Interaction of Gases and Coating

Hydrogen Saturation Study of TiZrV Non-Evaporable Getter Coating for Fixed Target Operation at LHCb

Fixed target experiments at LHC - STRONG-2020 workshop

5-7 January, 2023
Aussois, Vanoise Massif, France

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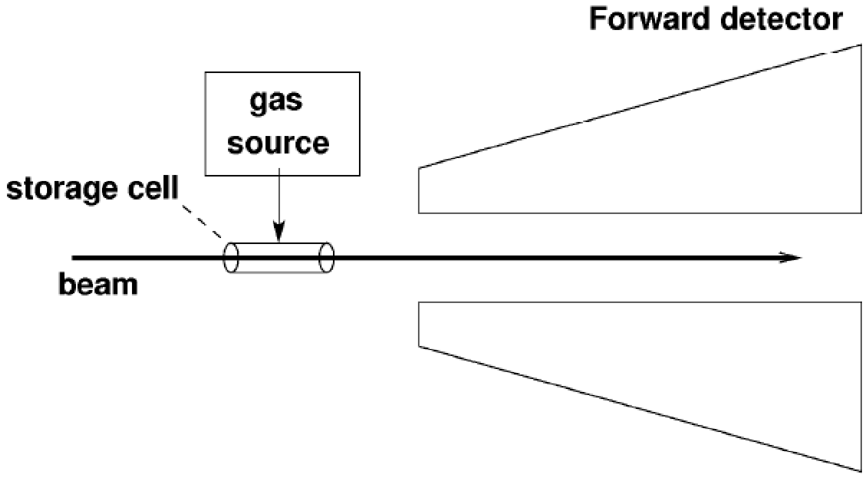
Content

Introduction

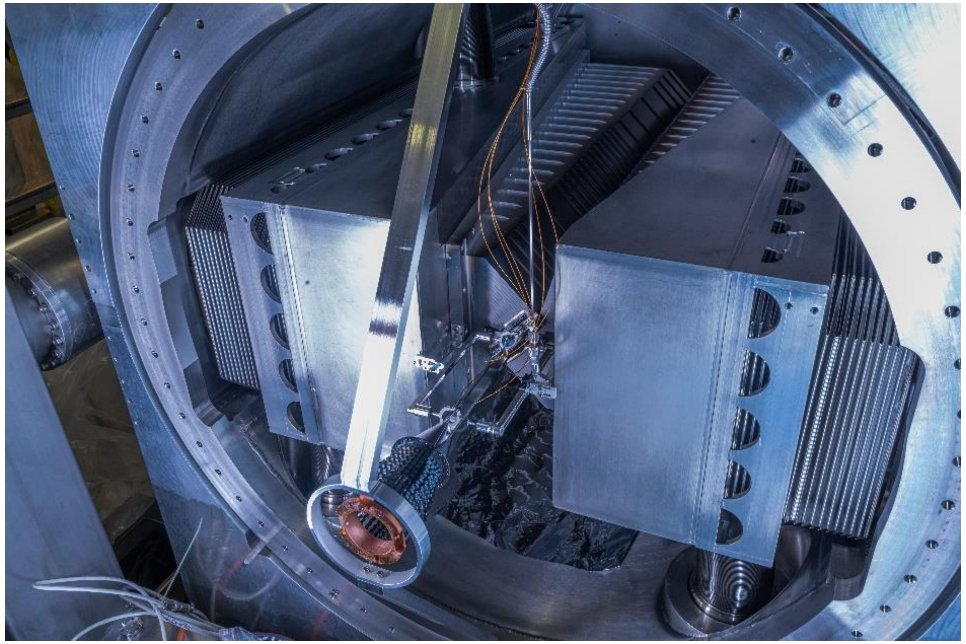
SMOG2 upgrade at LHCb

- Gas injection system for fixed target experiments
  - He, Ne, Ar, Kr, Xe, H_2, D_2, O_2, N_2



Schematic of the LHCb fixed target set-up with unpolarized gaseous target

Source: Barschel, Colin, et al. *LHC fixed target experiments: Report from the LHC Fixed Target Working Group of the CERN Physics Beyond Colliders Forum*. Diss. CERN Yellow Reports: Monographs Published by CERN, 2020.



SMOG2 installation in the LHCb cavern

Source: https://cds.cern.ch/record/2727007?ln=en

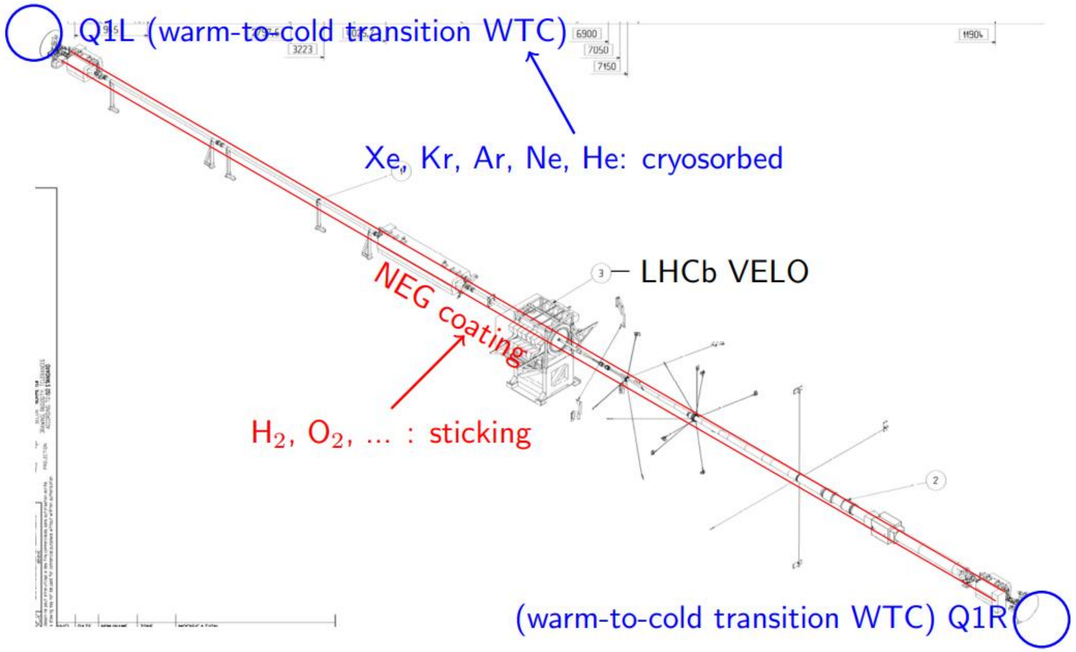
Introduction

SMOG2 upgrade at LHCb

- Gas injection system for fixed target experiments
  - He, Ne, Ar, Kr, Xe, **H_2**, D_2, O_2, N_2

Non-evaporable getter (NEG) coating

- Chemical pump for distributed pumping
- Low secondary electron yield (SEY) coating



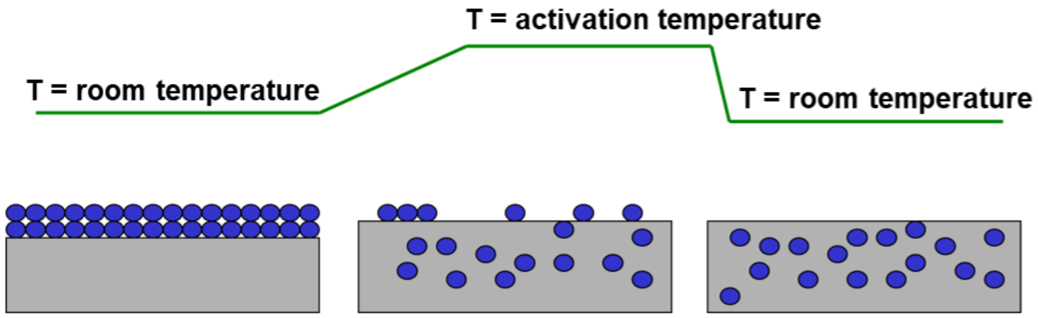
Technical drawing of LHCb vacuum system with NEG coated section highlighted

Source: Massimiliano Ferro-Luzzi: Fixed targets at the LHC: proposals and challenges
https://indico.cern.ch/event/1222068/contributions/5178127/attachments/2571819/4434293/MFL_LHCFixT_Aussois_5jan2023.pdf

Introduction

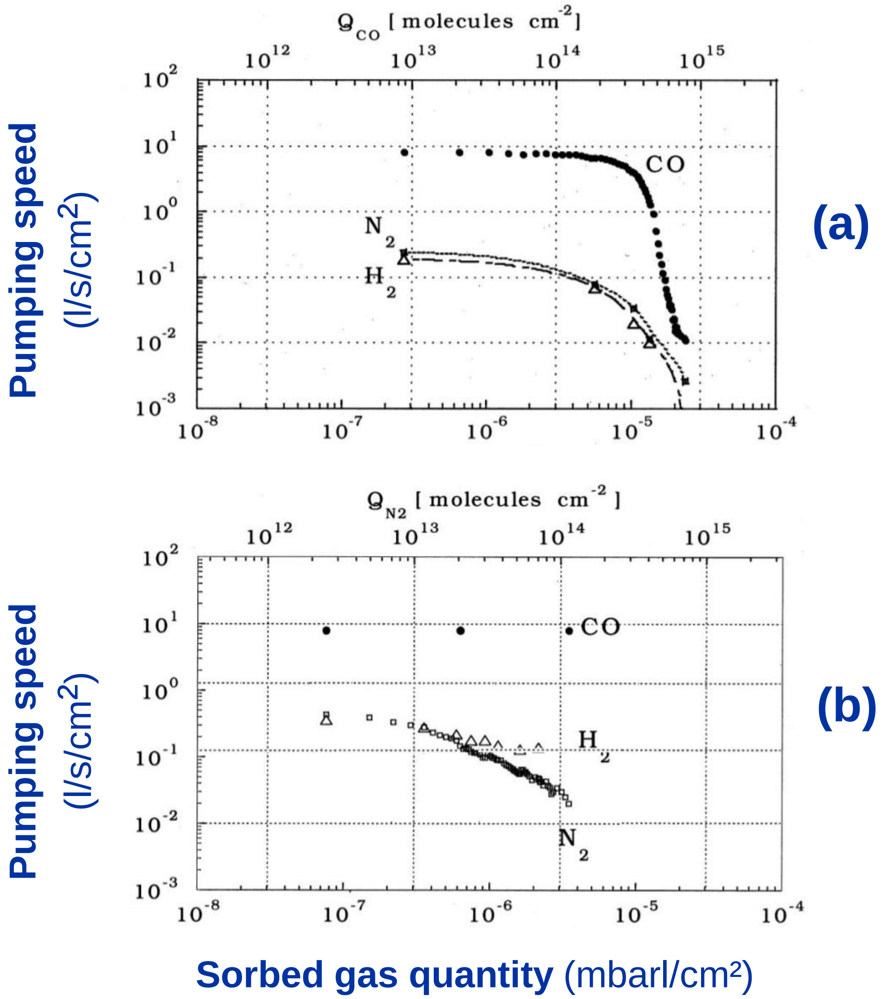
NEG coating operation

- Activation
- Sorption
    - H_2O, O_2, N_2, CO_2, CO: chemisorption
    - **H_2: dissociation and diffusion into bulk**
    - Noble gases and hydrocarbons: inert
- Saturation



Schematic of native oxide dissolution of getter during activation

Source: Costa-Pinto, P. Vacuum chambers for LHC LSS. No. TS-Note-2004-009. CERN-TS-Note-2004-009, 2004.



Pumping speed decrease of TiZrV coating during saturation with CO (a) and N_2 (b)

Source: Benvenuti, C., et al. "Influence of the elemental composition and crystal structure on the vacuum properties of Ti–Zr–V nonevaporable getter films." Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films 19.6 (2001): 2925-2930.

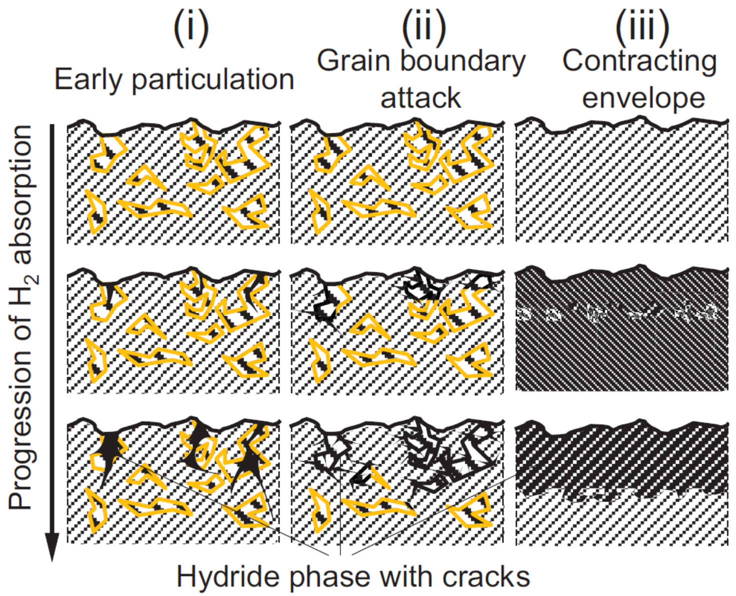
Introduction

NEG coating and H_2 interaction

- Excessive H_2 sorption can lead to decreased H_2 sticking coefficient, or in severe cases embrittlement of the layer potentially leading to peel-off of the thin film

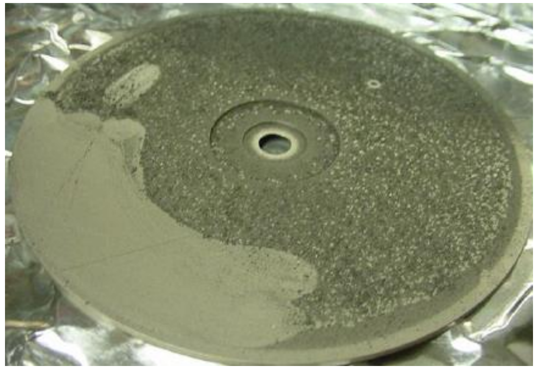
- Effect of H_2 sorption on aging

- Effect of H_2 sorption on SEY



Hydride phase and consequent crack formation mechanisms in gettering materials

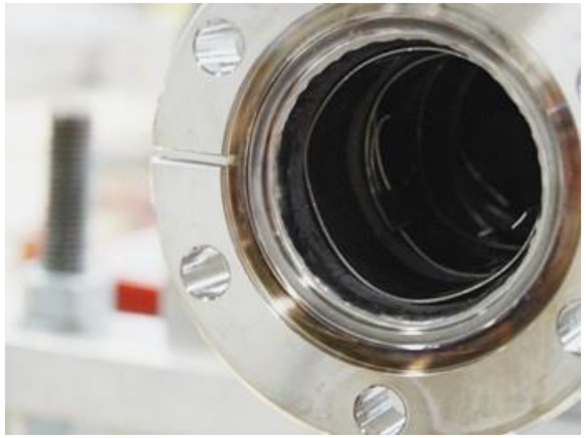
Source: Modi, Poojan, and Kondo-Francois Aguey-Zinsou. "Room temperature metal hydrides for stationary and heat storage applications: A review." Frontiers in Energy Research 9 (2021): 128.



