

# First Tritium Pumping Results by a Cryopump with a precondensed Argon Layer

Date: 19/09/2006

F. Eichelhardt\*, B. Bornschein<sup>†</sup>, L. Bornschein\*, O. Kazachenko<sup>†</sup>, N. Kernert\*\* and M. Sturm\*

\**Universität Karlsruhe, Institut für Experimentelle Kernphysik, Germany*

<sup>†</sup>*Forschungszentrum Karlsruhe, Tritiumlabor Karlsruhe, Germany*

\*\**Forschungszentrum Karlsruhe, Institut für Kernphysik, Germany*

**Abstract.** The Karlsruhe Tritium Neutrino experiment KATRIN employs a cryo pump at 4.2 K with pre-condensed Ar as adsorbent to keep its electrostatic spectrometers essentially free from tritium. The influence of the decay heat of tritium decay on the pumping properties of such a cryo pump is investigated with the test experiment TRAP (Tritium Argon frost Pump) at flow rate levels of  $10^{-7} - 10^{-14}$  mbar l/s. This publication presents results obtained by the TRAP experiment during the measurement run #6 and gives a first approximation for the tritium transmission probability through the Cryogenic Pumping Section (CPS) of KATRIN.

**Keywords:** tritium, cryopump, precondensed argon, silicon detector

**PACS:** 07.30.-t, 68.43.-h

## INTRODUCTION

The investigation presented in the following was undertaken within the scope of the KATRIN experiment, the next generation tritium decay experiment for determining the electron neutrino mass [1]. KATRIN investigates spectroscopically the electron spectrum from tritium beta decay near its kinematical endpoint of 18.6 keV. With a strong windowless gaseous tritium source ( $2.7 \cdot 10^{19}$  tritium molecules) and a tandem of two electrostatic spectrometers, KATRIN will allow a model independent measurement of neutrino masses with an expected sensitivity of 0.2 eV/c<sup>2</sup> (90% CL).

In the 10 m long source tube a constant tritium column density of  $5 \cdot 10^{17}$  molecules/cm<sup>2</sup> is maintained by continuous tritium inlet of 1.8 mbar l/s (STP) in the middle and by continuous pumping of the tritium at its ends. Between source and spectrometer the magnetic transport system is located, which guides the decay electrons adiabatically from the source to the spectrometers while at the same time suppressing the tritium flow rate below  $10^{-14}$  mbar l/s. A first flow rate reduction of  $10^7$  is achieved by a differential pumping section with turbomolecular pumps. The remaining  $10^7$  suppression will be accomplished by the Cryogenic Pumping Section (CPS), which is essentially a tube covered with pre-condensed Ar as adsorbent and kept at  $\approx 4.2$  K via cooling with liquid helium.

Gas binding pumps like getter pumps or cryogenic pumps are widely used for pumping hydrogen (and its isotopes). The cryogenic surface of the latter type is often covered with an adsorbent like a molecular thief, charcoal, zeolith or a pre-condensed heavier gas. Especially in fusion reactor research, cryo pumps (at  $\approx 77$  K) are employed in various types for removing traces of hydrogen isotopes from helium as reported in [2, 3, 4, 5, 6]. Yuferov et al. [7] and Benvenuti et al. [8] report on adsorption of H<sub>2</sub>, D<sub>2</sub> and HD at 4.2 K on pre-condensed gas layers like, for example, frozen Ar, CO<sub>2</sub> and Ne. Although Ar does neither provide the biggest monolayer capacitance nor the lowest equilibrium pressure of the available adsorbents [9], it is still the choice for pumping tritium in the CPS for the following reasons:

- As a pre-condensed gas Ar does not suffer from the drawbacks solid adsorbents have: e.g. difficulty of attachment to metallic surface with good thermal contact, sensitivity to poisoning and reduced life time with thermal cycling
- As an inert gas Ar is chemically inactive which is convenient with regard to tritium processing by means of catalysts employed in tritium infrastructure systems.
- Carbonaceous cryo deposits like CO<sub>2</sub> with better cryo pumping properties than Ar have the disadvantage that the tritium retention system of the Tritium Laboratory Karlsruhe (TLK), where KATRIN is being built, can only

cope with them inadequately.

