PAUL SCHERRER INSTITUT

UNIVERSITÄT BERN

Wir schaffen Wissen – heute für morgen

Gas phase chemistry with SHE – Experiments

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Outline SHE Chemistry

Constraints are:
- short half-lives 1 min-1 s and below
- low production rates

Theory:
- ask the right questions

SHE Experiment:
- data analysis, results
- thermodynamic & kinetic models

Adequate technique for single atoms:
- mulitstep processes
- separation, detection
- model experiments
- kinetic & thermodynamic model description

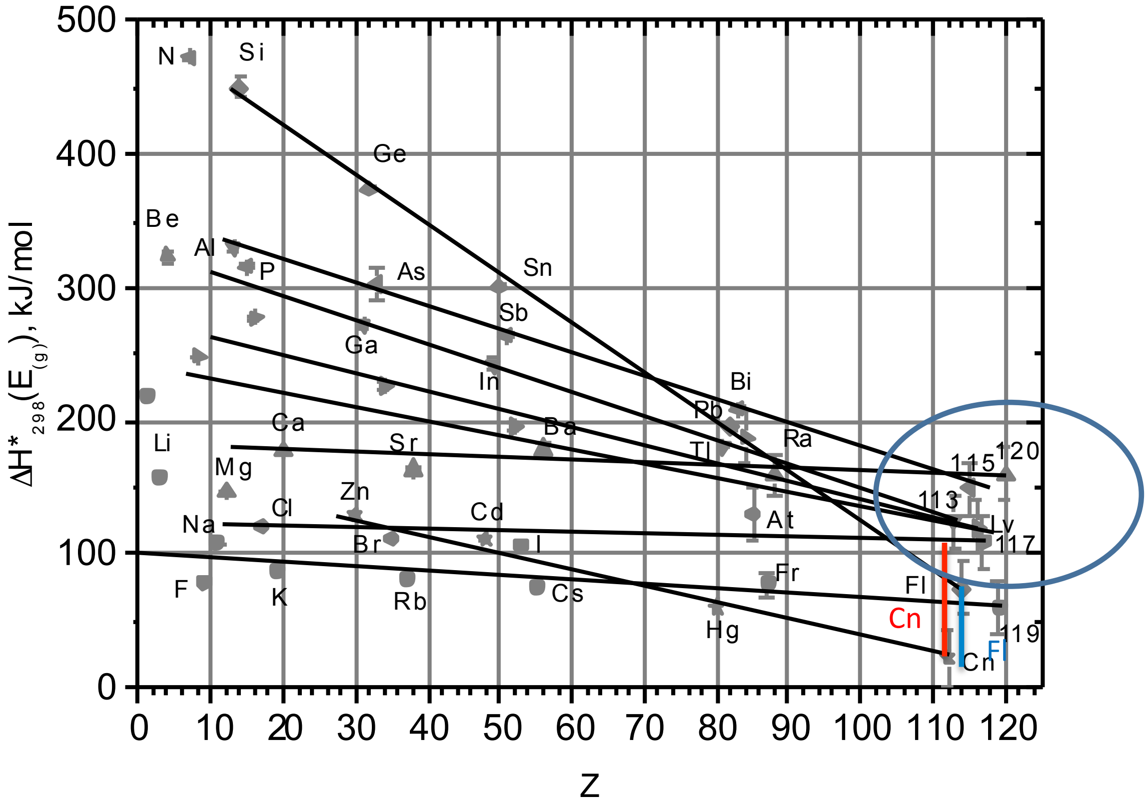
Future Transactinide Chemistry

Summary s- and p- elements

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	113	Fl	115	Lv	117	118
119	*120*																

