SYSTEM FOR DEEP PURIFICATION OF HYDROGEN IN MuCap EXPERIMENT

V.A. Ganzha, P.A. Kravtsov, V.A. Trofimov, G.N. Shapkin, A.A. Vasilyev, M.E. Vznuzdaev

1. Introduction

The MuCap experiment at PSI is a precision measurement of the rate Λ_S for the basic electroweak process of muon capture, $\mu^- + p \rightarrow n + \nu$. A measurement of 1% accuracy determines the last well-known of the nucleon charged current form factors, the induced pseudoscalar g_P, to 7%. The experiment is carried out at the πE3 muon beam of the 580 MeV proton accelerator at the Paul Scherrer Institute (PSI), Switzerland.

The capture rate is determined from a measurement of the disappearance rate $\lambda_- \approx \lambda_+ + \Lambda_S$ of negative muons in hydrogen and the world average of the free μ^+ decay rate λ_+. Muons are stopped in a time projection chamber (TPC) with sensitive volume $15 \times 12 \times 30$ cm^3 filled with ultrapure hydrogen at 10 bar. Electrons ionized by muons or other projectiles drift vertically in a homogeneous electrical field to the bottom of the TPC, where they are amplified with a multiwire proportional chamber (MWPC) and read out in two dimensions.

The experiment imposes strict and critical requirements on the hydrogen gas system supporting the active target – TPC detector. As the hydrogen gas density of the experiment of ten times higher than at standard temperature and pressure, the rates for muon transfer from μp to typical chemical impurities (N_2, H_2O, O_2) are 3 orders of magnitudes larger than the muon decay rate. Once a muon has been transferred to a μZ atom, nuclear muon capture proceeds more than 100 times faster than on a μp atom. Thus, gas impurities distort the observed lifetime spectrum, and transfer must be suppressed by keeping the gas contaminations below a level of typically 10 ppb. Isotopic purity is required as well, since muon transfers to deuterium lead to a difficult diffusion problem. A dedicated isotope separation device [1] was constructed to produce protium with a deuterium contamination of less than 70 ppb; this work will be published separately.

As the gas amplification in the MWPC of the TPC sensitively depends on the hydrogen pressure, the pressure inside the TPC must be stabilized on the level of 10 bar with 0.1% accuracy. This is required to keep the MWPC gas gain constant within 1%. There is also concern that large flow variation in the TPC might induce dust accumulation and breakdown of the chamber high voltage. Thus the flow has to be stabilized as well. Finally, the whole detection system operated with high voltage in a pure hydrogen environment, thus reliability and hydrogen safety were of utmost importance.

The "Circulating Hydrogen Ultrahigh Purification System" (CHUPS) was designed and built to provide continuous protium purification and achieved all design criteria over several experimental runs with typically 2 months of continuous operation per year.

2. Circulating system (CHUPS) design

2.1. CHUPS operation scheme

During the experiment the TPC can directly monitor the capture reactions on impurities, the yield roughly proportional to the impurity concentration. This technique was used in 2003 before CHUPS was installed, showing the yield increasing during several days after filling the TPC. The chamber was filled through the palladium filter and exposed to the muon beam. The experiment revealed accumulation rate of some tens ppb per day which is unacceptable for the experimental requirements. This is the main reason for development of CHUPS designed for continuous gas cleaning and circulation through the operating TPC.

Initially a circulation system consisting of a mechanical pump and a Pd filter was considered. Pumps with high vacuum rating operating at 10 bar pressure are not commercially available and Pd filters supporting the required flux of 3 slpm (standard litres per minute) are expensive. Instead the alternative CHUPS scheme was realized, which is based on an adsorption cryopump to maintain the hydrogen flow and a cryogenic adsorption filter for removing the impurities. The adsorption cryopump has essential advantages, such as intrinsic high purity and reliability due to the absence of moving parts. The circulating system was designed, mounted and tested at PNPI in Gatchina, Russia, and installed at PSI during the preparation period

of the MuCap experiment in 2004 [2]. It was upgraded in 2005 on the basis of the operating experience. A simplified diagram of the system is shown in Fig. 1. CHUPS consists of two main units which are mounted separately on a common frame: compressor and purifier.