- Since the adsorbent is freshly prepared at the beginning of each KATRIN measurement run, no tritium inventories are collected over time.

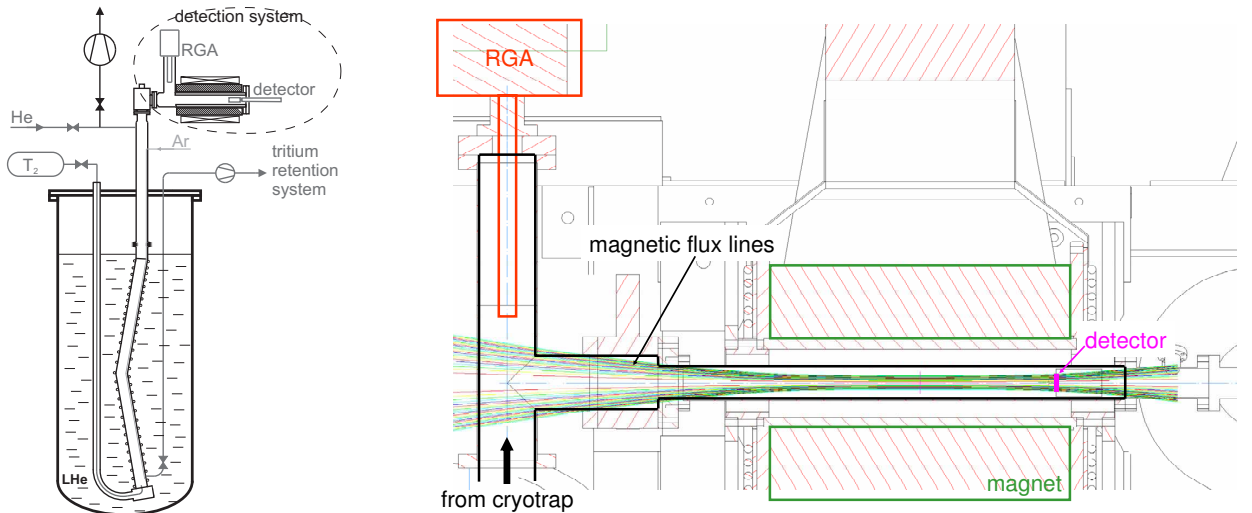
Yet, the pumping properties of the frozen Ar adsorbent are negatively influenced by the radioactive decay of tritium. Since the decay electrons from tritium beta decay with the average energy of  $\approx 6$  keV have a reach which equals approximately the thickness of the frozen Ar layer ( $1 - 2 \mu\text{m}$ ), almost the whole decay heat is being deposited in the adsorbent. This increases the desorption rate of both argon and tritium. Due to successive desorption-adsorption processes, tritium might migrate along the cryo surface and pass the whole CPS. The absence of experimental data on tritium pumping with an Ar covered cryo surface at 4.2 K in the flow rate regime of  $10^{-14} - 10^{-7}$  mbar l/s demands for the test experiment TRAP, which is being pursued at the TLK. TRAP is a model pump for the CPS with the same gas conductivity and tritium surface density. The main goal is to measure the tritium transmission probability  $K$  of a CPS-like cryo pump with Ar condensate as adsorbent.

This paper presents data obtained during the measurement run #6 with TRAP. Unlike the runs performed before, run #6 provides the possibility to calculate the tritium transmission probability of a CPS-like cryo pump from the knowledge of the ingoing and outgoing tritium flow rates.

## THE TRAP EXPERIMENT

The TRAP experiment is a cryogenic pump for measuring the pumping properties of Ar cryo deposits at 4.2 K. It can not only test Ar, but also other pre-condensed gas adsorbents. The TRAP test experiment is a model for the cryogenic pumping section CPS with the same cryogenic properties but without the electron guiding magnets. It has the following general features (see fig. 1 and refer to [10] for more details):