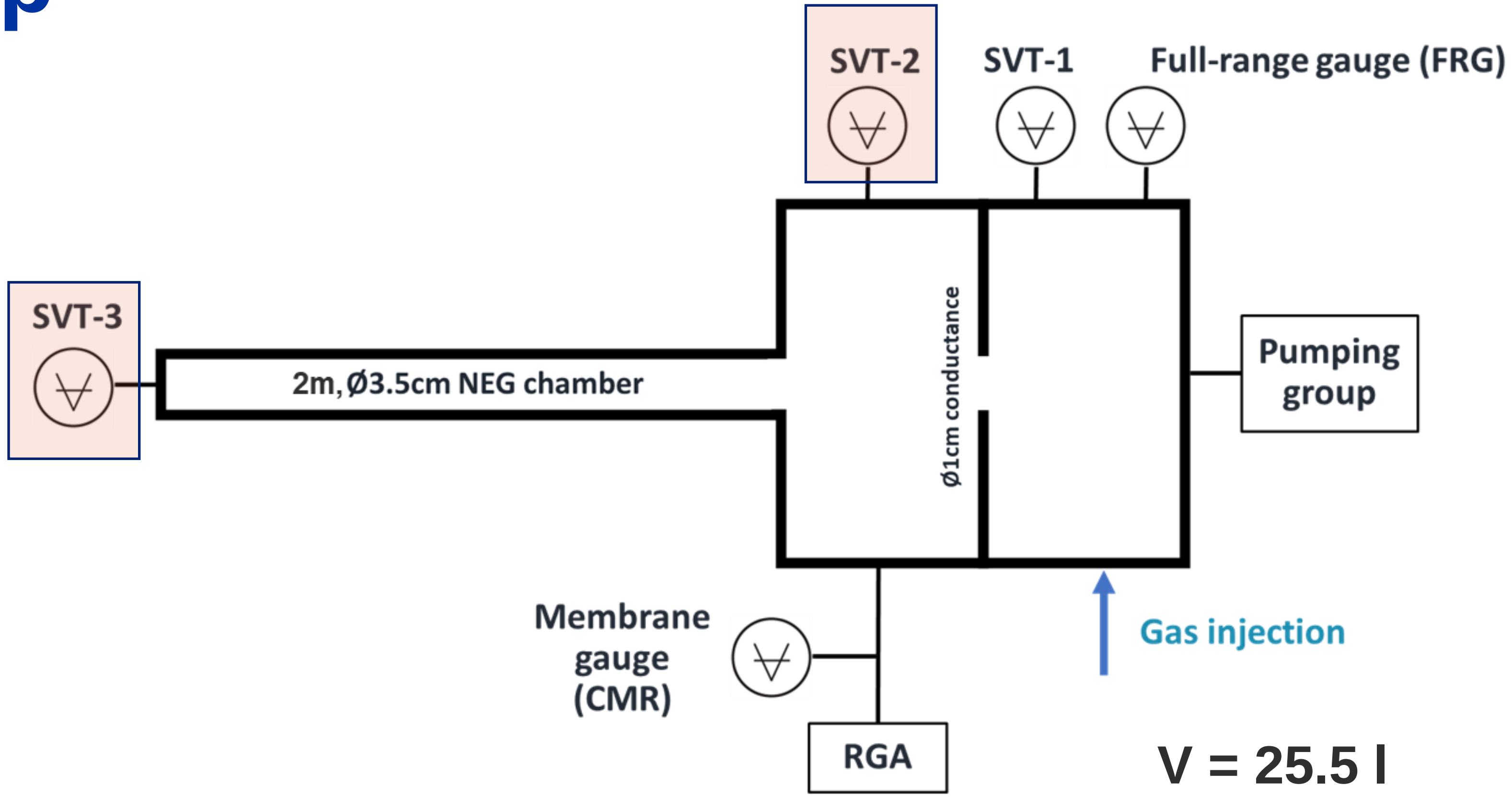
Getter powder peel-off after extensive H_2 gas sorption

Source: Li, Yulin, and Xianghong Liu. "Vacuum Science and Technology for Accelerator Vacuum Systems." Cornell University, Ithaca, NY (2013).

Experimental set-up



Witness samples placed at the entrance of the pipe



V = 25.5 l

2 m long NEG coated DN40 304L stainless steel pipes:

- 1.24452 g and 1.73885 g (0.02161 mol and 0.03021 mol) NEG coating
- 2200 cm² surface area

Experimental procedure

H_2 saturation experiments at 2 injection pressure conditions under vacuum at room temperature on TiZrV coated test pipes

1. **High pressure (HP) experiment**
   - 6 cycles involving instantaneous H_2 injection at 1 mbar
2. **Low pressure (LP) experiment**
   - 1 cycle involving continuous H_2 injection at $5 \cdot 10^{-7}$ mbar

→ Vacuum performance characterization in vacuum test bench

→ Materials characterization of witness samples

Experimental procedure: HP experiment

1. Bake-out and activation (230 °C for 24 h)
2. H_2 sticking coefficient measurement
3. High pressure H_2 injection (1 mbar)
   – afterwards: static vacuum (2 h) + 24 h pumping
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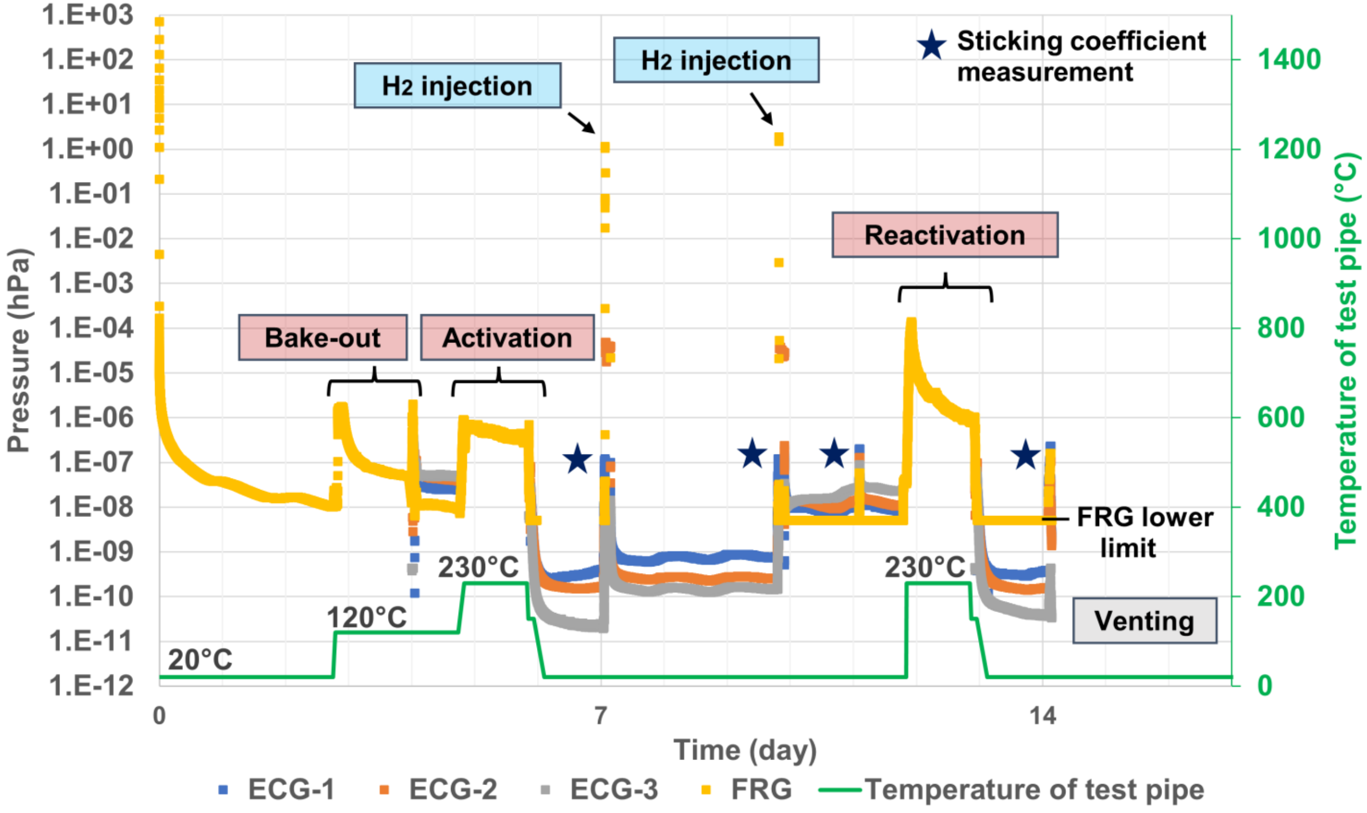
6. (H_2 sticking coefficient measurement)
7. Reactivation (230 °C for 24 h)
8. H_2 sticking coefficient measurement
9. Venting, witness sample retrieval, and visual inspection

→ **repeated 6 times**



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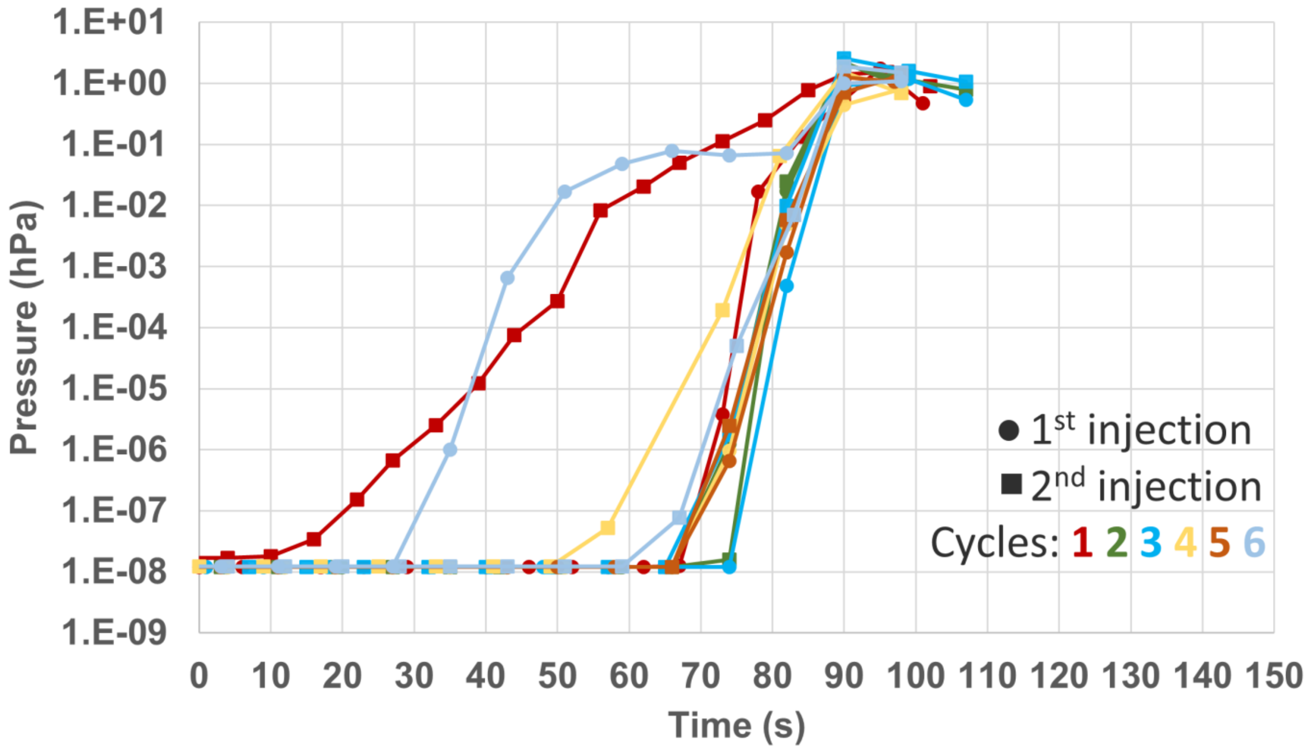

