

## H<sub>2</sub> EQUILIBRIUM PRESSURE WITH A NEG-COATED VACUUM CHAMBER AS A FUNCTION OF TEMPERATURE AND H<sub>2</sub> CONCENTRATION

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### Abstract

Non Evaporable Getter (NEG) coating is used in the Large Hadron Collider (LHC) room-temperature sections to ensure a low residual gas pressure for its properties of distributed pumping, low outgassing and desorption under particle bombardment; and to limit or cure electron cloud build-up due to its low secondary electron emission.

In certain regions of the LHC, and in particular close to the beam collimators, the temperature of the vacuum chamber is expected to rise due to energy deposition from particle losses. Hydrogen molecules are pumped by the NEG via dissociation on the surface, sorption at the superficial sites and diffusion into the NEG bulk. In the case of hydrogen, the sorption is thermally reversible, causing the dissociation pressure to increase with NEG temperature and amount of H<sub>2</sub> pumped.

Measurements were carried out on a stainless steel chamber coated with TiZrV NEG as a function of the H<sub>2</sub> concentration and the chamber temperature, to estimate the residual gas pressure in the collimator regions for various LHC operation scenarios, corresponding to different particle loss rates and times between NEG regenerations. The results are presented in this paper and discussed.

### INTRODUCTION

The beam vacuum chambers in the Large Hadron Collider (LHC) ambient temperature sections are coated with TiZrV Non Evaporable Getter (NEG) [1, 2]. After activation, gas molecules, like H<sub>2</sub>, CO and CO<sub>2</sub>, are chemically pumped on the NEG surface. In the case of hydrogen, the dissociated molecule can diffuse into the bulk. The sorption is thermally reversible, causing the residual pressure to increase with NEG temperature and amount of H<sub>2</sub> pumped, i.e. its concentration into the NEG layer. The hydrogen equilibrium pressure is known to follow Sievert's law.

In the collimation regions, particle losses are expected to induce a temperature raise of about 150°C in the neighbouring vacuum chambers. Works reporting on thermal properties of the single transition elements and some of their alloys have been carried out for they are widely used as nuclear materials of light water and heavy water reactors, and as tritium storage in nuclear fusion devices [3-12].

In this paper measurements to evaluate the Sievert's law parameters for the TiZrV alloy are described and discussed. These measurements will allow forecasting the contribution to the residual gas pressure due to the increased temperature in the collimator regions and establish if any additional pumping or cooling is required.

### EXPERIMENTAL SETUP

Hydrogen pressure measurements were performed using a stainless steel chamber, 1.2 m long and 100 mm inner diameter, deposited with 1.5 µm Ti(30)Zr(30)V(40) and provided with two Bayard Alpert gauges (BA), one Balzers QMG112 residual gas analyser (RGA), an injection and evacuation system.

Bayard Alpert gauges are directly connected to the NEG coated chamber at a distance of 1 m one from each other. The injection system is designed to quantify the amount of hydrogen injected and therefore to evaluate the variation of hydrogen concentration in the NEG layer. The pumping system is composed of two turbo-molecular pumps (TMP), one used for the system evacuation and conditioning, and the other used to pump (via a limited conductance of 1 l/s) during measurements gas species which are not pumped by the NEG.

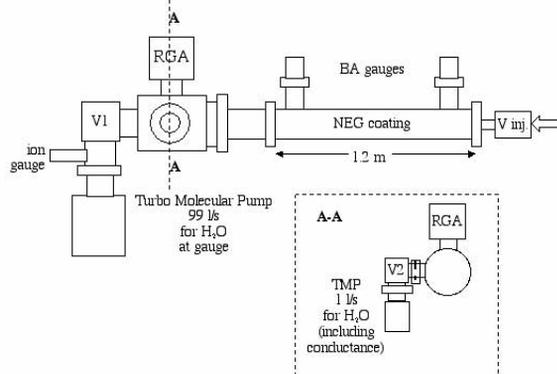


Figure 1: Layout of the experimental setup.

TiZrV is deposited on the chamber by magnetron sputtering with krypton as discharge gas. The cathode is obtained inter-twisting three Ti, Zr and V wires of 3mm diameter [1, 2]. After the coating, the chamber is kept under nitrogen atmosphere and opened to air only during the assembly of the measuring system.

