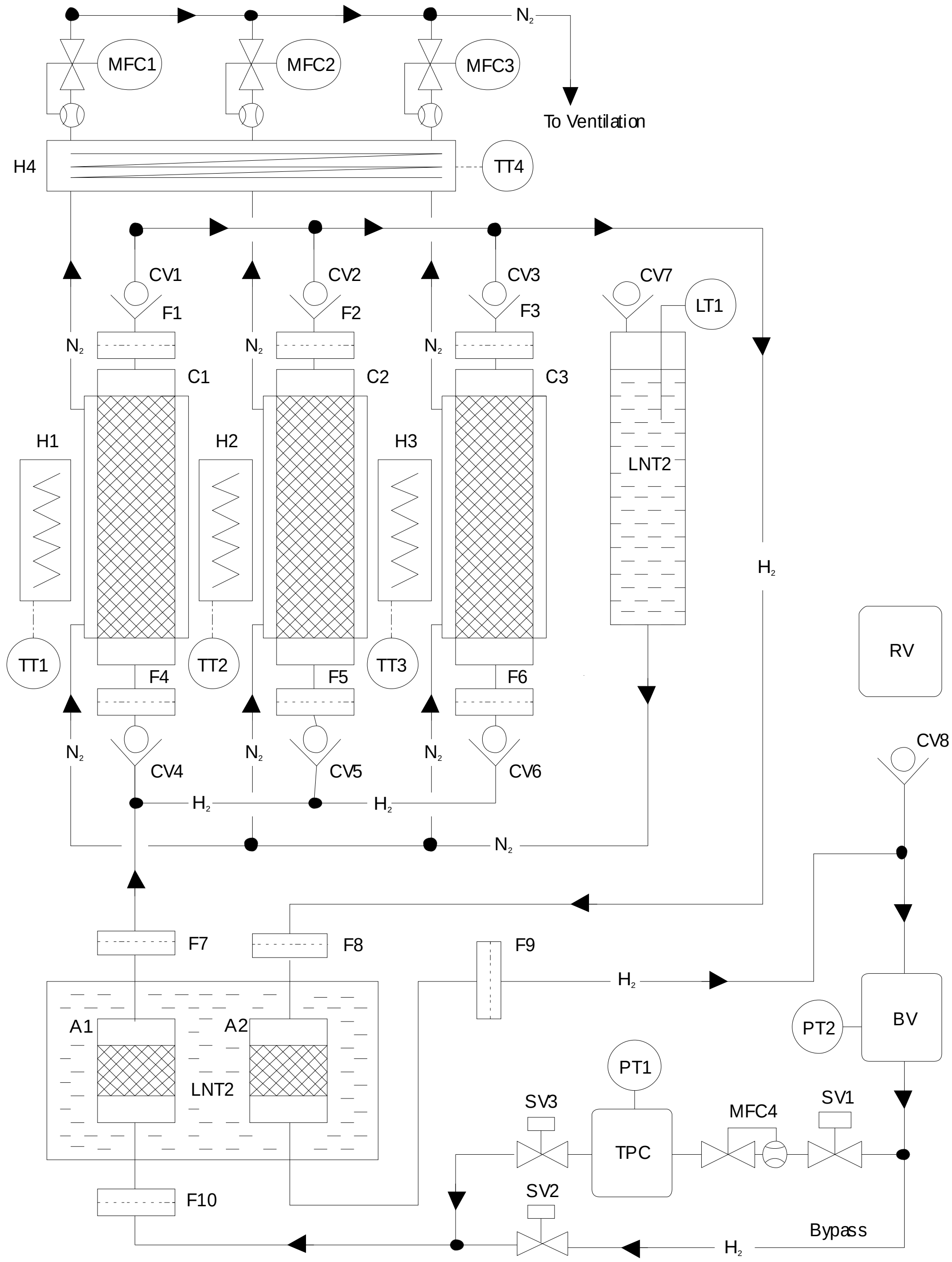

Figure 1. CHUPS schematic diagram.

The system consists of three base units: Compressor, Purifier and automatic Control System. The Compressor provides a constant flow of hydrogen through the Purifier with a sufficiently high rate to support the specified purity of the gas in the TPC.

The Compressor is the main part of the system. By principle of the operation, the Compressor is a cryopump. Its operation is based on the ability of special substance (adsorbent) to absorb considerable amount of gas and extract it by subsequent heating. In this case, activated carbon is used as the adsorbent. Cooling of the adsorbent is provided by liquid nitrogen, heating – by electrical heaters.

Activated carbon Norit RB is cased in cylindrical cartridges – columns. The compressor has three such cartridges (on the scheme – C1-3). Every column is equipped by a spiral copper pipe – heat exchanger for liquid nitrogen. Heat exchanger is fixed on the compressor by a solid solder. Coils of the electrical heaters (H1-3) are wound between the heat exchanger's turns. Temperature sensors (PT-100 type) are fixed directly on the heat exchangers surface.

Simplified P-T diagram (Figure 2) shows the principle of the column operation.