1. The cryo surface consisting of the two lower tubes is cooled in a LHe bath cryostat to a temperature of  $\approx 4.2$  K.
2. The cryo surface is a bent tube ( $20^\circ$ ) with approximately the same gas conductivity which is planned for CPS ( $\approx 4.5$  l/s). Its surface ( $1571 \text{ cm}^2$ ) is about 1/10 of the CPS surface.
3. Ar cryo deposit is prepared on the cryo surface via a capillary. The Ar gas exits the capillary through tiny holes (diameter  $\approx 0.2$  mm) along its length and is distributed approximately homogeneously over the cryo surface.
4. Tritium is injected from a calibrated buffer vessel into the cryo trap. From the pressure drop in the vessel the average inlet flow rate can be calculated.
5. Tritium passing through the cryo trap can be detected with a residual gas analyzer (*Spectra Microvision PLUS "Smart Head"*: Open Source, Tungsten Filaments) or a silicon solid state detector (LN<sub>2</sub> cooled,  $300 \text{ mm}^2$  surface,



**FIGURE 1.** Left: Scheme of the complete TRAP setup. Right: Scheme of the TRAP detection system with magnetic flux lines.

**TABLE 1.** Experimental parameters of gas inlet during run #6

	Inlet amount	Inlet flow rate	Layer thickness	Inlet mixture	Activity
Argon preparation*	74.9 mbar l	0.009 mbar l/s	1 – 2 $\mu\text{m}$	—	—
Tritium inlet	0.134 mbar l	$\approx 10^{-7}$ mbar l/s	—	$\approx 44\%$ tritium <sup>†</sup>	5.6 GBq

\* cryosurface temperature  $\approx 5.2$  K, achieved by over-pressurizing the cryostat to  $\approx 1.4$  bar

<sup>†</sup> Isotopic inlet mixture: DT 43%, T<sub>2</sub> 19%, HT 7%, HD / H<sub>2</sub> / D<sub>2</sub> 31%

500  $\mu\text{m}$  thickness, energy resolution (FWHM)  $\approx 2.1$  keV, noise level in energy window 6 – 20 keV  $\approx 70$  mHz). See fig. 1 for a scheme of the detection system. A magnetic field of 0.1 T can be applied to guide the tritium beta decay electrons onto the Si detector.

- Connections for gaseous He are foreseen for the regeneration procedure: To avoid an increase of contamination of the weakly contaminated (upper) parts of the cryo surface, the cryo trap is purged with He gas in the direction from low to high contamination during the warming up.

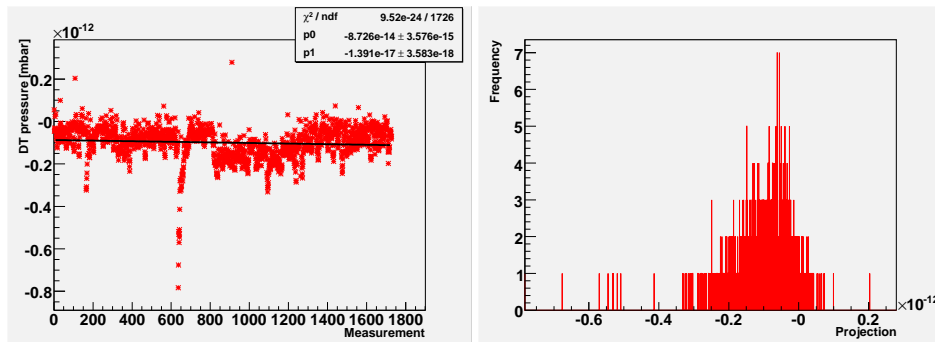
## TRAP MEASUREMENT RUN #6

Table 1 shows the experimental parameters of the measurement run #6 which was performed during five weeks. The tritium detection was done by observing mass 5 (DT) on the RGA and by measuring the count rate on the Si detector in the energy window 6 – 20 keV. A continuous measurement with both methods is not possible since the RGA's filament casts light on the Si detector which increases significantly the noise on the detector. Therefore, the RGA and the Si detector were operated in turns. Since the tritium partial pressure (DT pressure) is below the RGA's detection limit (see next section), the tritium detection was mostly done with the Si detector.