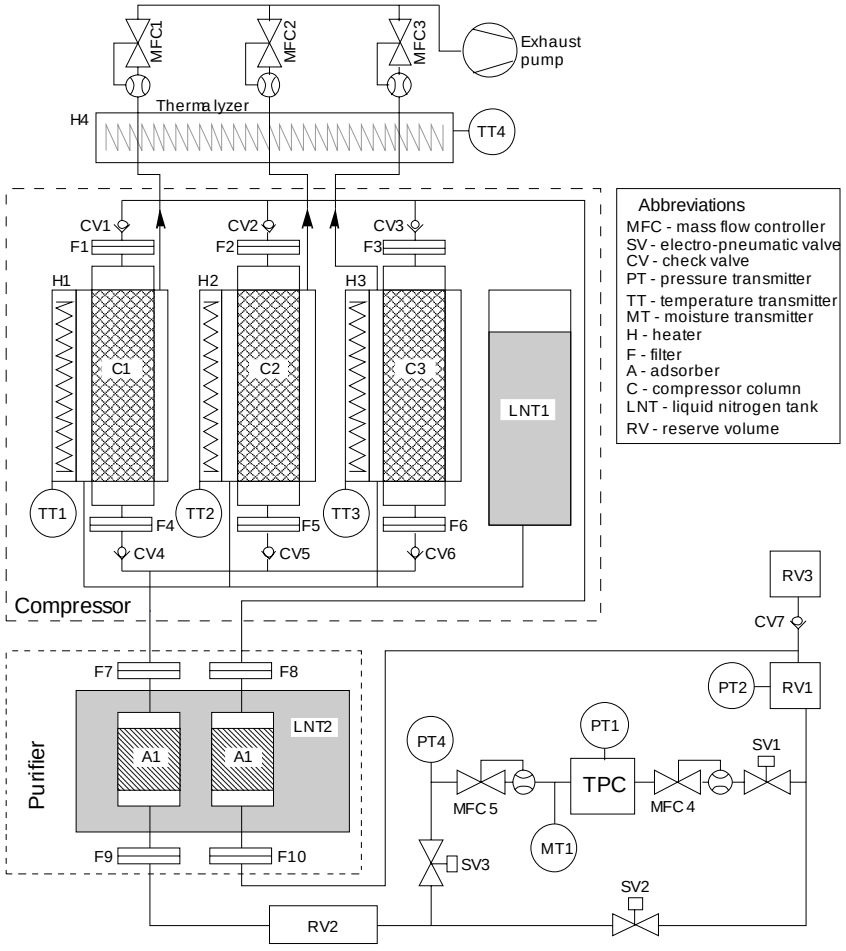

Fig. 1. Simplified CHUPS diagram

2.2. Compressor

The compressor is a triplex adsorption cryopump. It has three identical cartridges (columns) filled with activated carbon [1] and connected in parallel. The cartridges are made from thick-walled stainless steel tube and designed to keep high pressure. The volume of each column is about 1 liter and contains with respect to packed density of the carbon up to 0.6 kg of the adsorbent. Each column (C1, C2, C3 in Fig. 1) has a heat exchanger made of copper tube. It is coiled around the column and soldered to its outer surface by silver hard alloy. The heat exchanger is used to cool down the compressor column using liquid nitrogen flowing from the 40-litre supply vessel (LNT1). Liquid nitrogen flow is provided by an exhaust pump connected to the heat exchangers manifold. After cooling the columns nitrogen flows through the thermalizer (H4), which heats the gas up to the room temperature. Mass-flow controllers [2] MFC1, MFC2, MFC3 control the flow rate of the gaseous nitrogen and consequently regulate the liquid nitrogen flow rate and the cooling rate of the column. An electric heater is coiled around the column between the heat exchanger turns to provide the column heating. Each column has two check valves with the actuation pressure of 50 mbar that are installed in the inlet and outlet pipelines (CV1−CV6). Inlet and outlet lines of the columns are combined to the inlet and the outlet manifolds, respectively.

[1] Norit Nederland B.V. Nijverheidsweg Noord 72, 3800 AC AMERSFOORT, The Netherlands.
[2] Aalborg (http://www.aalborg.com/) stainless steel GFC series mass-flow controllers.