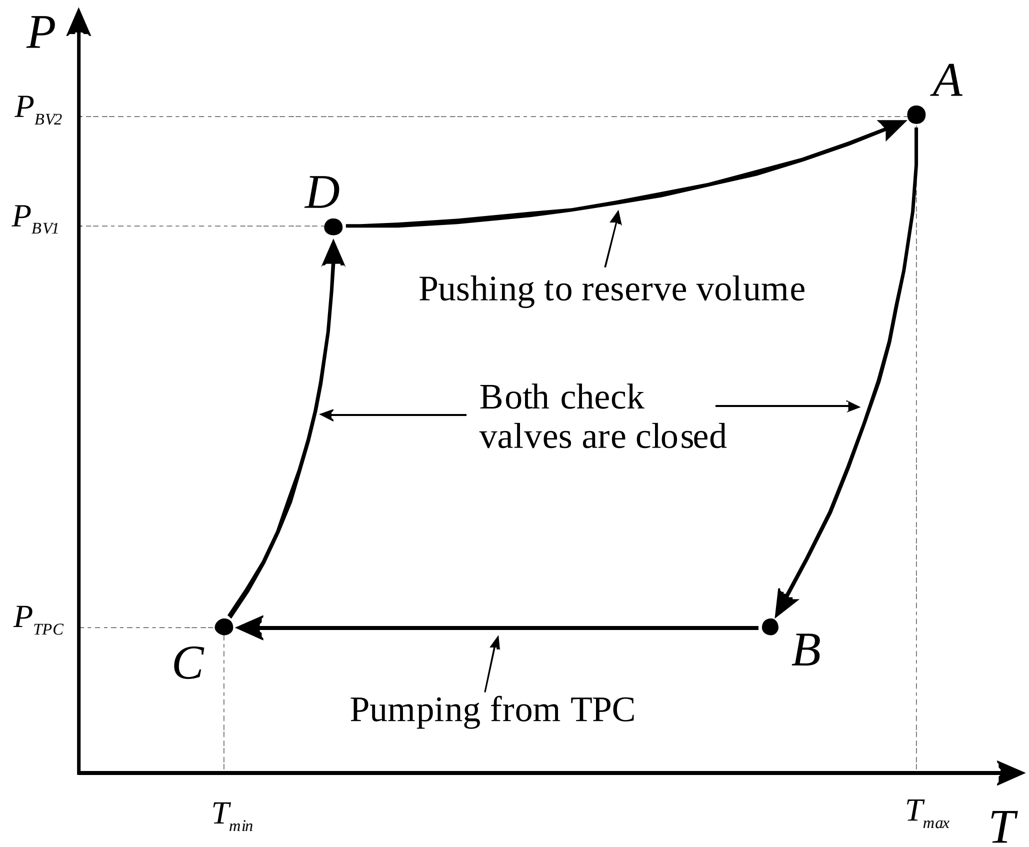

Figure 2. Compressor column P-T diagram.

A working cycle of every column consists of a stage of cooling and stage of heating. At the stage of cooling, liquid nitrogen from the tank LNT1 proceeds into the heat exchanger. Overpressure inside the tank is made by native vaporization of liquid nitrogen. The level of this pressure during the operation does not exceed 0.3 bar. It is limited by the check-valve CV7 that release the excess of nitrogen into the atmosphere.

Nitrogen boils in the heat exchanger and cools down the adsorbent in the column. The most part of liquid nitrogen evaporates in the heat exchanger. The gaseous nitrogen flow is controlled by the mass-flow controllers MFC1-3. Each compressor has its own mass-flow controller; thereby it is possible to regulate the cooling process of each column separately.

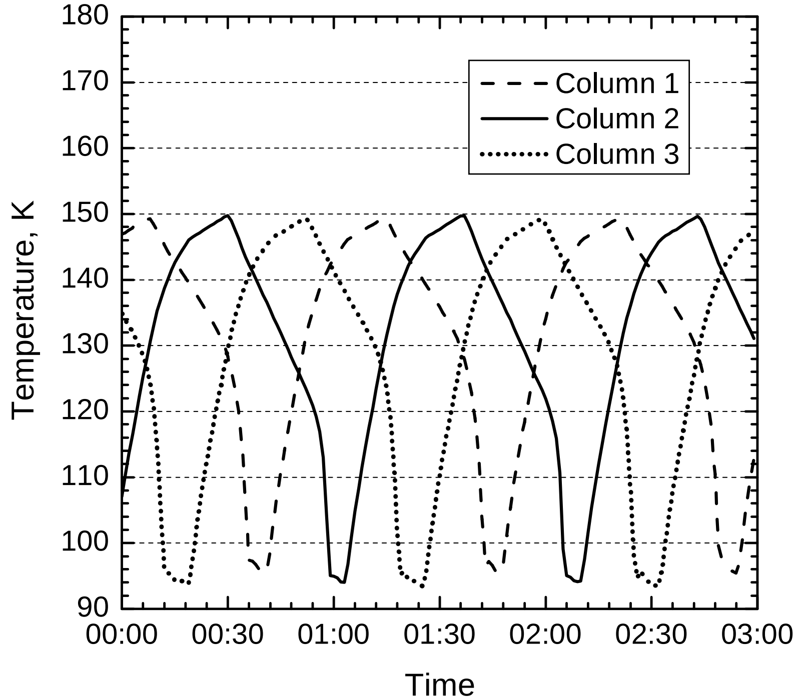

Figure 3. Temperature cycles of compressor columns at hydrogen flow of 1.6 slpm.

To provide normal operating conditions of the mass-flow controllers MFC1–3 (Aalborg CFG37), heater (thermalizer) H4 is used. In the thermalizer, gaseous nitrogen from the heat exchangers is finally warmed up to room temperature. Thermalizer design is very similar to design of the heat exchangers of compressor columns. It has electric heater coiled between copper rounds and fixed by solid solder. Gaseous nitrogen produced in compressor is released to atmosphere.

Adsorption capacity of the activated carbon increases during the cooling. Starting from the high temperature T_{max} (Figure 2), column adsorbs hydrogen during cooling at A–B phase. Simultaneously column pressure decreases down to TPC internal pressure. By reaching of the pressure difference 0.1 bar and more, intake check-valve (CV 4–6) opens, and hydrogen from the TPC proceeds into the column. Then, temperature of the adsorbent goes down to T_{min} during the B–C phase. Column pressure is stabilized at the TPC internal pressure level by compensation of the TPC outlet hydrogen adsorbed by the compressor column using clean hydrogen from the buffer volume. Thus the pressure is constant during B–C phase. When T_{min} is achieved, corresponded mass-flow controller closes nitrogen flux. The column turns into heating stage

During the heating stage activated carbon inside the compressor is warmed up to the T_{max} temperature. This leads to intensive desorption of the hydrogen from the adsorbent. This stage begins from C–D phase. Pressure increases quickly in the small column volume up to buffer volume pressure level.

