PSI Center for Nuclear Engineering and Sciences

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SHE chemistry

Developments at the Laboratory of Radiochemistry PSI Switzerland

Robert Eichler, P Steinegger, G. Tiebel, J. Wilson, R. Dressler, D Herrmann, A. Vögele

SHE Cold Fusion 50, EREVAN, 20.-23. November 2024

Cold fusion: an early Dubna – Swiss collaboration topic

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A study of fusion reactions between ^{206}, ^{207}Pb nuclei and ^{40}Ar ions near the coulomb barrier

Published: December 1979

Volume 289, pages 415–420, (1979) Cite this article

H. Gaeggeler*, A.S. Iljinov**, G.S. Popeko, W. Seidel,
G.M. Ter-Akopian, and S.P. Tretyakova
Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Dubna, USSR

Received September 4, 1978

Excitation functions for the reactions ^{206}Pb(^{40}Ar, $2n$)^{244}Fm and ^{207}Pb(^{40}Ar, $3n$)^{244}Fm have been measured and analysed in terms of a statistical model. The optical potential parameters have been found to be as follows: $V_0 = -70$ MeV, $r_0 = 1.26 \times 10^{-13}$ cm and $d = 0.36 \times 10^{-13}$ cm. Some data on the properties of the excited compound nucleus of fermium have been obtained.

Table 2. Experimental results

Thickness of degrading foils, mg/cm^2	Bombarding energy, MeV	Integral ion flux, $\times 10^{16}$	Target thickness, mg/cm^2	Number of detected fission events	Corrected number of s.f. events	σ, cm^2
			^{206}Pb(^{40}Ar, $2n$)^{244}Fm			
2.2 Al + 1.4 Ni	175 ± 5	2.8	1.2	0	0	$< 1.3 \times 10^{-35}$
2.2 Al + 1.0 Ni	179 ± 3	2.1	0.7	1	1	$(6 \pm 6) \times 10^{-35}$
1.6 Al + 1.4 Ni	182 ± 5	5.6	1.2	96	96	$(1.2 \pm 0.2) \times 10^{-33}$
1.3 Al + 1.3 Ni	188 ± 3	5.8	0.5	64	64	$(1.7 \pm 0.2) \times 10^{-33}$
2.2 Al	190 ± 3	1.4	0.4	18	17	$(2.8 \pm 1.4) \times 10^{-33}$
2.3 Ni	195 ± 3	3.0	0.7	16	11	$(4.4 \pm 1.3) \times 10^{-34}$
1.4 Ni	204 ± 4	6.7	1.2	23	7	$(7 \pm 3) \times 10^{-35}$
			^{207}Pb(^{40}Ar, $3n$)^{244}Fm			
1.6 Al + 0.8 Ni	189 ± 3	2.5	0.7	44	39	$(1.9 \pm 0.4) \times 10^{-33}$
2.2 Ni	196 ± 3	2.2	0.7	70	64	$(4.2 \pm 0.7) \times 10^{-33}$
1.8 Ni	200 ± 3	4.2	0.7	118	109	$(3.5 \pm 0.4) \times 10^{-33}$
0.8 Ni	211 ± 3	3.3	0.7	53	49	$(1.8 \pm 0.4) \times 10^{-33}$
Without degrader	220 ± 2	5.4	0.7	6	6	$(1.2 \pm 0.6) \times 10^{-34}$

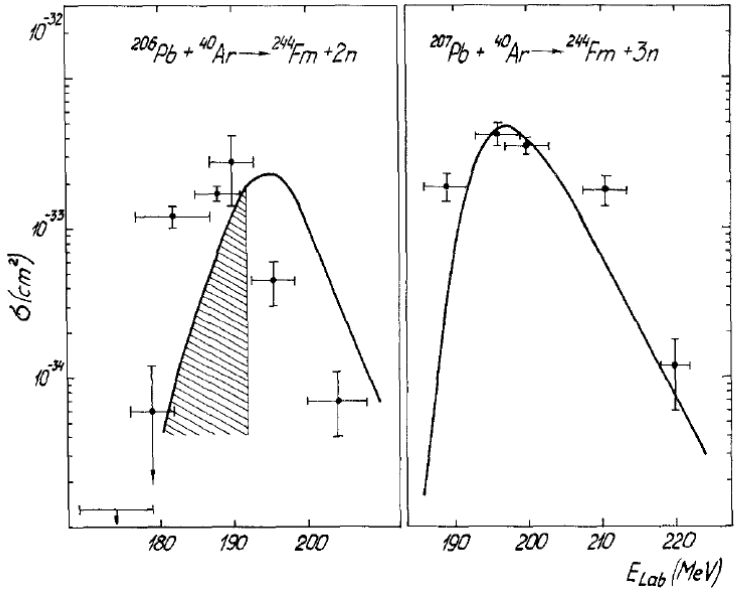


Fig. 2. Excitation functions for the reactions ^{206}Pb(^{40}Ar, $2n$)^{244}Fm and ^{207}Pb(^{40}Ar, $3n$)^{244}Fm. The solid lines show the calculation with fitted absolute values of σ_x in the maxima of excitation functions by the selection of the fissility parameter $\langle \Gamma_n/\Gamma_f \rangle$ (see Eq. (10)). The shaded area corresponds to the subbarrier energies of argon ions

Requirements to the chemical method for SHE

PSI

- efficiency
  - velocity
  - sensitive detection of SHE decay

- formation of defined reversible chemical states:
  - stable compound formation, interaction type
  - requires stable surfaces, stable resins, stable reactant concentrations etc.

Activated Adsorption:
3 parameters

- excellent separation of interfering by-products:

→ everything with SF or high energy a-particle emission

* lighter TA (a, SF)
* Po (precursor Bi and Pb or At and Rn (a)
* heavy actinides (Cf, Cm, Md, Fm) (SF)
* removal/destruction of aerosol particles, colloids

Requirements to the chemical method

PSI

macroscopic amounts: → Equilibrium concentrations,

⇒ 1 step

single atoms: → Probabilities

⇒ **multi-step** „*chromatographic*" "reversible" systems

ion exchange *chromatography* → Distribution coefficients

extraction *chromatography* → Distribution coefficients

gas *chromatography* → Adsorption properties

Demand from „Physical Chemistry":
To determine thermodynamic constants we have to be able to either:
- change concentrations of reactants in case of investigating molecule or complex formation
or/and:
- change of adsorption temperature in case of pure molecular/elemental adsorption

Group 13: Homolog Studies

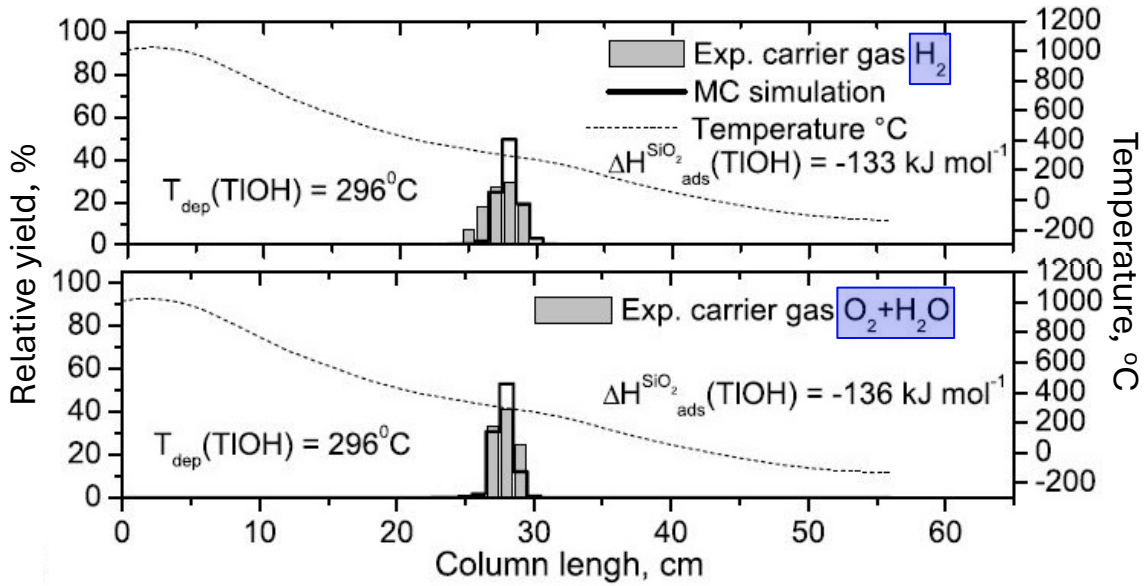
Experiments on <u>fused silica</u> surfaces

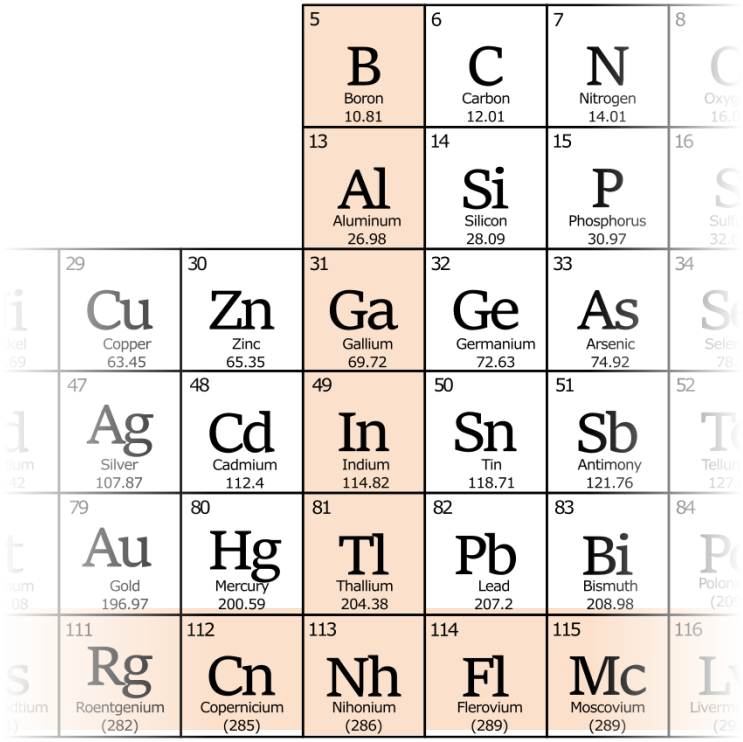
→ In/InOH: Offline thermochromatography

→ Tl/TlOH: Offline thermochromatography

→ **Tl: Online* isothermal chromatography**

→ Nh/NhOH ?