At the cooling stage the adsorbent inside the column adsorbs hydrogen, and the internal pressure drops below the pressure in the inlet line of the compressor. Consequently, the inlet check valve opens and passes hydrogen into the column. During the heating stage the adsorbent desorbs hydrogen, and the column internal pressure rises above the outlet line pressure. The outlet check valve opens and passes hydrogen into the outlet line. Thus, the combination of the check valves provides a pulsating flux of hydrogen in one direction. Cooling and heating rates are regulated by the balance of liquid nitrogen flow (controlled by MFC1, MFC2, MFC3) and heating power (managed by pulse-width modulation of the power supplies).

The temperature phases of the columns are shifted with respect to each other. Upper and lower temperatures and cycle frequency are regulated in accordance with the required average flow rate. A single compressor column pumps approximately 32 standard liters of hydrogen in one cycle employing a temperature range of 80–150 K. The maximum hydrogen flow of 3 slpm through the compressor is defined by the maximum cooling rate and the thermalization time of the activated carbon.

2.3. Purifier

The purifier contains two cartridges filled with NaX-type zeolite [3] (indicated as adsorbers A1 and A2 in Fig. 1). The adsorbers are immersed into liquid nitrogen vessel (LNT2) and permanently kept under temperature of 77 K during the whole experimental run. Low temperature is essential to increase the adsorption ability of zeolite for high-boiling contaminants (oxygen, nitrogen) against the main gas (hydrogen). Two adsorbers contain about 40 g of the sorbent in total, enough to accumulate up to 1 g of adsorbed water. The total amount of water supplied by the flow of 3 slpm during the two-month experimental run was about 2×10^{-2} g (allowing for 100 ppb constant humidity), which is 2% of the adsorber capacity. The adsorption capacity for other contaminants is comparable. Thus, the adsorbers guarantee full impurity removal in the hydrogen flow during the long term experiment. The adsorbent has to be exchanged or can be regenerated by heating up to 400°C and pumping before each experimental run.

The liquid nitrogen vessel is contained in the vacuum case and protected from external thermal radiation by a copper shield mounted on the secondary liquid nitrogen vessel which is also used to cool down the incoming hydrogen. This technique decreases liquid nitrogen consumption and prevents heating the zeolite adsorbers. The two adsorbers of the purifier (A1 and A2) are mounted on the hydrogen lines upstream and downstream of the TPC, respectively, providing two stage purification of the gas. The inlet zeolite adsorber takes out most of the impurities, which helps to avoid any decrease of the compressor capacity due to the accumulation of impurities in the activated carbon.

The system is equipped with mechanical 2 μm filters installed before and after the compressor and in the detector pipelines that prevent carbon or zeolite dust penetration to the clean part of the system. The final purification is provided by a special gas purifier [4] of limited capacity installed at the TPC inlet.

2.4. Control system

A special stand-alone microcontroller block provides all necessary regulation algorithms. The control block is connected to a PC *via* RS-232 or RS-485 serial interface. Due to implementation of all control algorithms in the independent control block, the system remains operational in the case of computer failure. Computer software is used for adjusting the parameters of the regulation algorithms, collecting and visualizing the process variables and keeping them in the database. All system events (including software messages and alarms) are also saved in the database. Special software was developed to access the parameters history and event log in the database.

The control block measures and controls all system devices like pressure sensors, valves, mass-flow controllers, *etc.* It provides the following regulation procedures:

- temperature stabilization of the three compressor columns and the thermalizer by regulating the heating and cooling provided by the heaters and the nitrogen mass-flow controllers, respectively;

[3] CECA company, http://www.adsorbents.com/sites/ceca/en/home.page.
[4] SAES Pure Gas - MicroTorr Ambient Temperature Gas Purifiers. http://www.puregastechnologies.com/microt.htm.

- cyclic operation of the compressor columns with phase shift;
- TPC internal pressure stabilization using mass-flow controllers;
- temperature stabilization of the humidity sensor.

Alarm and interlock functions are also implemented in the control block firmware (see Table). It protects the detector from underpressure and overpressure and controls differential pressure between the TPC and reserve volume RV1 to avoid hydrogen flux variations. Also, the software tracks liquid nitrogen level in the compressor tank (LNT1) and compressed air pressure that is used for the electro-pneumatic valves (SV1–SV3) actuation. All alarm events are attended with light and sound signal.