Experimental procedure: LP experiment

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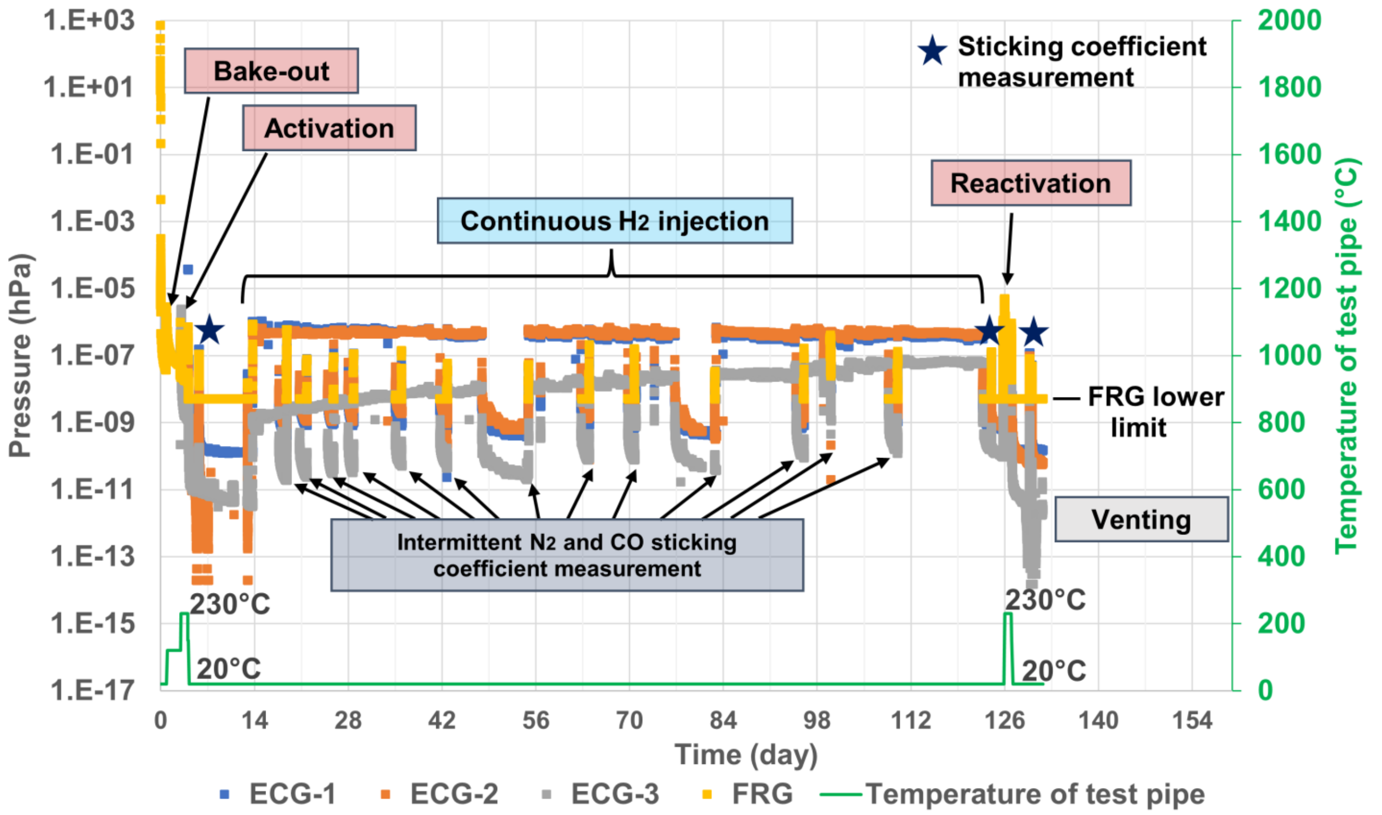
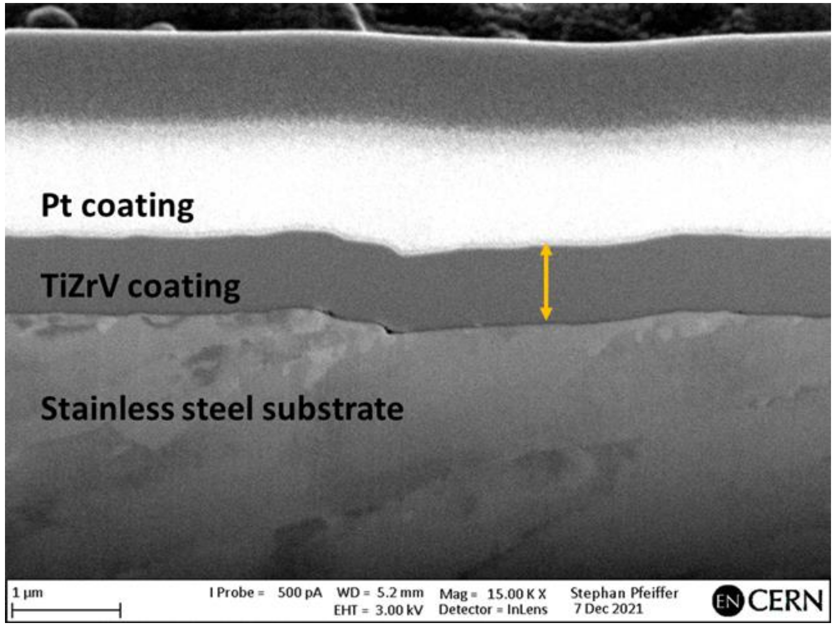
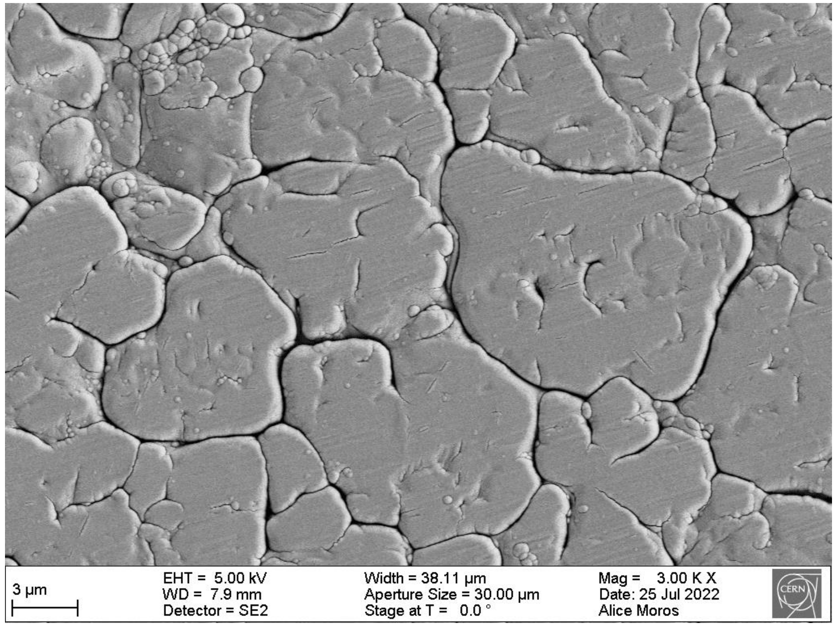


As-deposited samples

FIB cross-section, SEM top view, and XRF composition

	Thickness (µm)	Ti (at%)	Zr (at%)	V (at%)
LP-0	1.4±0.4	33.2	19.0	47.8
HP-0	1.0±0.4	29.0	16.4	54.6





High pressure experiment

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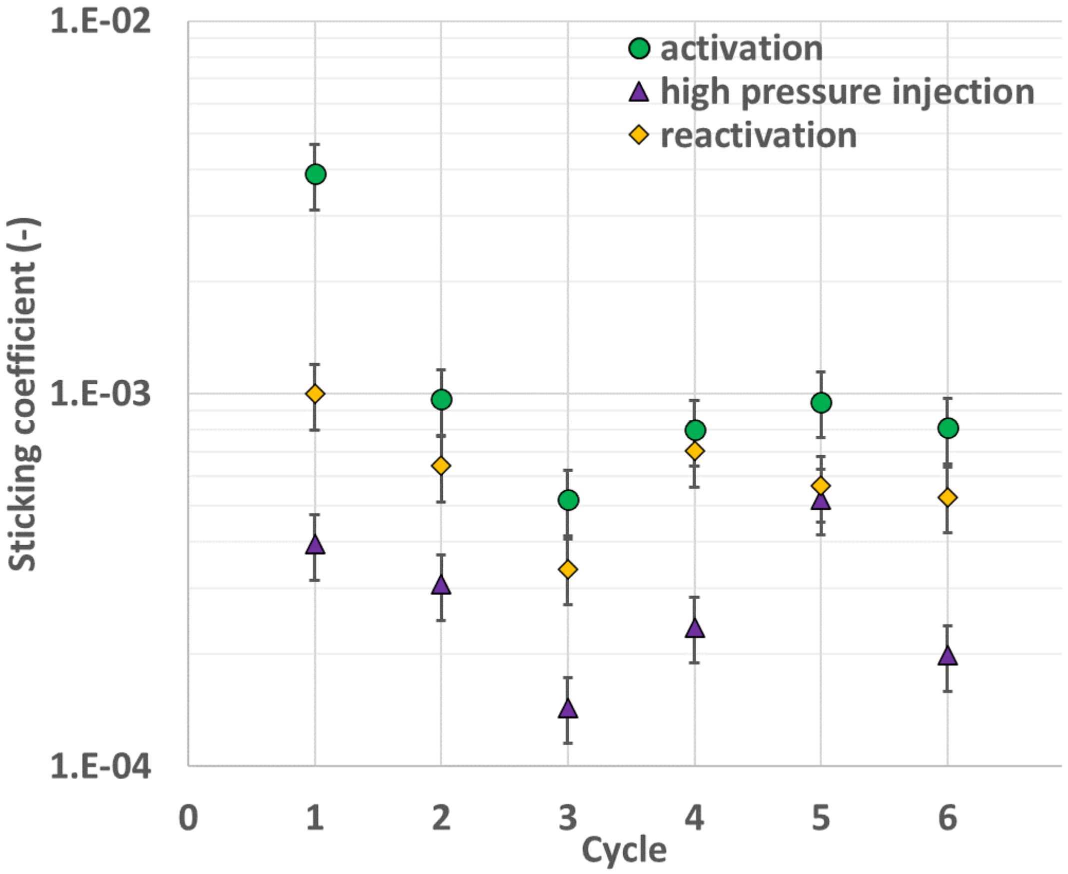

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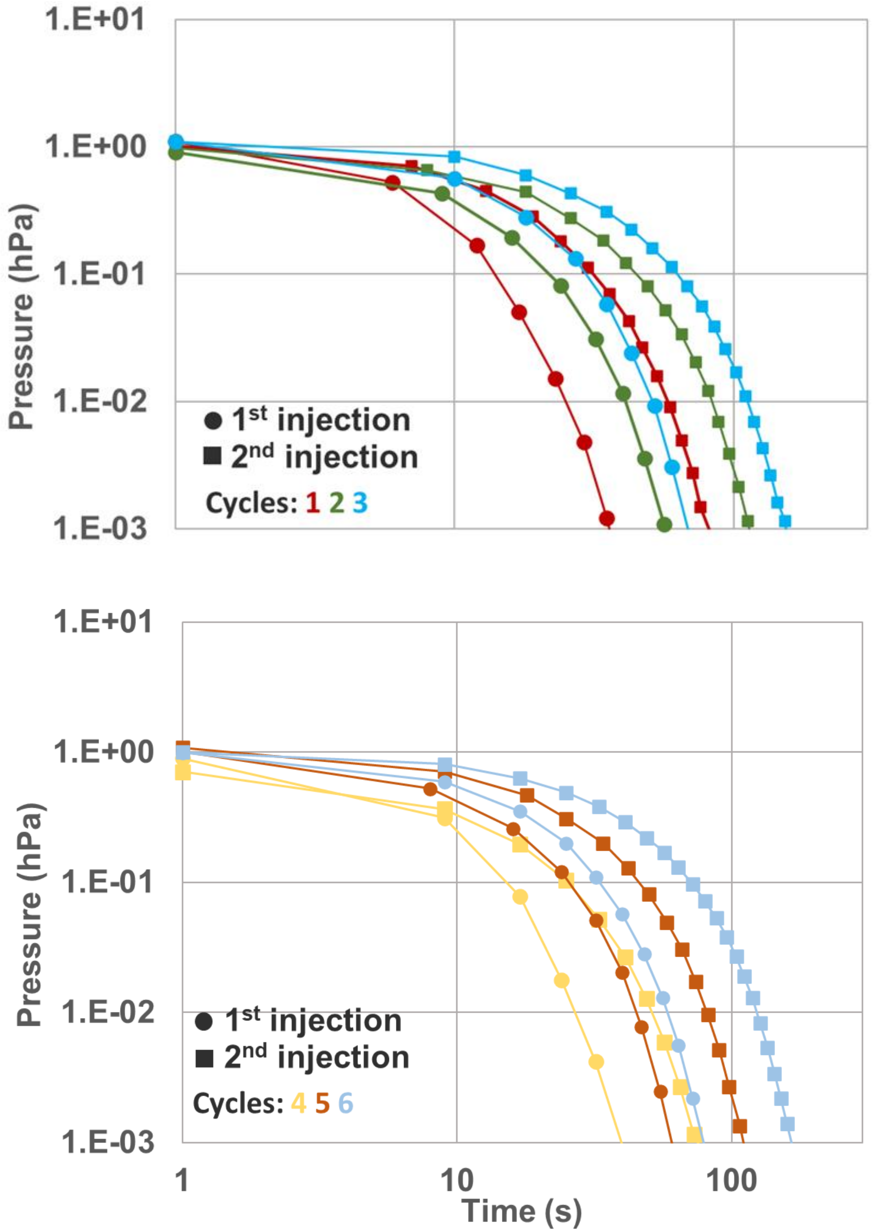

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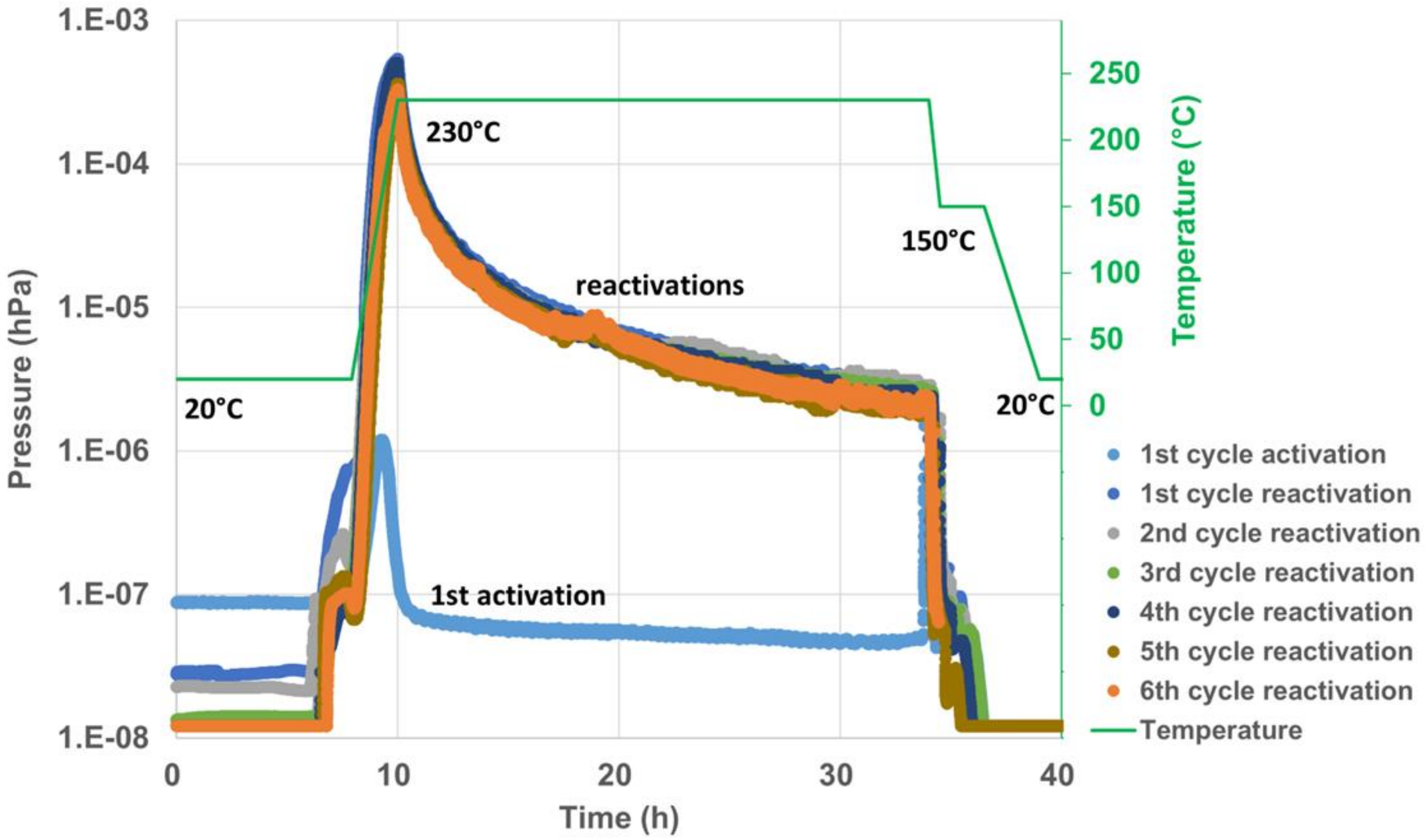
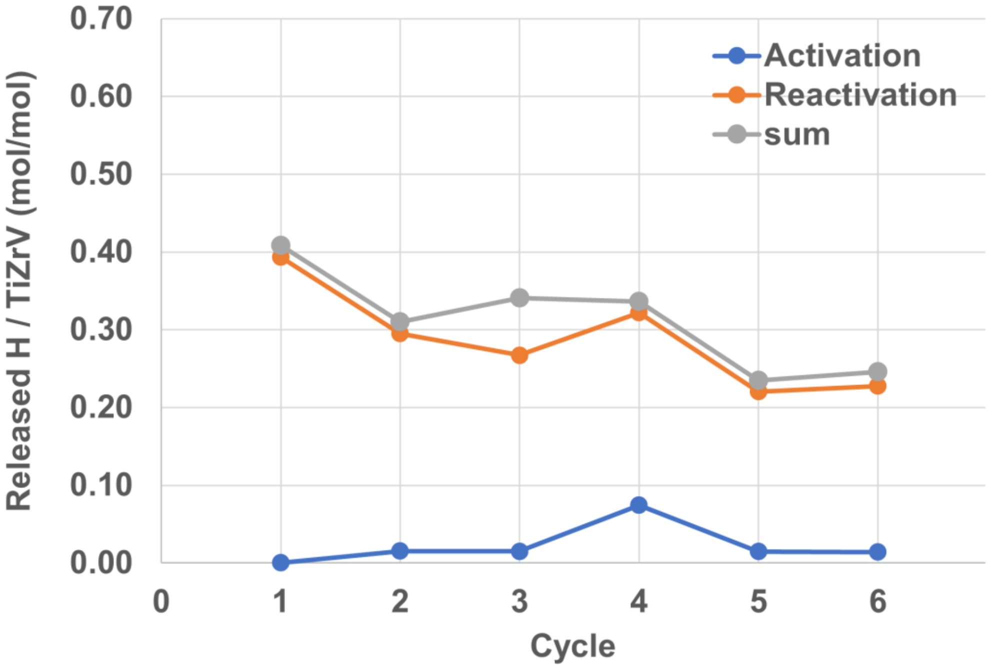