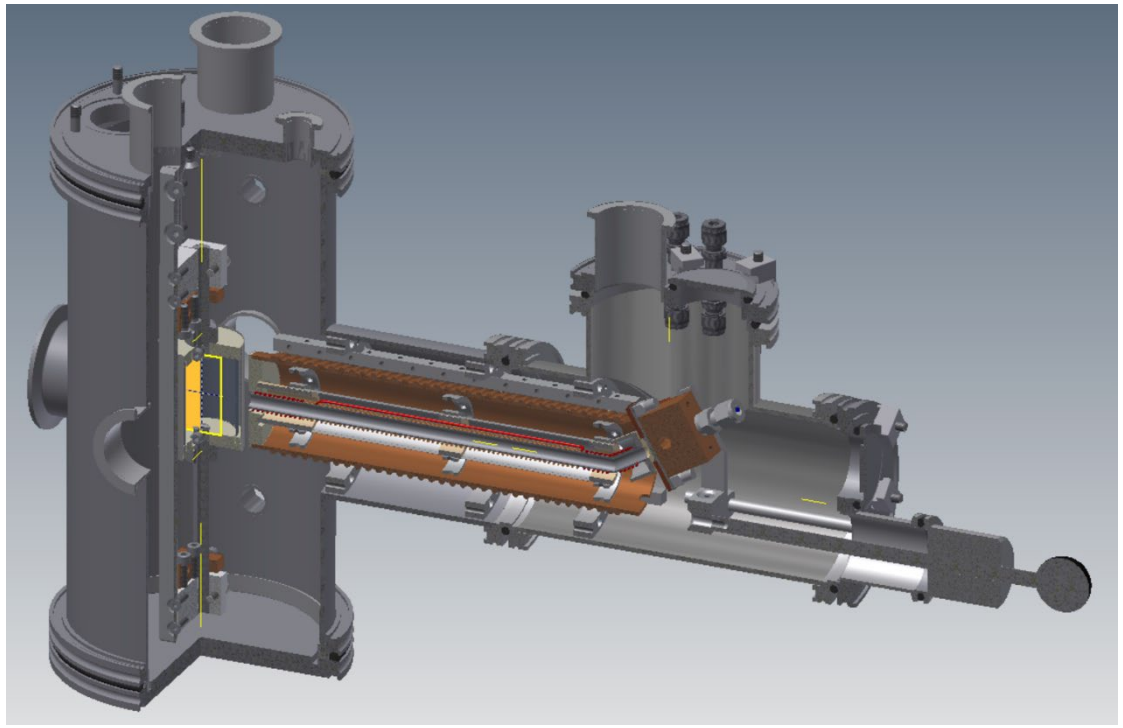




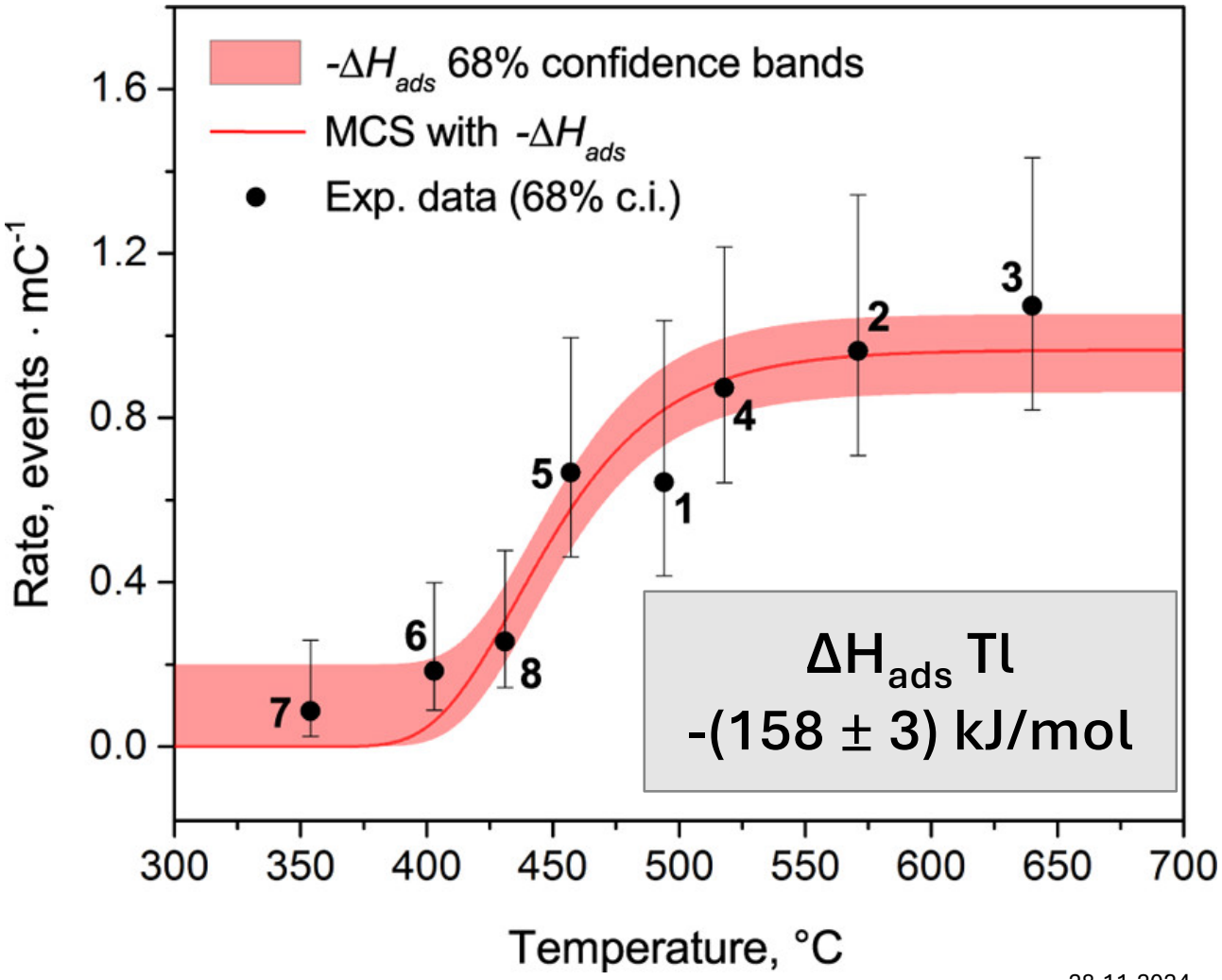
Serov et al.(2011&2013)

Adsorption of Thallium on Quartz Results

@ JAEA PSI



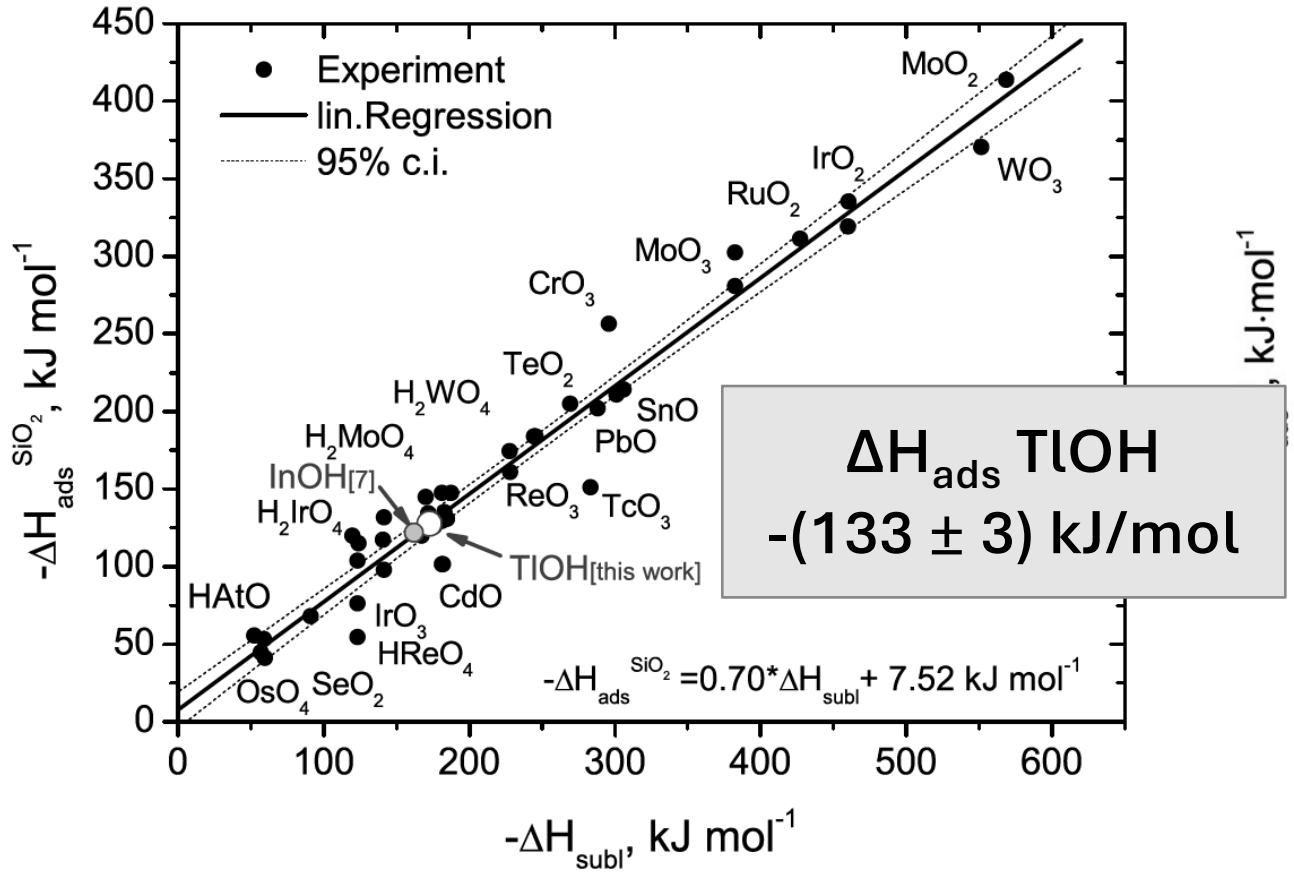
ΔH_{ads} TlOH
-(133 ± 3) kJ/mol



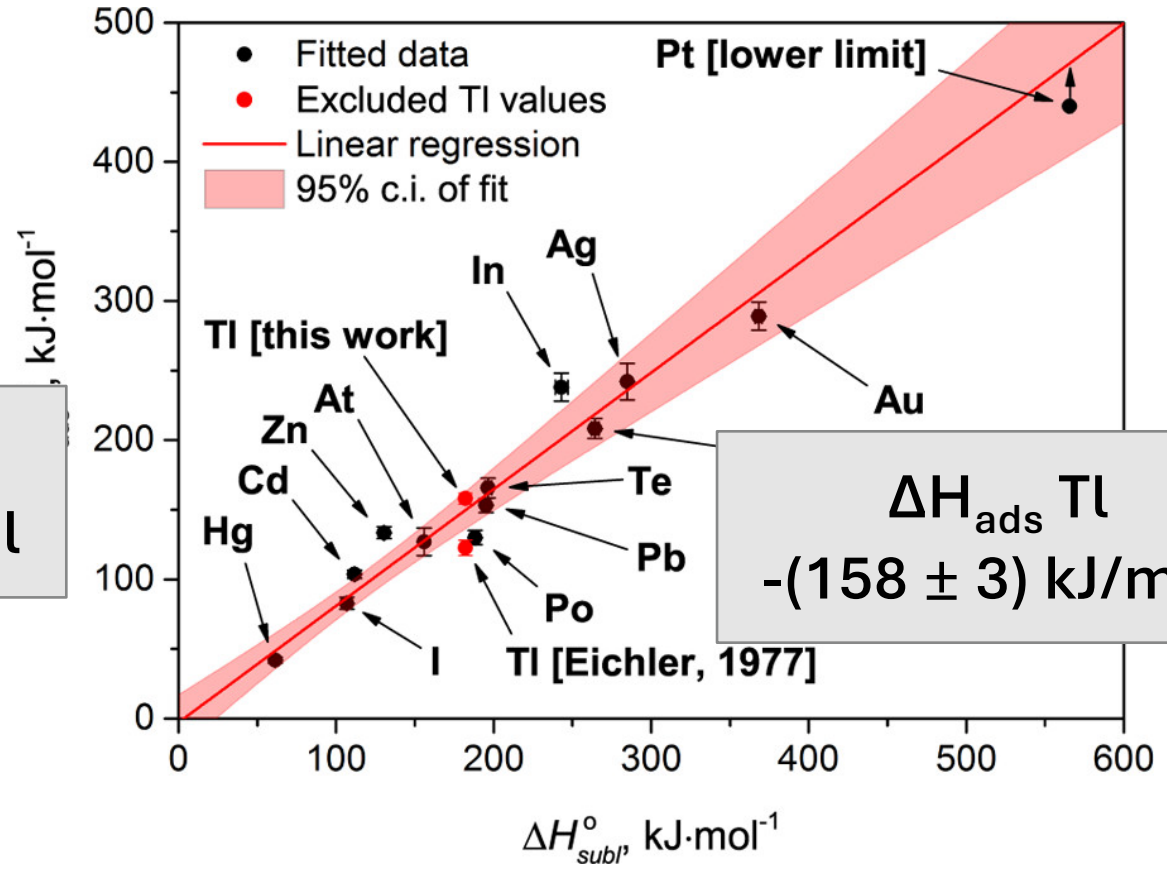
P. Steinegger et al.: *Physical Chemistry C* 120 (2016) 7122

