PAUL SCHERRER INSTITUT

PSI

ETH zürich

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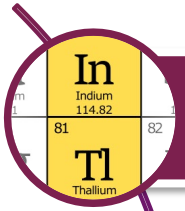
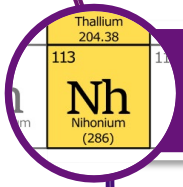
Gas-Phase Chemistry of Group 13 Elements

TASCA Workshop ‘23, ZOOM, April 26, 2023

Outline

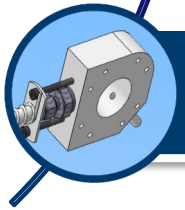


In/Tl background [online/offline]



Status of Nh experiments

Status of most recent Tl studies



Future plans

Previous Group 13 Homolog Studies

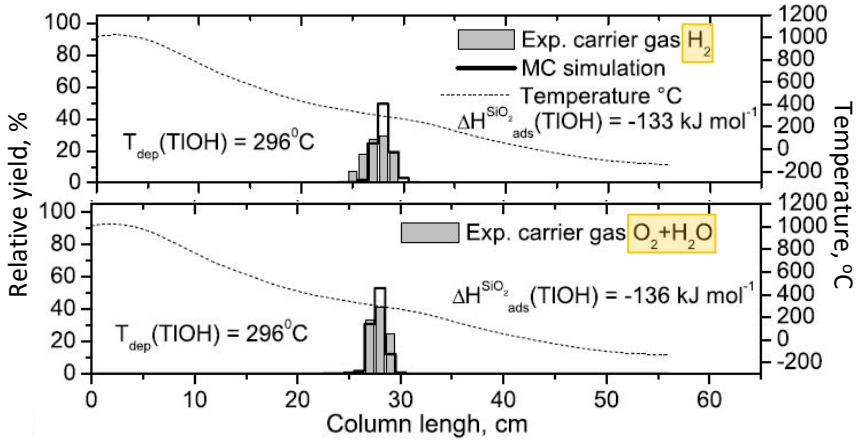
Experiments on fused silica surfaces

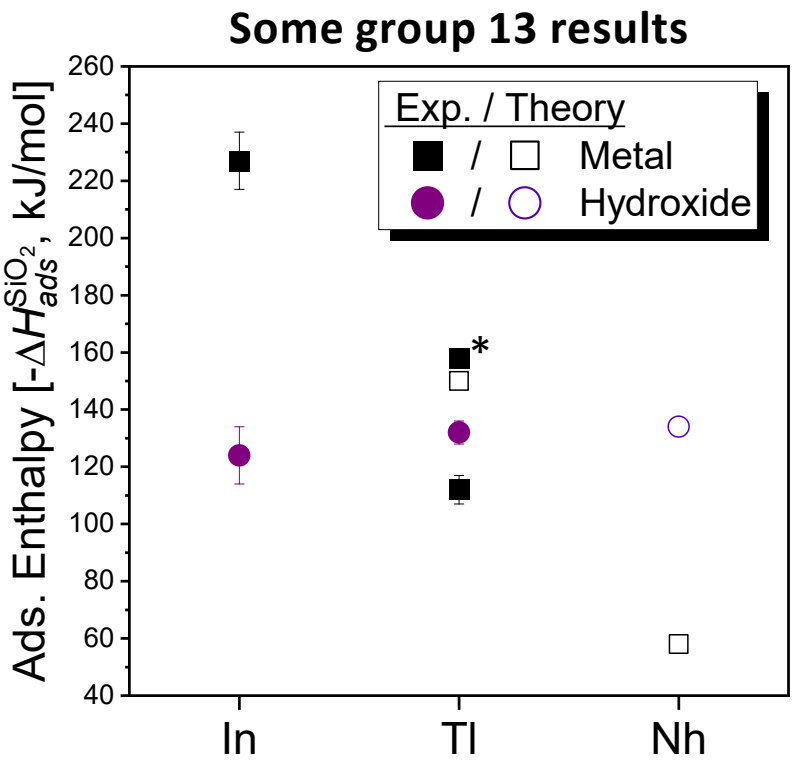
→ In/InOH: Offline thermochromatography

→ Tl/TlOH: Offline thermochromatography

→ **Tl: Online* isothermal chromatography**

→ Nh/NhOH: only theoretical studies





Eichler et al. (1977), Serov et al.(2011&2013), Steinegger (2016), Pershina et al. (2016 & 2018)