### EXPERIMENTAL PROCEDURE

Before starting the measurements, the NEG chamber was activated. All uncoated parts were baked before hand to limit any contamination to the NEG while the NEG chamber was kept at 80°C; after instrumentation degassing, the NEG was brought to 250°C for 18 h for the activation. At 250°C, 2 to 4 hours would be sufficient to activate the NEG. This duration was chosen to perform the whole conditioning during working hours, and be present during temperature ramp-up and cool-down, when the risk of leaks opening is the greatest. The

instrumentation was degassed a second time at the end of the activation plateau.

The hydrogen equilibrium pressure was measured increasing the NEG temperature in steps of 50°C from 100°C to 250°C, the maximum temperature being limited by the maximum pressure measurable with BA type gauges. The temperature was kept constant for the time necessary to reach equilibrium and to allow taking measurements (with typical plateaus of 2 hours). During these measurements the rest of the system was at ambient temperature. After each temperature cycle the NEG chamber was cooled down to ambient until the initial base pressure was recovered. The first temperature cycle was carried out just after NEG activation (i.e. before any additional hydrogen injection) to be able to determine the initial hydrogen content.

The hydrogen concentration,  $c_H$  (in atomic ratio, H/NEG) was varied by injecting known quantities of hydrogen in the system and waiting until the system base pressure was recovered (i.e. until all the hydrogen injected was pumped into the NEG coating). During this time the whole system was at ambient temperature and isolated (V1 and V2 closed) from the pumps. The maximum pressure measurable with BA type gauges in the temperature range of interest limited the maximum hydrogen concentration.

During the temperature cycles, a small fraction of the krypton implanted into the NEG layer during the coating (estimated to about 1 to 10 ppm depending on the deposition parameters) was released. Helium and argon (coming from the hydrogen bottle) and methane were also observed. Neither noble gases nor hydrocarbons are pumped by the NEG, and accumulate in the experimental set up, while hydrogen is pumped and released. Due to the conductance in between the RGA and the NEG coated chamber, the gas proportions estimated at the location of the RGA are not representative of the real composition inside the NEG chamber. To overcome this problem, valve V2 was open to pump via a conductance of 1 l/s, after making sure that the amount of hydrogen pumped was negligible, limiting to < 2% the gas species other than hydrogen. This operation was not carried out for the measurements at 100 °C (except for the highest hydrogen concentration), when the pressure inside the NEG chamber was of the same order or better than behind V2. The data taken at 100 °C were not used for the data fitting.

In one case ( $c_H=1.5E-02$  H/NEG) the measurements were repeated twice. The pressure variation was found to be ~ 50%.

### RESULTS AND DISCUSSION

All data for hydrogen solubility in pure metals and solid solution have been found to obey Sievert's law, which reads:

$$c_H = K_S \sqrt{P_{H_2}}$$

where

$$\ln K_S = \left( A + \frac{B}{T} \right) B = -\frac{\Delta H}{R}$$

$c_H$  is the hydrogen content in atom ratio (concentration),  $P_{H_2}$  is the hydrogen equilibrium pressure,  $\Delta H$  is the enthalpy of solution,  $R$  is the molar gas constant and  $A$  depends on the excess entropy of solution. The equation parameters are constant over the solubility range.

The hydrogen equilibrium pressure is plotted as a function of the temperature, at constant hydrogen concentration in Fig. 2 and as a function of the hydrogen concentration, at constant temperature in Fig. 3. The symbols correspond to the measured data and the lines represent the calculated Sievert's curves. Each of the points shown in the figures were obtained averaging the measurements taken during the time when valve V2 was open, to minimise errors, and subtracting the base hydrogen pressure measured at ambient temperature, just before each temperature cycle.

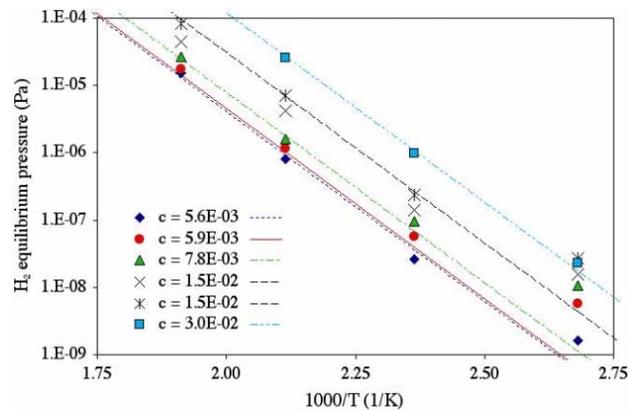


Figure 2: Hydrogen equilibrium pressure measured at constant hydrogen content as a function of NEG temperature.