Chemical identification

macroscopic vs. microscopic behavior

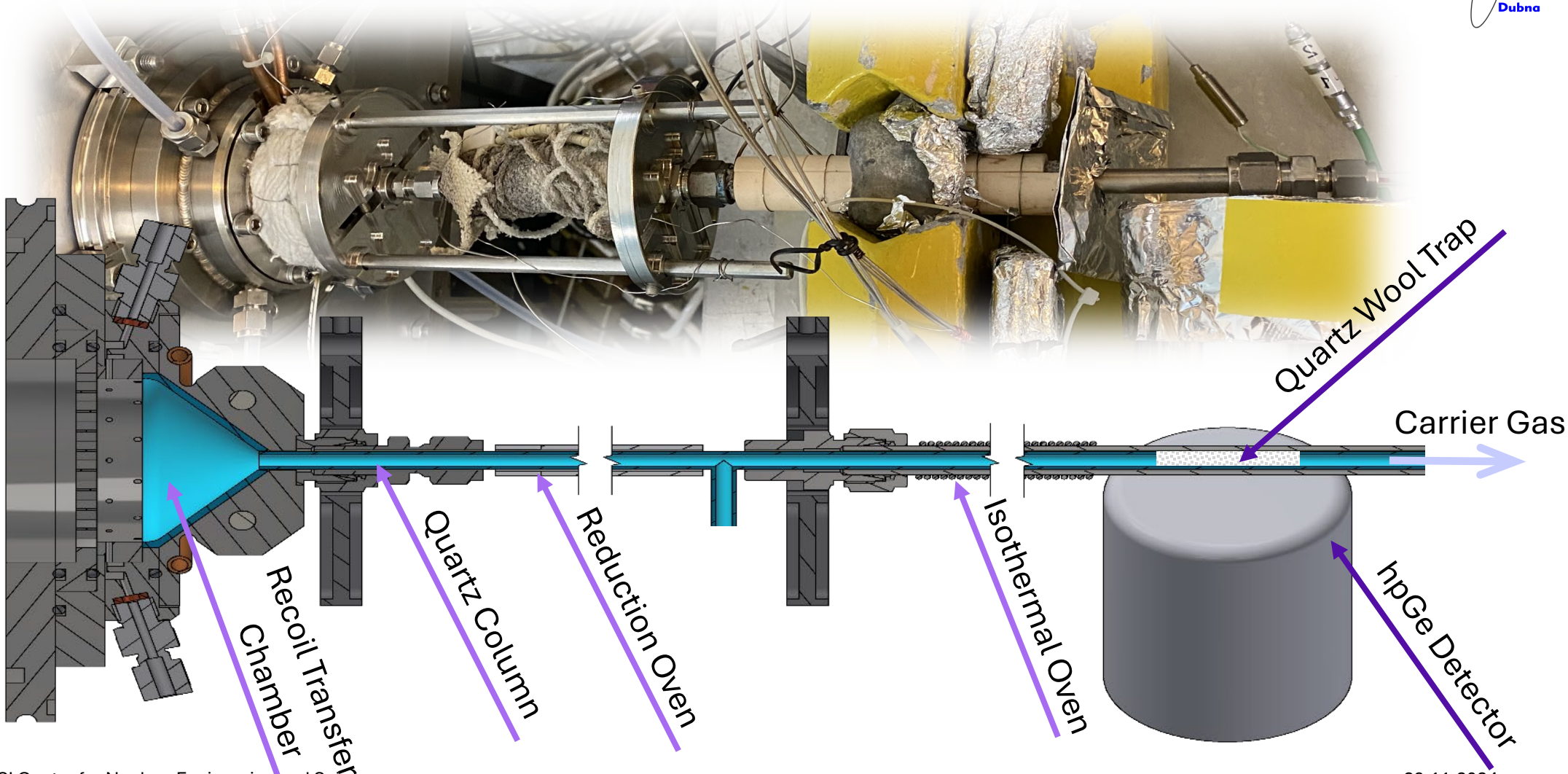


ΔH_{ads} TlOH
-(133 ± 3) kJ/mol

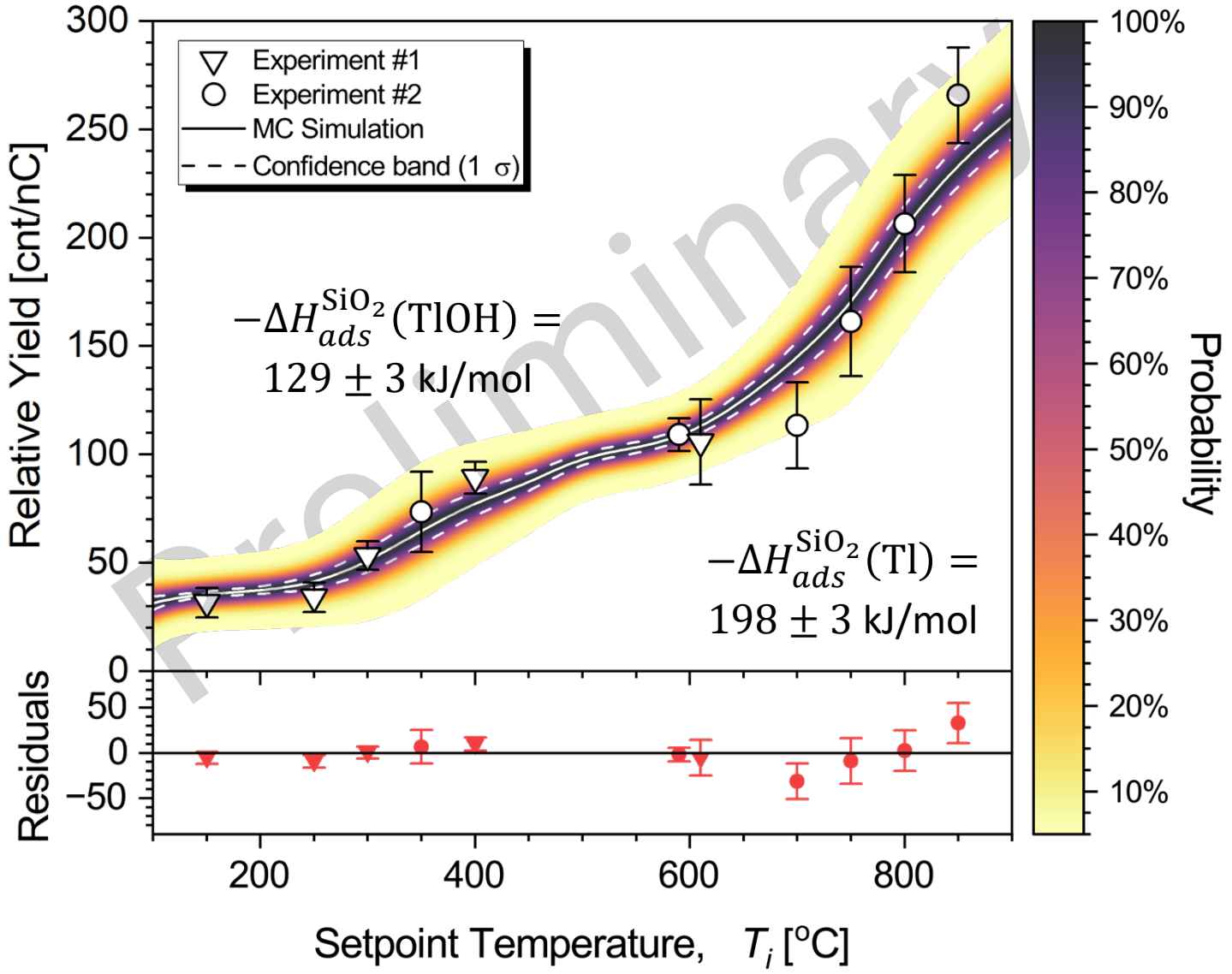


ΔH_{ads} Tl
-(158 ± 3) kJ/mol

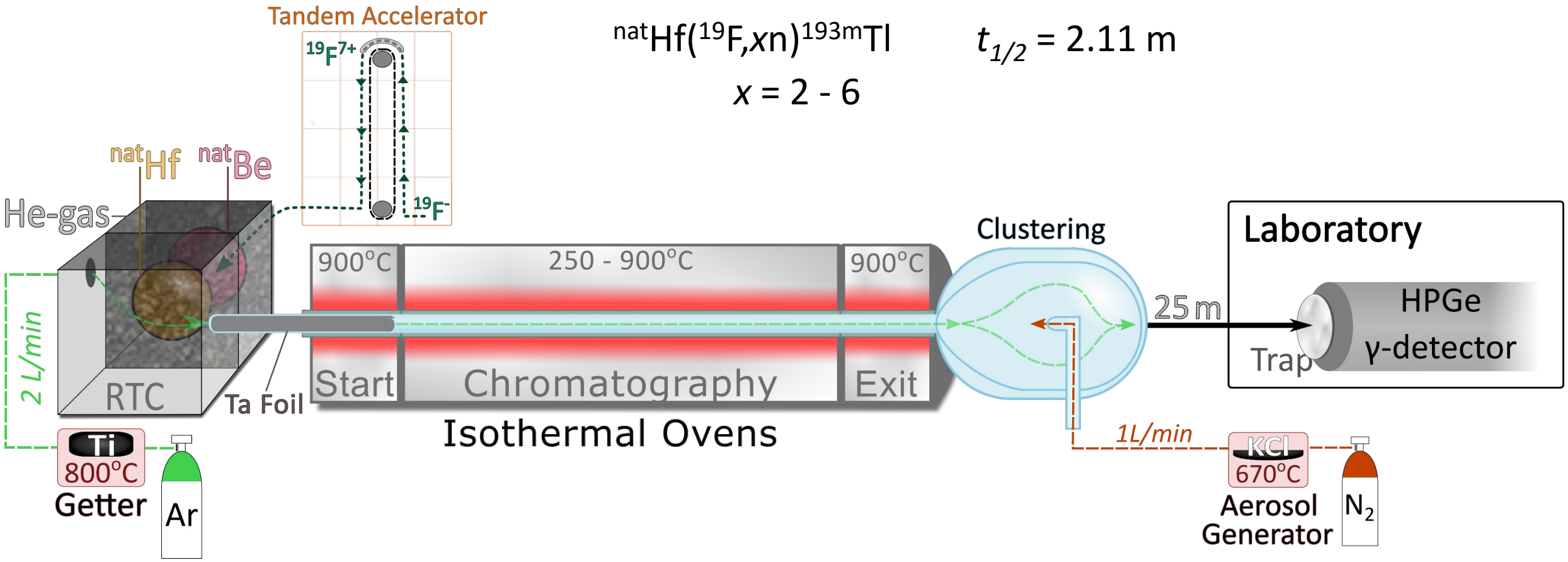
Adsorption of Thallium on Quartz @ JINR
"Isothermal" Gas-Chromatography (2021)



Online Experiment at FLNR/JINR



Online Isothermal Chromatography



$^{nat}Hf(^{19}F,xn)^{193m}Tl$ $\quad t_{1/2}$ = 2.11 m

x = 2 - 6

Online Isothermal Chromatography

$-\Delta H_{ads}^{SiO_2}(TlOH) = 129 \pm 3$ kJ/mol

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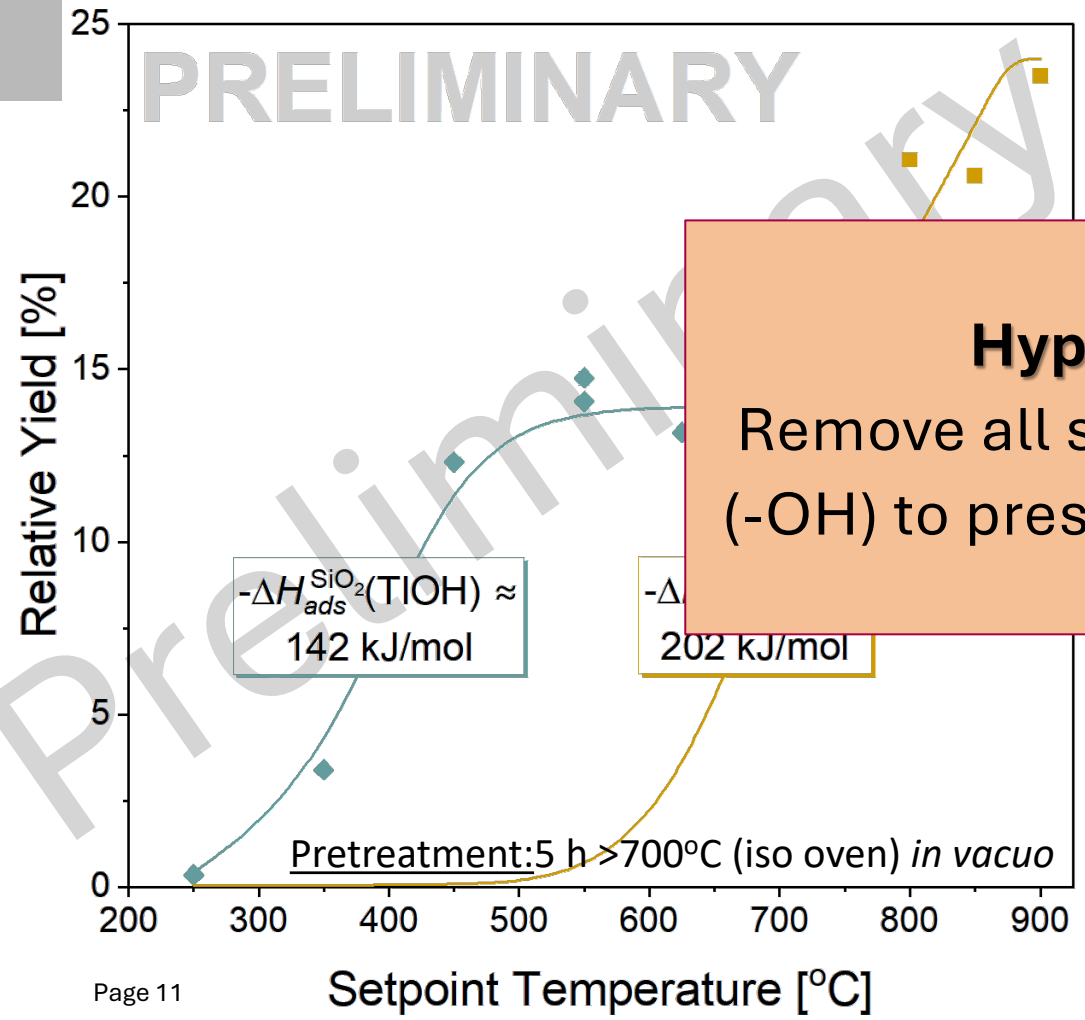
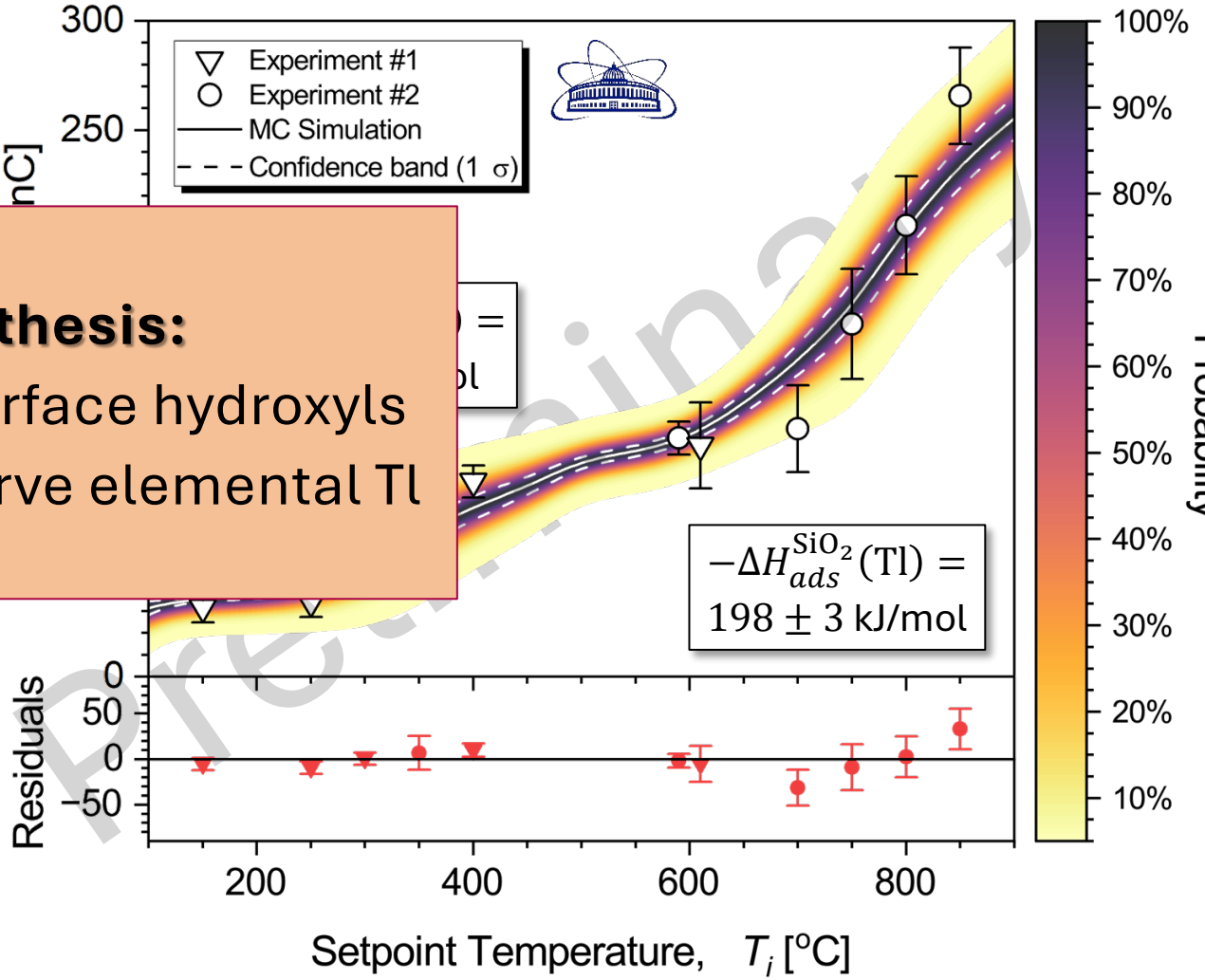
JAEA