Previous Nh Chemistry Experiments

S. N. Dmitriev et al. (2014)

→ 5 decay chains observed

→ $-\Delta H_{ads}^{\mathrm{Au}}(\mathrm{Nh\ species}) > 60$ kJ/mol

N. V. Aksenov et al. (2017)

→ Repetition of S. N. Dmitriev's experiments

→ Exchanged SiO_2 surfaces to PTFE

→ $-\Delta H_{ads}^{\mathrm{PTFE}}(\mathrm{Nh}) > 45$ kJ/mol

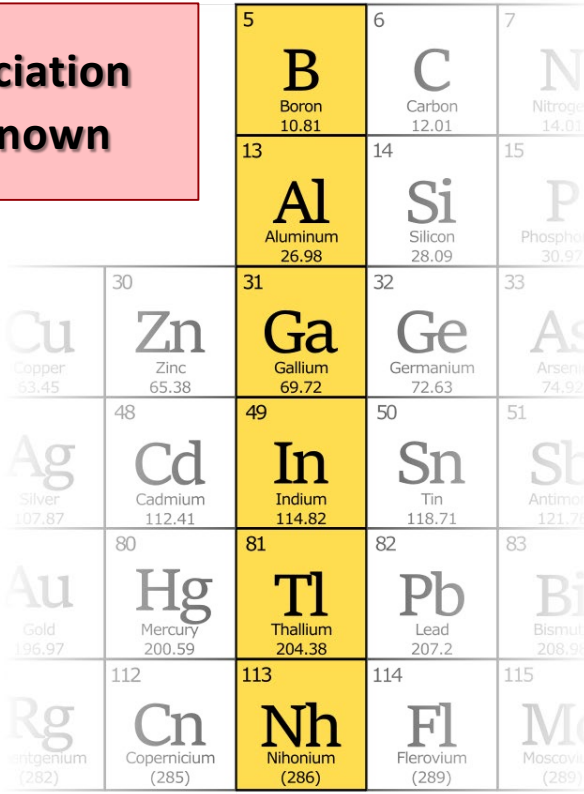
A. Yakushev et al. (2021)

→ No decay chains observed

→ possible relatively non-volatile Nh species

→ Confirmed adsorption limits of 2017 experiment

Chemical speciation remains unknown



Towards the Next Nh Experiment

Chemistry preparations

→ On- & offline homolog experiments

→ Secure one chemical species for evaluation

→ Defined and stable stationary phase

Setup preparations

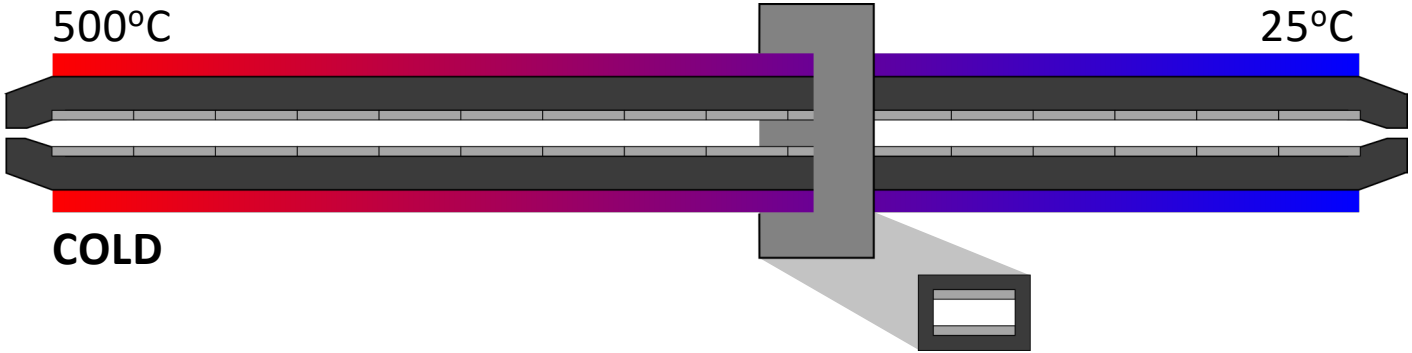
→ $-\Delta H_{ads}^{SiO_2}$(Nh/NhOH) ≈ 60 / 130 kJ/mol

→ need $T_{surface}$ > 80°C & 350°C

→ Adaptation of COLD towards high temperatures (500°C)

→ Detectors: 4H-SiC (Georg Tiebel's talk)