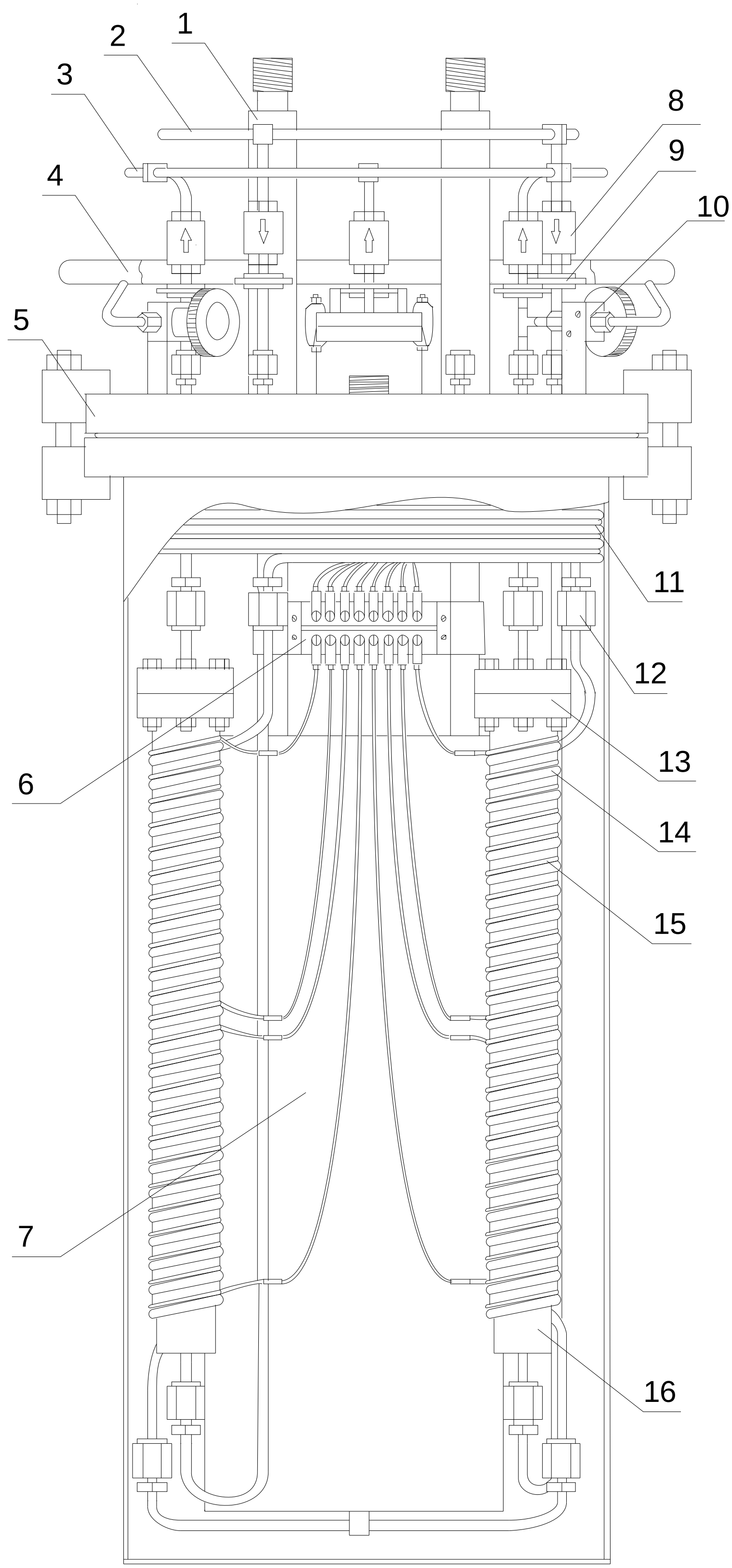

Figure 4. Scheme of the Compressor.

Sufficient pressure difference on the corresponding outlet check-valve (CV1-3) opens it and hydrogen proceeds into the buffer volume. Then, after opening of the outlet check valve, further heating of the compressor column increases the buffer volume pressure slowly at D–A stage.

Each compressor operates in a periodic mode and produces pulsing hydrogen flux. The Compressor block has three columns. Their temperature stages are shifted (Figure 3) to make the hydrogen flux smooth.

Vacuum case (Figure 4) containing the compressors (pos. 16) and liquid nitrogen tank (pos. 7) provides temperature insulation. It is a cylindrical vessel with a removable upper flange (ISO K250, pos.5). Three rails are installed on the upper flange. The vacuum rail (pos. 4) is connected to the columns through exhaust valves (pos. 10). During the preparation of the system, the vacuum rail is connected to the vacuum pump unit and the columns are evacuated through the valves. The inlet (pos. 2) and outlet (pos. 3) rails combine inlet and outlet lines of the columns through the check-valves (pos. 8).

Mechanical filters (pos. 9) are intended to protect gas fittings of the system from adsorbent's dust. They are produced from porous stainless steel.

As described above, each column is equipped by a spiral heat-exchanger (pos. 14) and electric heater (pos. 15). All columns have detachable caps (pos. 13). Hydrogen and nitrogen lines of the columns are connected to the system lines by VCR connectors (pos. 12).

Thermalizer (pos. 11) is fixed on the bottom side of the upper flange. To improve thermo-insulation, inner parts of the compressor are wrapped by 200 μm aluminum-backed Mylar.

A buffer volume (BV in Figure 1) is included into the system to smooth variation of the hydrogen flow. Reserve of hydrogen accumulated in buffer volume is necessary to support pressure stabilization in the TPC. The second function of the volume is to keep entire amount of hydrogen (from the CHUPS and the TPC) between MuCAP runs and in emergency cases. Therefore it is designed to a high (100 bar) pressure. The buffer volume consists of three thick-walled vessels combined into one block (Figure 5). Total volume of the buffer volume is 15 l. It is cased into a thermo-insulation shell with vacuum-processed walls. This enables cooling the buffer volume by liquid nitrogen to gather a maximal amount of hydrogen.

Principle of Purifier operation is based on prevalent (in comparison with the main component – hydrogen) adsorption of contaminants (nitrogen, oxygen, water etc.) in the adsorption filter. Synthetic zeolite is used as the adsorbent for this goal. To increase a rate of purification, filter is frozen by liquid nitrogen (the same as for the Compressor). Adsorption method of purification guarantees a high level of purification at very wide range of species. The Purifier has two adsorption filters: A1 and A2 (Figure 1). They are installed in a reservoir with liquid nitrogen LNT2. During the circulation, contaminated gas is being purified twice. First, it passes through preliminary cleaning in A1 before compressor. On the next stage, final cleaning in A2 occurs. Filter A2 is installed on the outlet line of the Compressor.