$$p_{H_2} = 2 \ln x_H + 14.2 - \frac{8468}{T}$$

High pressure experiment

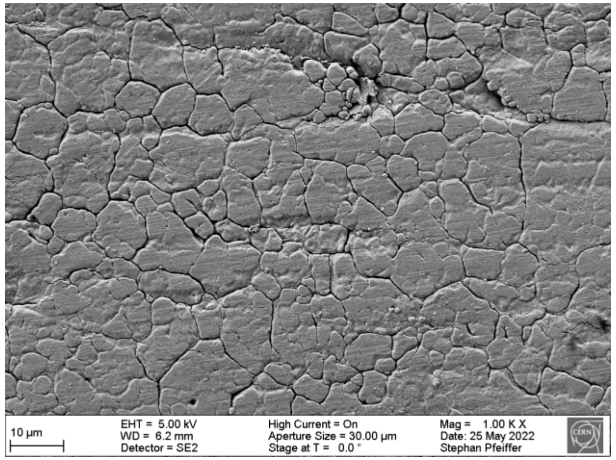
Calculated released hydrogen quantity



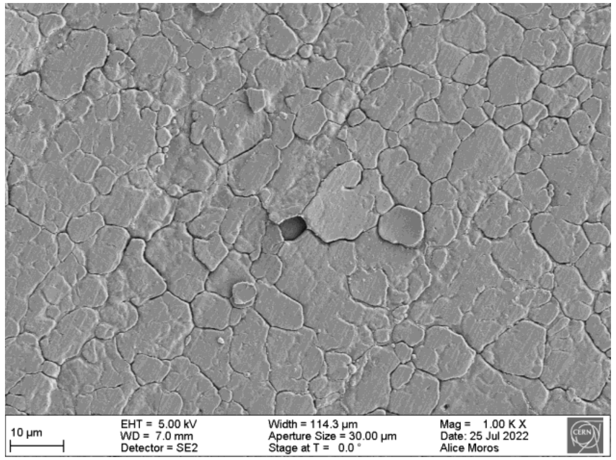
Released hydrogen from NEG coating during (re)activations

High pressure experiment

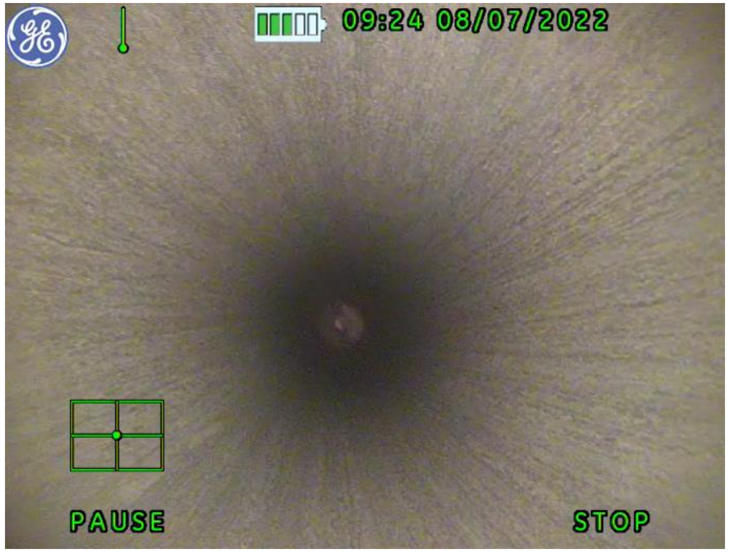
SEM and endoscopy



Top SEM image of HP-0 as-deposited sample



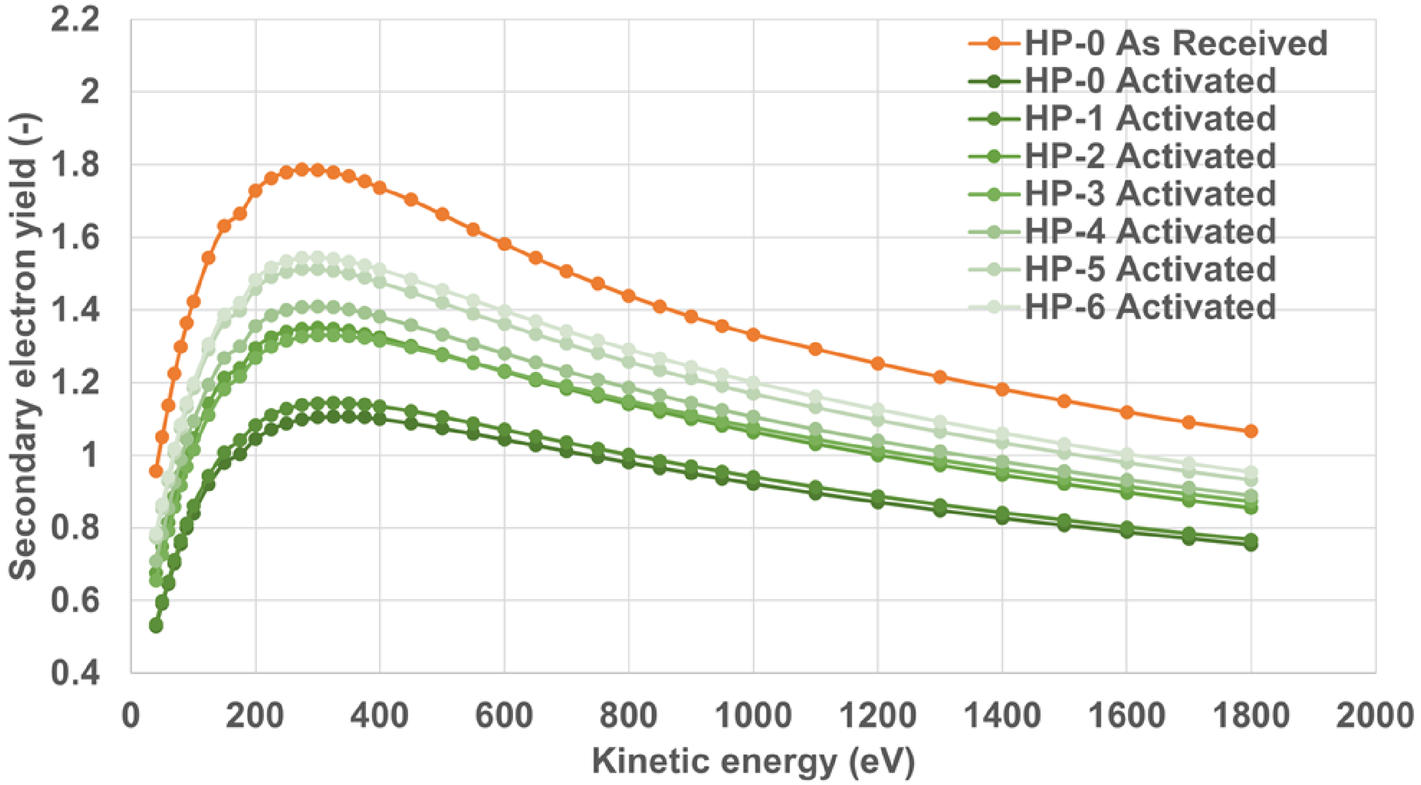
Top SEM image of HP-6 witness sample



Endoscopic view of the TiZrV coated test pipe after the 6th cycle

High pressure experiment

SEY

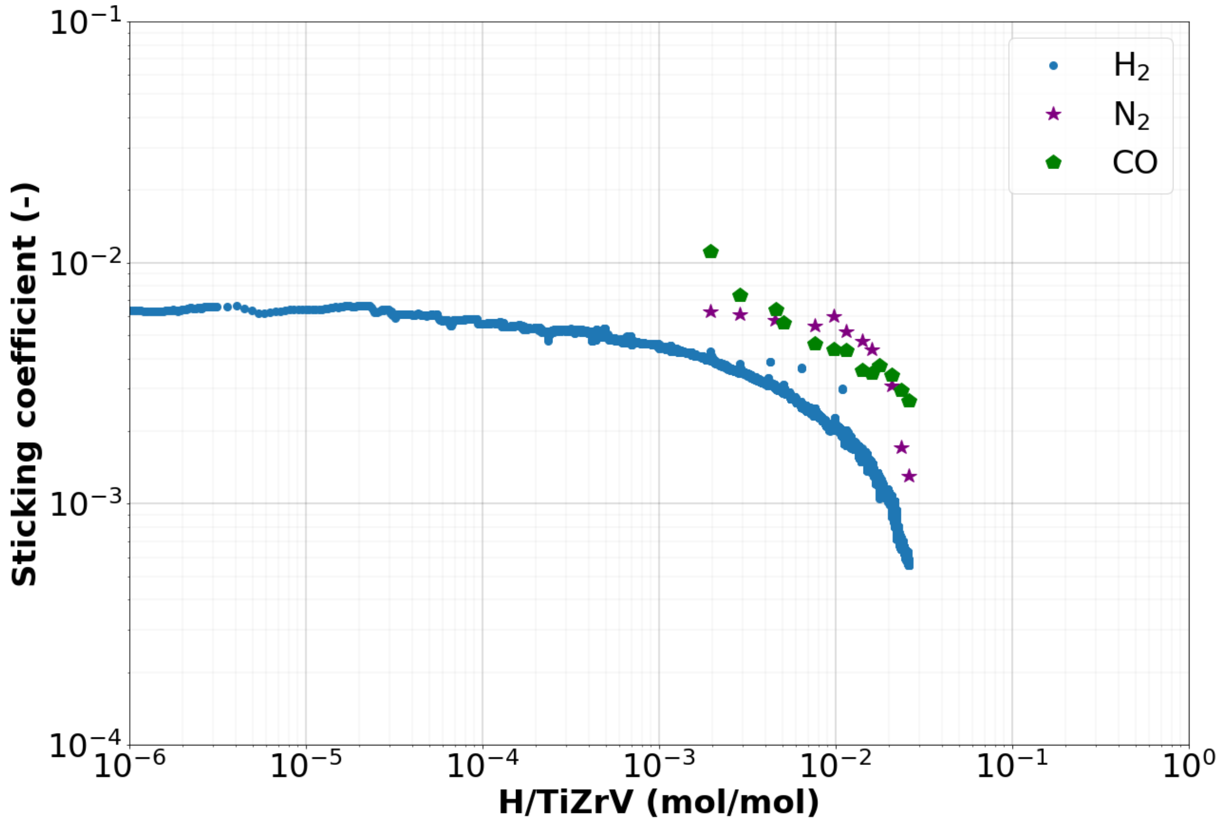


Secondary electron yield of witness samples after 1 h 250 °C in-situ activation

Low pressure experiment

Sticking coefficient evolution

- H_2, N_2, and CO sticking coefficient evolution as a function of sorbed hydrogen quantity



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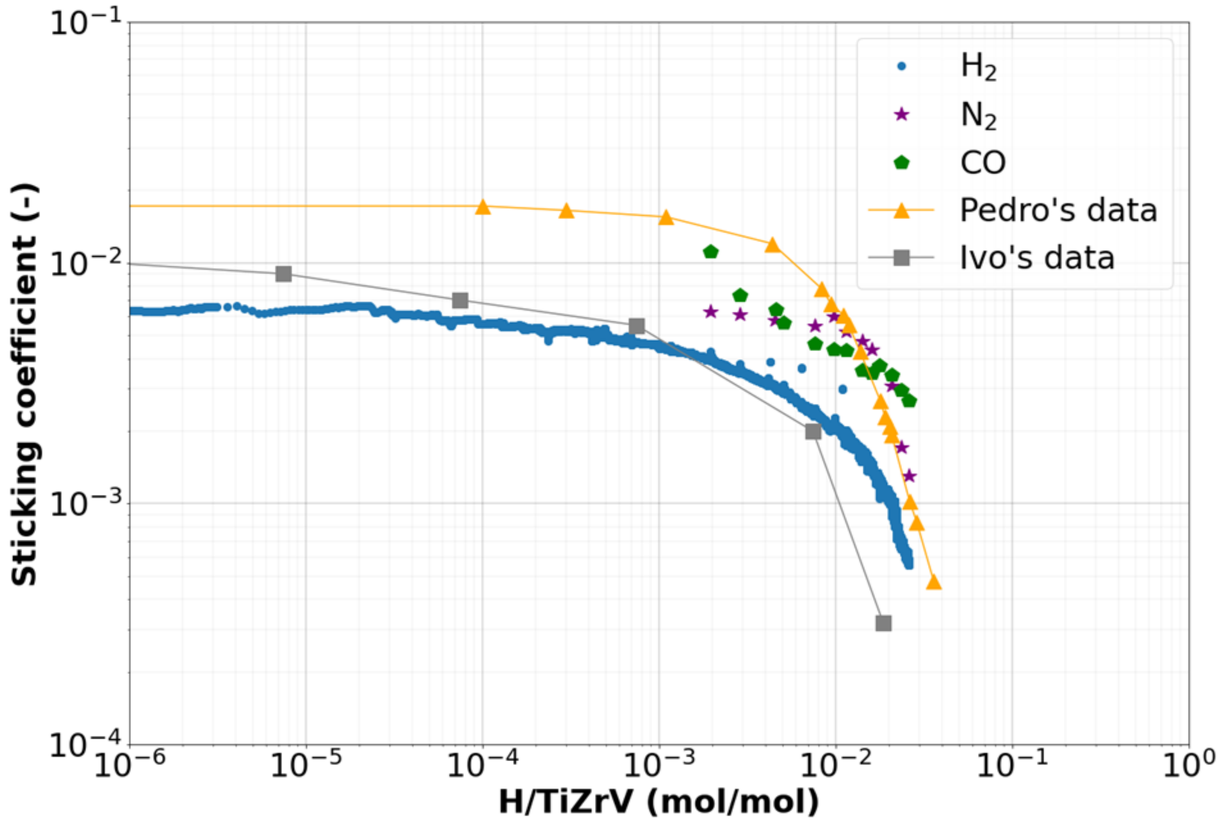