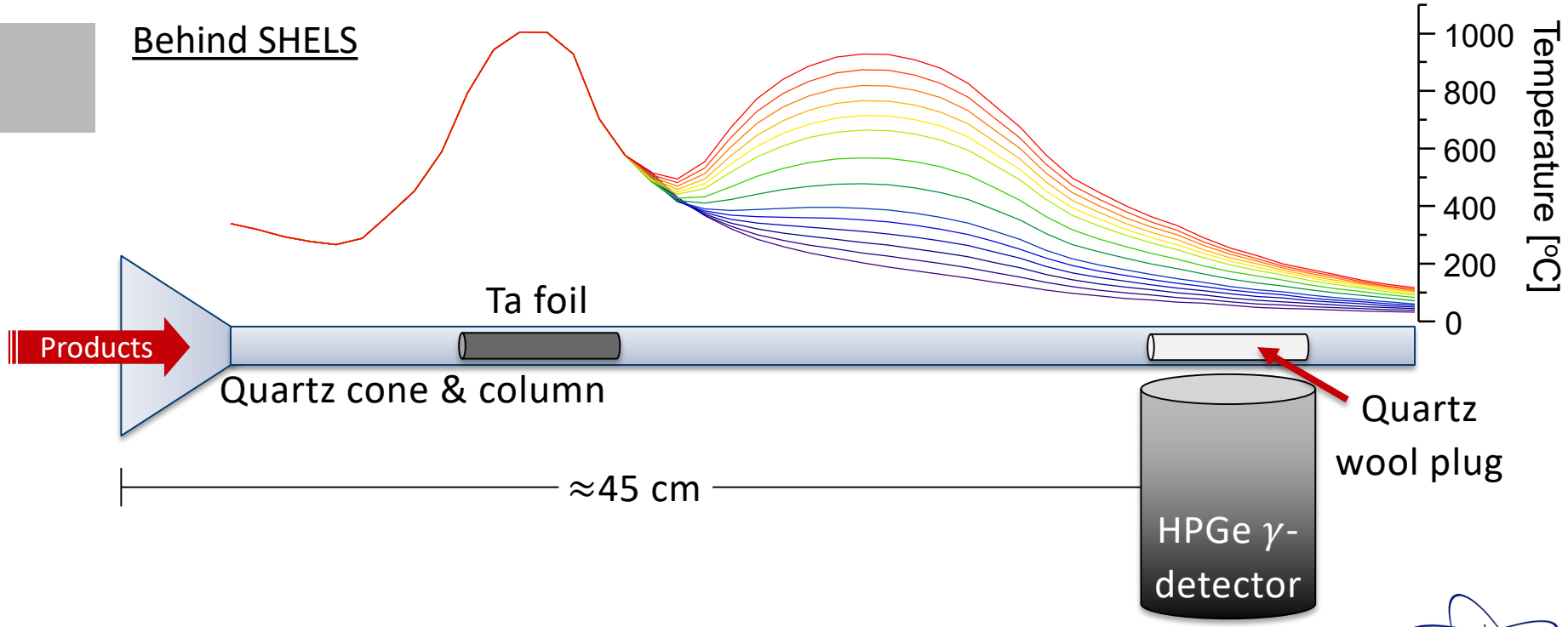
→ Hardware upgrades



Cross section of COLD

Pershina et al. (2016 & 2018)

Online Experiment at FLNR/JINR



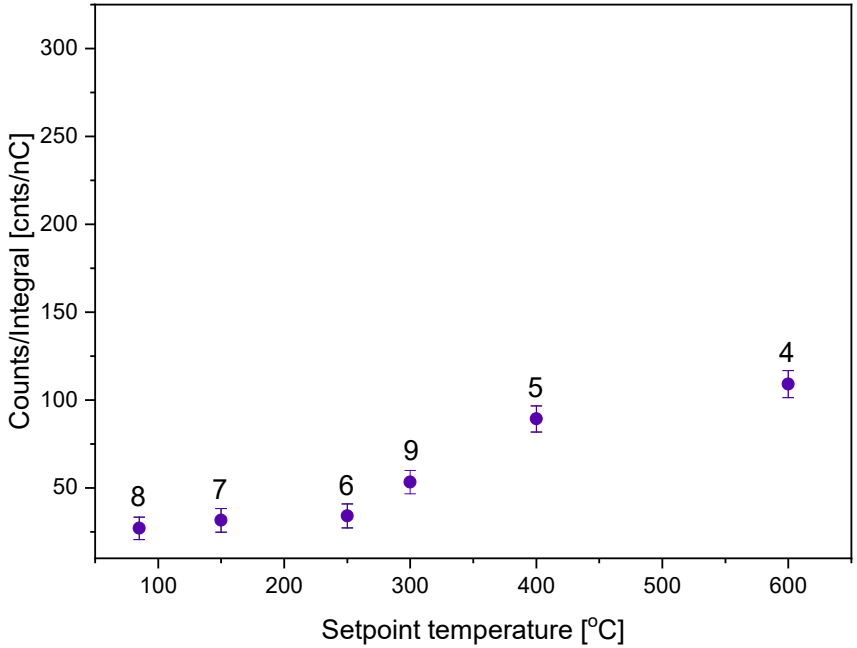
Reaction: $^{141}Pr(^{48}Ti, xn)^{189-x}Tl$ (x = 4,5), 255 MeV ion-beam energy

At room temperature (no Ta foil) a volatile species was seen!