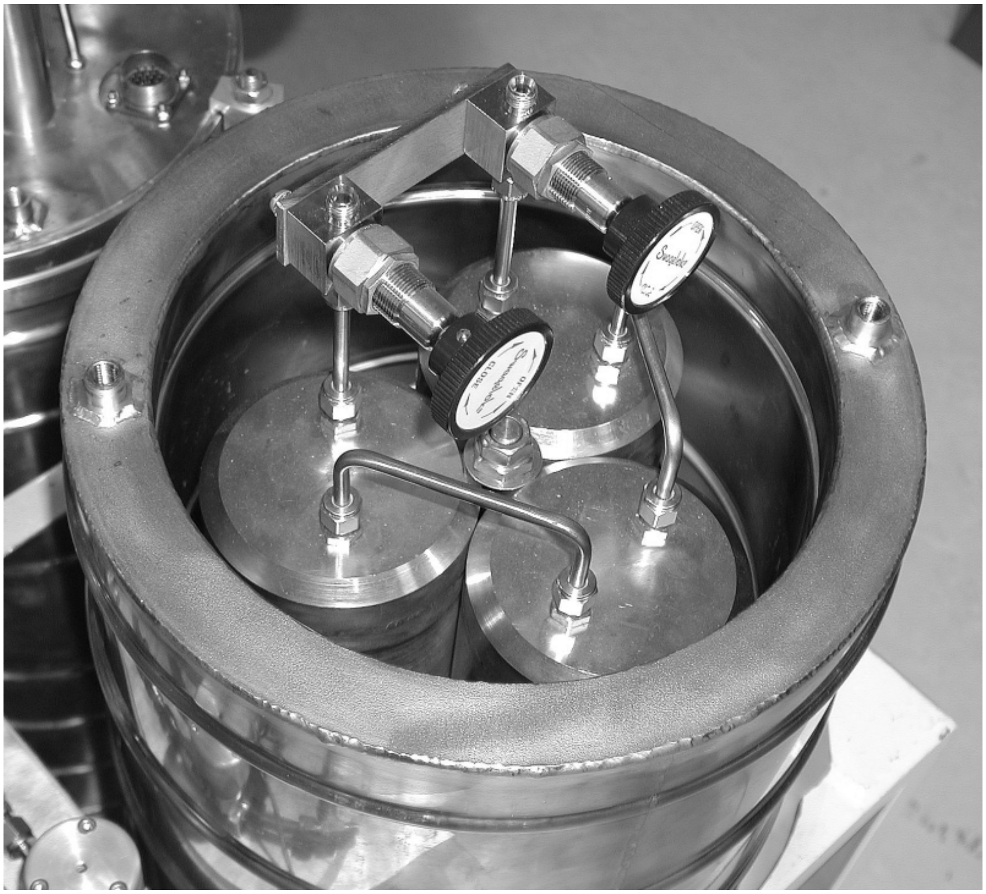

Figure 5. Buffer volume in the thermo-insulating shell.

3. Data acquisition and control

CHUPS is controlled by a specially developed microcontroller board. All electronics and power supplies for the mass-flow controllers (MFC) and electro-pneumatic valves are installed in the usual ATX PC case. Controller board is based on a Atmel AT89S8252 processor working at 22.1184 MHz frequency. Sensor signals come through two 16-channel Burr-Brown multiplexers (MPC506) to the 16-bit ADC (ADS7813). One multiplexer is used for temperature measurements, second – for all other sensors. Communication between control computer and controller is done via standard RS-232 (or RS-485) serial channel (Figure 6). Mass-flow controllers setpoints are generated by 16-bit DAC (DAC7634). Control board input-output capabilities are:

- 16 temperature sensors (PT-100);
- 5 pressure sensors (with 4-20mA current output);
- 10 other sensors (MFC, levelmeters etc.);
- 4 digital outputs for heaters;
- 12 digital outputs for valves and other devices;
- 4 analog outputs for hydrogen MFCs (16-bit, 0-5V)
- 4 analog outputs for nitrogen MFCs (12-bit, 0-5V)

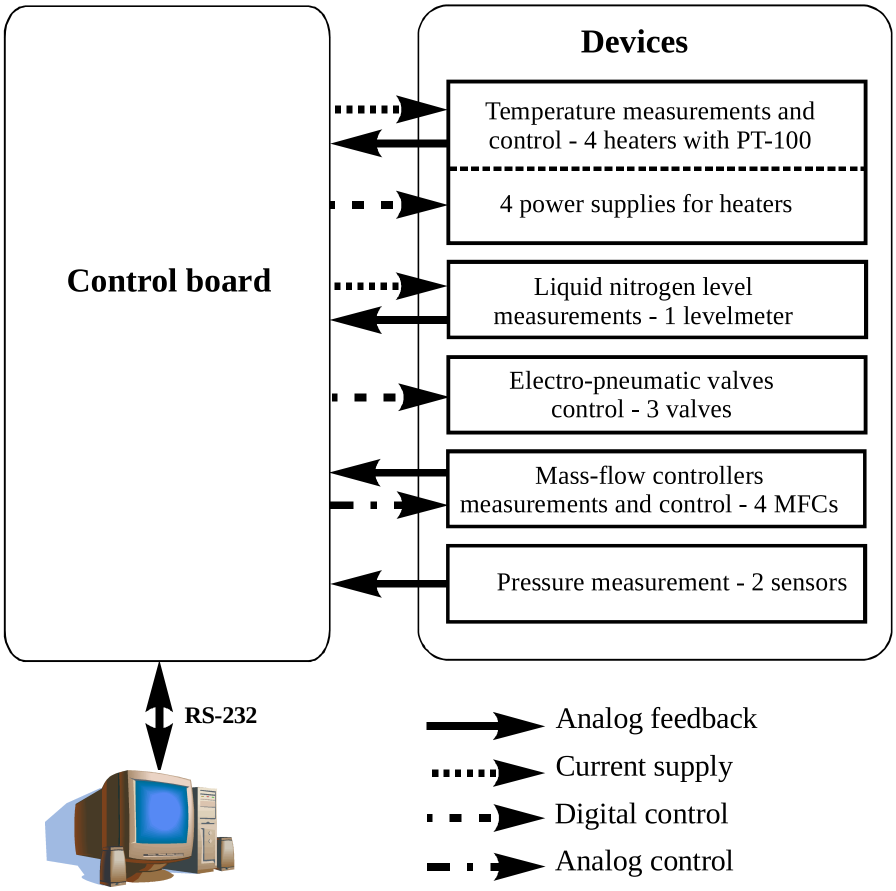

Figure 6. DAQ and control diagram.