## Partial pressure data from RGA

Mass 5 (DT) was chosen as observation mass for the tritium pumping behavior of the cryo pump for the following reasons:

- The background measurements before tritium inlet showed, that mass 5 (DT) had the lowest background reading on the RGA.
- DT is pumped worse on Ar frost than T<sub>2</sub>, since the saturated vapor pressure of condensation is lower for T<sub>2</sub> than for DT [11]. The DT pressure will thus give an upper limit for the T<sub>2</sub> pressure.
- HT although being the worst of the tritium containing hydrogen molecules to be pumped could not be used for observation, since mass 4 could be HT, D<sub>2</sub> and He and it was not possible to separate all of them even with the high resolution 1-6 amu residual gas analyzer.



**FIGURE 2.** DT partial pressure measured with residual gas analyzer (left) and frequency of pressure bins (right: bin width  $10^{-16}$  mbar).

- DT was the most abundant molecule in the isotopic inlet mixture (refer to tab. 1).

The RGA often shows invalid pressure readings below zero as can be seen in fig. 2. This is due to the fact that the DT partial pressure is below the RGA's detection limit of  $\approx 10^{-13}$  mbar, which means that the RGA's electronic noise exceeds the ionization current. Yet, an increase of the DT partial pressure should nonetheless be visible as an increase of the frequency of the valid pressure readings (in the range of  $10^{-13} - 10^{-12}$  mbar). A linear fit to the pressure data yields a slightly negative slope of  $(-1.4 \pm 0.4) \cdot 10^{-17}$  mbar, which is compliant with zero when taking the order of magnitude into account. Thus, the RGA does not detect an increase in the DT partial pressure.

### Silicon detector data

The Si detector is used to detect tritium nuclei via their radioactive decay. The tritium beta decay electrons hit the Si detector creating a count rate  $A$  [1/s] in the energy window 6 – 20 keV. The lower cut suppresses electronic noise from the detector, the upper one marks approximately the endpoint of the tritium  $\beta$  spectrum taking the detector energy resolution into account. The TRAP experiment has two operational modes when measuring with the Si detector:

1. **Magnetic field off:** The count rate at the detector is  $A_{off}$ , originating from beta electrons hitting the detector surface by chance.
2. **Magnetic field on:** The count rate at the detector is  $A_{on}$ , originating from beta electrons which are guided onto the detector surface along the magnetic field lines. Generally  $A_{on} > A_{off}$  is true.

The count rates over time measured with these two operational modes during the tritium inlet are presented in fig. 3. The main goal of the TRAP experiment is to obtain the tritium transmission probability through the cryo trap:

$$K = \frac{\text{outgoing flow rate}}{\text{incoming flow rate}} = \frac{Q_{out}}{Q_{in}} \quad (1)$$

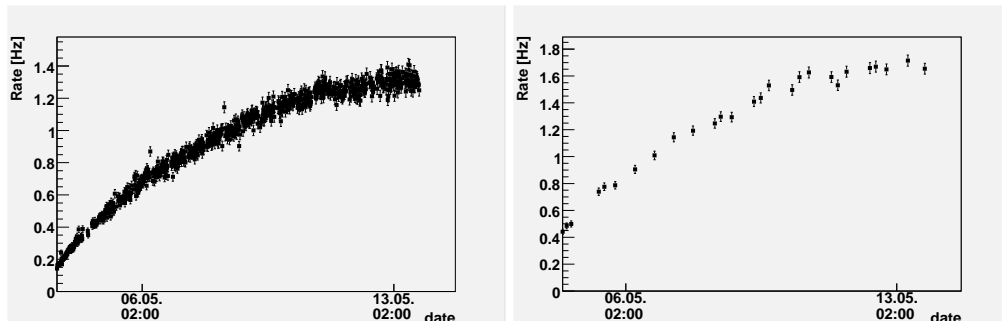
In run #6 the tritium inlet was performed over  $\approx 10$  days with an average inlet flow rate of  $Q_{in} \approx 10^{-7}$  mbar l/s. The outgoing tritium flow rate  $Q_{out}$  [mbar l/s] during the same 10 days consists of several parts, since tritium coming from the cryo trap into the detection system is either adsorbed on the detector (adsorption rate  $Q_D$  [mbar l/s]), adsorbed on the walls of the detection system (adsorption rate  $Q_W$  [mbar l/s]), kept in the gas phase (accumulation rate  $Q_G$  [mbar l/s]) or buried in the bulk material of the stainless steel walls in the detection system (diffusion rate  $Q_B$  [mbar l/s]):

$$\Rightarrow Q_{out} = Q_D + Q_W + Q_G + Q_B \quad (2)$$

These different contributions are investigated in the following to obtain  $Q_{out}$  and hence  $K$ .