Table

Alarm events and interlock actions

Alarm description	**Condition**	**Action**
TPC pressure high	PT1>PT1max	Cut-off TPC and open bypass
TPC pressure low	PT1<PT1min	Cut-off TPC and open bypass
Differential pressure high	(PT2-PT1)>DPmax	Cut-off TPC and open bypass
Differential pressure low	(PT2-PT1)<DPmin	Cut-off TPC and open bypass
Compressed air pressure low	PT3<PT3min	Alarm signaling
Liquid nitrogen level low in LNT1	N2Level<N2Levelmin	Alarm signaling

2,5, TPC pressure and flow control

The three columns of the compressor induce the hydrogen flow to exchange the gas in the TPC. This flow is pulsating because of the periodical mode of the column operation. The pressure inside the TPC must be stabilized with 0.1% accuracy. In order to smooth the pressure variations caused by compressor, the CHUPS system is equipped with two reserve volumes of 15 liters content each (RV1 and RV2), pressure sensors (PT1, PT2, PT4) and mass-flow controllers (MFC4 and MFC5).

The reserve volumes are installed in the inlet and outlet lines of the TPC. They are used as buffering volumes. The RV1 also provides a hydrogen reserve to support the pressure stabilization algorithm. Each volume is equipped with a pressure sensor (PT2 and PT4, same model as PT1). Two mass-flow controllers with a maximal flow of 20 slpm are mounted at the inlet (MFC4) and outlet (MFC5) lines of the TPC.

The internal pressure of the detector (measured by PT1) is stabilized using PID (Proportional-Integral-Derivative) regulation. The mass-flow controller at the detector outlet (MFC5) is set to a constant flow rate. The TPC inlet mass-flow controller (MFC4) is operated by PID algorithm in the control software. The MFC5 set point defines the average flow rate through the TPC vessel. The pressure distribution histogram (Fig. 2) for the long term operation during 50 days shows excellent pressure stability on the level of 0.024% at a mean hydrogen flow of 3 slpm. The histogram bars correspond to the ADC discretization in the control system (1.2 mbar). Thus the pressure is kept within ±2 least significant bits of the ADC. The second reserve

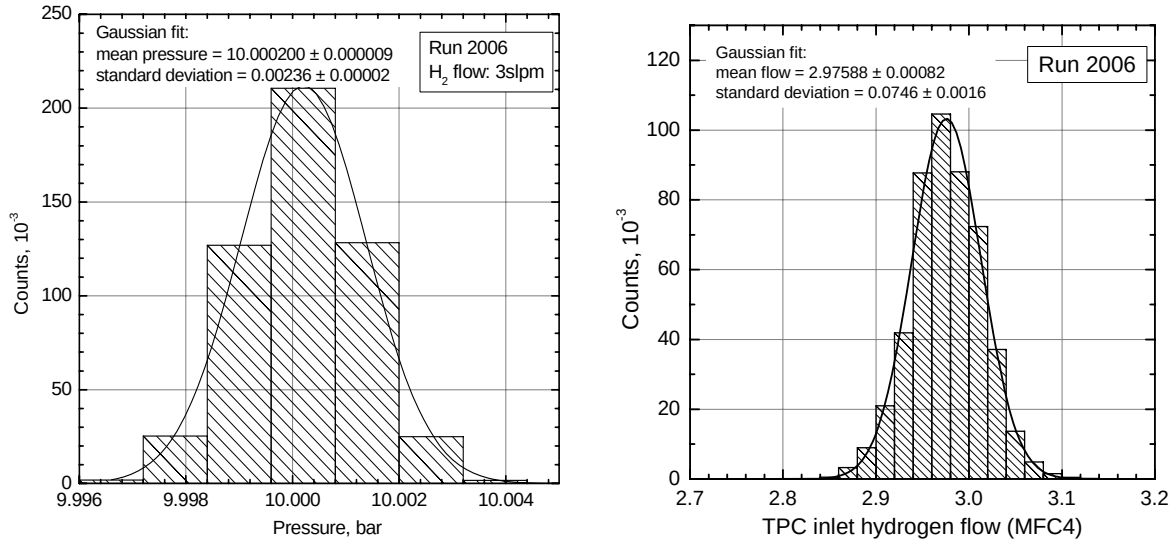

Fig. 2. TPC pressure and inlet flow distribution histograms. The gaussian fits gives a full width of 2.4 mbar and 0.075 slpm that correspond to 0.024% and 2.5% stability, respectively

volume (RV2) and the mass flow controller (MFC5) at the compressor inlet were installed to prevent pressure drops caused by the fast opening of the check valve. This results in a flow stability of 2.5% at the mean flow level of 3 slpm during 50 days of CHUPS operation (Fig. 2).