Software for the microcontroller is written in C language and provides most of the control and monitoring tasks of the system. The program reads all sensors, control mass-flow controllers and valves, and handles alarm actions. A number of stabilization loops are implemented in the software.

Temperature stabilization of the compressor columns and thermalizer is done using pulse-width modulation of the electrical heaters (during heating phase) and proportional regulation of the nitrogen flux through the heat exchanger (during cooling phase). Four identical power supplies for the electrical heaters (1000W, 24VAC output) are mounted in two separate cases and controlled using solid state relays. Pulse-width modulation of the control signal makes it possible to vary heating rate of the compressor, and, correspondingly, flux of hydrogen through the system.

Compressor column cycles are implemented using internal timer of the CPU and temperature stabilization. Cycle parameters are cooling time, heating time, high temperature and low temperature. Phase shift for the three columns is calculated automatically to provide smooth compressor operation.

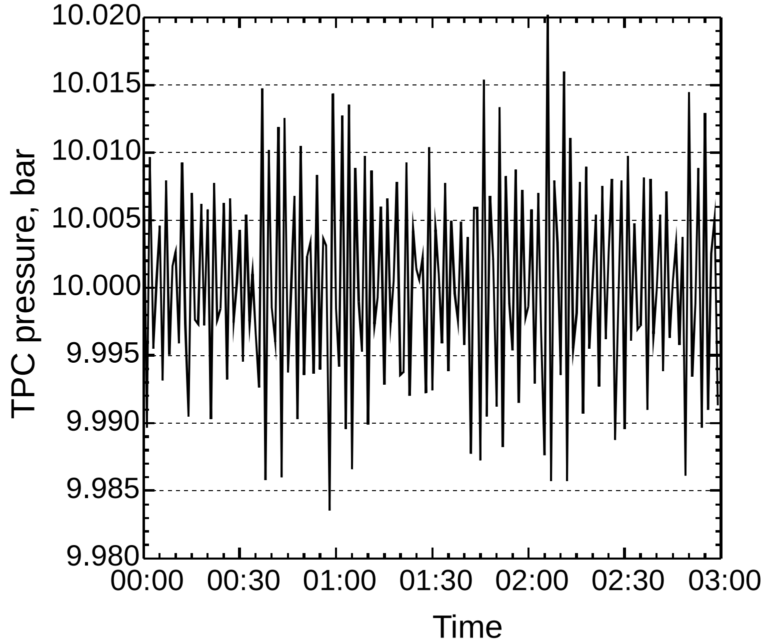

Figure 7. TPC internal pressure behavior.

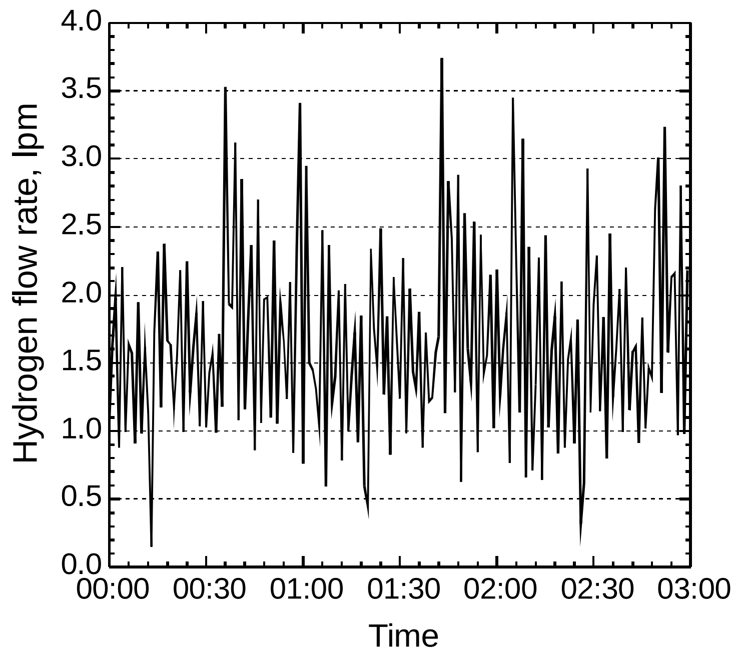

Figure 8. TPC inlet hydrogen flux.

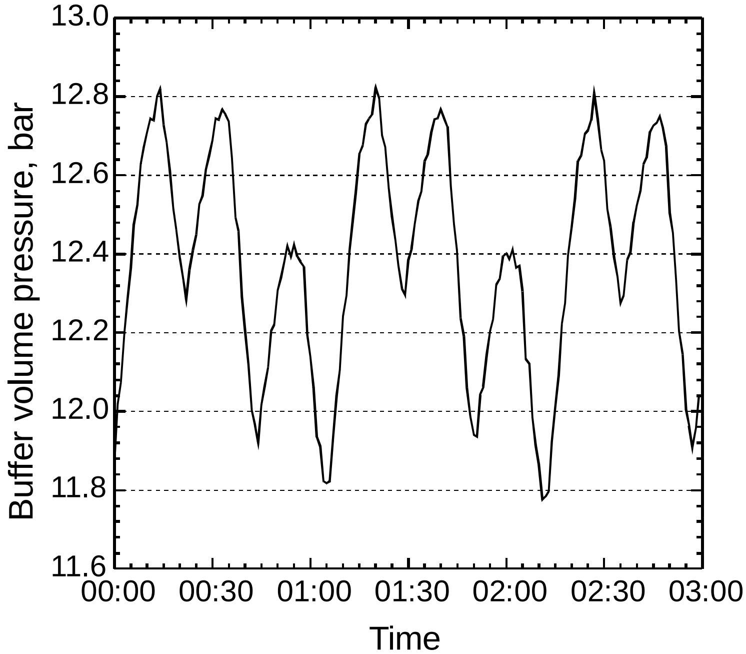

Figure 9. Buffer volume pressure behavior.