**$Q_G$ :** The signal rate contribution from the gas phase must be low ( $< 1$  mBq), since we know from the residual gas analyzer data that the tritium partial pressure (DT pressure) is below  $10^{-13}$  mbar. If one assumes a steady increase of the tritium partial pressure in the detection system (volume  $< 3$  l) from zero to  $10^{-13}$  mbar during the 10 days tritium inlet, one obtains the following upper limit for the accumulation rate:

$$\Rightarrow Q_G < 3 \cdot 10^{-19} \text{ mbar l/s} \quad (3)$$



**FIGURE 3.** Count rate measured with the Si detector during tritium inlet in the energy window 6 – 20 keV over time with magnetic field off (left:  $A_{off}$ ) and magnetic field on (right:  $A_{on}$ ). [12]

**Q<sub>B</sub>**: The biggest unknown in the TRAP measurement runs so far is the tritium diffusion rate into the stainless steel bulk material of the detection system. Until now, we had no possibility to determine  $Q_B$ . The usage of gold plated walls in the detection system will suppress the burying of tritium and should increase the other contributions to the total outgoing tritium flow rate. For run #7 this gold plating will be applied.

**Q<sub>D</sub>** and **Q<sub>W</sub>**: The tritium adsorption rates on the detector surface and on the chamber walls of the detection system can be determined from the count rates  $A_{off}(t)$  and  $A_{on}(t)$  shown in fig. 3. To achieve this, we first need to calculate the time-dependence of the tritium surface contamination on the detector  $C_D(t)$  [Bq] and on the walls of the detection system  $C_W(t)$  [Bq]. These contaminations transfer to the count rates with and without magnet in the following way:

$$A_{off} = e \cdot C_D + a \cdot C_W \quad \text{and} \quad A_{on} = r \cdot e \cdot C_D + b \cdot C_W \quad (4)$$

The constants a, b, e and r were determined by Monte Carlo simulations [12]:

$$\begin{aligned} a &= 7.0 \cdot 10^{-4} && \text{(fraction of the wall contamination detected by the detector when magnet is off)} \\ b &= 1.4 \cdot 10^{-3} && \text{(fraction of the wall contamination detected by the detector when magnet is on)} \\ e &= 7.2 \cdot 10^{-2} && \text{(fraction of the detector contamination detected by the detector when magnet is off)} \\ r &= 1.19 && \text{(reflection coefficient due to the reflection of decay electrons in the magnetic field)} \end{aligned}$$

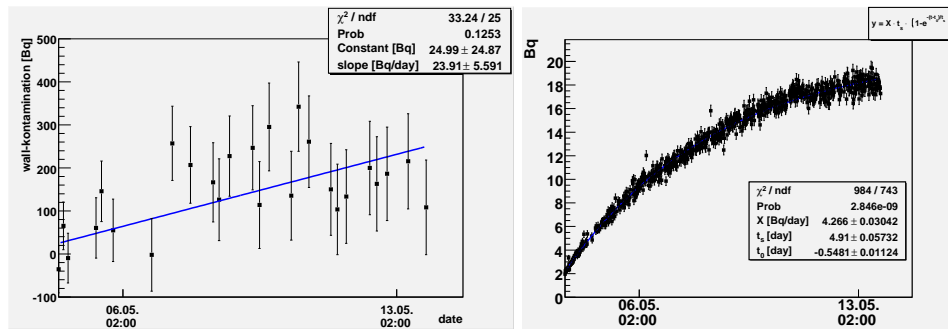
Using the representation above for the measured rates  $A_{on}$  and  $A_{off}$ , one can calculate the surface contaminations:

$$C_W = \frac{r \cdot A_{off} - A_{on}}{r \cdot a - b} \quad \text{and} \quad C_D = \frac{A_{off}}{e} - \frac{a}{e} \cdot \left[ \frac{r \cdot A_{off} - A_{on}}{r \cdot a - b} \right] \stackrel{<20\% \text{ error}}{\approx} \frac{A_{off}}{e} \quad (5)$$