Low pressure experiment

1. Bake-out and activation (230 °C for 24 h)
2. **Sticking coefficient measurement** (H_2, N_2, and CO)
3. Continuous H_2 injection ($5 \cdot 10^{-7}$ mbar)
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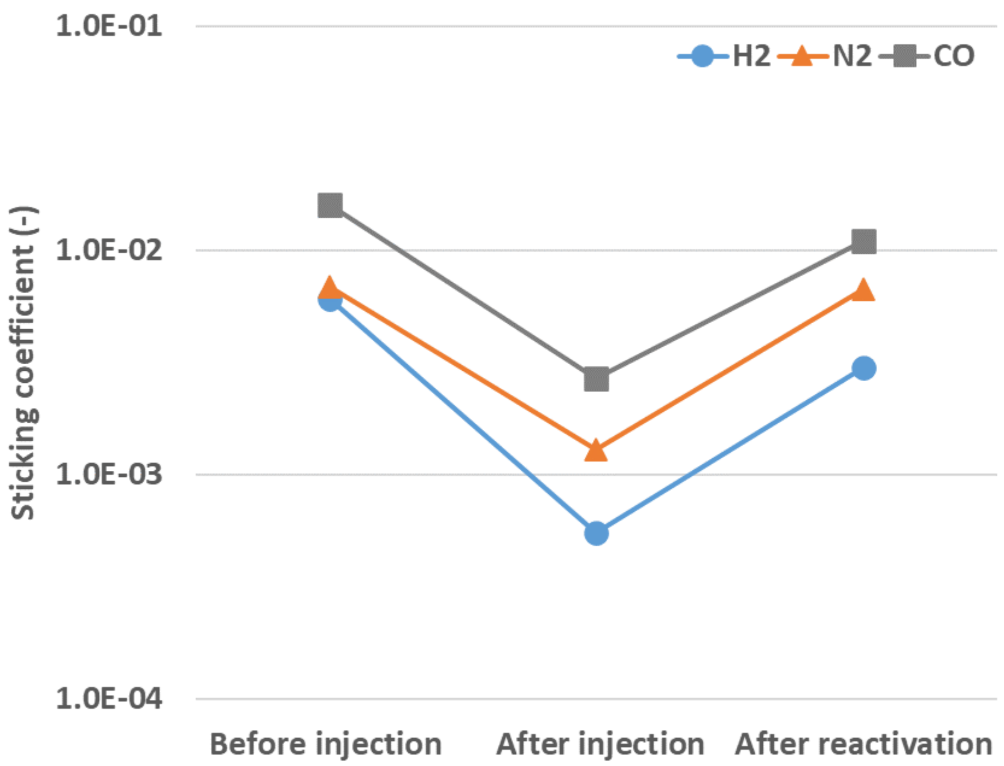
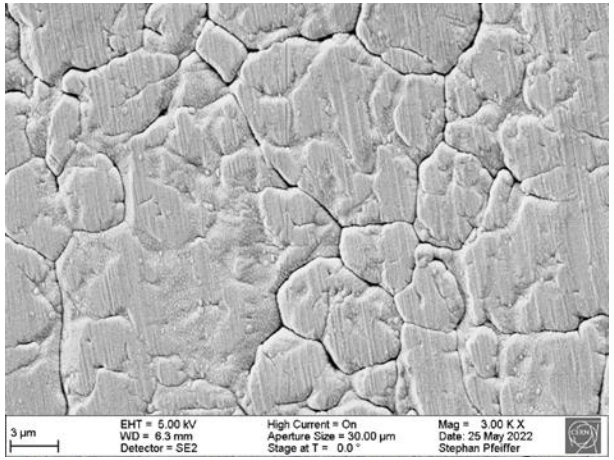
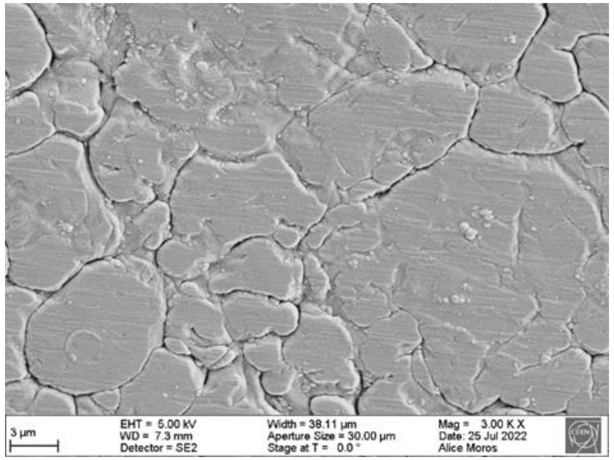


Low pressure experiment

SEM and endoscopy



Top SEM image of LP-0 as-deposited sample



Top SEM image of LP-1 witness sample

Endoscopic view of the TiZrV coated test pipe after the LP cycle

Conclusion

- 6 cycles of high pressure (1 mbar) H_2 injections with 230 °C and 24 h activations
- 1 cycle of low pressure ($5 \cdot 10^{-7}$ mbar) H_2 injection with 230 °C and 24 h activation

→ Absorption of more than 0.20 H / NEG possible repeatedly without observing visual signs of peel-off in the beam pipe at 1 mbar injection pressure

→ Saturation above 0.01 H / NEG observed at $5 \cdot 10^{-7}$ mbar injection pressure

→ Sticking coefficients decrease with increasing sorbed hydrogen quantity

→ Sticking coefficient is only partially regainable with the 230 °C and 24 h activation

- Witness sample peel-off / coating delamination needs further investigation using flat samples
- Effect of “aging” needs further investigation, as progressing in the high pressure cycles, after 1 h in-situ activation at 250°C:
  - Decrease of O dissolution and increase of SEY
  - Lower carbon-to-carbide conversion
  - Lower degree of metal reduction

Acknowledgements

Supervision: Giuseppe Bregliozzi, Josef Sestak and Ivo Wevers

Coating deposition: Yorick Delaup and Pedro Costa Pinto

Materials characterization and analysis: Ahmed Cherif, Mickael Denis Crouvizier, Alice Moros, Stephan Pfeiffer, Martino Rimoldi

Lab activities: Rowan Cape Hill-James, Jerome Gilles Chaure, Cesar Vazquez Pelaez, Orlando Santos

the **Physics Beyond Colliders – Fixed Target working group** for providing support for this work

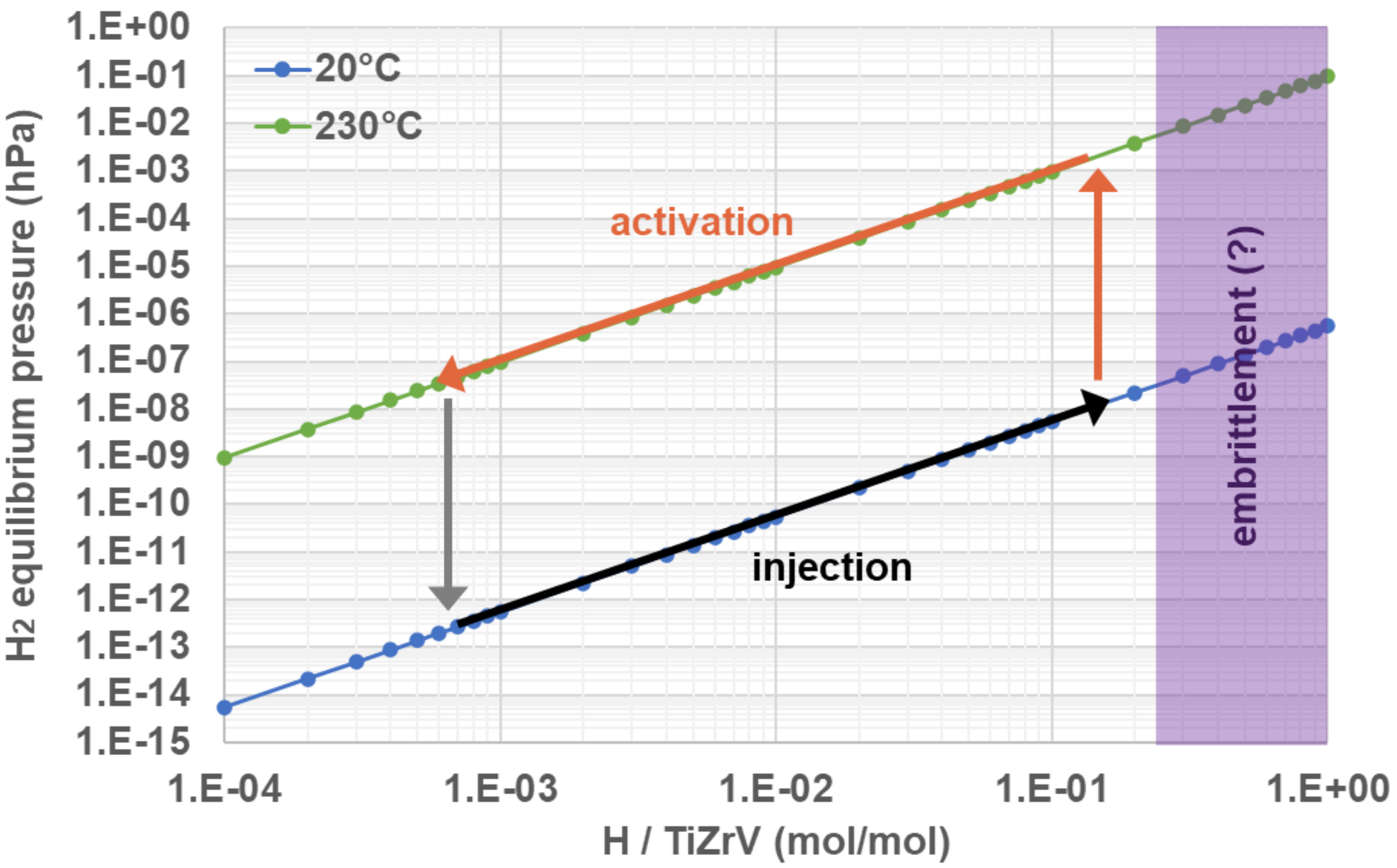
and many others…

CERN

Additional info

Sievert's law [1]

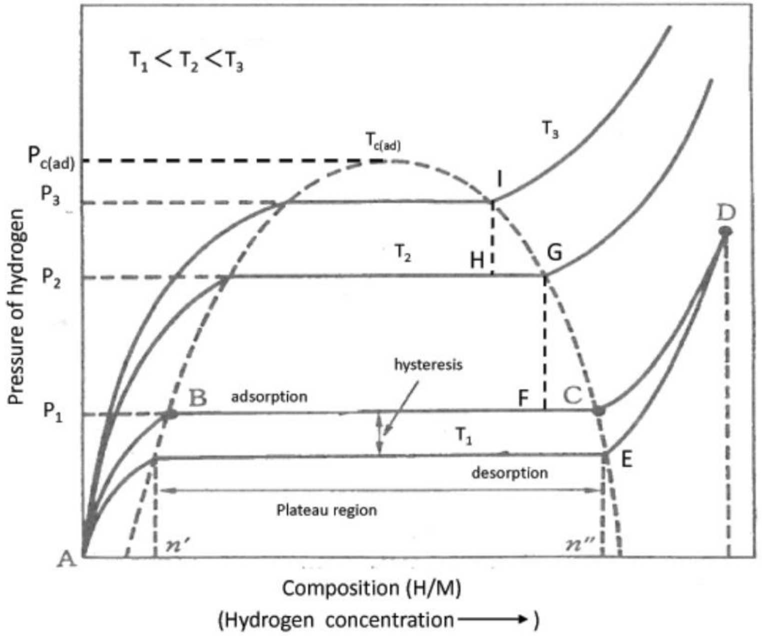
$$p_{H_2} = 2 \ln x_H + 14.2 - \frac{8468}{T}$$



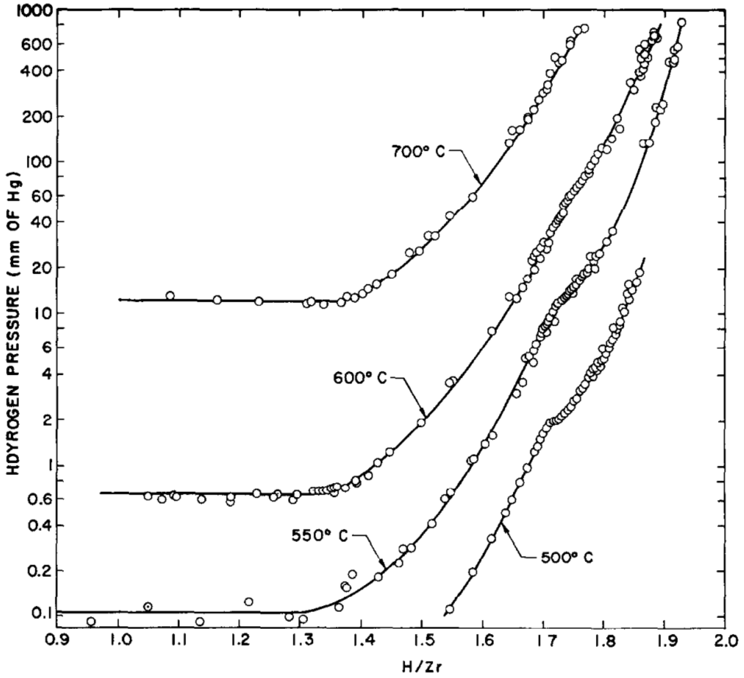
[1] Chiggiato, P., and P. Costa Pinto. "Ti–Zr–V non-evaporable getter films: From development to large scale production for the Large Hadron Collider." *Thin Solid Films* 515.2 (2006): 382-388.