3. Monitoring impurities

The initial purity of the evacuated TPC system was provided by continuous pumping and baking of the TPC vessel. The residual gas contents were controlled by a quadrupole mass-spectrometer in the mass range from 2 to 100 atomic mass units (a.m.u.). During the protium runs atmospheric gases (oxygen and nitrogen) were analyzed by off-line gas sample chromatography, and the moisture of the gas was directly monitored with an on-line humidity sensor. The total amount of impurities was continuously monitored by direct measurements of the yield of muon capture on impurities as observed in the TPC.

3.1. Chromatographic analysis of nitrogen and oxygen

The oxygen and nitrogen content is measured by gas chromatography with a thermal conductivity sensor. After being filtered and purified in a cryogenic adsorption purifier, helium carrier gas is distributed in two directions with equally adjusted flow rates. The first flow passes through the reference chromatographic column. The second flow can be either routed to the working chromatographic column directly or through the accumulating column using the volume batcher which injects the fixed volume into specified direction. Both chromatographic columns are filled with a specially treated adsorbent (zeolite). The adsorbent in the working column separates the admixtures, while the one in the reference column provides a hydraulic resistance equal to the working column.

Preparatory enrichment is required for the very low concentrations of impurities in the hydrogen gas samples. Hydrogen from the sample bottle is directed to the accumulating column by the batcher. This column is filled with zeolite and immersed into a liquid nitrogen vessel at 77 K temperature. The amount of gas passed through the accumulating column is measured by a rotary drum gas meter. Impurities from the gas stream are adsorbed by the zeolite and remain in the accumulating column. After the desired amount of sample gas has passed through the adsorber, the accumulating column is removed from the cryogenic vessel and heated. Then the batcher directs the helium carrier gas flow through the accumulating column which washes the impurities out of the adsorbent.

In the next step, the carrier gas enriched by contaminants flows through the working column, where contaminants are separated on the adsorbent. Both reference and working flows pass through the detector in parallel, that measures their differential heat conductivity. The differential output signal is proportional to the admixture concentration. Signal peaks are registered by a PC and processed by special software. The enrichment coefficient (the sample volume passed through the accumulating column) and parameters of the measuring scheme are adjusted with respect to the admixture concentration. The final calculation of the concentration is based on a calibration measurement. A serial dilution method was used to obtain the set of calibrating samples with decreasing concentration of nitrogen and oxygen. With this method a sample of the air was serially attenuated by the hydrogen. The total calibration error, including non-linearity, is 10%. The traces of the main air components were monitored by gas samples of 8−10 normal liters from the TPC using the chromatographic method. A sensitivity of 5 ±1 ppb for oxygen and 7 ±1 ppb for nitrogen was obtained.

3.2. Humidity analysis

During the MuCap experiment 2004 we observed a capture yield corresponding to about 60 ppb impurities, significantly higher than the oxygen and nitrogen concentration derived from the gas chromatography

In 2005 an online humidity sensor [5] was installed to investigate whether the additional observed yield can

[5] Pura PUR-TX-120 Gas Dew-point Transmitter.MichellInstruments. http://www.michell.co.uk/cat/view/pura.html

be explained by residual water vapor in the hydrogen gas. The sensor has sensitivity of 0.02 ppb with error of +30–50% in the 2–100 ppb range. It was mounted in a temperature-stabilized box in order to reduce any influence of the ambient temperature to the sensor reading. Sensor and inlet pipeline were kept at 21°C with 0.2°C stability. As shown in Fig. 1, the humidity sensor (MT1) was installed in the gas circuit such that it could measure either the humidity in the outlet TPC flow or in the isolated CHUPS system (while hydrogen is circulating through the chamber bypass). Continuous bypass circulation gave 3 ppb humidity. During the production run the humidity sensor clearly demonstrated that humidity was the main additional impurity. To calibrate the effect of H_2O impurities on the experiment, a water permeation tube [6] was used for generation of the known water concentration in the hydrogen flow. The tube was placed in the temperature stabilized vessel. Stable hydrogen flow was passed through the volume with the tube.