Pressure stabilization is done making use of hydrogen mass-flow controller MFC4. Setpoint for this MFC is generated on the base of PT1 sensor reading (TPC pressure) and pressure setpoint using proportional-derivative regulation. Outlet hydrogen flux from the TPC is defined by the compressor operation and varies in a wide range (0÷6 slpm). This flux is compensated by the inlet TPC flux from the buffer volume. Figures 7 and 8 show the TPC pressure behavior and inlet hydrogen flux regulated by MFC4. TPC pressure fluctuations meet the experiment requirements. Buffer volume pressure fluctuations shown in Figure 9 are much higher. The main condition of the stable pressure inside the TPC is permanent difference between buffer volume pressure and TPC not less than 0.5 bar.

Alarms handling

The TPC detector is very sensitive to internal pressure deviation. Chamber operation faults and even mechanical damages can result from a significant pressure variation. Besides, total volume of hydrogen in CHUPS and TPC is about 1000 standard liters, which makes this equipment explosive and dangerous. Control electronics takes some preventive actions for the alarm conditions.

The most crucial alarm conditions are changing of TPC internal pressure and increasing of buffer volume pressure. These alarms cause the control electronics to cut off the TPC and open bypass valve (SV2). This configuration is stable, CHUPS can pump hydrogen through the compressor and purification units without TPC until liquid nitrogen is used up. CHUPS recovery after these alarm conditions can be done only manually by operator. In case of further system

pressure increase (above 16 bar) electro-pneumatic valves can open spontaneously. This can happen for example if liquid nitrogen finishes and all three columns of the compressor heat up to room temperature. To protect TPC volume in such cases an emergency check valve CV8 is installed. It opens at 16 bar and release exceeding hydrogen to a special previously pumped volume.
Additional two alarm conditions are only informative. System will give a warning to operator in case of low level of liquid nitrogen in compressor tank or low air pressure in valve actuators manifold.

PC software

PC software for the CHUPS provides operator interface for tuning the regulation parameters of the control electronics, process variables monitoring and history accumulation. The software is developed using Borland Delphi and works in Windows environment.
Operator interface consists of four windows. A manual control window with simplified diagram of the system contains all actuating devices are displayed in a symbolic form. Device symbols are active, mouse click on any device symbol changes corresponding device state. All system process variables are shown in the diagram. Compressor columns status is also shown in this window. Separate window displays four charts. Three of them correspond to three columns of the compressor. Column temperature, heater power and nitrogen flux are shown for each compressor column on the chart. Fourth chart shows TPC internal pressure, buffer volume pressure and hydrogen flux. Two additional windows represent system messages log and TPC internal pressure histogram chart.
The software reads CHUPS process variables from the control electronics every second and writes them into MS Access database. Data history visualization and analysis is provided by a special program. It allows to display up to 4 system parameters on the chart for the given time period. Database viewer program also shows system messages directly on the chart. Unlimited number of system parameters could be exported to a text file or MS Excel spreadsheet for further analysis.

4. The results

The system was installed and tested during the MuCAP experimental run of 2004 (Figure 10). Total duration of the CHUPS stable operation mode is more than 700 hours. The main criterion of CHUPS efficiency is impurities level behavior in the TPC during the run. The best characteristic of hydrogen purity, from the point of view of MuCAP experiment, is impurities capture yield. This value is calculated during the run by the TPC data. It gives the information about effective concentration of all contaminants. The behavior of this value during the run is shown in Figure 11.