Additional info

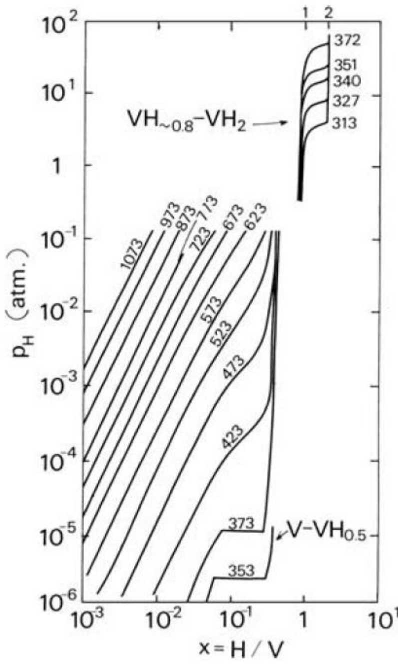
Metal-hydrogen pressure-composition isotherm diagrams



(a) [1]



(b) [2]



(c) [3]

Ti-H (a), Zr-H (b), and V-H (c) pressure-composition isotherm diagrams

[1] Henry-Couannier, Frederic. "From Dark Gravity to LENR." Journal of Condensed Matter Nuclear Science 18 (2016): 1-23.
[2] Libowitz, George G. "A pressure-composition-temperature study of the zirconium-hydrogen system at high hydrogen contents." Journal of Nuclear Materials 5.2 (1962): 228-233.
[3] Fukai, Yuh. The metal-hydrogen system: basic bulk properties. Vol. 21. Springer Science & Business Media, 2006

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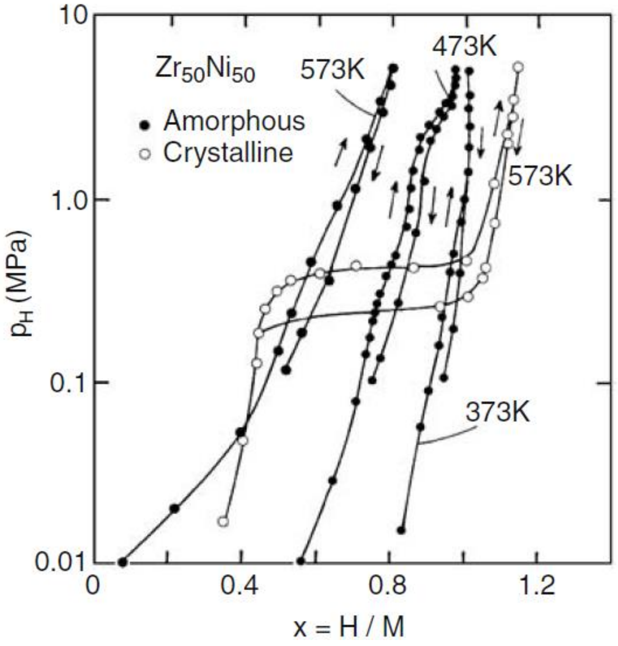


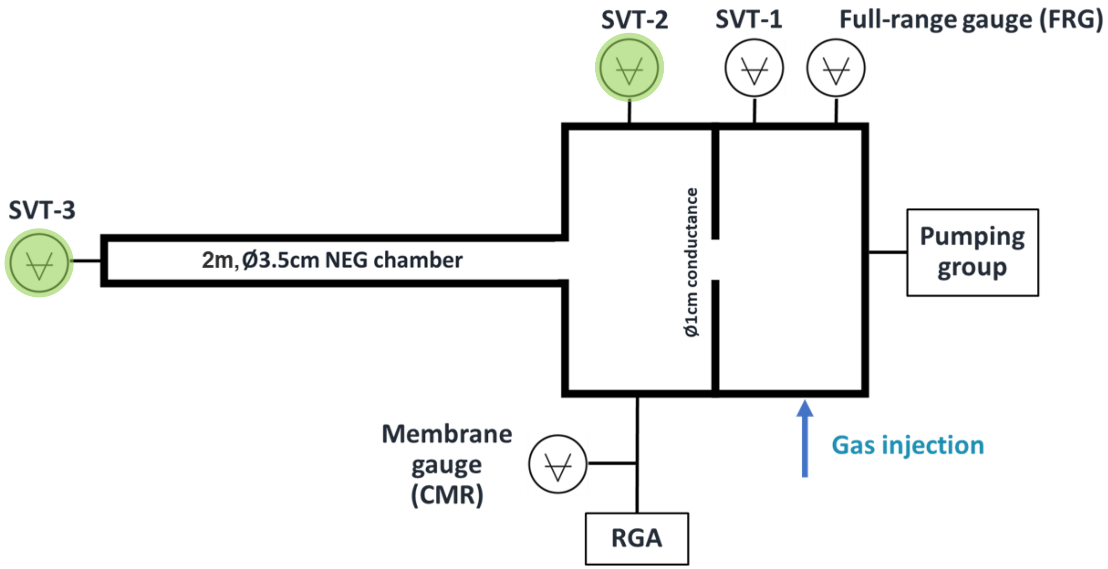
Pressure-composition isotherm diagram of amorphous and crystalline $Zr_{0.5}Ni_{0.5}$ alloy [1]

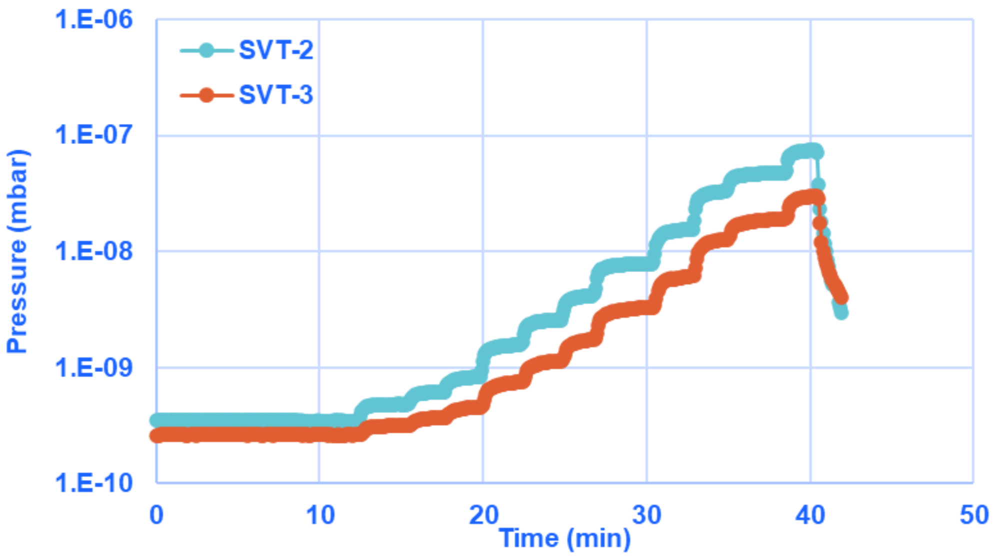
[1] Fukai, Yuh. The metal-hydrogen system: basic bulk properties. Vol. 21. Springer Science & Business Media, 2006

Additional info
transmission method

Sticking coefficient measurement

- Transmission ratio: **p_2/p_3** measured during transmission measurement





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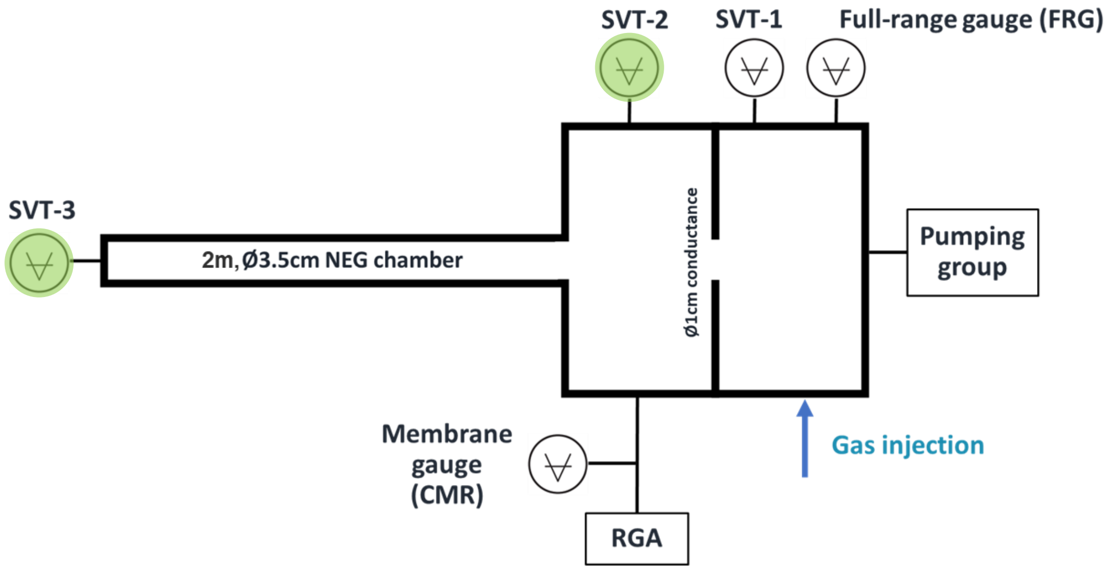

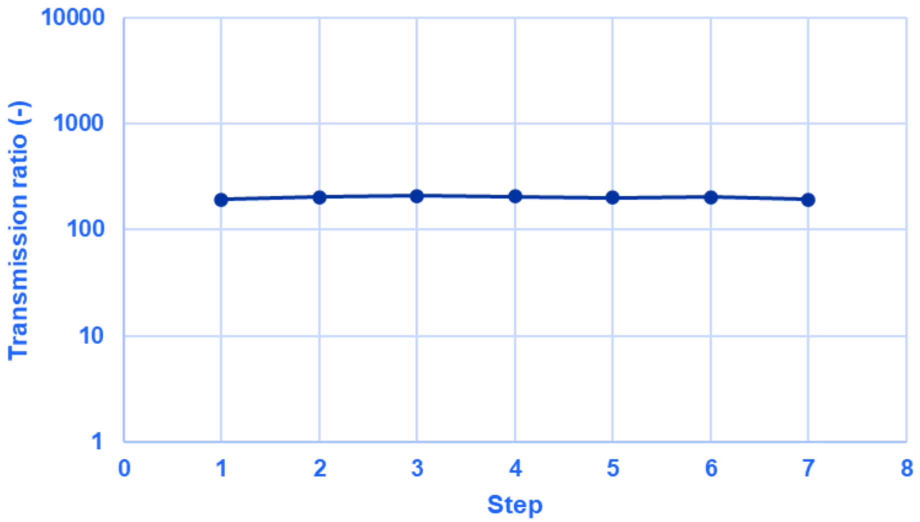

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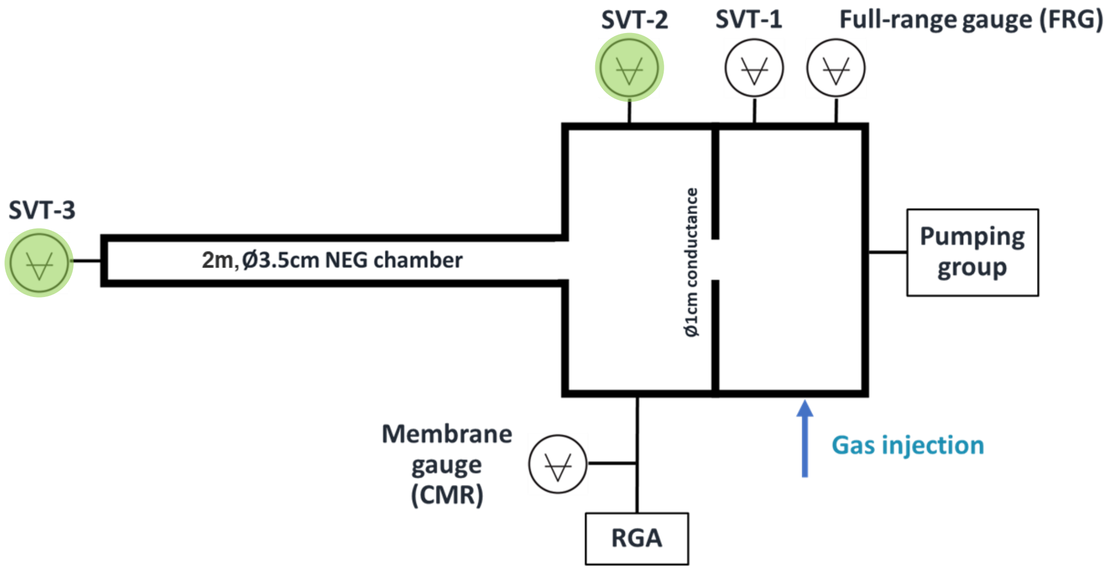
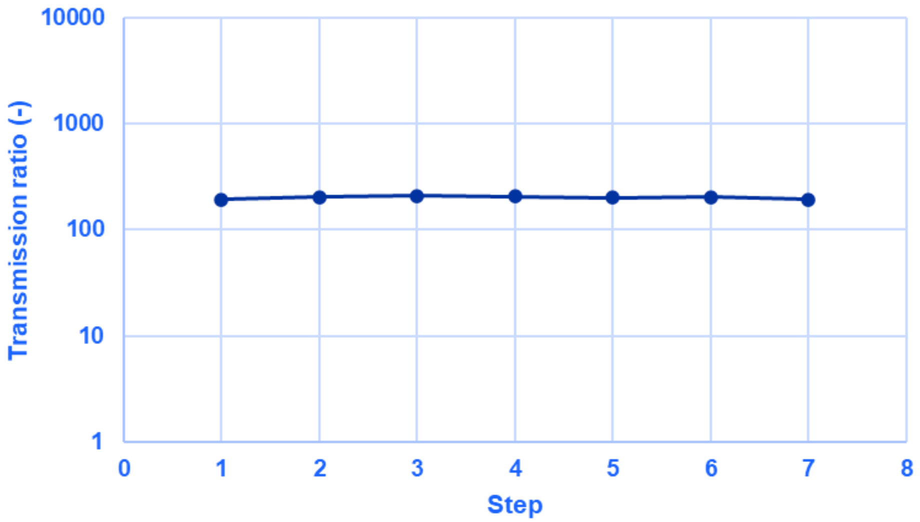
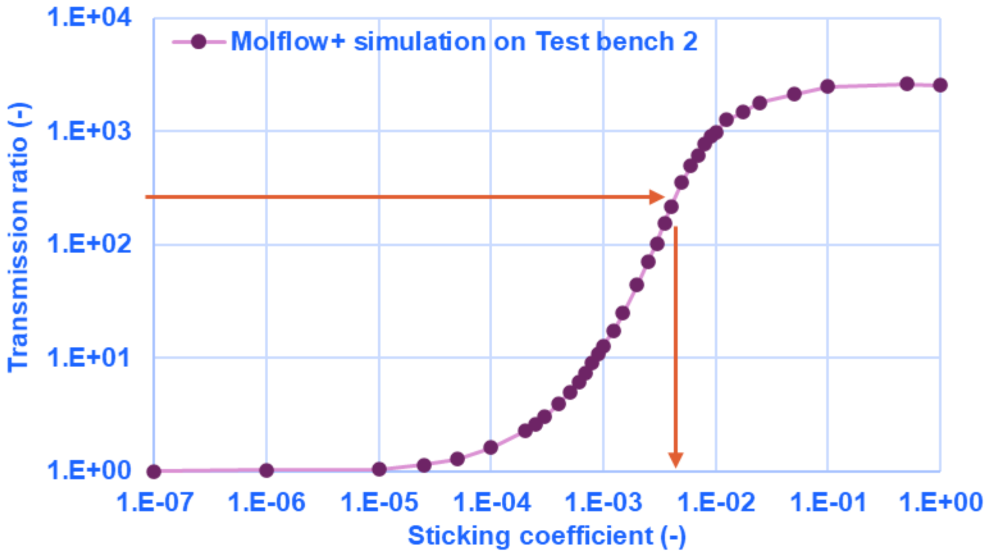
Sticking coefficient measurement