3.3. Impurity capture rates

The specifics of the MuCap experiment allow unique in-situ measurements of impurity concentrations during the experimental runs. While the vast majority of negative muons remain in muonic hydrogen atomic (μp) and molecular ($p\mu p$) states during their lifetime, a small fraction can be captured on impurities in the following steps: $\mu p + N_Z \rightarrow \mu N_Z + p$; $\mu N_Z \rightarrow N_{Z-1} + \nu$. Here N_Z is a nucleus of an impurity atom, like N or O. The first transfer reaction proceeds with a rate proportional to the impurity concentration c_Z. The second capture reaction occurs with a probability $\Lambda_Z/(\Lambda_Z + \lambda_+)$, which is the result of the competing processes of capture with rate Λ_Z and free muon decay with rate λ_+. The charged recoil nuclei N_{Z-1} are detected in the TPC. The observed yield of capture recoils per muon is $Y_Z = c_Z k_Z$, where k_Z are coefficients which depend on Λ_Z and the detection efficiency for recoil N_{Z-1}. They are determined in dedicated calibration runs where hydrogen is doped with a single impurity N_Z of well measured concentration c_Z, typically 100–1000 times higher than in the clean run conditions. Typical values are $k_N = 70$ and $k_O = 400$ for both Y_Z and c_Z in ppm (the detection efficiencies slightly differ from run to run). In summary, the capture yield measurement allows a continuous and precise measurement of the overall observed capture yield from impurities, but it does not distinguish between their elemental composition.

4. Operating experience

4.1. Clean fills for production data

Before the 2004 experiment the TPC was baked for several weeks. The performance of the gas system over the whole run period is documented in Fig. 3. The chromatographic analysis allows monitoring the change of oxygen and nitrogen concentrations during the MuCap experimental run. Fig. 3 shows the behavior of nitrogen concentration for the MuCap experimental run after starting the CHUPS. The final chromatographic measurements resulted in nitrogen concentrations less than the method sensitivity (7 ppb). Oxygen traces dropped below the method sensitivity of 5 ppb within two days after starting the circulation.

The most reliable drying experience was obtained during the spring 2006 experimental run. Before the run, the TPC had been exposed to continuous (about 2 months) vacuum pumping with a simultaneous baking at ~120°C. This procedure led to an initial humidity level at the moment of CHUPS connection of 60 ppb (Fig. 4, left). During 400 hours of continuous cleaning with a mean hydrogen flux rate of 3 l/min the humidity exponentially decreased to ~18 ppb and remained at this level till the end of the main μ^- data run, providing a stable operation over more than 1000 hours. The minor fluctuations of the humidity are explained by temperature fluctuations in the experimental hall. The change of hall temperature affects the adsorption-desorption equilibrium in the chamber and, consequently, its outgassing rate. Then CHUPS was disconnected from the TPC and run through bypass line. The humidity decreased exponentially and reached the 10 ppb level in 20 hours (Fig. 4, right). The final CHUPS stand-alone humidity result is 3 ppb.

[6] Permeation tube providing 500 ppb ± 10% at 0.5 slpm flow. Valco Instruments Company Inc. P.O. Box 55603, Houston, TX 77255, USA.