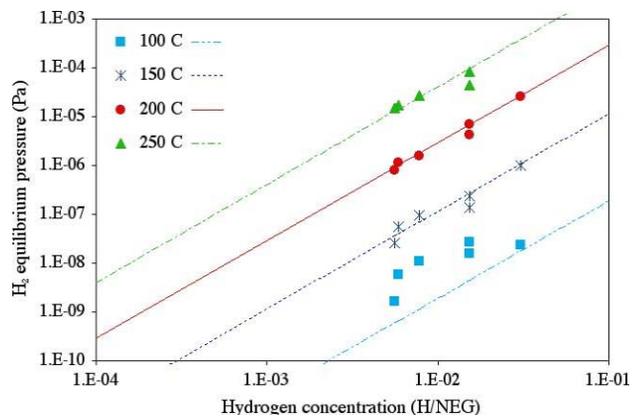


Figure 3: Hydrogen equilibrium pressure at constant temperature as a function of NEG hydrogen content.

The Sievert's law constants and the initial concentration were evaluated using the least square fit method:  $(c_H)_0 = 5.5E-03$  H/NEG,  $\Delta H = -54.0$  kJ/mol,  $A = -12.0$ .

These values are compared in Table 1 to those found in literature [3-13].

Table 1: Data comparison

	$\Delta H$ (kJ/mol)	A	T (°C)	
TiZrV	-54.0	-12.0	100 to 250	<b>present study</b>
TiZrV	-81.0	-13.9	100 to 250	references [13]
Ti	-52.7 to -44.5	-13.1 to -11.6	500 to 1200	references [3-12]
Zr	-64 to -37.8	-11.8 to -9.3	500 to 1200	references [3-12]
V	-32.5 to -28.1	-13.1	500 to 1200	references [3-12]
Zr alloys	-30.6 to -42.3	-12.3 to -11.4	100 to 500	references [3] and [12]

The values in [4-12 and referenced in 10] are relative to elemental metals (Ti, Zr or V). The temperature range (between 500 and 1200°C) also differs from the present study. The film grain size for the materials used in [3-12] is of the order of the micrometer, i.e. larger than in sputtered NEG (where in order to increase the pumping speed and capacity the total NEG bulk and surface area is maximised, and the grain size is about 3 to 5 nm [13]). Zr alloys [4, 12] exhibit lower values compared to pure Zr. Studies on the influence of oxygen content have not given conclusive answers.

The enthalpy of solution estimated in the present study falls in the range of elemental Zr and close to the values for elemental Ti.

The constant A is within the data spread. A is independent of the bulk structure since it is related to the variation of entropy between the gas phase and the solid solution, the latter being negligible compared to the first.

## CONCLUSIONS

The thermal hydrogen equilibrium pressure in a TiZrV NEG coated chamber has been measured and the Sievert's law constants determined. The enthalpy of solution compares well with other data found in literature and so does the Sievert's constant A.

The present study was carried out to try and estimate the pressure close to the LHC collimators in the case of particle losses causing the vacuum chamber temperature to rise to about 150°C. The maximum tolerable hydrogen pressure in these regions is  $10^{-5}$  Pa. This limit would be attained for a hydrogen concentration of the order of 0.1 H/NEG, which is one order of magnitude higher than what is expected to accumulate in the NEG coating even after several years of operations. It is concluded that the temperature rise should not constitute a problem to the functioning of the LHC.

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## REFERENCES

- [1] C. Benvenuti et al., Vacuum 60 (2001) 57-65.
- [2] C. Benvenuti et al., J. Vac. Sci. Technol. A 16(1) Jan/Feb 1998.
- [3] P. Vizcaino et al., Thermochemica Acta 429 (2005) 7-11.
- [4] P. Vizcaino et al., Metal Mat. Trans. A 35(8) (2004) 2343.
- [5] M. Steinbrück et al., J. Nuc. Mat. 334 (2004) 58-64.
- [6] G. Andersson et al., J. Alloys Comp. 334 (2002) 14-19.
- [7] S. Yamanaka et al., J. Alloys Comp. 231 (1995) 503-507
- [8] S. Yamanaka et al., J. Nuc. Mat. 247 (1997) 224-248.
- [9] S. Yamanaka et al., J. Nuc. Mat. 270 (1999) 233-241.
- [10] S. Yamanaka et al., J. Alloys Comp. 293-295 (1999) 38-41.
- [11] S. Yamanaka et al., J. Alloys Comp. 293-295 (1999) 42-51.
- [12] D. Setoyama et al., J. Nuc. Mat. 344 (2005) 291-294.
- [13] P. Chiggiato and P. Costa-Pinto, Thin Solid Films, Articles in Press.