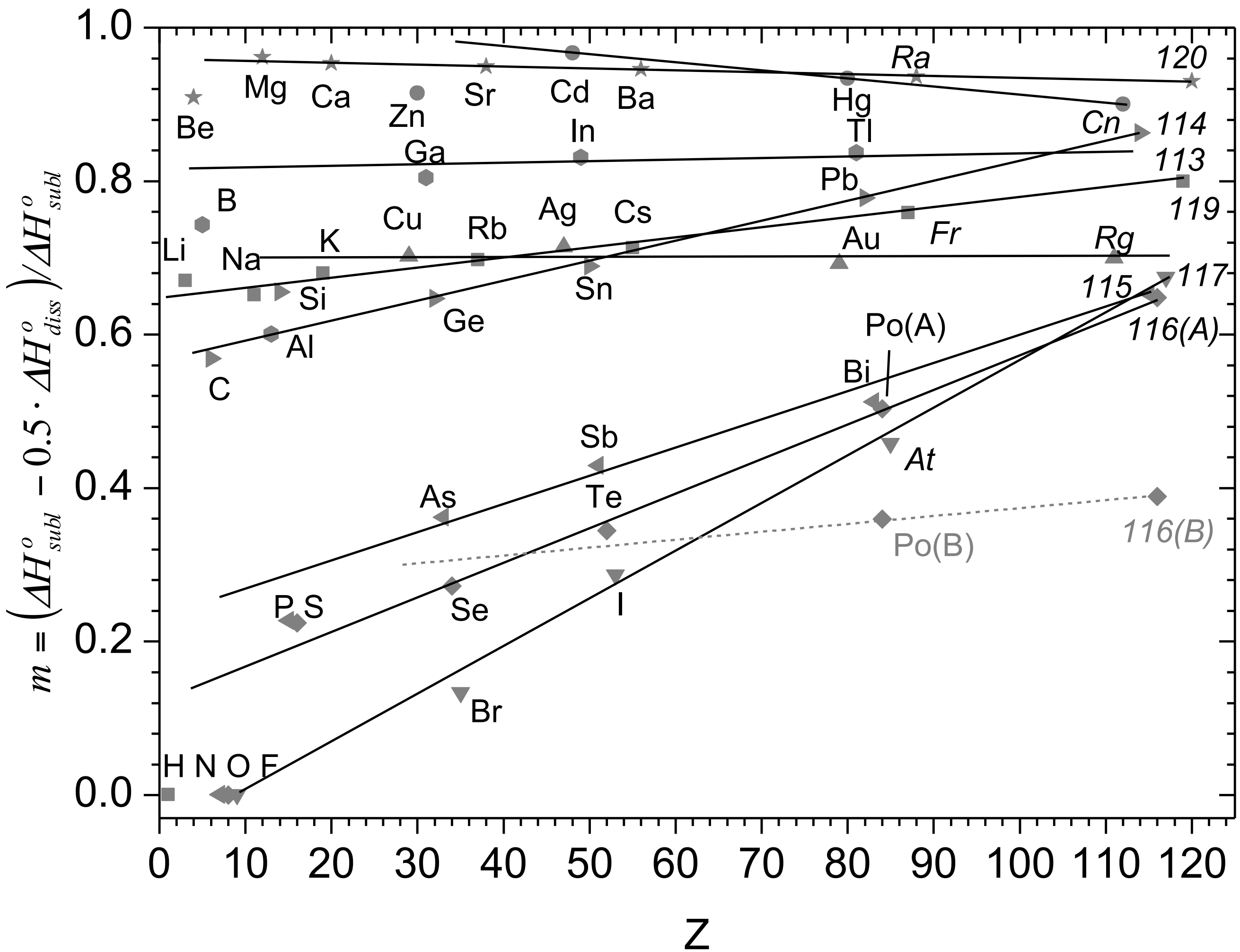
*	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
**	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Elemental Volatility