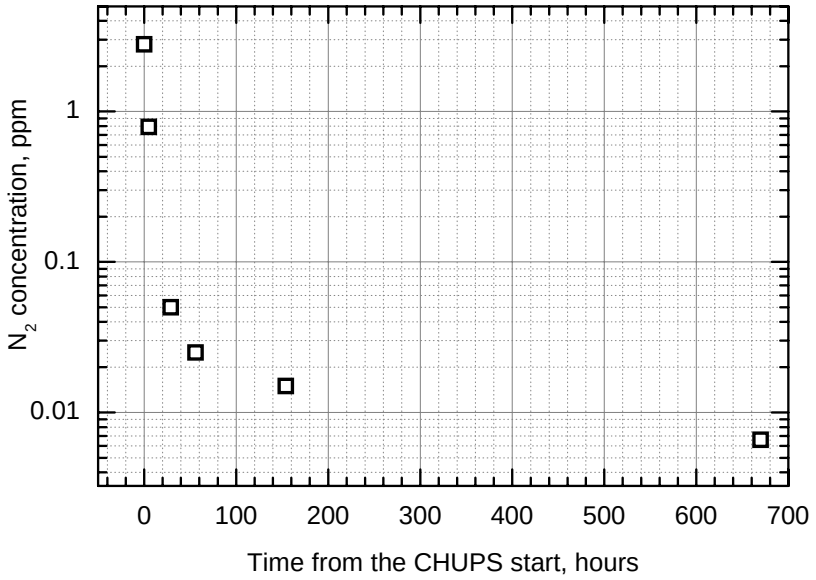

Fig. 3. Nitrogen concentration change during MuCap run 2004. Average hydrogen flow is 1.6 l/min

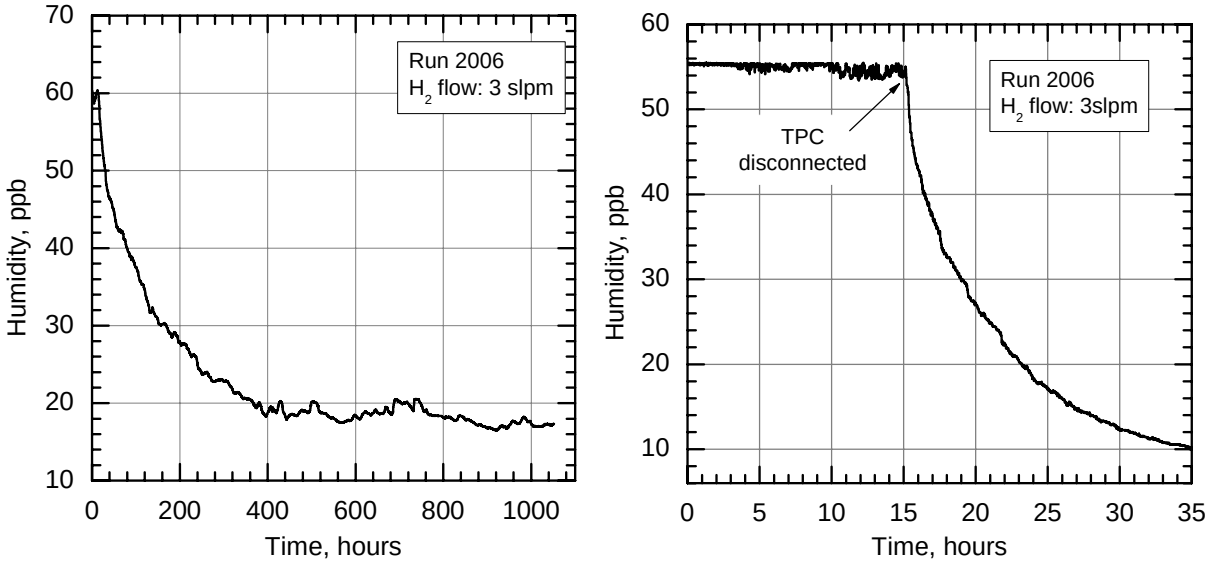

Fig. 4. Humidity decrease in the TPC after CHUPS connection (left) and after bypassing the TPC (right)

4.2. Impurity doped fills for calibration data

The cleaning power of CHUPS was greatly confirmed during systematic calibration studies of the TPC detector which were carried out during the last weeks of the MuCap experimental run in spring 2006. Several experiments were carried out for the TPC calibration and the determination of the major impurities. The high cleaning power of the CHUPS system makes it possible to return the detector to the normal clean operation in a short time.

The first experiment was a "water doped run". During this test the humidity inside the TPC was increased up to 2000 ppb approximately by a specially prepared humidity generator. Then CHUPS was connected and the gas was cleaned down to 400 ppb in one day (Fig. 5). The slower cleaning speed can be explained by wetting of the inner surfaces of the chamber.