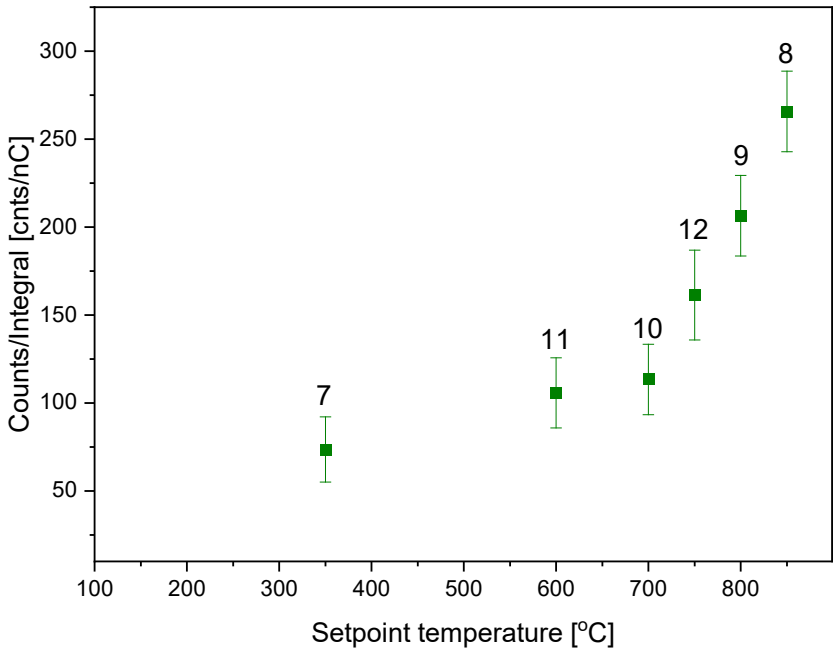
Experiment Evaluation

External chromatograms on a dehydroxylated surface

Experiment #1



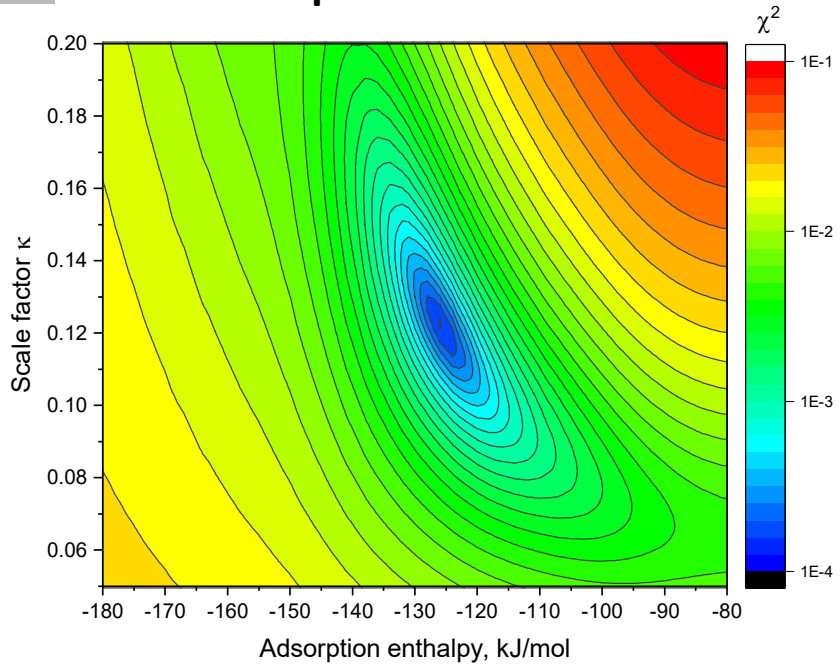
Experiment #2



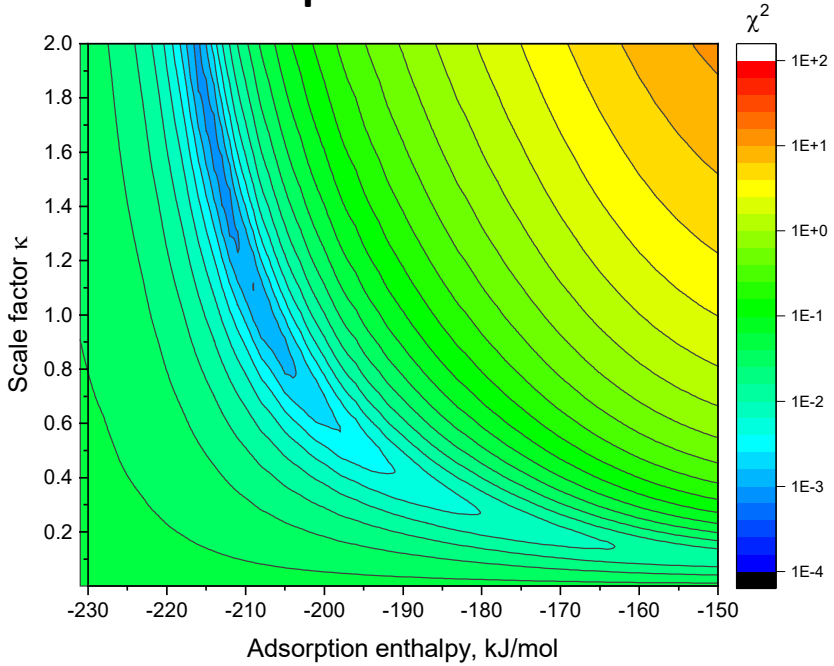
Experiment Evaluation

MCS of experimental results + χ^2-minimization

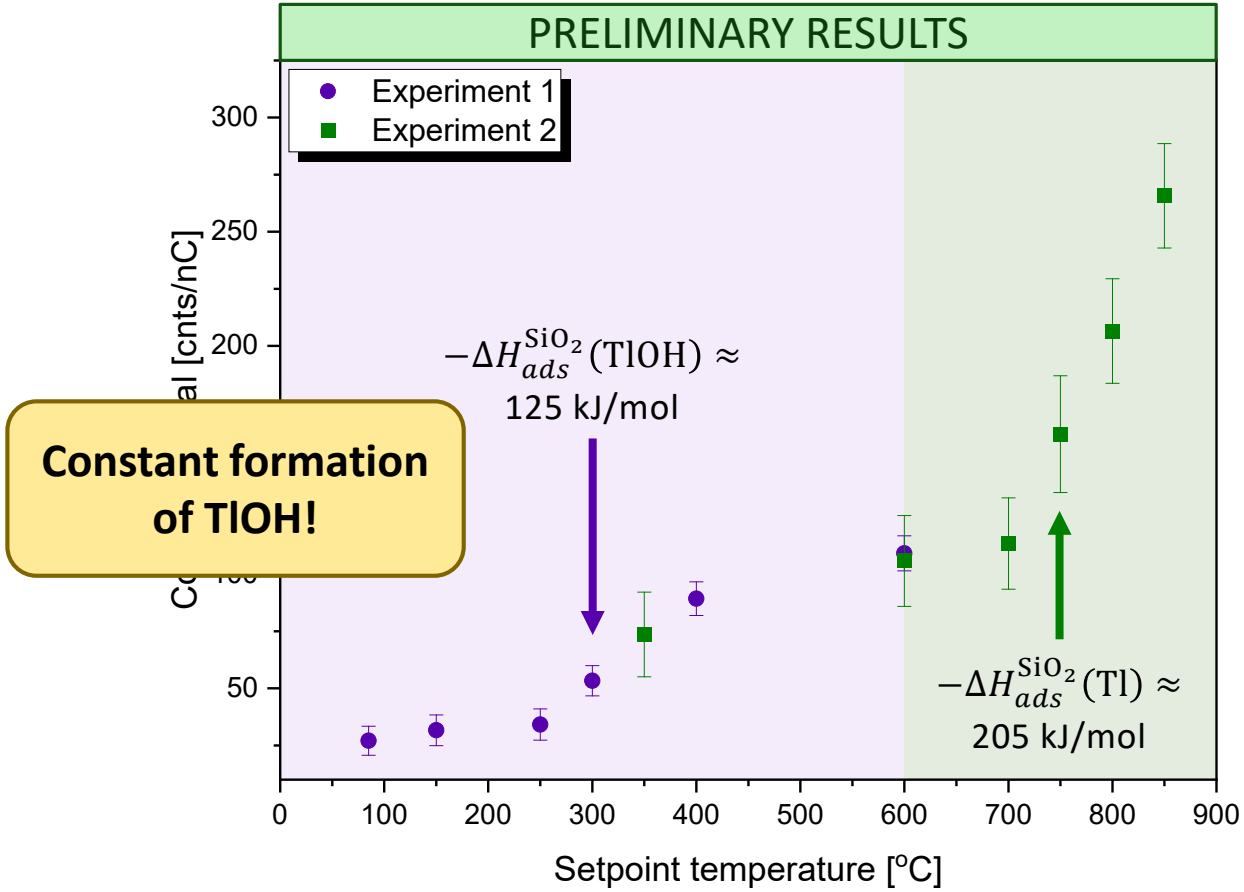
Experiment #1



Experiment #2