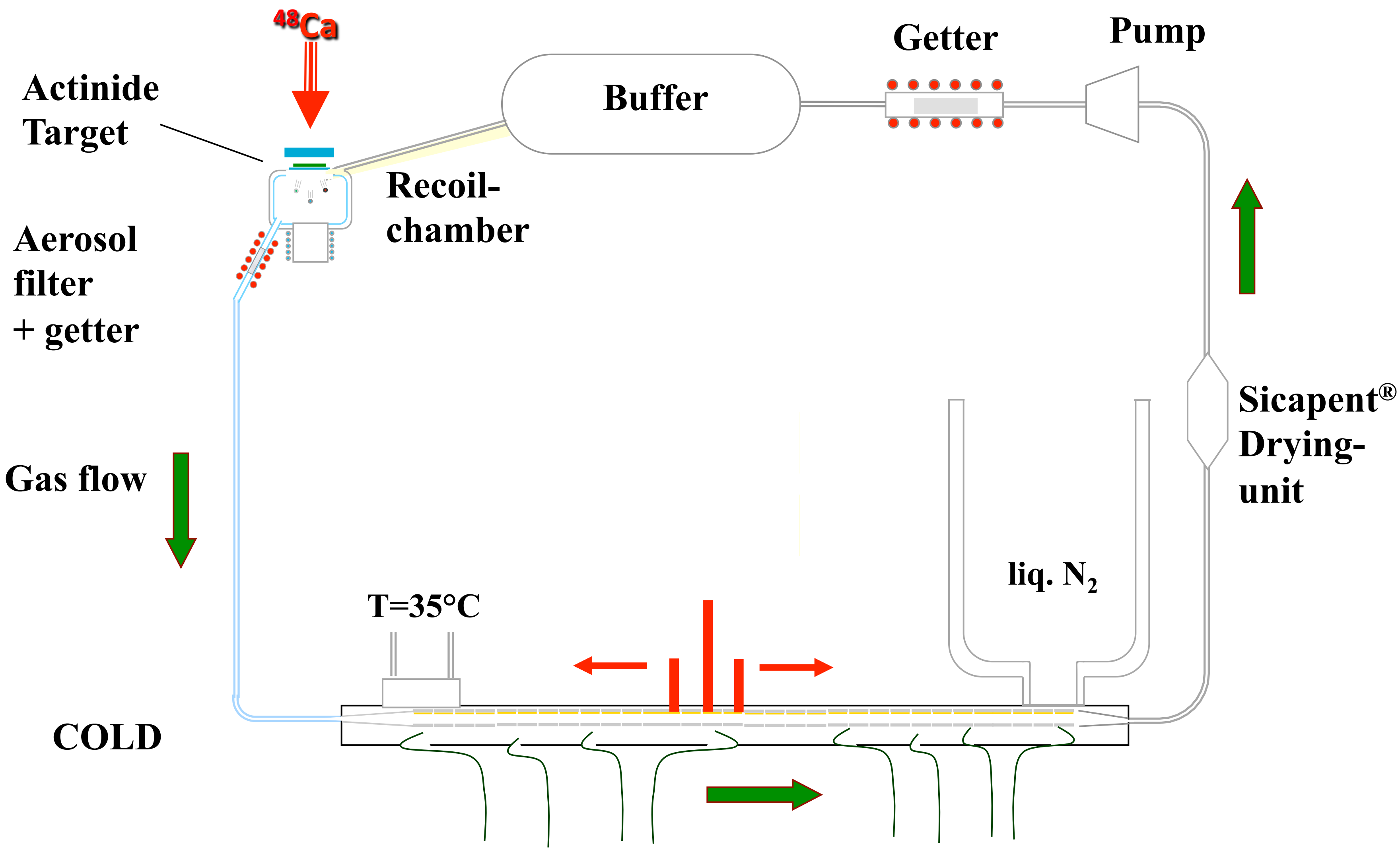
Eichler, B., Kernenergie 19 (10), (1976) 307

Metallic character

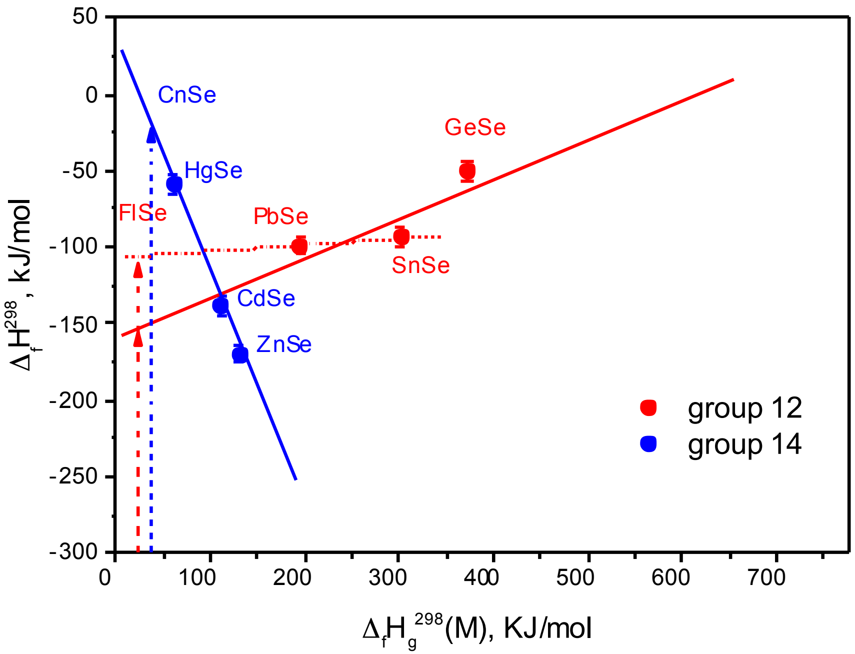


Eichler, B.: Kernenergie 19, 307 (1976).