The time dependence of  $C_W$  and  $C_D$  is shown in fig. 4. The contamination increase on the stainless steel walls can be due to physisorption, chemisorption and isotopic exchange reactions, where tritium is bound on the wall surface as single atoms. The contamination increase on the detector is probably exclusively due to adsorption of tritium on  $\text{SiO}_2$  and pre-condensed  $\text{H}_2\text{O}$ , since the removal of the tritium from the detector surface does not need excessive heating<sup>1</sup>. The wall adsorption rate  $Q_W$  can be calculated approximately from the slope of a linear fit to the data points  $C_W(t)$  in fig. 4 on the left using the proportionality of the number of nuclei  $N$  and their decay rate  $dN/dt$ :

$$\frac{dN}{dt} = \lambda \cdot N \quad (6)$$

Herein  $\lambda = 1.785 \cdot 10^{-9}$  1/s is the tritium decay constant. A more thorough investigation is difficult, since it is unknown to what degree physisorption, chemisorption and isotopic exchange are involved in the process of collecting tritium on the stainless steel<sup>2</sup>. In the next measurement run (run #7), where gold plated walls will be employed in the detection



**FIGURE 4.** Total tritium surface contamination during tritium inlet over time on the tube walls of the detection system (left:  $C_W$  calculated according to eq. 5) and the Si detector (right:  $C_D$  calculated according to the approximation in eq. 5). [12]

<sup>1</sup> A temperature of  $\approx 70^\circ\text{C}$  is enough to get the signal rate due to tritium contamination below the background signal rate of about 70 mHz in the energy window 6 – 20 keV.

<sup>2</sup> this depends strongly on the history of the stainless steel [13]

system and the measurement time with and without magnet will be equalized providing more data points for  $C_W$ , a more sophisticated analysis should be possible. Yet, in first approximation, one obtains  $(23.9 \pm 5.6)$  Bq/d which transforms to

$$Q_W \approx 6 \cdot 10^{-15} \text{ mbar l/s} \quad (7)$$

Assuming that only adsorption is involved in the increase of tritium contamination on the detector, we can do a more thorough analysis for  $C_D$ : If steady state is not yet reached, the effective adsorption rate of hydrogen molecules<sup>3</sup> on the detector surface is given by

$$\frac{dn}{dt} = \frac{dn_A}{dt} - \frac{dn_D}{dt}, \quad (8)$$

where  $n$  is the number of  $Q_2$  molecules adsorbed on the detector and

$$\frac{dn_A}{dt} = 3.51 \cdot 10^{22} \cdot \frac{S \cdot P \alpha}{\sqrt{MT}} \left[ \frac{\text{molecules}}{\text{s}} \right] \quad \text{and} \quad \frac{dn_D}{dt} = \frac{N}{t_s} \left[ \frac{\text{molecules}}{\text{s}} \right] \quad (9)$$

are the adsorption and desorption rates for physisorption processes [14] with the adsorption surface  $S$  [cm<sup>2</sup>], the partial pressure of the pumped gas  $P$  [Torr], the adsorbate molecular mass  $M$  [g/mol], the adsorption temperature  $T$  [K], the sojourn time  $t_s$  [s] and the sticking coefficient  $\alpha$ . One obtains a linear differential equation for the time dependence of the detector contamination  $n(t)$ , which is solved in terms of surface contamination<sup>4</sup> by:

$$C_D(t) = 5.5 \cdot 10^{13} \cdot \frac{P \alpha S}{\sqrt{MT}} \cdot t_s \cdot \left( 1 - e^{-\frac{t-t_0}{t_s}} \right) \quad (10)$$

The data shown on the right in fig. 4 can be fitted to eq. 10. If one sets  $T \approx 100$  K and  $M \approx 5$ , since DT is the most abundant isotopic mixture in the inlet gas, one obtains  $t_s = (4.24 \pm 0.05) \cdot 10^5$  s and  $P \cdot \alpha \approx 6.7 \cdot 10^{-18}$  Torr. Since  $\alpha$  is in the range from 0.1 – 1 for adsorption on surfaces with temperatures around 100 K [14], we can give the following boundaries for the DT partial pressure during tritium inlet:  $8 \cdot 10^{-18} \text{ mbar} < P < 8 \cdot 10^{-17} \text{ mbar}$ .