External Chromatogram



External Chromatogram [1]



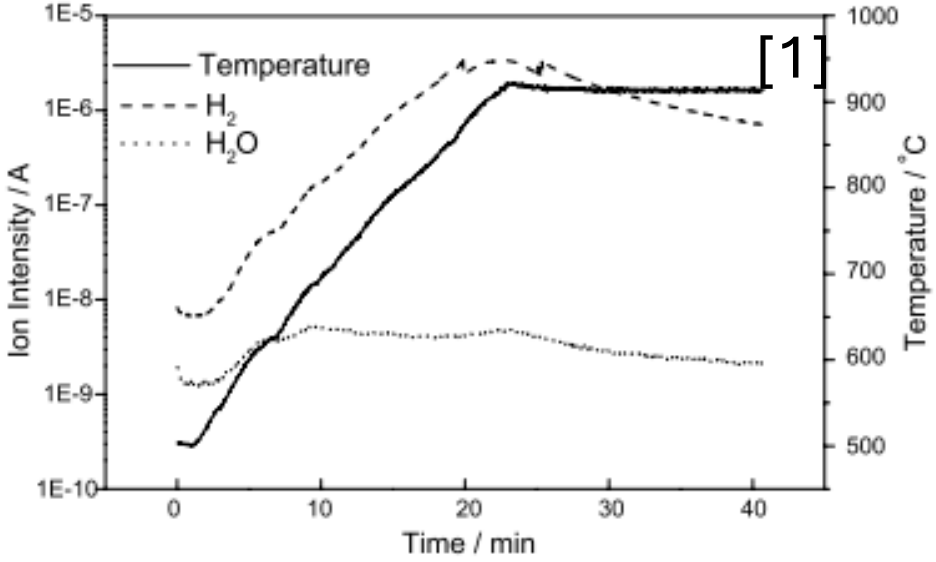
Hypothesis:
Remove all surface hydroxyls (-OH) to preserve elemental Tl

[1] Wilson *under review* (2024)

Surface Dehydroxylation

- Primary TlOH formation occurs (likely) due to adjacent hydroxyls

- Surface changes with temperature [1,2]
  - ➢100-200 °C: surface dehydration
  - ➢>200 °C: dehydroxylation
  - ➢400 °C: rehydroxyation strongly hindered with H_2O or air



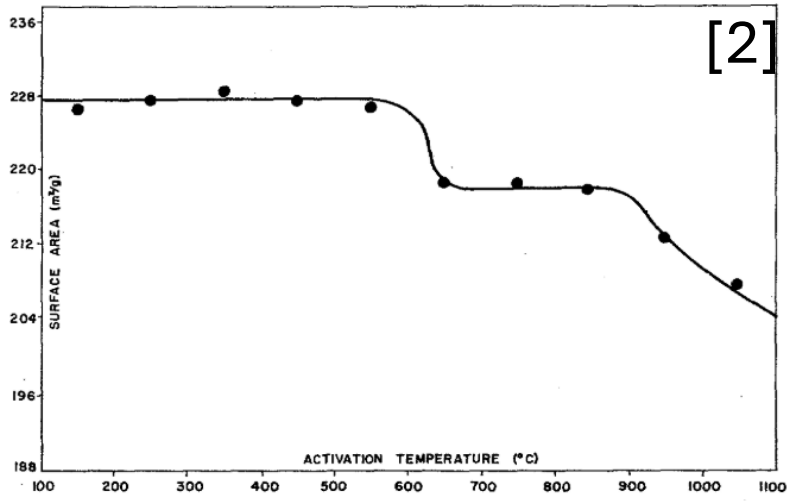


FIG. 3. Change in surface area with heat treatment of sample.

[1] Youngheng, Z., Zhenan, G. *J. Non-Cryst. Solids* (2006) **352** pg. 4030-4033.
[2] Young, G. J., *Journal of Colloid Science* (1958) **13** pg. 67-85.

Theoretical Adsorption Studies 2022

PSI

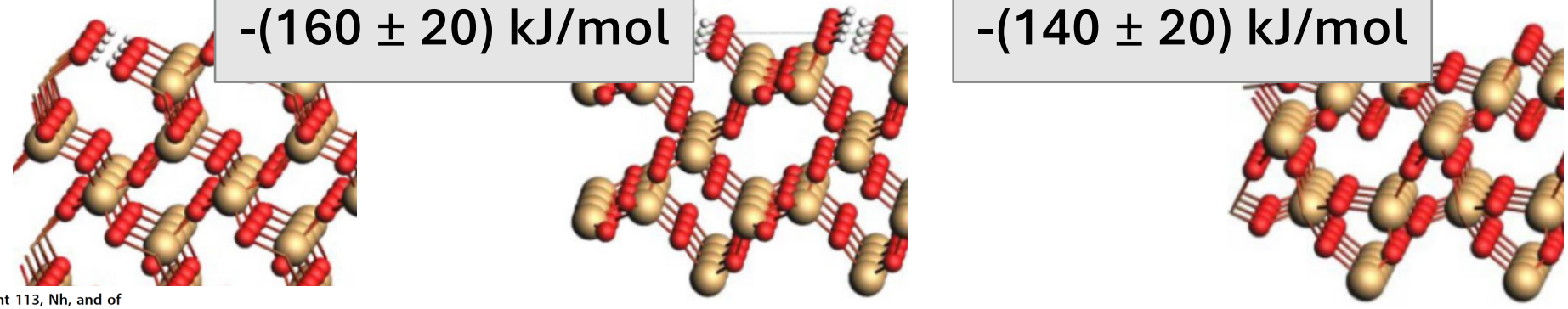
	$-\Delta H_{ads}^{SiO_2}$ [kJ/mol]			
	Tl	TlOH	Nh	NhOH
Geminal surface (G)	20.1	133.1	4.7	127.1
Vicinal surface (V)	44.2	157	27.1	140.7
Dehydroxylated (B)	80.9	324.5	24.3	237.8

?

Fully hydro ally hydroxylat ated (B)

ΔH_{ads} Tl
-(160 ± 20) kJ/mol

ΔH_{ads} TlOH
-(140 ± 20) kJ/mol



Reactivity of Group 13 Elements Tl and Element 113, Nh, and of Their Hydroxides with Respect to Various Quartz Surfaces from Periodic Relativistic DFT Calculations

Miroslav Iliaš and Valeria Pershina*

Cite This: *Inorg. Chem.* 2022, 61, 15910−15920 Read Online

Tl in room temperature isothermal gas chromatography

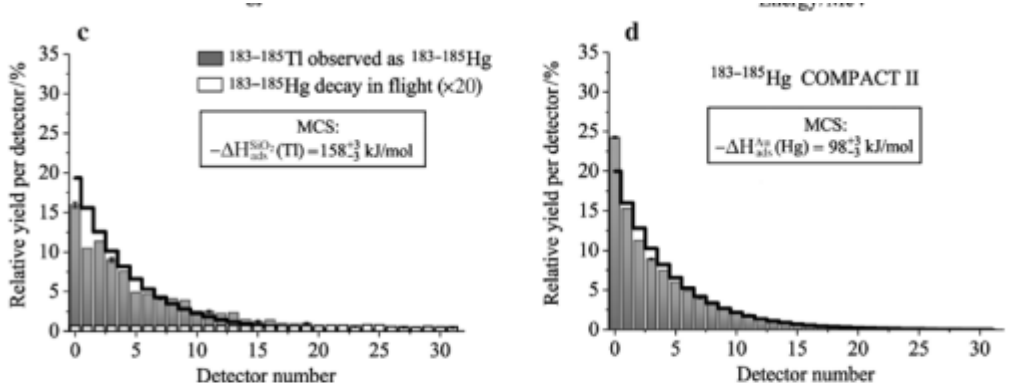


Figure 8: Panels (a and b) show the sum spectra of Hg in COMPACT I and II. In panels (c and d) the distribution (with statistical uncertainties indicated as error bars in every fourth experimental bar) of Hg (3.5–6.5 MeV) inside the COMPACT detector arrays, together with the result of the MCS using the $-\Delta H_{ads}$ for Tl from Ref. [32] is shown. The white bars shown in panel c, show Hg decay in flight, determined from the total sum of Hg, subtracted the deposition of Tl.

These experiments will *not tell us*:

1) what species deposits (maybe two or three?).
2) and what are the absolute adsorption properties.

They will allow to determine adsorption interaction limits assuming a hypothetical species.

DE GRUYTER Radiochim. Acta 2018; 106(12): 949–962

Lotte Lens*, Alexander Yakushev, Christoph Emanuel Düllmann, Masato Asai, Jochen Ballof, Michael Block, Helena May David, John Despotopulos, Antonio Di Nitto, Klaus Eberhardt, Julia Even, Michael Götz, Stefan Götz, Hiromitsu Haba, Laura Harkness-Brennan, Fritz Peter Heßberger, Rodi D. Herzberg, Jan Hoffmann, Annett Hübner, Egon Jäger, Daniel Judson, Jadambaa Khuyagbaatar, Birgit Kindler, Yukiko Komori, Joonas Konki, Jens Volker Kratz, Jörg Krier, Nikolaus Kurz, Mustapha [illegible], Bettina Lommel, M[illegible]

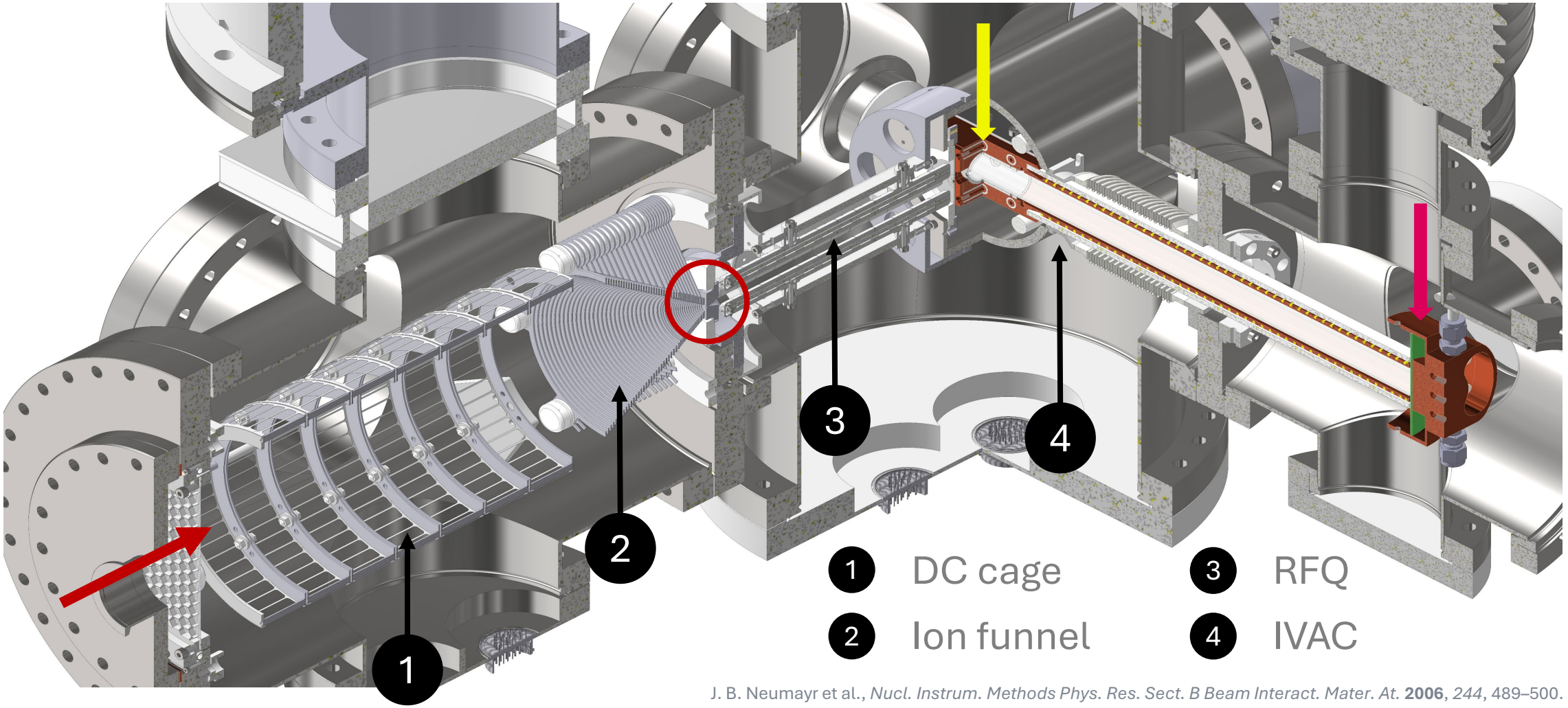
Such experiments (also with SHE) are extremely useful:

1) to assess the required temperature interval for future experiments!