The efficiency of CHUPS cleaning a large contamination of nitrogen in the detector hydrogen was tested by adding a "high" 22 ± 1 ppm nitrogen admixture from a known amount of nitrogen previously diluted

in a vessel with high purity protium. This "nitrogen doped" condition was intended as a detection efficiency calibration of nitrogen impurity capture events in the TPC. After measuring with the large nitrogen contamination (flat region) the CHUPS circulation through the TPC vessel was re-established and the nitrogen was removed. The cleaning progress observed on-line *via* capture events in the TPC is shown in Fig. 6. The cleaning started with a yield of more than 1000 ppb and proceeded until leveling off around ~30 ppb, a value established before the doping. The purification process took about 17 hours.

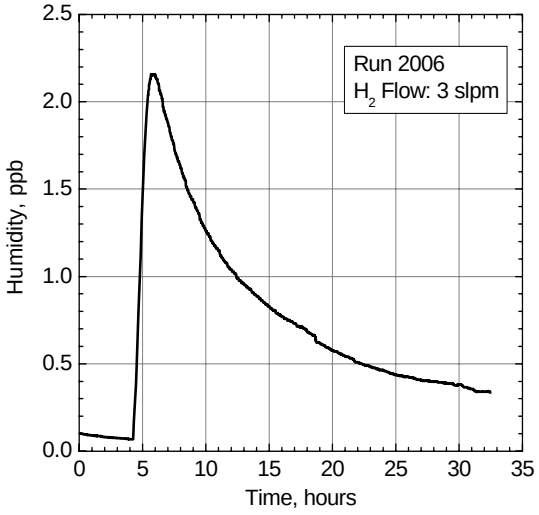

Fig. 5. Drying of TPC after the water doped run

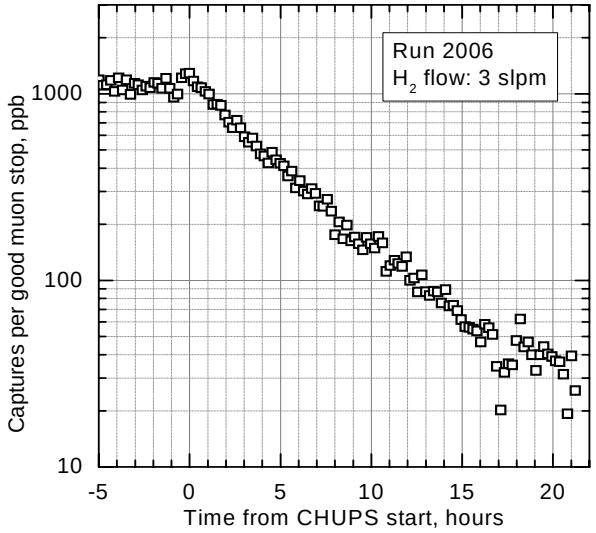

Fig. 6. Impurity captures per good muon stop during the nitrogen cleaning

4. Results

The CHUPS system was installed in the MuCap experiment at PSI and first connected to the TPC detector in 2004. The system proved to be very reliable and flexible during three experimental runs in 2004–2006.

The experience obtained during development and use of the CHUPS justifies the initial design decisions. The main result was a stable clean working gas of the TPC during the experimental runs. A smooth hydrogen flow of 3 l/min was kept during the whole TPC operation time (more than 1000 hours in one experimental run). The best purity reached was 18 ppb for moisture content and 8–10 ppb for nitrogen concentration. This result is sufficient to reach the main goal of the MuCap experiment and allows precise corrections of the proton capture rate for these impurities. In addition to regular cleaning, the CHUPS provided fast cleaning of the working gas after calibrations with contaminants doping. This feature was very useful for the nitrogen and water calibration experiments intended for systematic studies of the TPC detector. Also, variations of the CHUPS flow and correspondingly the equilibrium humidity in the chamber gave important additional calibration points for the systematic investigations.

With regards to the stable pressure requirement the CHUPS also yielded a good result. The pressure inside the TPC was kept at the appropriate level of 10 bar with 0.024% stability during all the operation modes. Pulsations of the hydrogen flow through the TPC chamber were also minimized to the level of 2.5% at the mean flow of 3 l/min.

References

1. I. Alekseev *et al.*, Preprint PNPI-2702, Gatchina, 2006. 26 p.
2. B. Bezymyannykh *et al*., Preprint PNPI-2611, Gatchina, 2004. 17 p.