- Transmission ratio: **p_2/p_3** measured during transmission measurement
- p_2/p_3 → sticking coefficient (via Molflow+ simulations)







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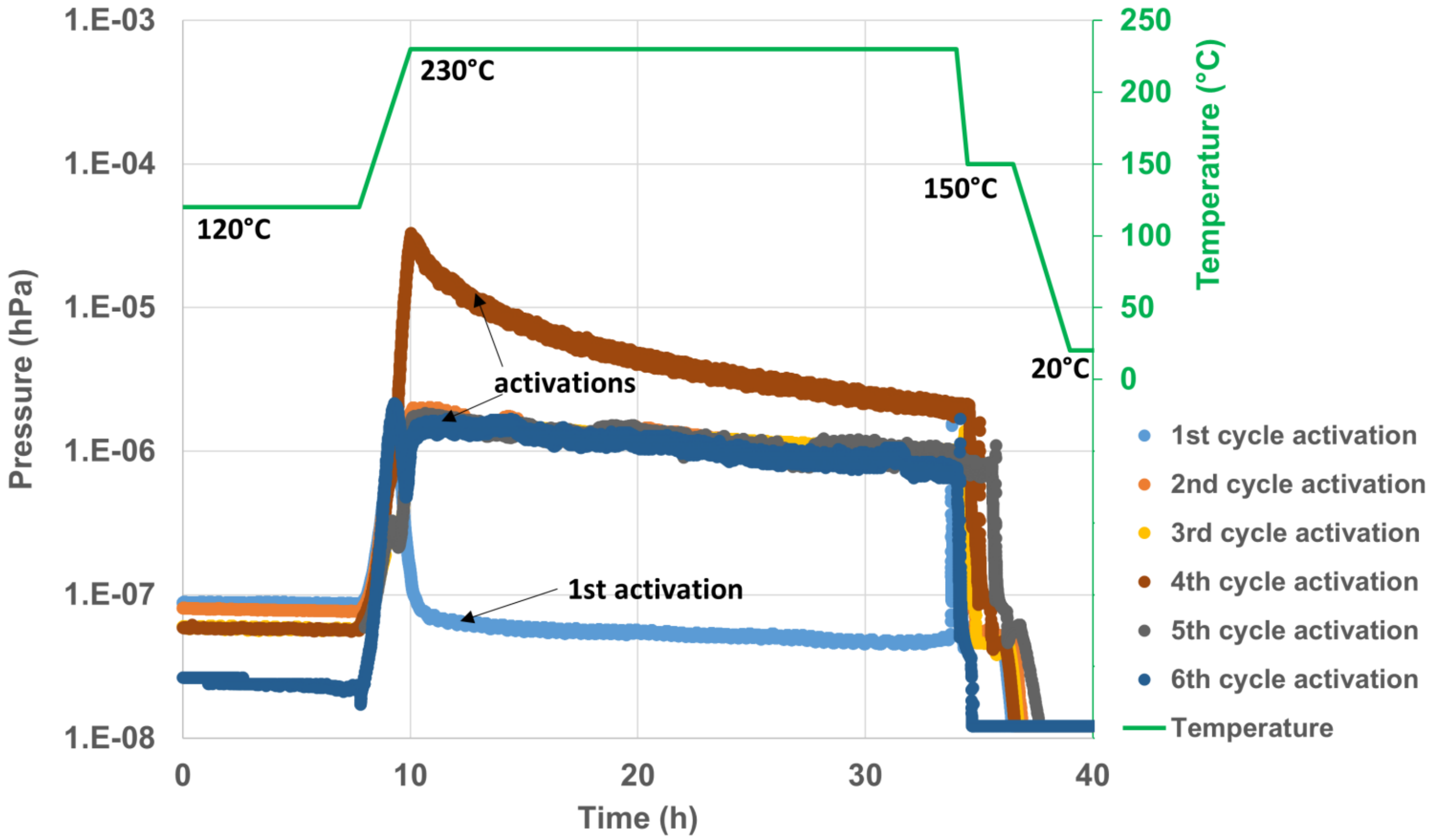

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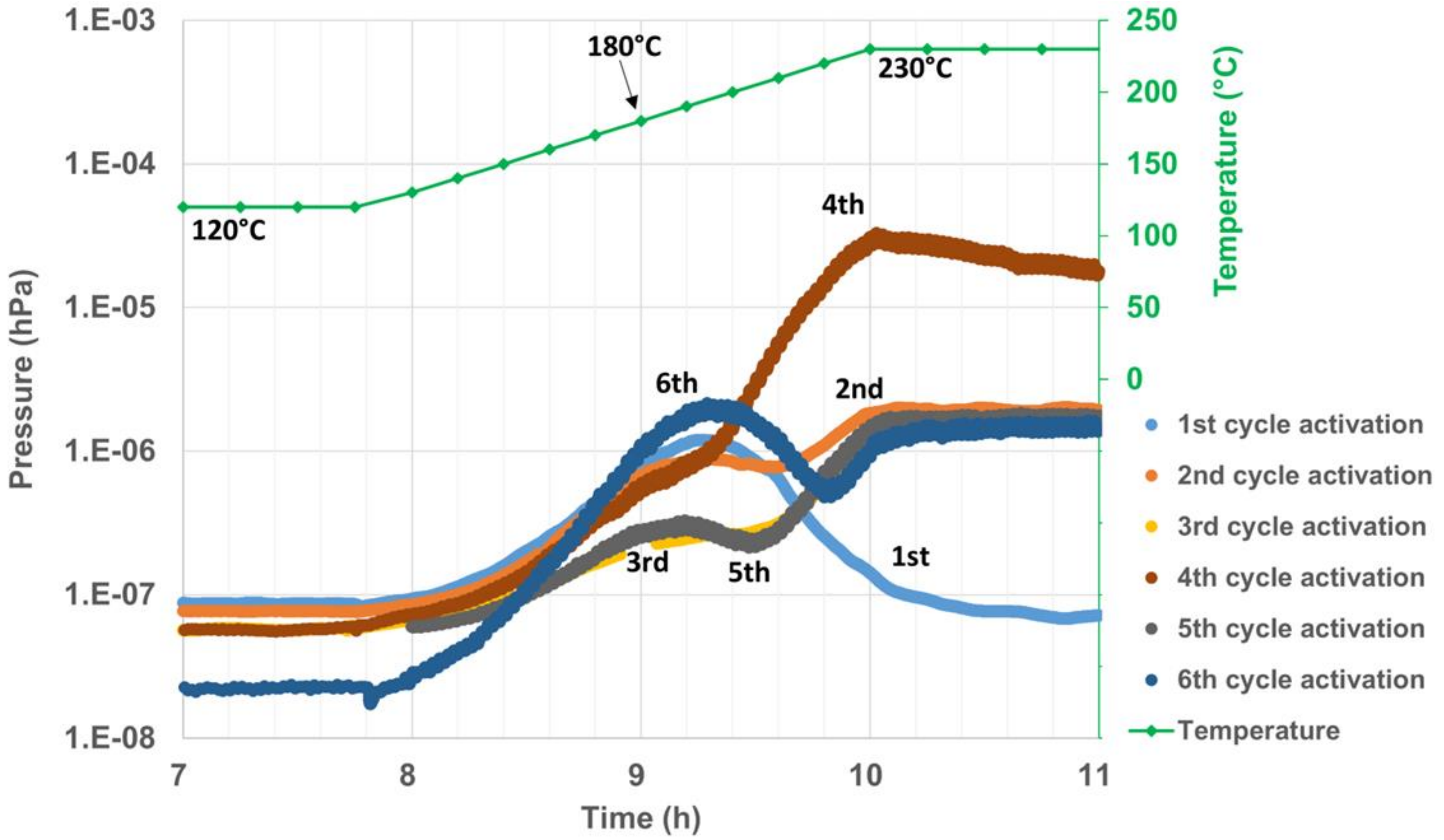
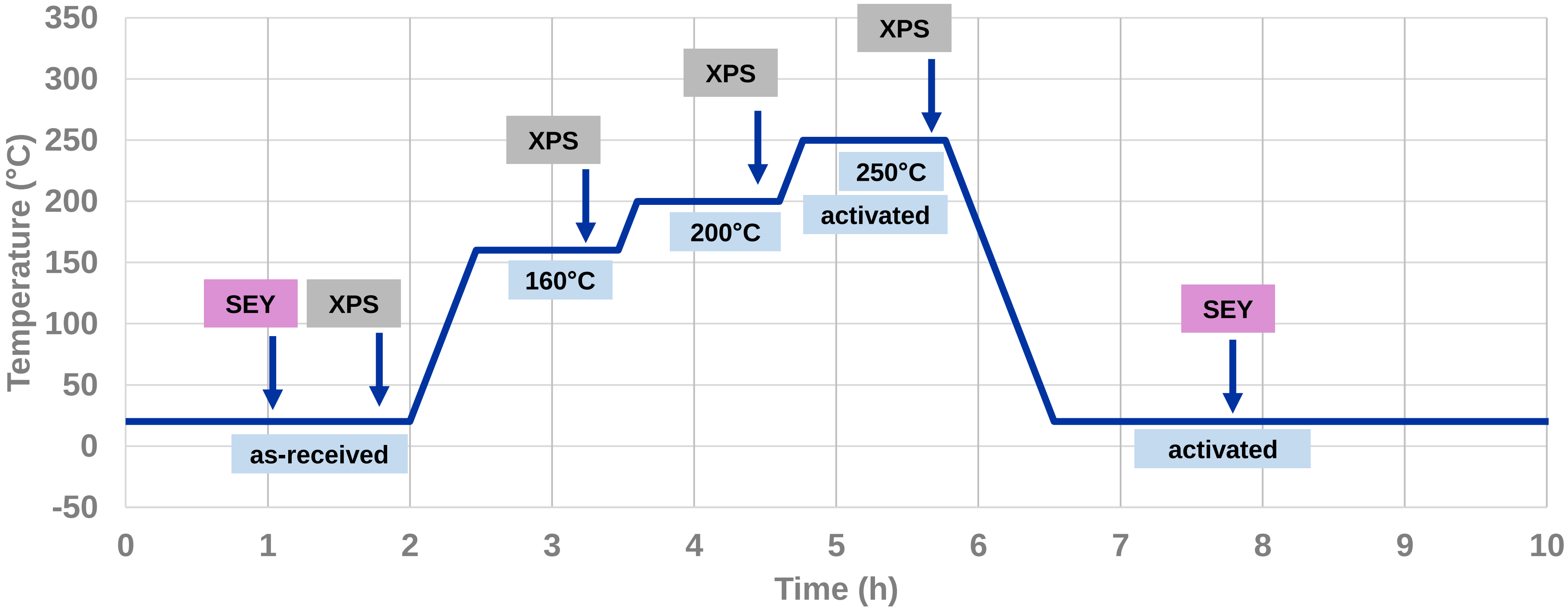


Additional info
high pressure experiment

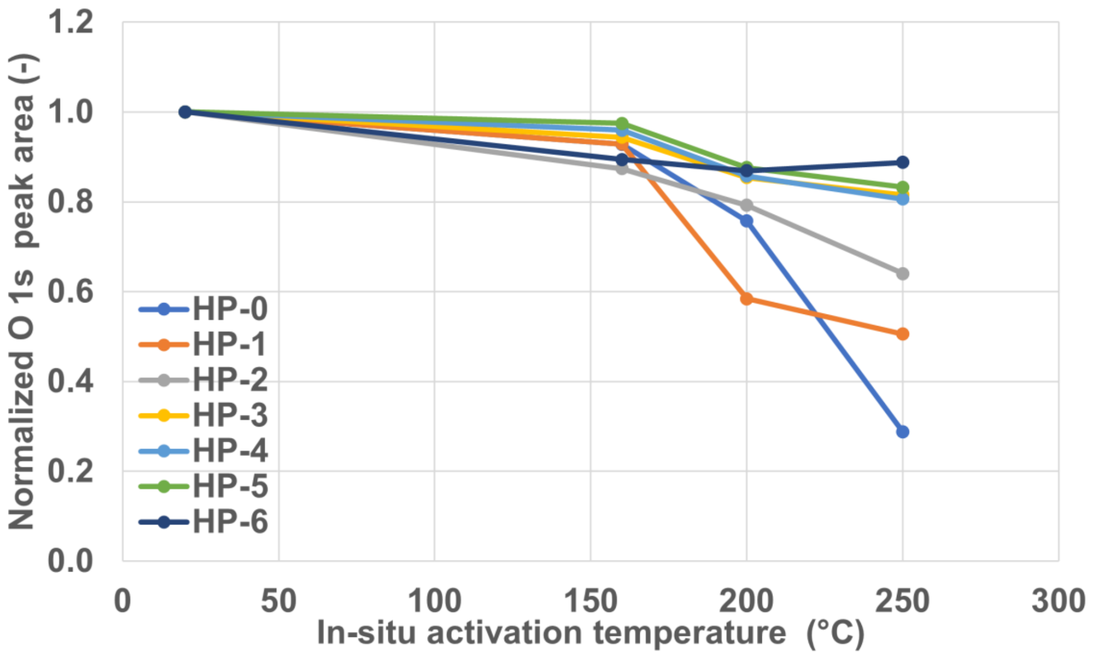
XPS and SEY procedure

→ compare SEY and top surface composition (XPS) before and after an in-situ activation of witness samples from the HP experiment