2) If two or more separated deposition interactions are observed, this can only be interpreted as formation of several chemical species if the adsorption process is defined.

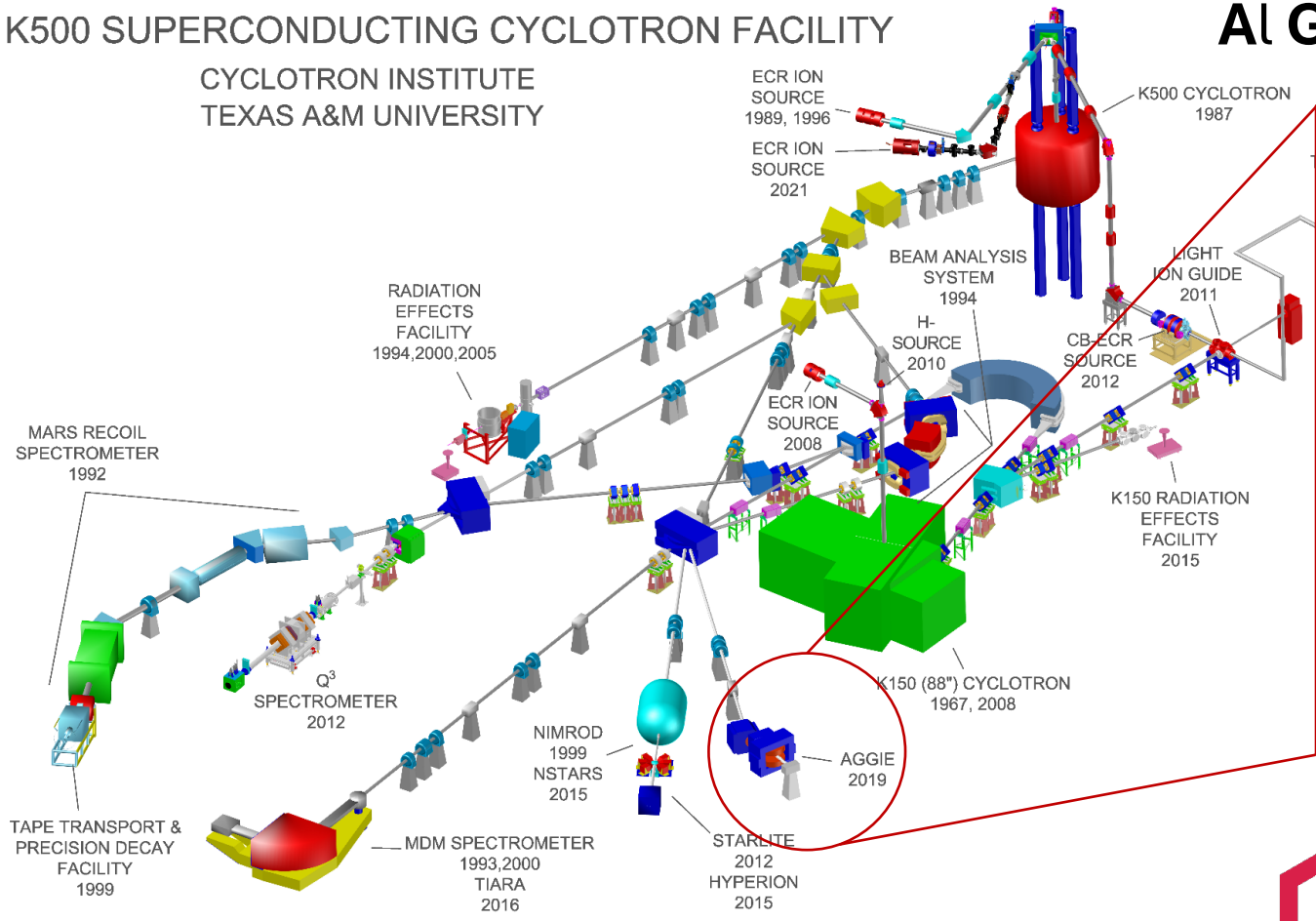
Vacuum seems more promising?