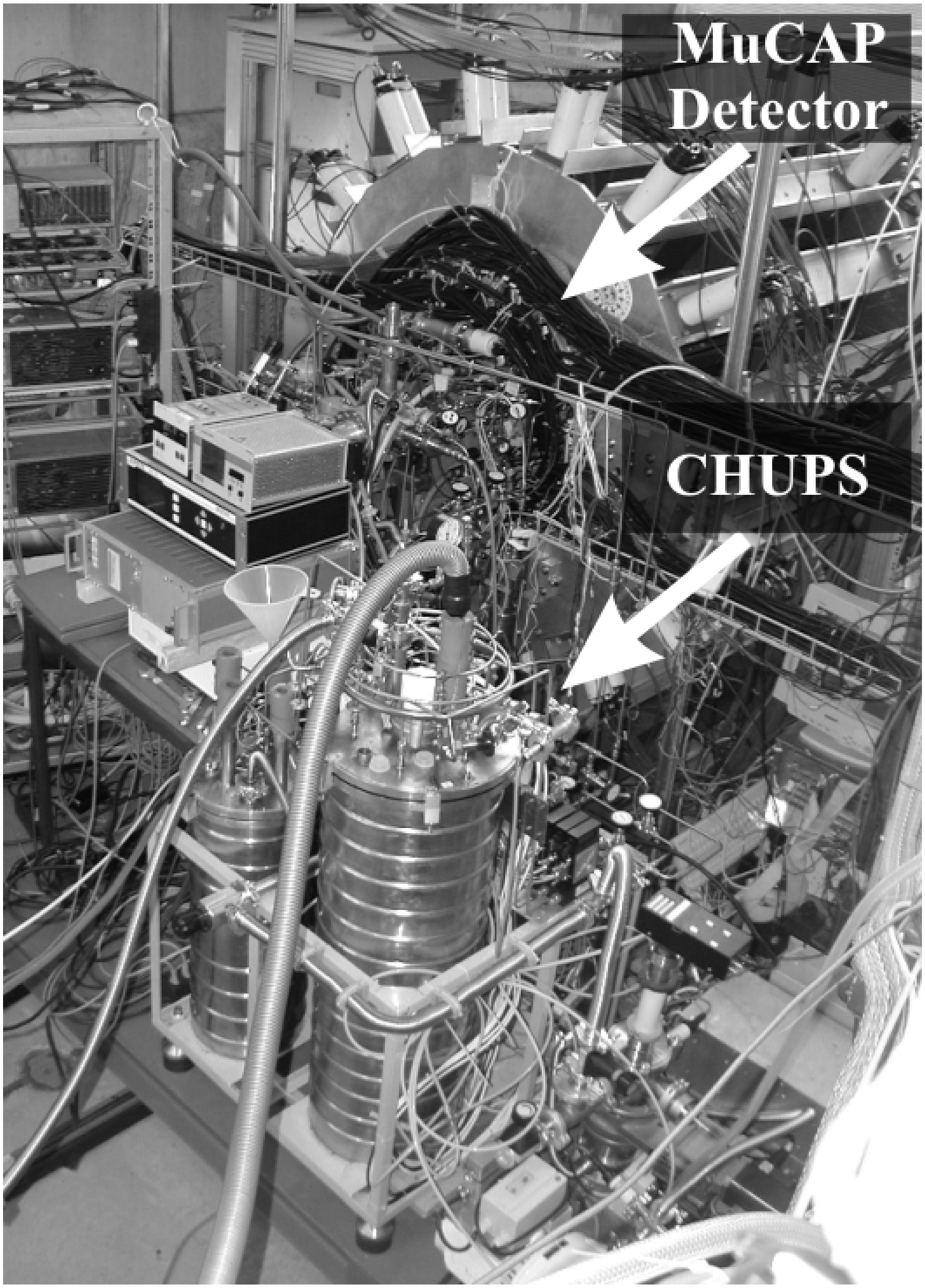

Figure 10. CHUPS in MuCAP experimental area.

The capture yield does not allow to distinguish individual contaminant concentrations. In order to verify cleaning efficiency for main impurities (nitrogen and oxygen), additional chromatographic analysis of hydrogen was realized. For this task, gas samples of about 10 l (at the normal conditions) were periodically taken into special probe volumes through the sampling line.

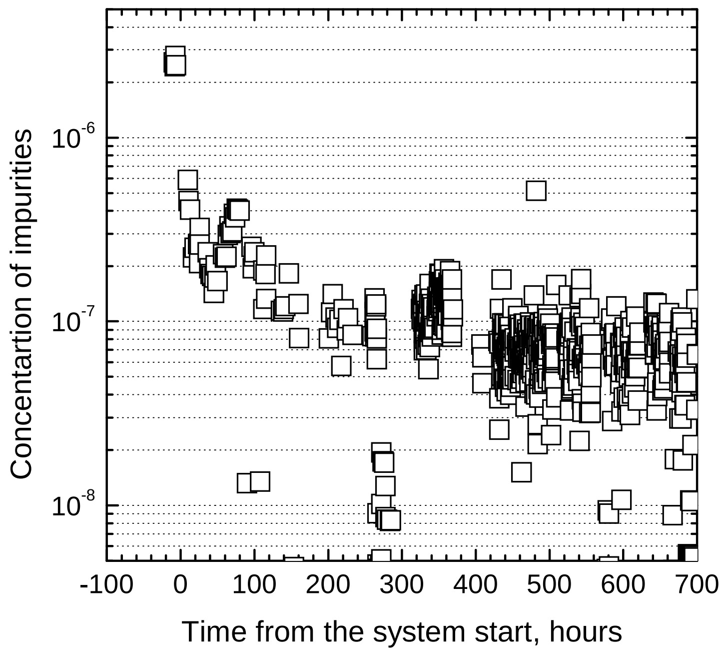

Figure 11. Impurity concentration in the TPC (capture yield data).

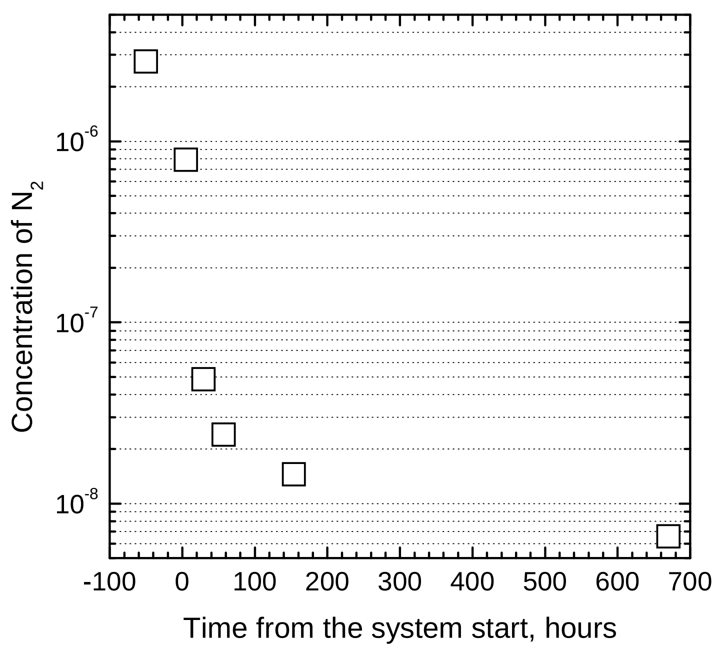

Figure 12. Nitrogen concentration in the TPC (chromatographic data).

The concentration of oxygen in a source gas (in two days before the CHUPS start) was 2.7 ppm. After 24 hours after the CHUPS start, concentration of oxygen in hydrogen dropped below the sensitivity of the chromatographic method (0.01 ppm) and did not exceeds this level later. Meanwhile, the concentration of

nitrogen permanently decreased. Results of the chromatographic measurements for nitrogen during the period of normal operation are shown in Figure 12.
Keeping the TPC pressure stable is the second important task of the system. The main condition of good pressure stability is the right choice of proportional and derivative parameters of PD-control. Figure 13 shows the pressure histograms at various hydrogen fluxes.