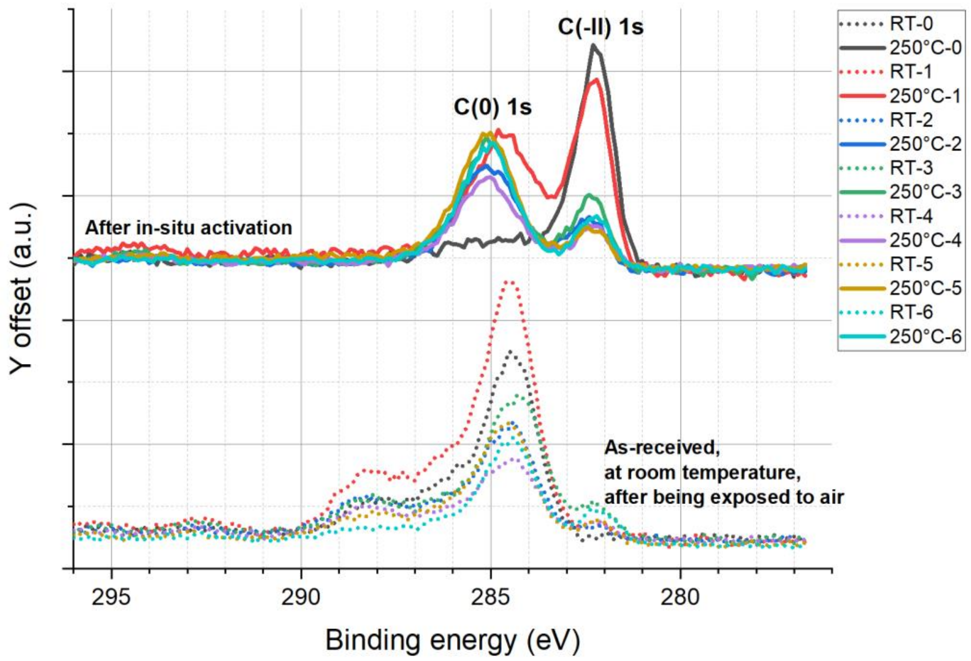


Additional info
high pressure experiment

XPS



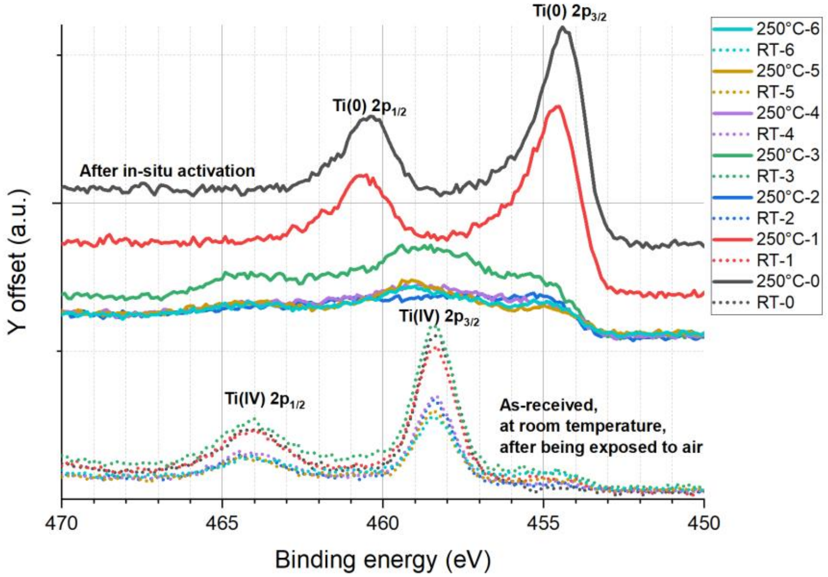
Relative surface O content decrease



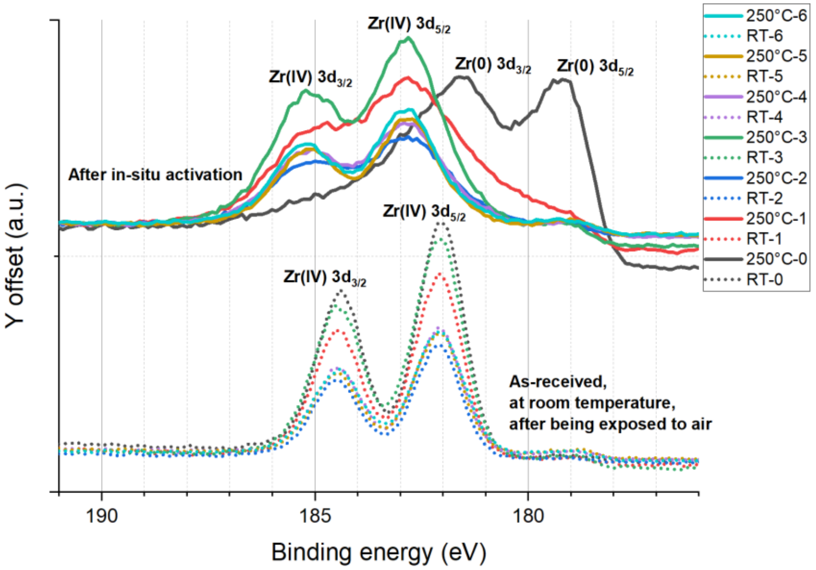
C 1 s peak: Carbon-to-carbide conversion

Additional info
high pressure experiment

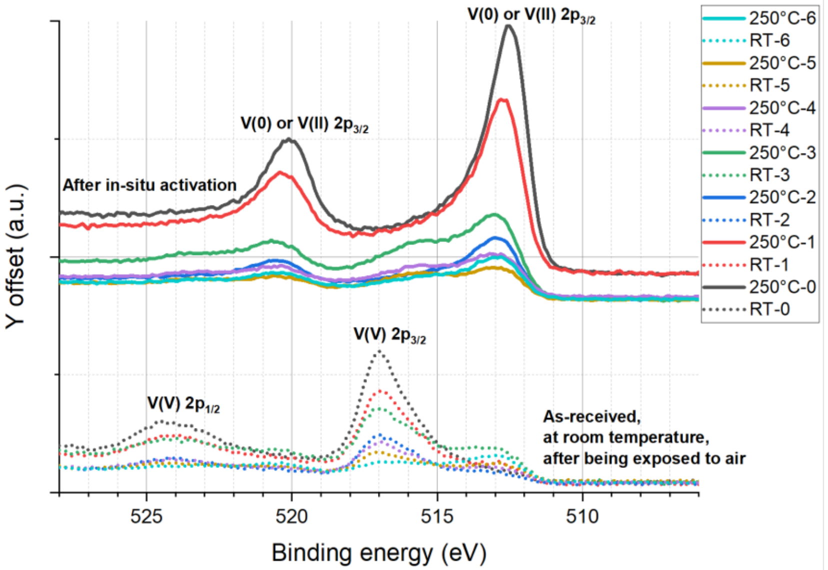
XPS



Ti 2p peak



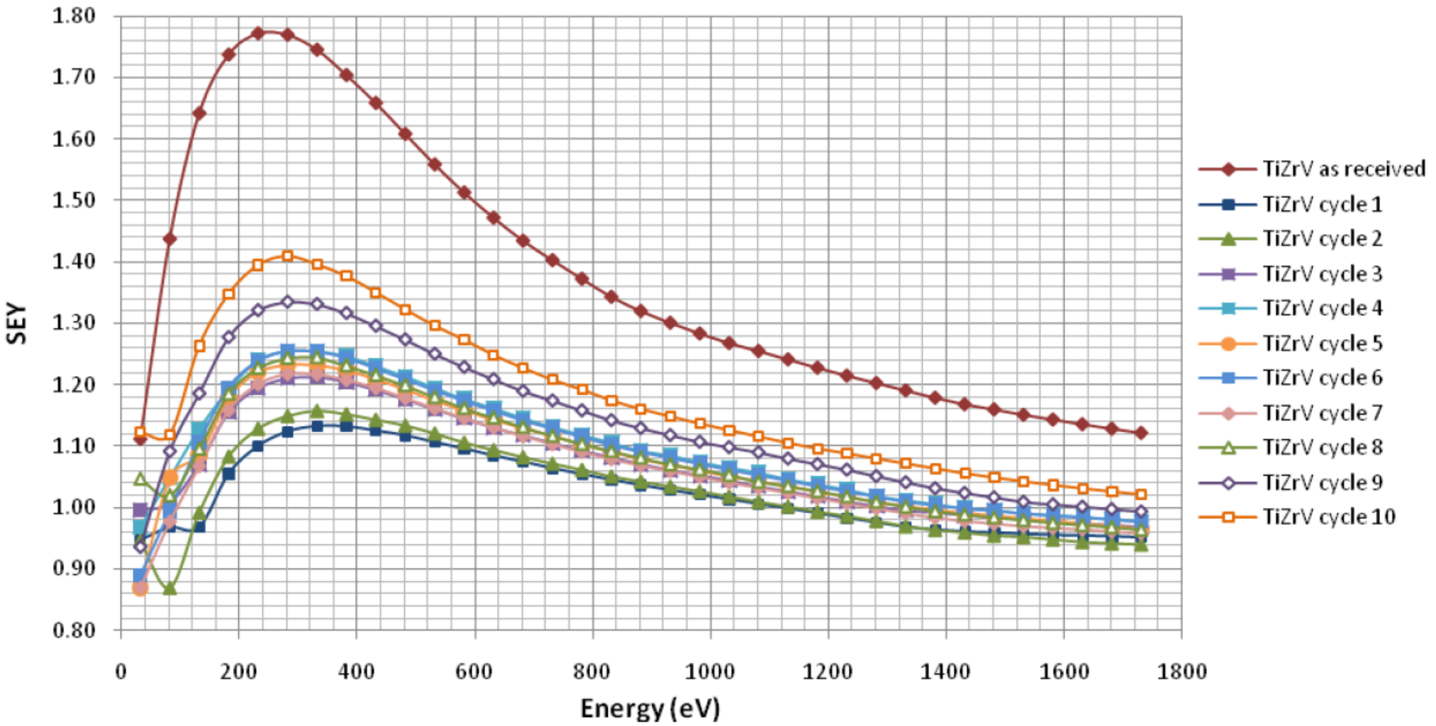
Zr 3d peak



V 2p peak

Additional info

effect of aging on secondary electron yield



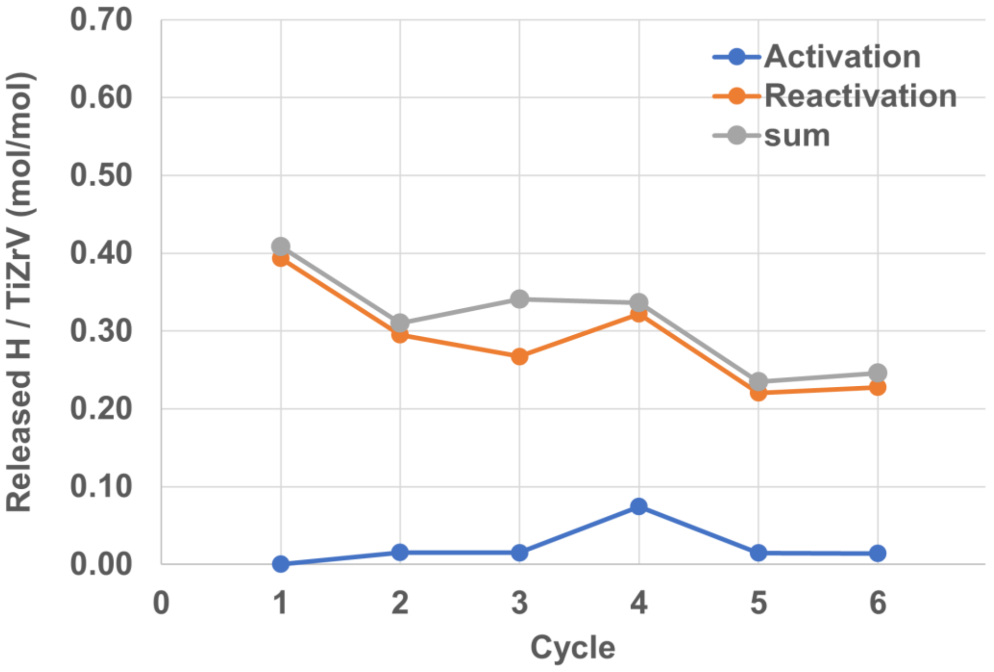
Secondary electron yield of TiZrV coating after repeated venting and activation cycles with 2 h 250 °C activation condition

Source: T. French: The secondary electron yield of non-evaporable getter films for repeated activation/venting cycles

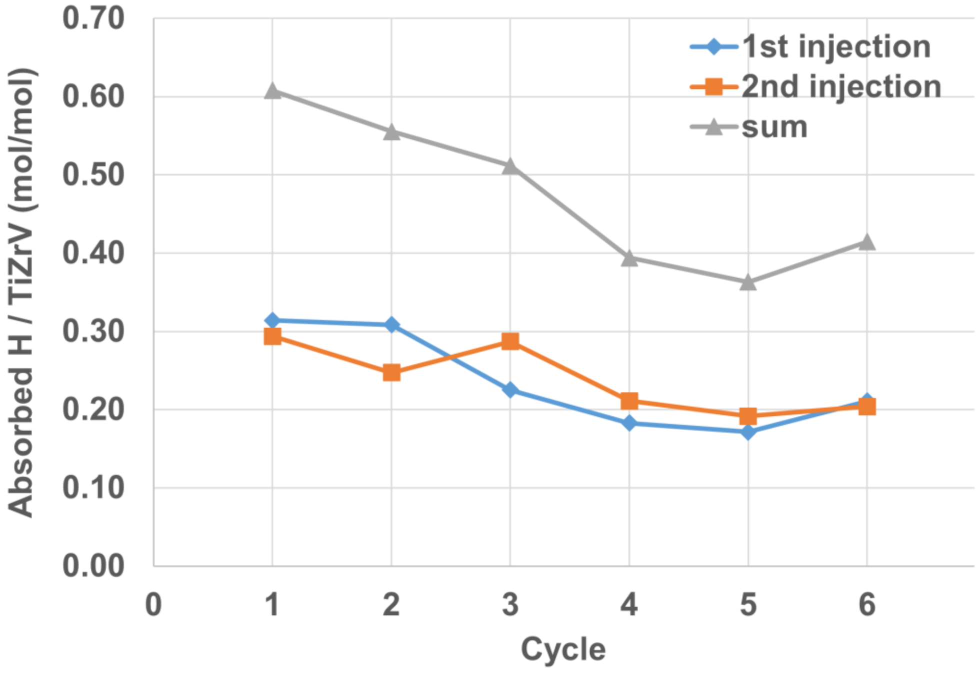
CERN

Additional info
high pressure experiment

Mass balance



Calculated released hydrogen quantity



Calculated estimated injected hydrogen quantity