The IF-to-IVAC System

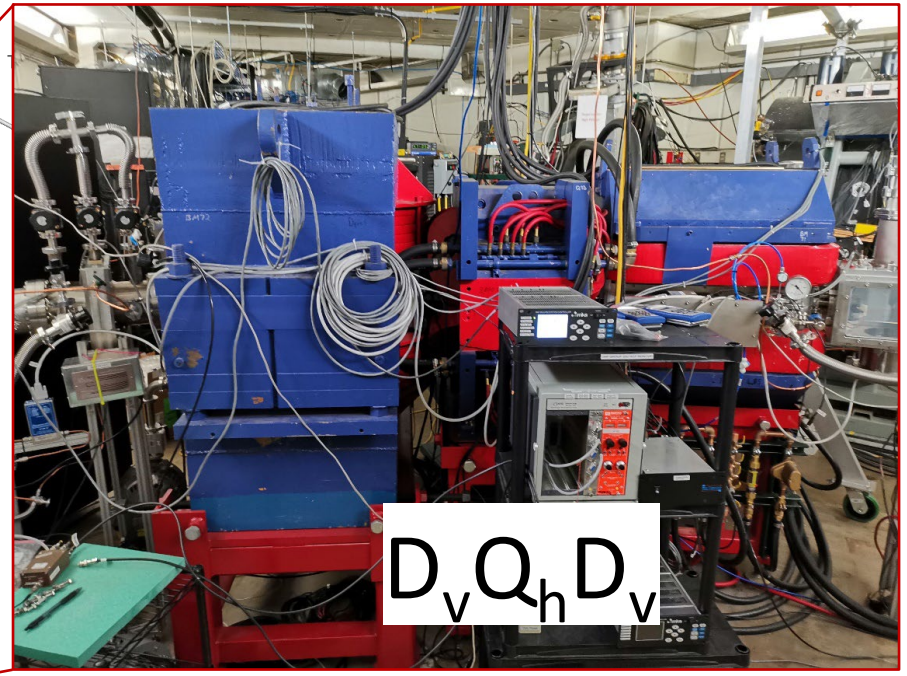


J. B. Neumayr et al., *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. At.* **2006**, *244*, 489–500.

Facilities @ Cyclotron Institute, TAMU

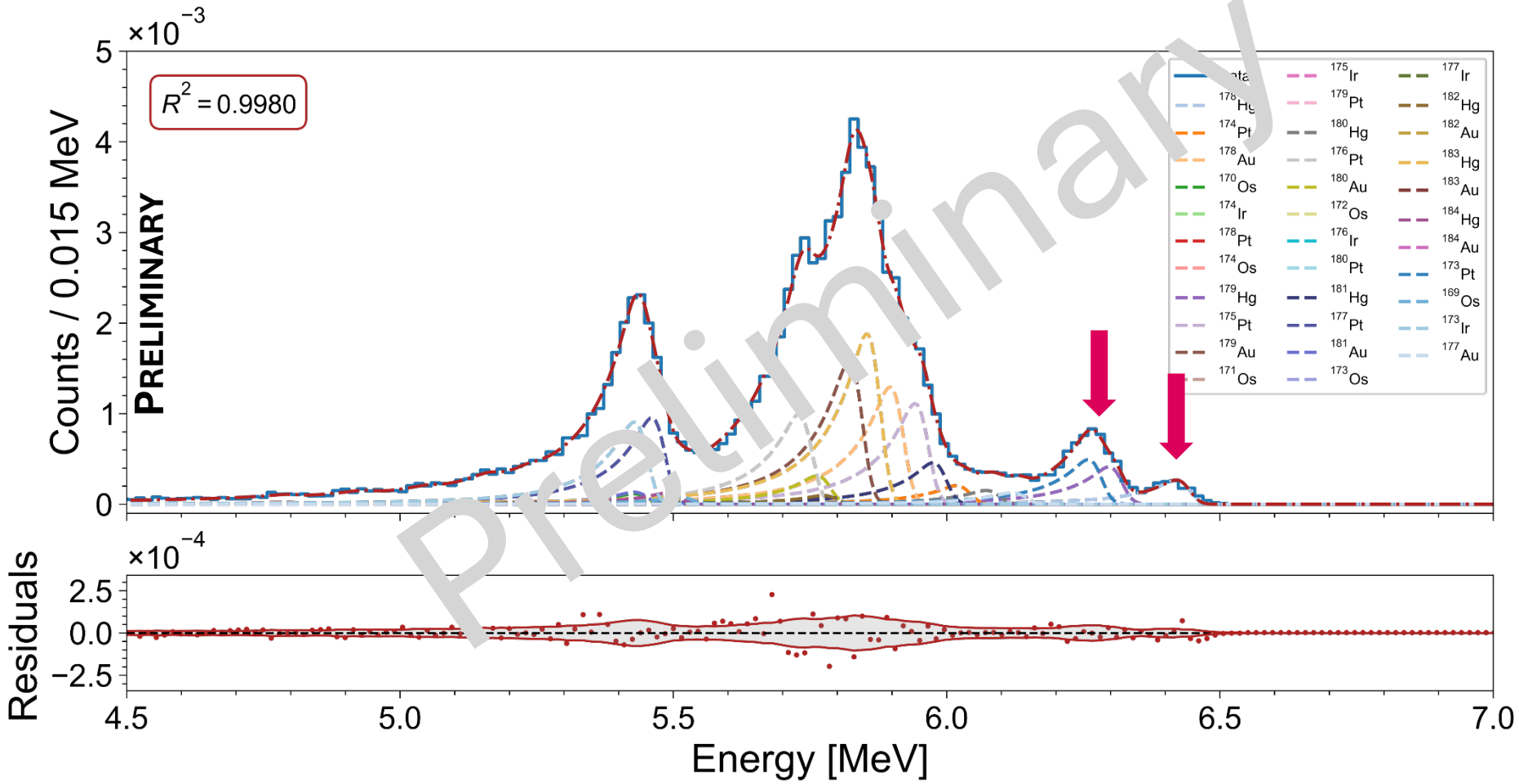


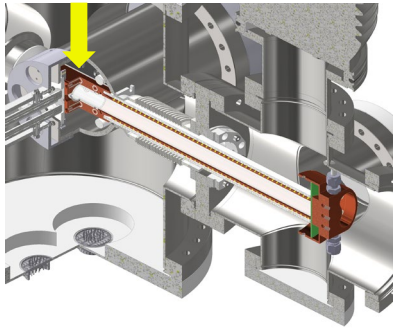
Al Ghiorso's Gas-filled Ion Equipment



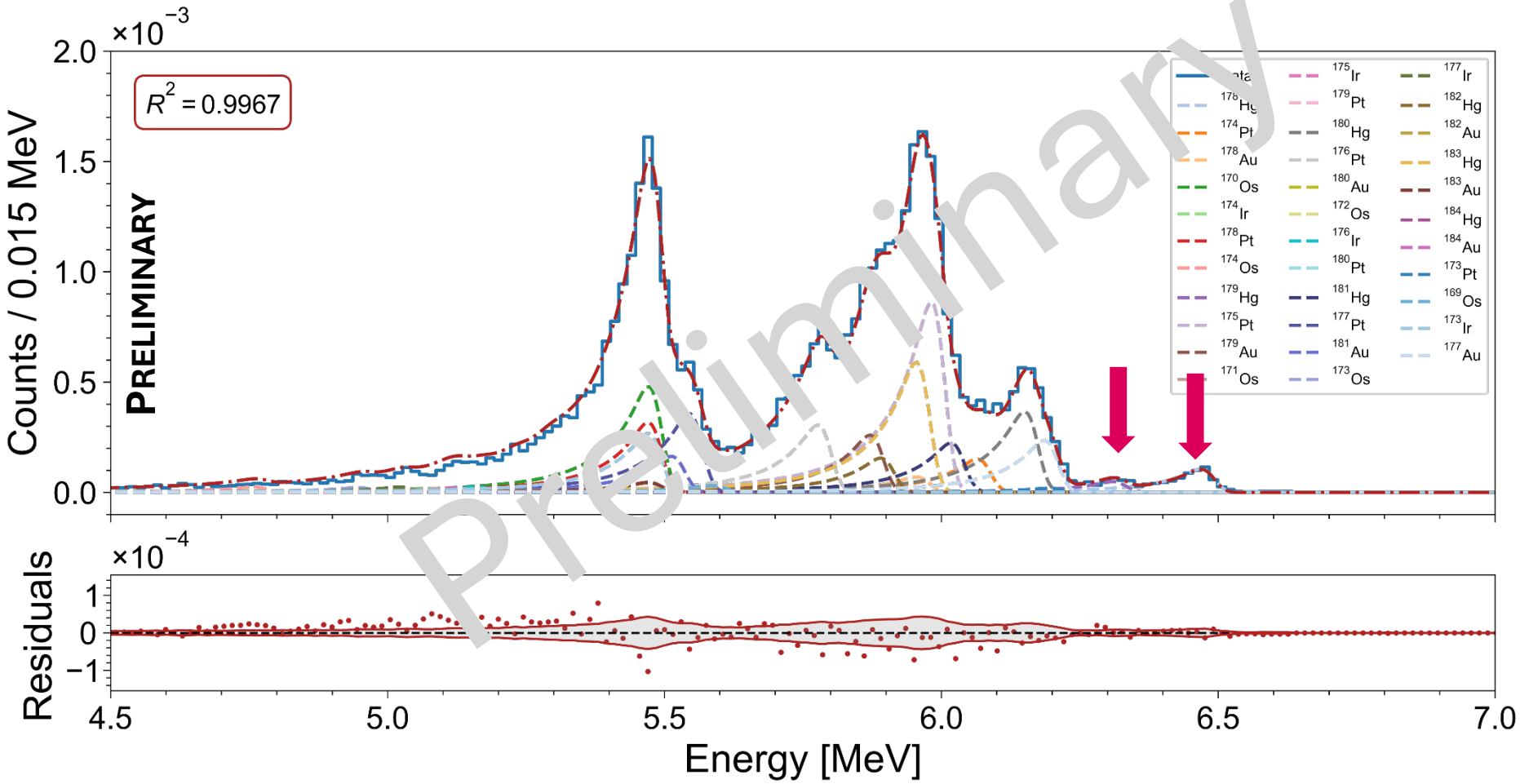
Project grant 200020_196981/1

Transport through Buffer Gas Cell @ E_{cot} = 184 MeV

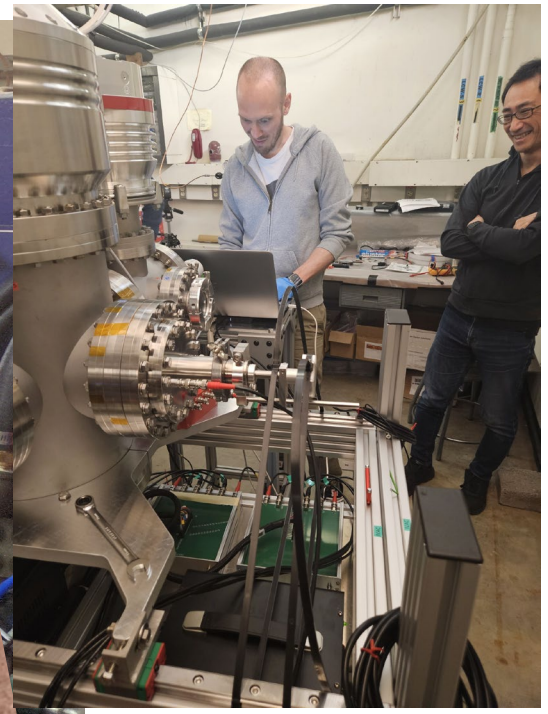




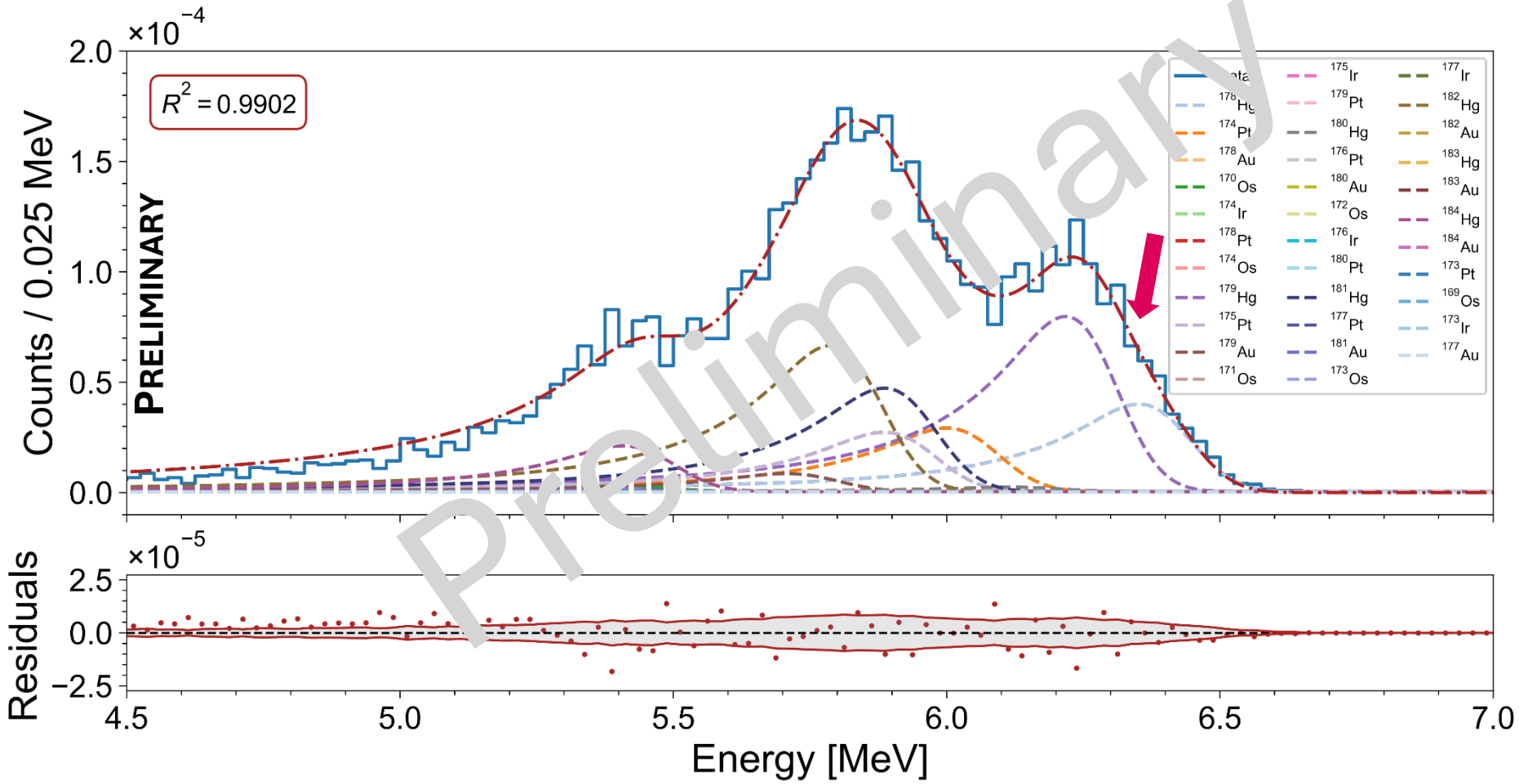
Transport through Buffer Gas Cell @ E_{cot} = 197 MeV

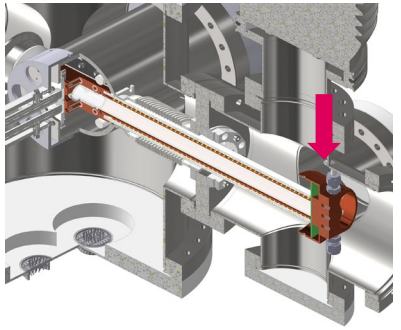


The real thing… ~2t of material

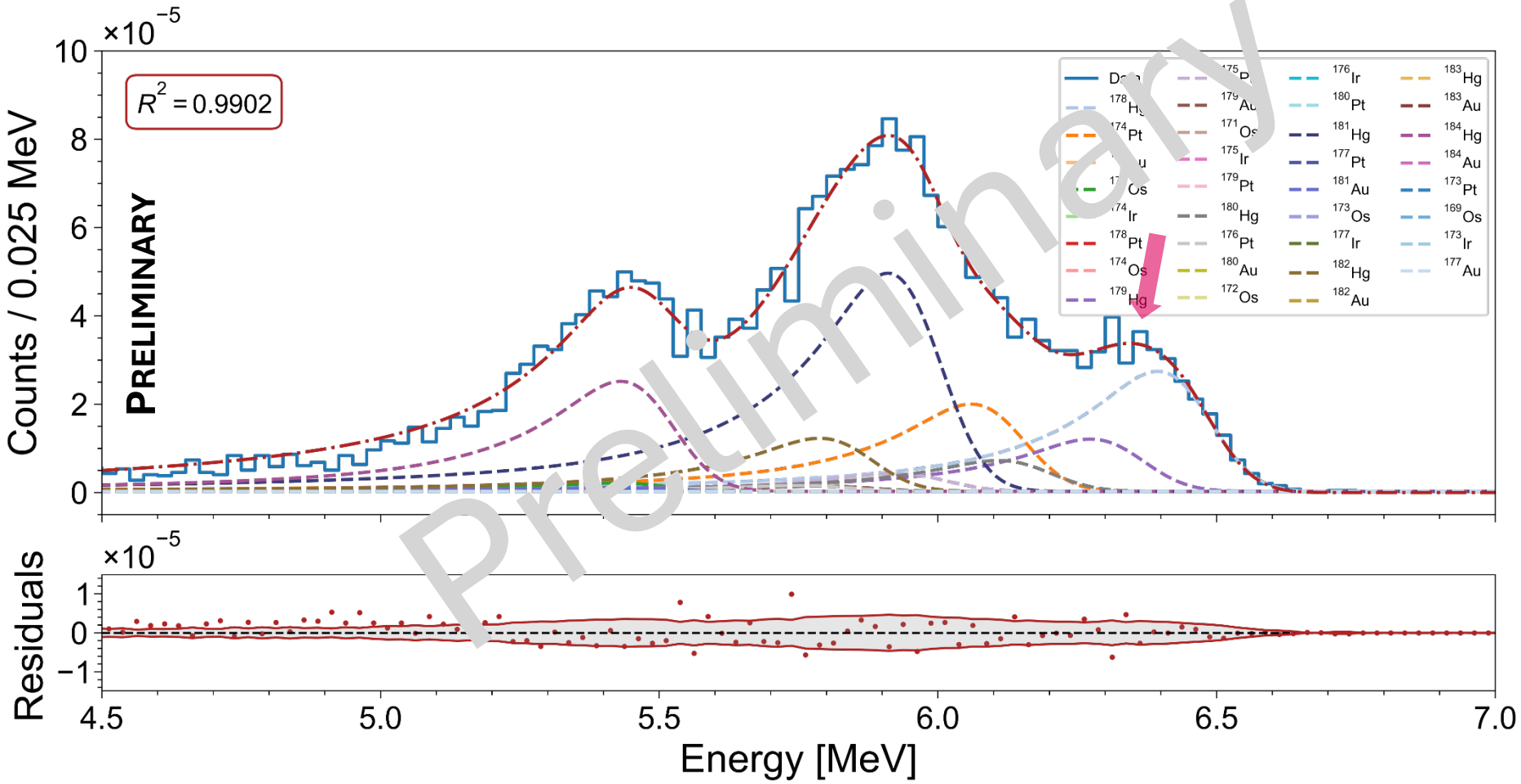


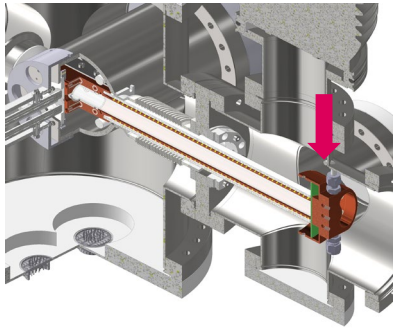
Transport through IVAC @ E_{cot} = 184 MeV