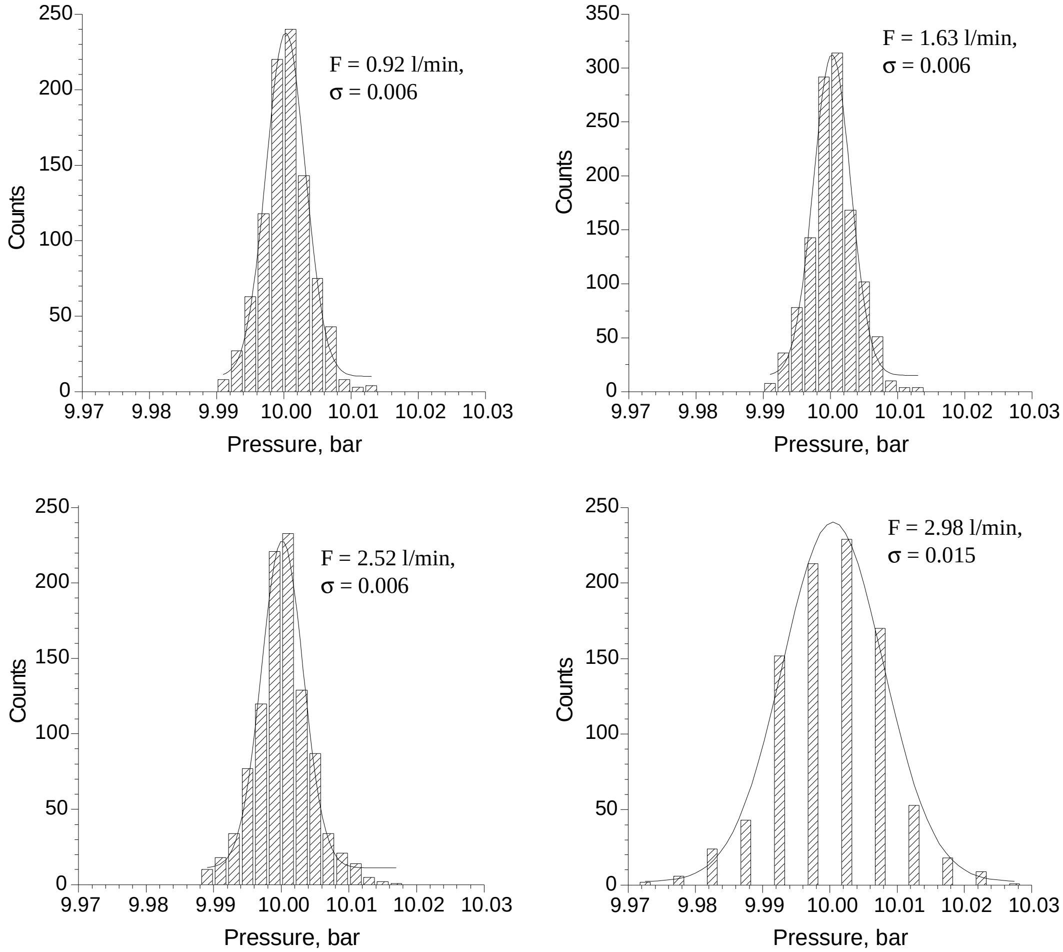

Figure 13. TPC pressure histograms at various hydrogen fluxes.
Average flux is denoted by F, histogram width – by σ.

For the normal operation CHUPS requires liquid nitrogen filling for compressors and filters tanks. It is convenient to have the time between these fillings of 8 hours or more. Experimental run showed that CHUPS accomplishes this requirement even at maximal average hydrogen flux (3.0 l/min). At lower flux the consumption of nitrogen is significantly less (Figure 14).

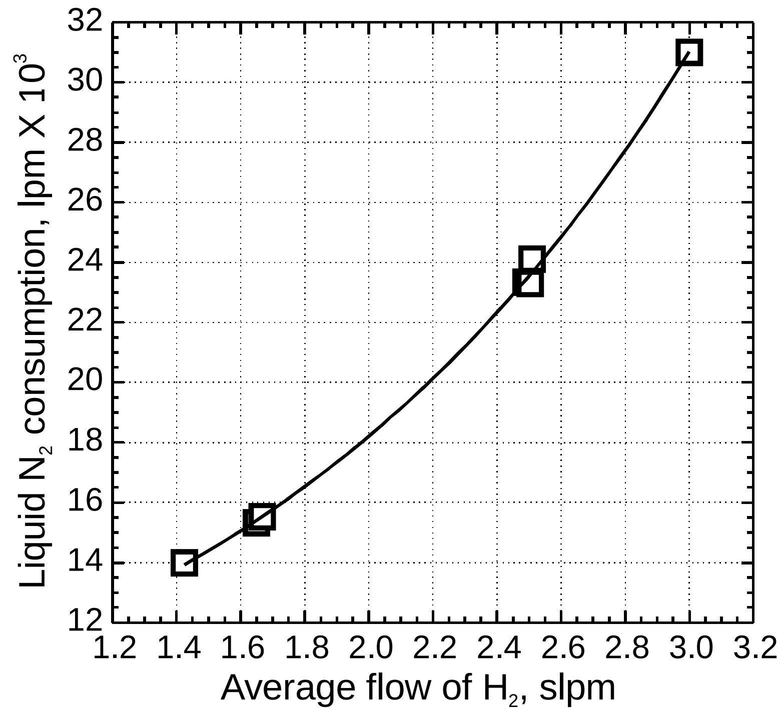

Figure 14. Liquid nitrogen consumption at various hydrogen flow rates.

5. Conclusion

The system showed stable operation during the real experimental run with more than 700 hours of working at the detector. Required purity of hydrogen in the TPC for the main expected contaminants (nitrogen and oxygen) was achieved. The requirements of detector pressure stability and liquid nitrogen consumption was also realized. These results confirmed applicability of cryopumps and adsorption cleaning method for ultrahigh hydrogen purification. Further evolution of the CHUPS may be associated with increasing of hydrogen flow rate and decreasing of liquid nitrogen consumption.

References

[1] F. Gray et al., Precision muon lifetime and capture experiments at PSI, 6th International Workshop on Neutrino Factories & Superbeams (NuFact'04), 2004, Osaka, Japan, nucl-ex/0410042.