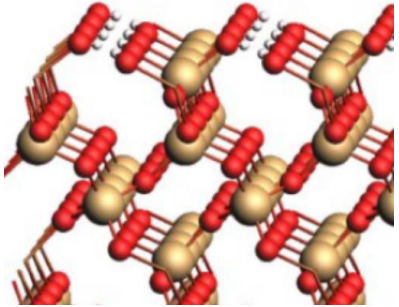
Online Experiment at FLNR/JINR



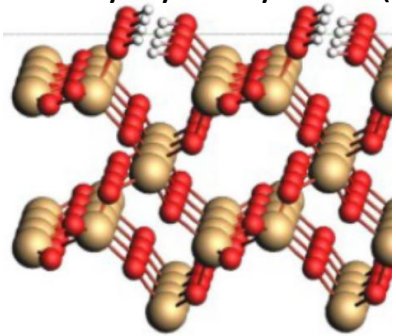
Theoretical Adsorption Studies

	$-\Delta H_{ads}^{SiO_2}$ [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

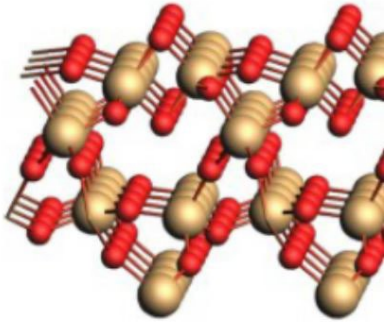
Fully hydroxylated (G)



Partially hydroxylated (V)



Dehydroxylated (B)



Ilias, M. et Pershina, V. (2022).