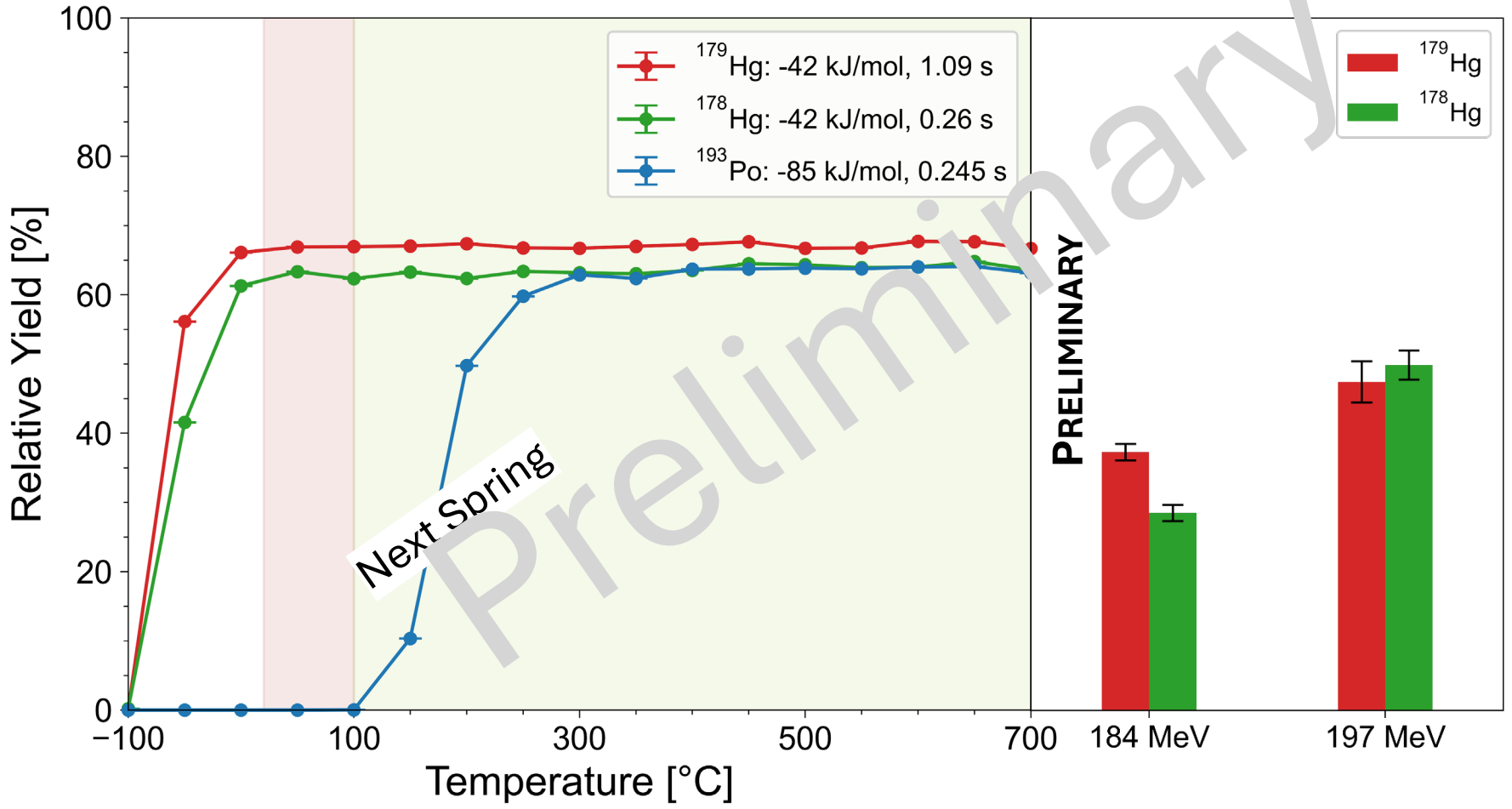


Transport through IVAC @ E_{cot} = 197 MeV





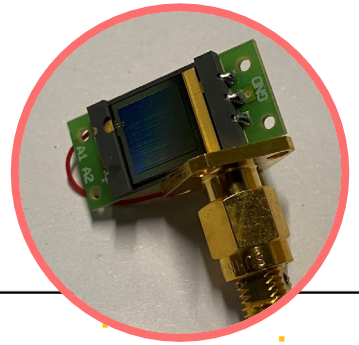
Comparison with SiO_2

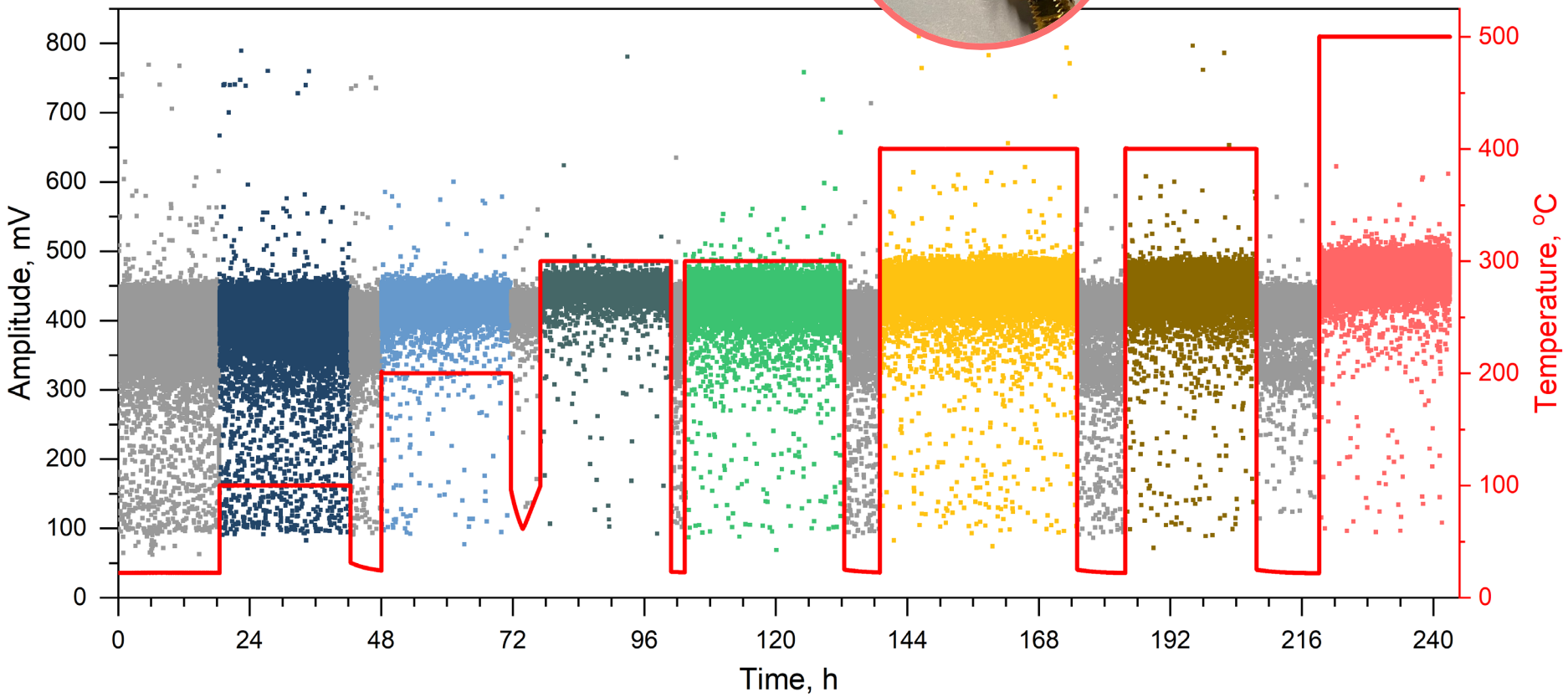


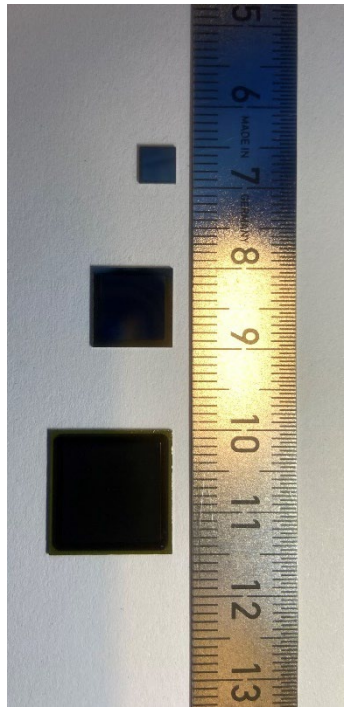
S. Soverna, Attempt to Chemically Characterize Element 112, Universität Bern, **2004**.
K. Hermainski et al., Poster @ NRC10, Brighton, **2024**.

High-temperature α-spectroscopy

After scCVD diamond: **4H-SiC**-based semiconductor solid-state detectors.





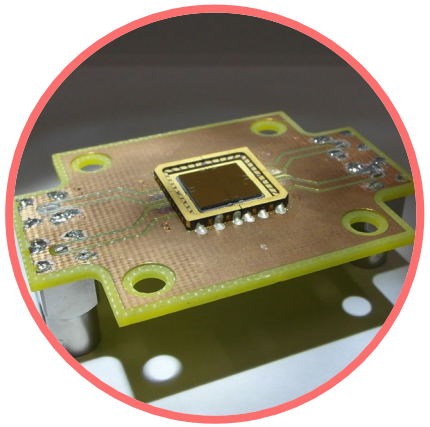


High-temperature α-spectroscopy

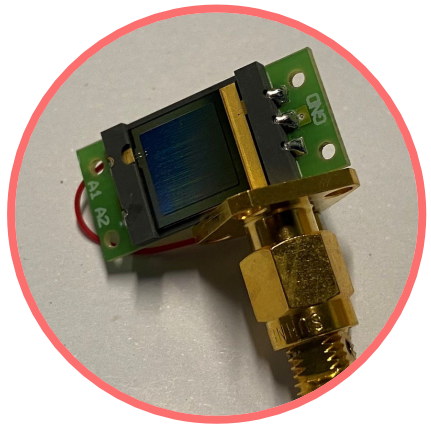
Looking at the predictions for Nh on SiO_2: **Higher stationary phase temperatures** needed for online **thermochromatography**

High-temperature α-spectroscopy

Diamond

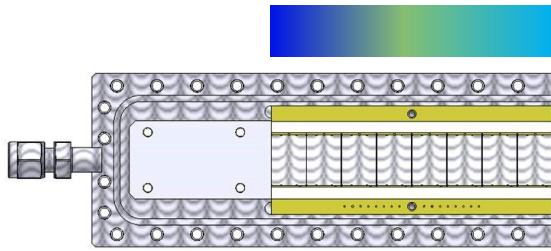


4H-SiC

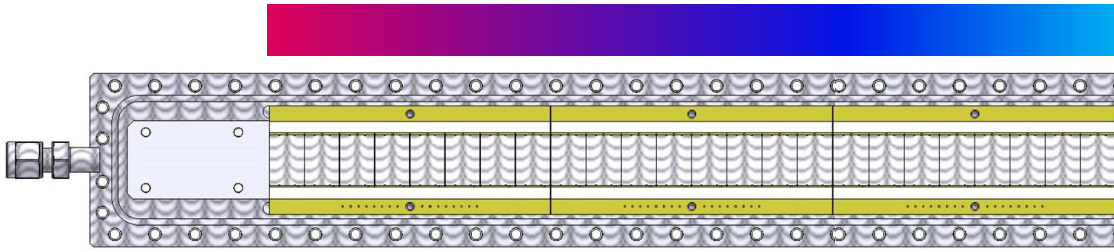


≈ 50°C

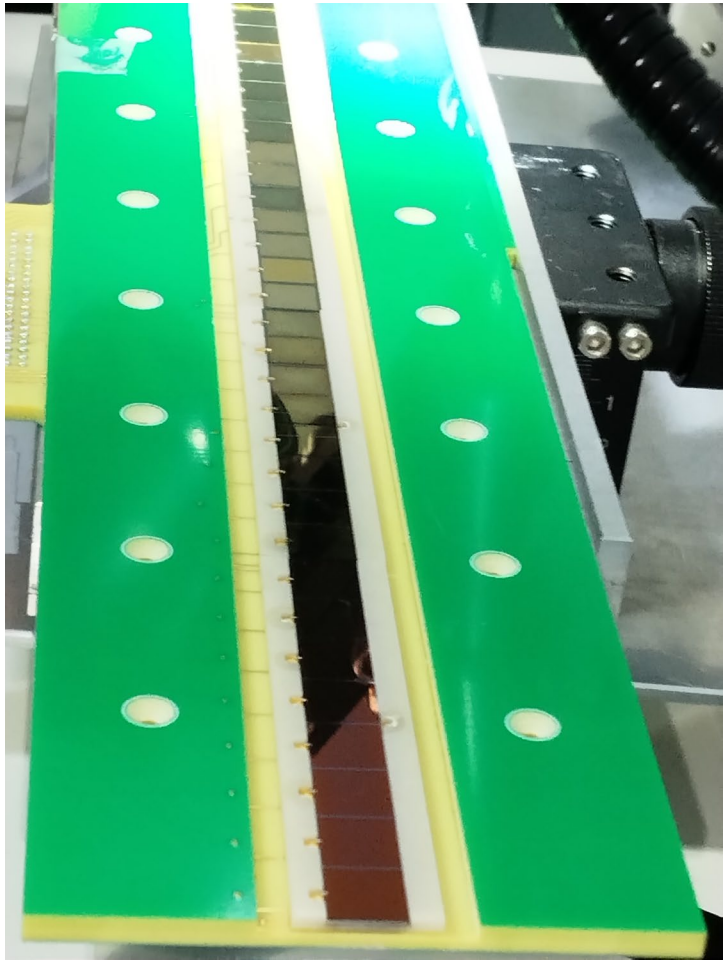
SHEs



≈ 500°C



LEGEND - first-generation SiC Thermochromatography (65°C → -40°C) for Nh



…see talk by Qin Zhi

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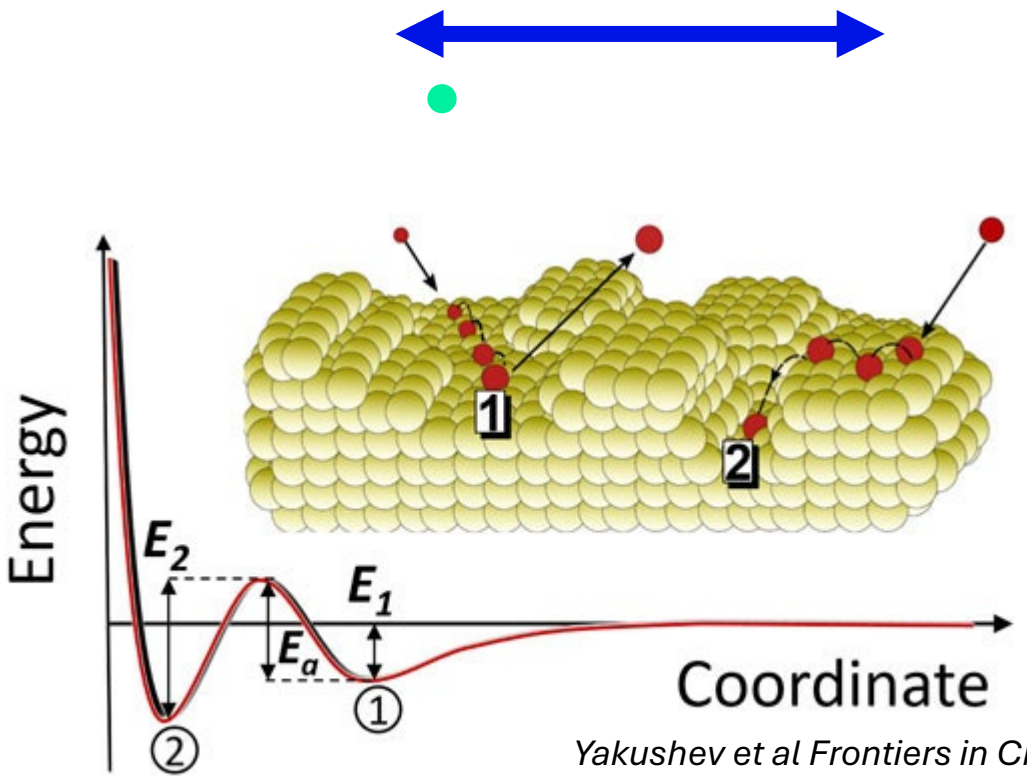
The “zero” jump between adsorptions