The adsorption rate of tritium on the detector is the first derivation of eq. 10, which is not constant over time. The maximum adsorption rate (for  $t = t_0$ ) yields an upper limit (using eq. 6 again):

$$Q_D < 8 \cdot 10^{-16} \text{ mbar l/s} \quad (11)$$

From all the contributions derived above and ignoring  $Q_B$ , we can now calculate an upper limit for the transmission probability

$$K = \frac{Q_{out}}{Q_{in}} = \frac{Q_D + Q_W + Q_G}{Q_{in}} < 6.7 \cdot 10^{-8}. \quad (12)$$

Yet, this boundary is only valid, if  $Q_B$  is negligible.

## CONCLUSION & OUTLOOK

For KATRIN the value for the transmission probability  $K$  is sufficient, especially when taken into account that the cryo surface in CPS will be ten times larger than the cryo surface in TRAP, which will increase the total pumping speed of the cryo surface and decrease  $K$ . Nonetheless, there is still the uncertainty of the amount of tritium trapped in the stainless steel bulk material of the walls ( $Q_B$ ). A further measurement run (run #7) with gold plated walls in the detection system is required to address this issue.

---

<sup>3</sup> H<sub>2</sub>, D<sub>2</sub>, T<sub>2</sub>, HD, HT, DT

<sup>4</sup>  $C_D(t) = \lambda \cdot 2n(t) \cdot 43\%$ . The pressure  $P$  was assumed to be constant during the adsorption.

## ACKNOWLEDGMENTS

The authors gratefully acknowledges support from the German Bundesministerium für Bildung und Forschung under contracts 05CK1VK1/7, 05CK1UM1/5, 05CK2PD1/5.

Our thanks also go to our colleagues who supported us during the TRAP runs: L. Anselment, J. Bohn, H. Frenzel, W. S. Gil, H. Gutzeit, S. Horn, H. Krause, M. Kuhl, M. Mark, K. Mehret, R. Neeb, H. Rapp, M. Riegel, D. Stern

In addition, the authors like to thank their colleagues from the TLK infrastructure team for supporting the TRAP experiment.

## REFERENCES

1. The KATRIN Collaboration, *KATRIN Design Report*, FZKA Scientific Reports 7090, 2004  
B. Bornschein, "The KATRIN experiment - direct measurement of neutrino masses in the sub-eV region," in *the International Workshop on Astroparticle and High Energy Physics (AHEP) 2003*, JHEP Proc. AHEP2003/064, Valencia, 2003
2. N. Bekris, C. Caldwell-Nichols, E. Hutter, *Fusion Engineering and Design* **69** (2003) 21
3. Y. Kawamura et al., *American Nuclear Society* **37** (2000) 54
4. A. Mack et al., *Fusion Engineering and Design* **61** (2002) 611
5. F. Toci et al., *Fusion Engineering and Design* **28** (1995) 373
6. R. S. Willms, "Cryogenic Adsorption of Low-Concentration Hydrogen on Charcoal, 5A Molecular Sieve, UOP S-115, ZSM-5 and Wessalith DAY," in *the 15th IEEE Symposium on Fusion Engineering-1993*, Proceedings of the 15th IEEE Symposium on Fusion Engineering, Hyannis, 1993
7. V. B. Yuferov et al., *Low Temp. Phys.* **19** (1993) 413
8. C. Benvenuti, R. S. Calder, G. Passardi, *J. Vac. Sci. Technol.* **13** (1976) 1175
9. R. A. Haefer, *Kryo-Vakuumtechnik*, Springer Verlag, 1981
10. O. Kazachenko et al., *TRAP - a Tritium Argon Frost Pump for KATRIN*, to be published
11. P. Clark Souers, *Hydrogen Properties for Fusion Energy*, University of California press, 1986
12. M. Sturm, *diploma thesis*, in preparation
13. R. Laesser, *Tritium and Helium-3 in Metals*, Springer-Verlag, 1989
14. A. Roth, *Vacuum Technology*, Elsevier Science B.V., 1990