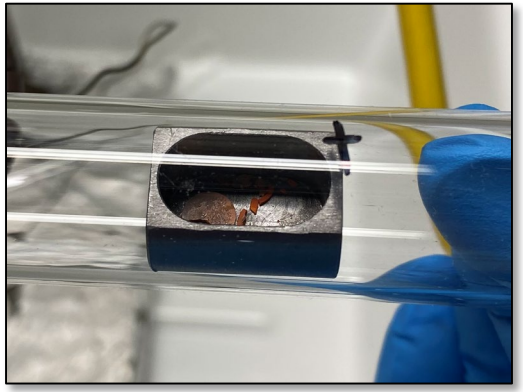
New Offline Studies at PSI

$^{nat}HgO(p,xn)^{206-x}Tl$ & $^{nat}HgO(p,pxn)^{205-x}Hg$, x = 3,4

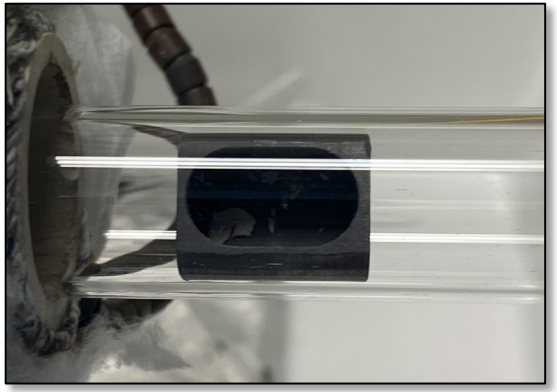
Separation: $2HgO \xrightarrow{heat} 2Hg + O_2$ at 525°C in Ta crucible under Ar flow → Tl remains

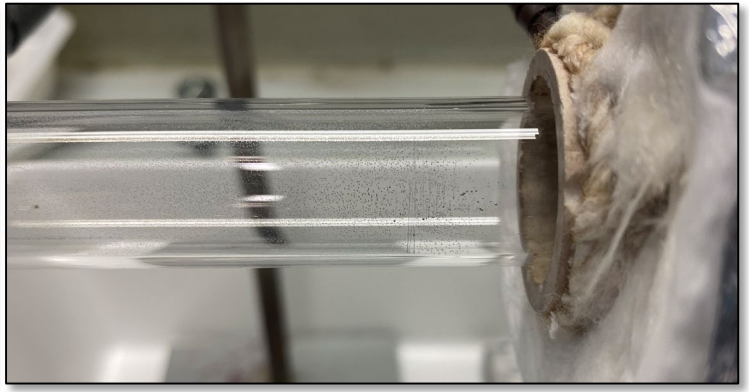
Tl transferred to Ta foil for thermochromatography experiments

Tl/Hg identified in different fractions via decay of ^{203}Hg and ^{202}Tl with HPGe detector