Wall collision number: $N_i = \frac{a+b}{\bar{V}(p,T)} \cdot \sqrt{\frac{2 \cdot \pi \cdot R \cdot T}{M_a}} \simeq$ 1000-150 [cm^{-1}] @ 298-80 K



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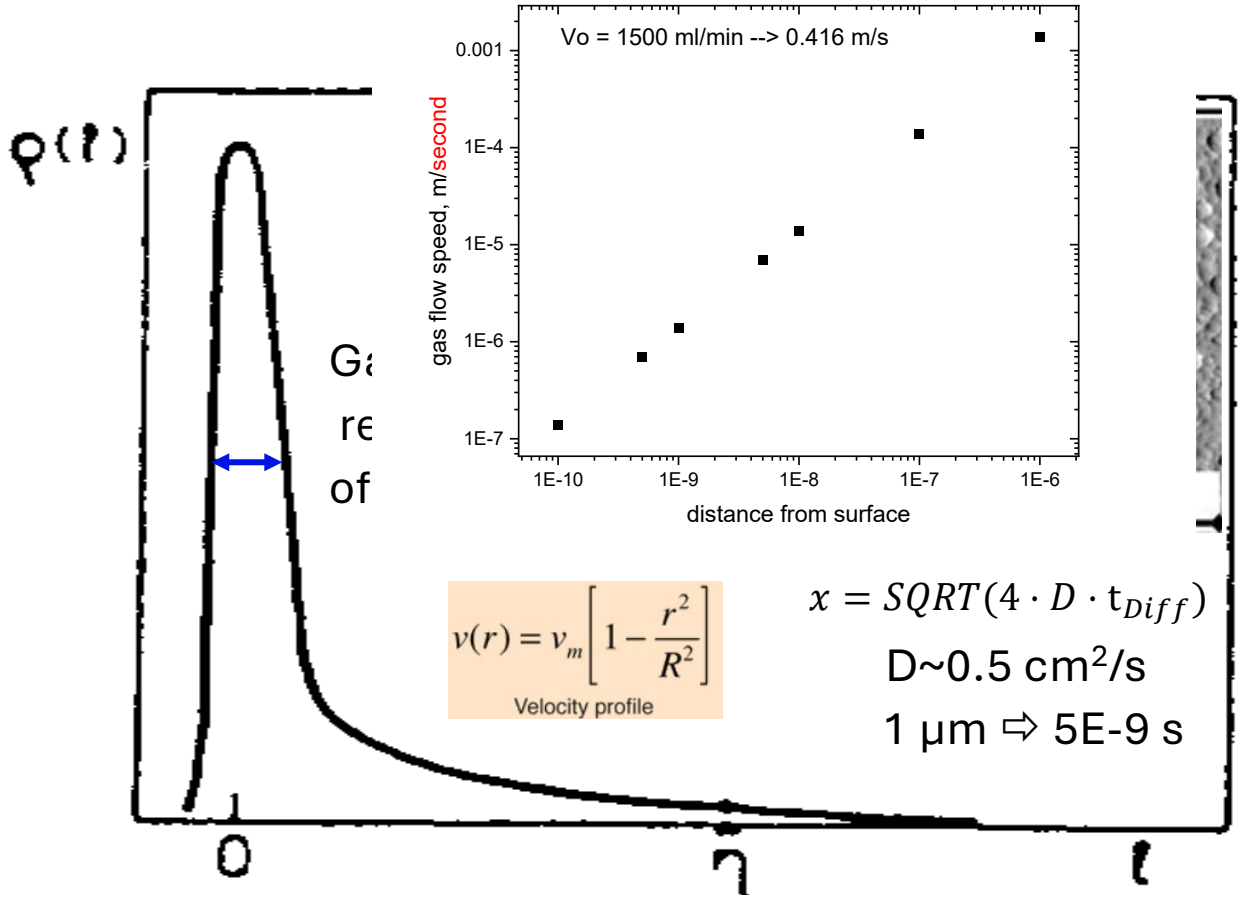
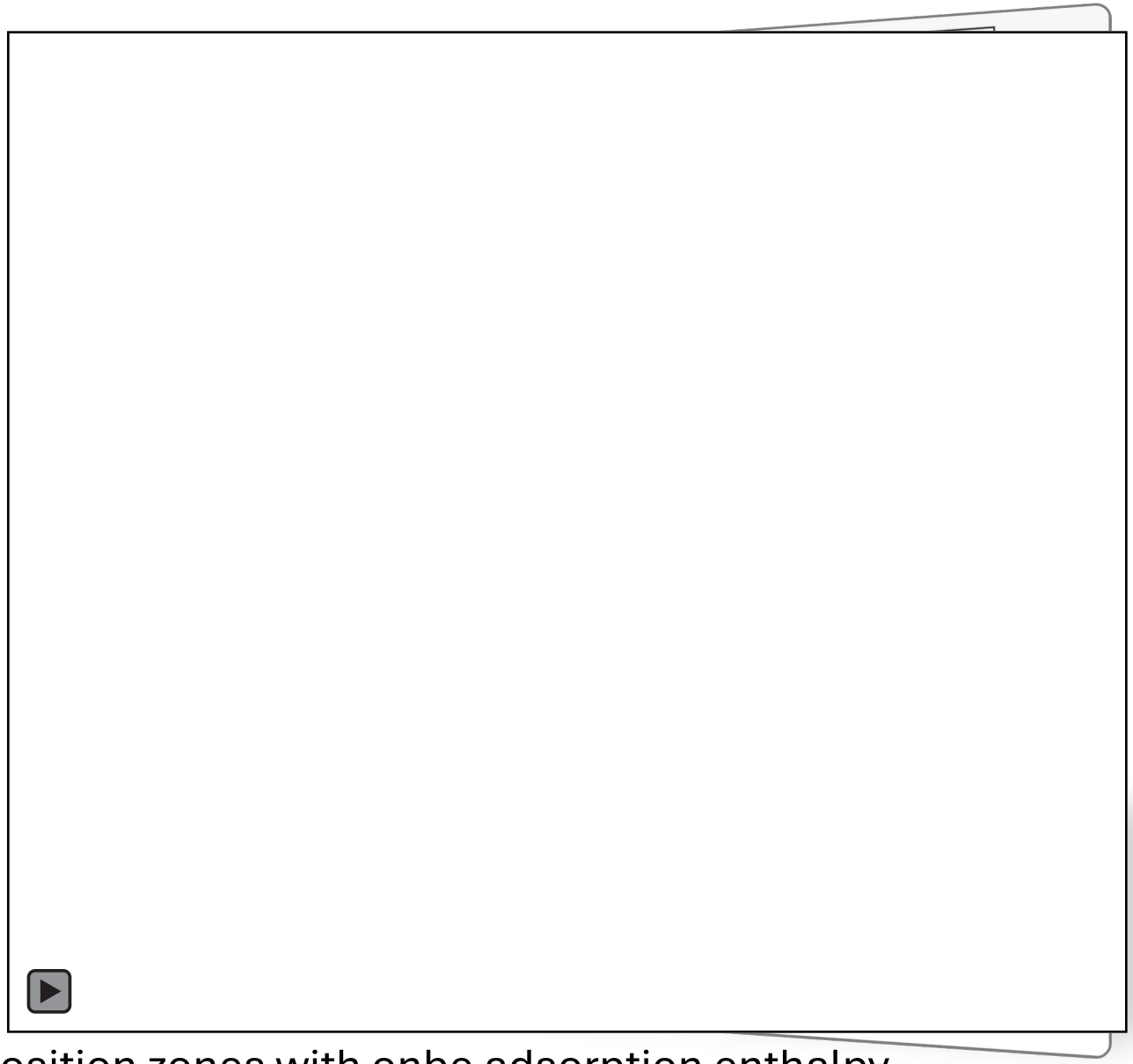
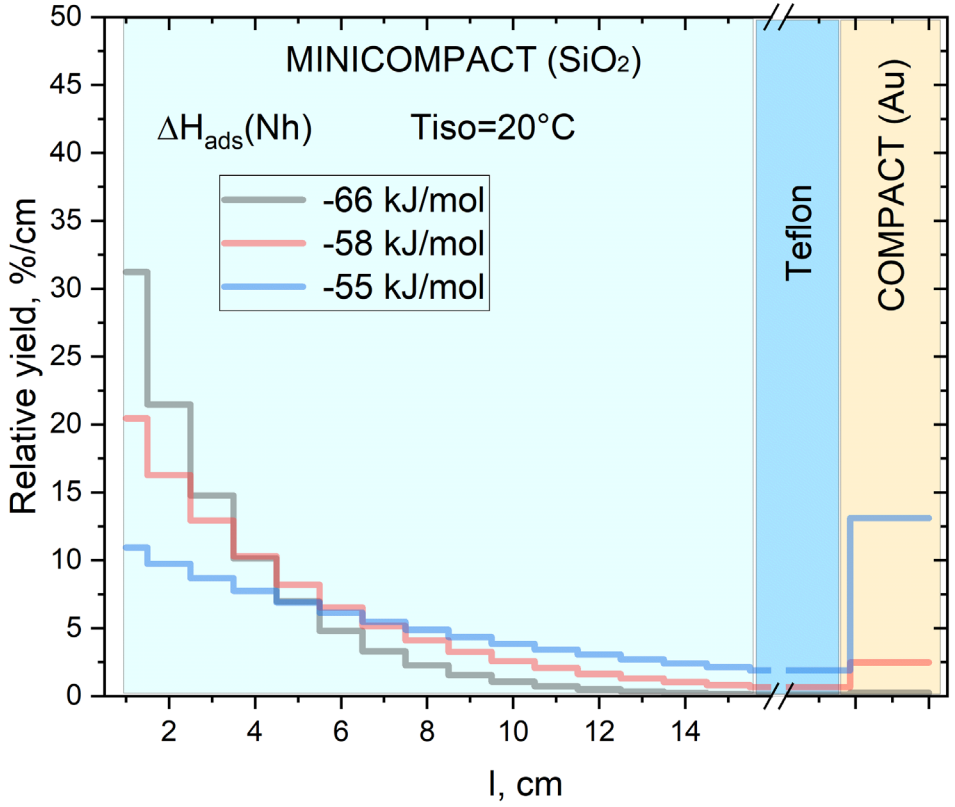


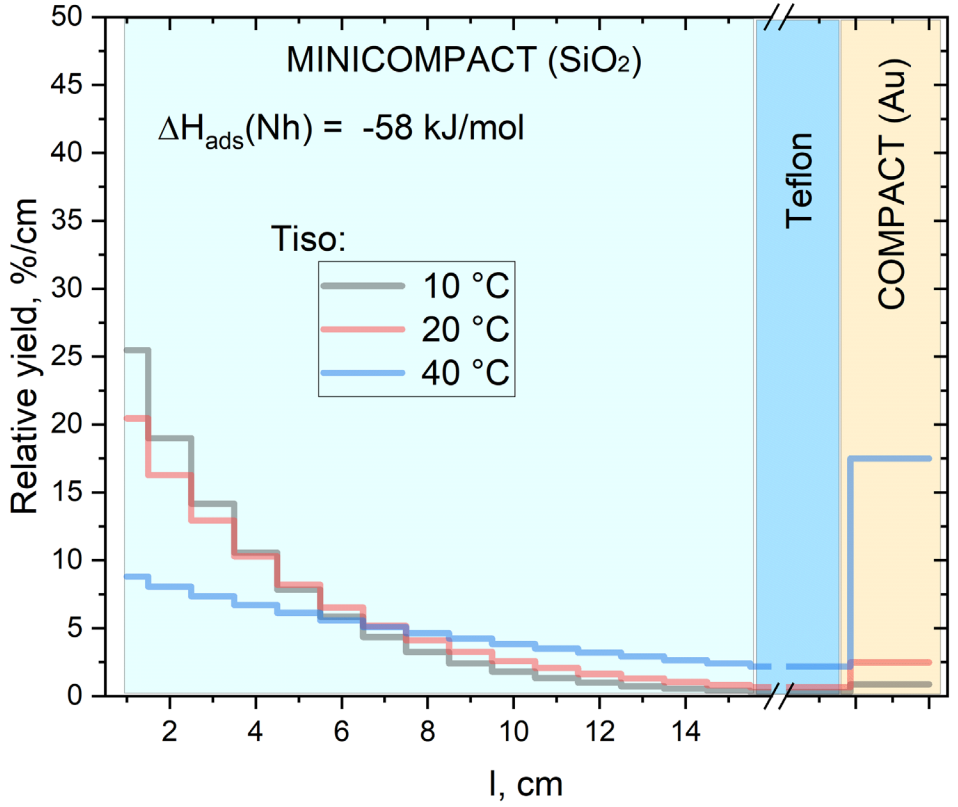
If there are inhomogeneities of the surface the atoms will “find” them efficiently and adsorb,
- this is defined as sticking coefficient!

$$SC = sc \cdot \exp\left(-\frac{E_a}{RT}\right)$$



There cannot be two deposition zones with onbe adsorption enthalpy


MINICOMPACT (SiO2)
ΔHads(Nh)
Tiso=20°C
-66 kJ/mol
-58 kJ/mol
-55 kJ/mol
Teflon
COMPACT (Au)
Relative yield, %/cm
l, cm


MINICOMPACT (SiO2)
ΔHads(Nh) = -58 kJ/mol
Tiso:
10 °C
20 °C
40 °C
Teflon
COMPACT (Au)
Relative yield, %/cm
l, cm