Before

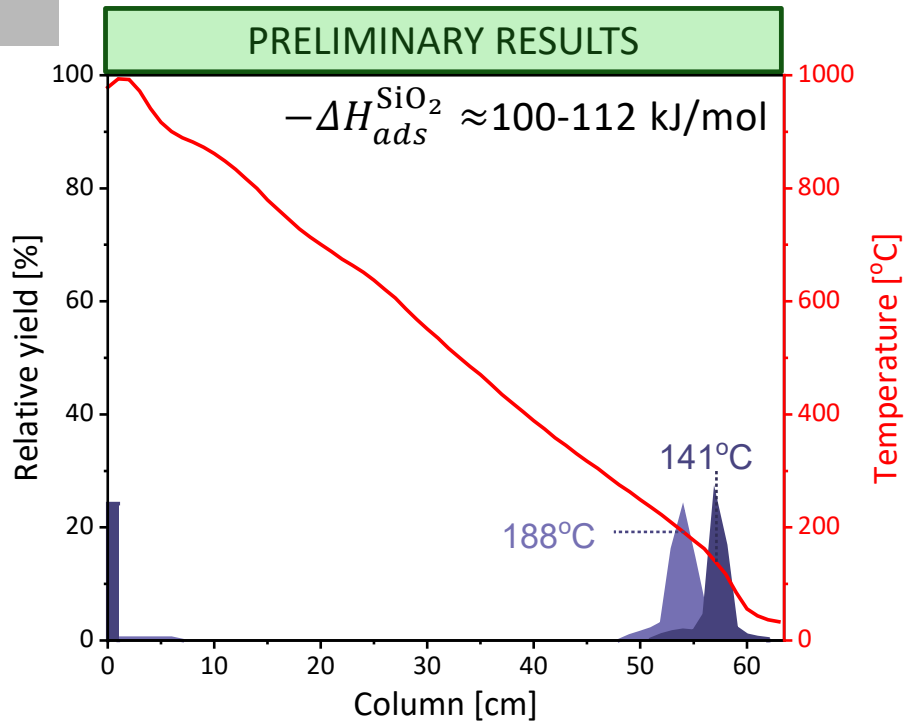




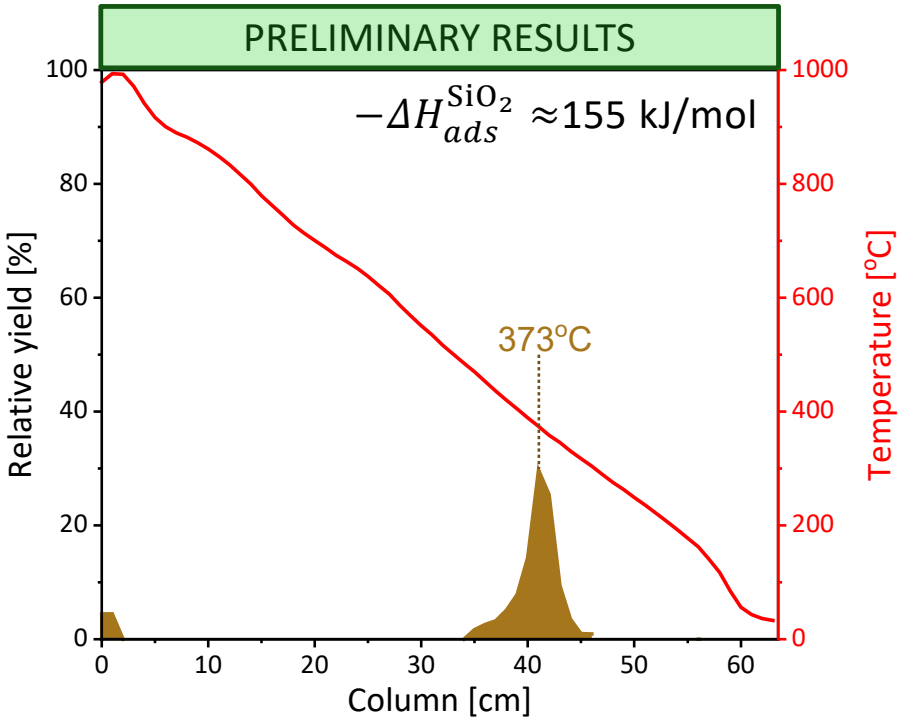
After

New Offline Studies at PSI

Dehydroxylated Surface



Untreated Surface



Dehydroxylation: fused silica column >620 °C for 3 h

Next Studies at JAEA

Isothermal chromatography

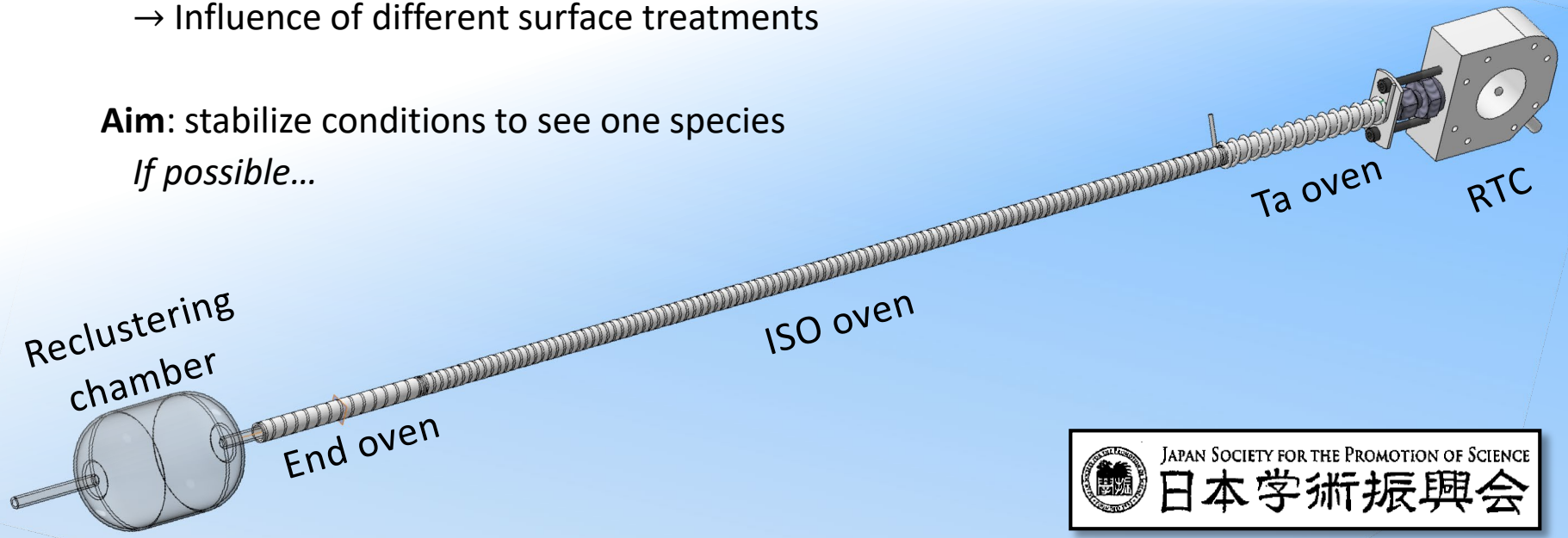
→ Better ISO ovens (more uniform temperature profiles)

→ Ta oven & escape oven

→ Influence of different surface treatments

Aim: stabilize conditions to see one species

If possible…



Thank you for your attention

My thanks go to

- Prof. Dr. Patrick Steinegger
- Prof. Dr. Robert Eichler
- PSI: D. Herrmann, P. V. Grundler, N. P. van der Meulen, A. Sommerhalder, R. Dressler, V. Zobnin
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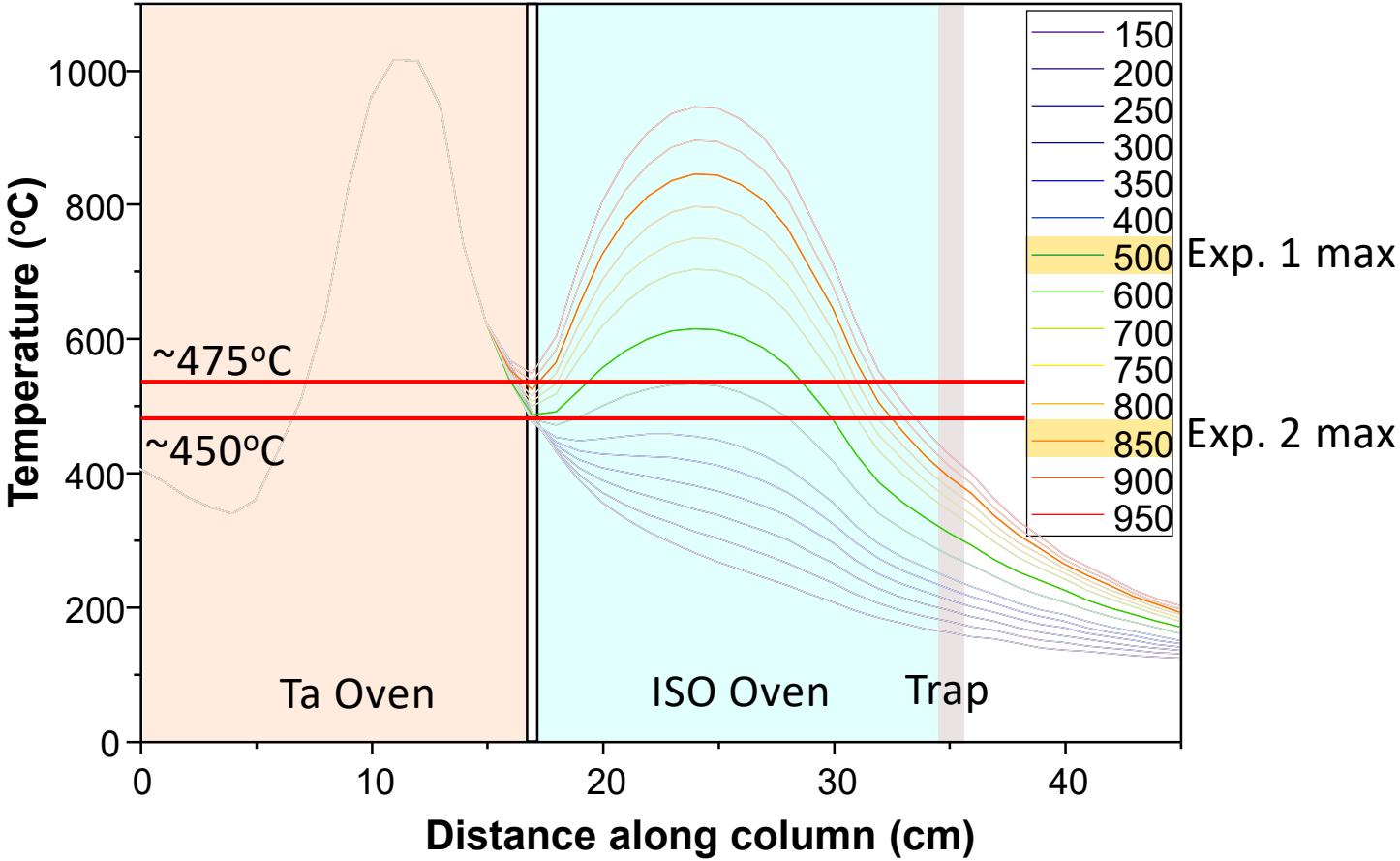
- University of Strasbourg: Z. Asfari, B. Gall
- JAEA: T. Sato

Thank you for your attention

Temperature Profiles from FLNR/JINR



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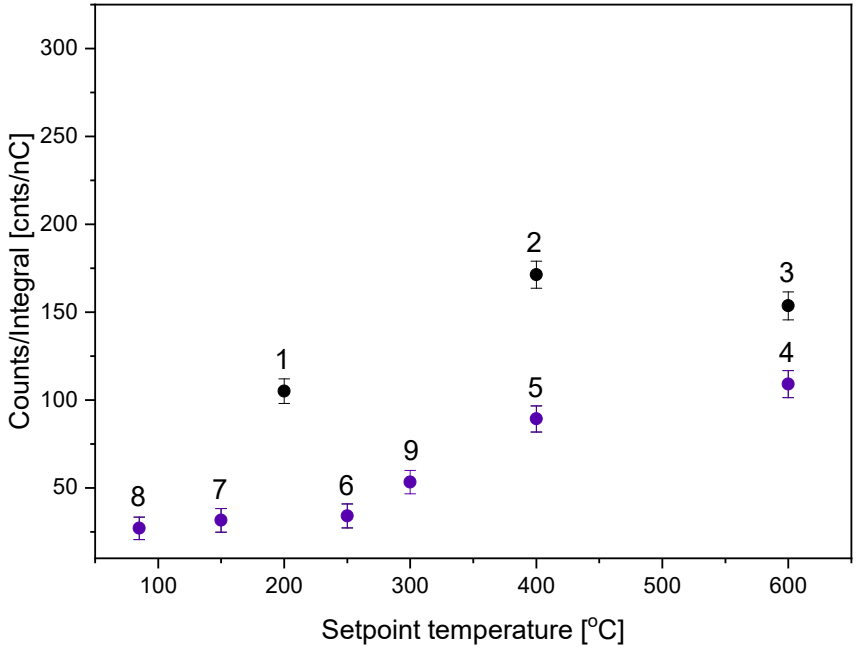

Experiment #2