Reaction chromatography with SHE using covered detectors

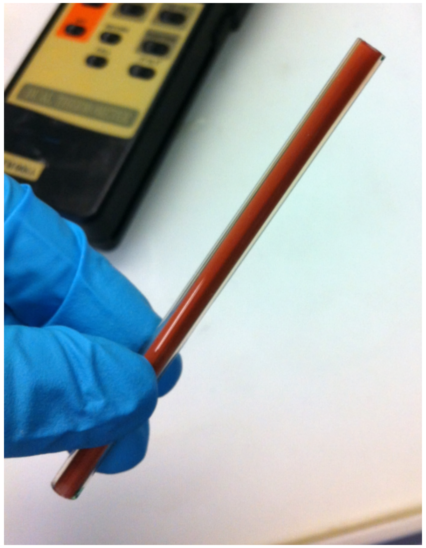


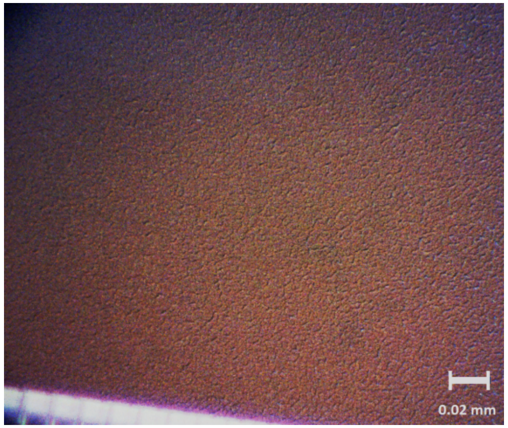
Reaction chromatography with SHE

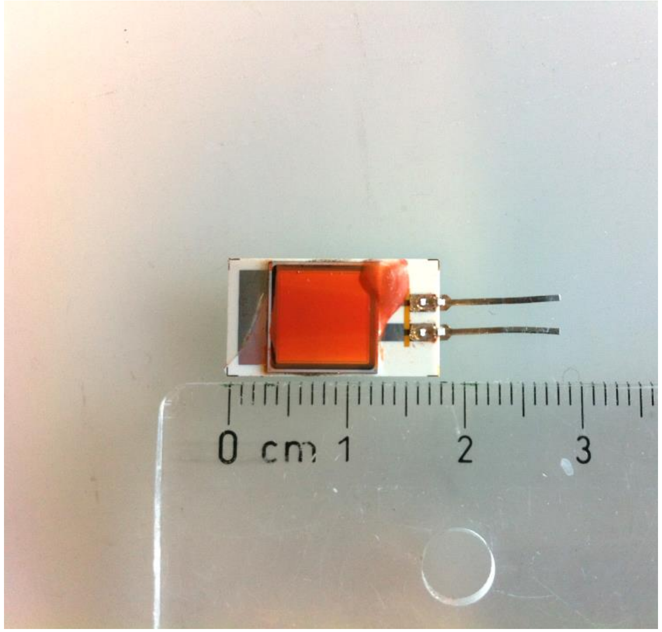


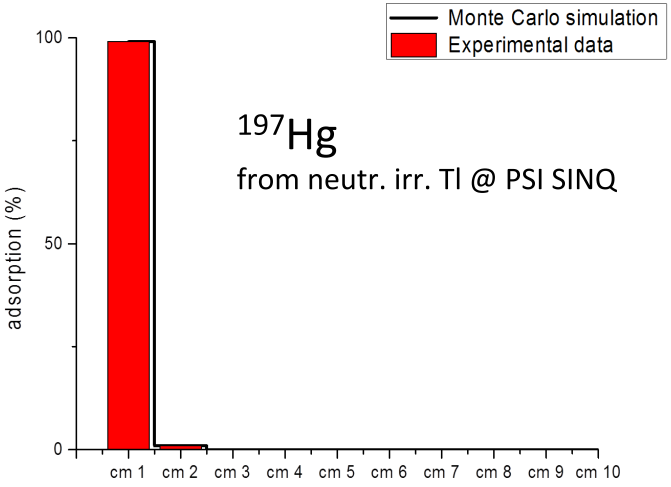
Nadine Chiera

Se columns and PIN diodes







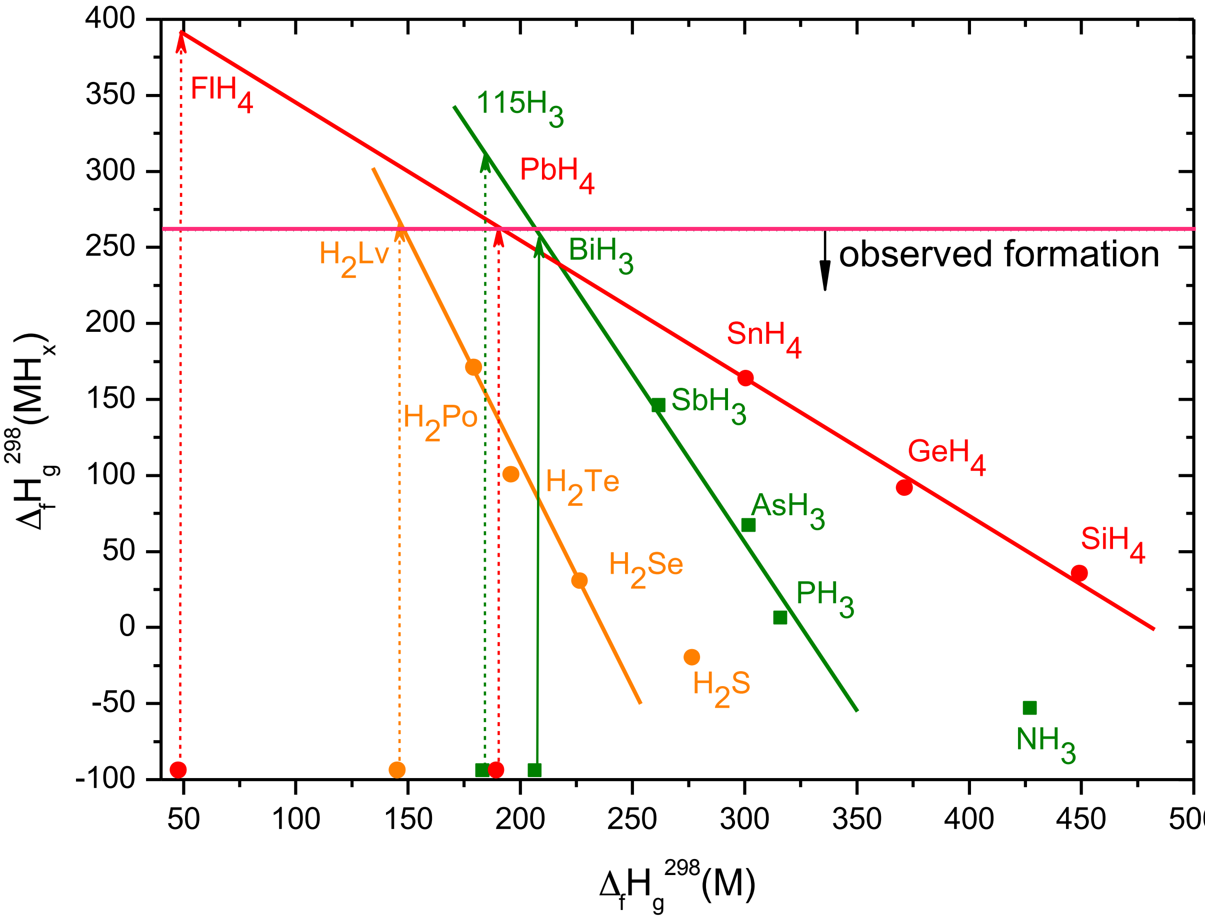


N. Chiera (2014)

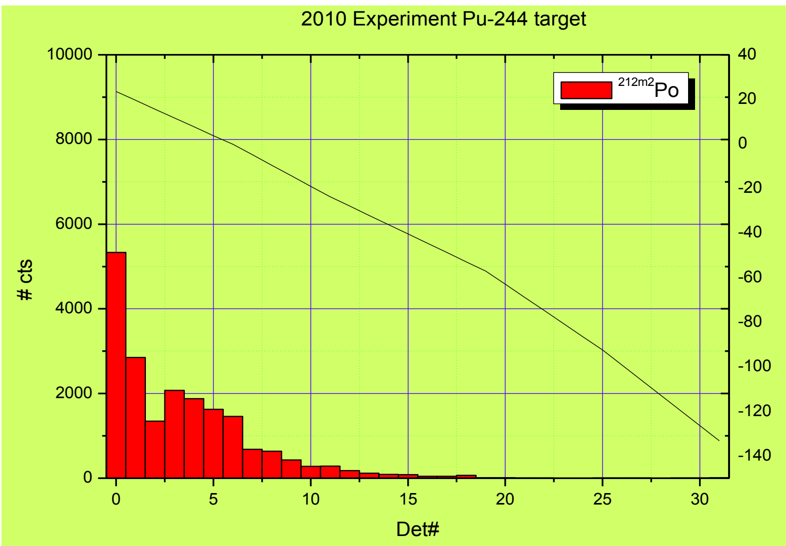
Experiment in collaboration with FLNR scheduled April 13.-26.2015 @FLNR

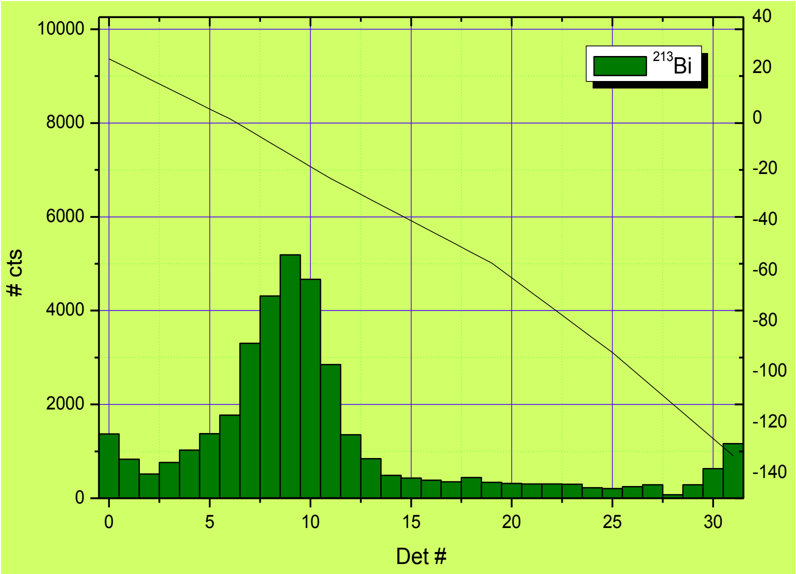
Reaction chromatography with SHE

Stability Trends Hydrides of Groups 14-16



Observation of volatile PoH_2 and BiH_3





Interesting Organo-Metal Chemistry

Prediction of properties of alkyl compounds of the elements 112, 114, astatine and 117

Voraussage von Eigenschaften der Alkylverbindungen der Elemente 112, 114, Astat und 117

Von P. HOFFMANN, Fachbereich für Anorganische Chemie und Kernchemie, Eduard-Zintl-Institut, Technische Hochschule Darmstadt

Mit 10 Abbildungen. (Eingegangen am 22. November 1972)

Summary

Using extrapolation methods, the following properties were determined of previously unknown methyl and ethyl compounds of the heavy-heavy elements 112, 114, 117, and of astatine: binding energy for $112(CH_3)_2$, $114(CH_3)_4$, $117CH_3$, and $AtCH_3$, atomic heat of formation and dissociation energy for $114(CH_3)_4$, heat of evaporation, plot of vapor pressure, and boiling point for $114(CH_3)_4$, $117CH_3$, and $AtCH_3$, and ionization potential for $114(CH_3)_4$, $117CH_3$, $117C_2H_5$, as well as for $AtCH_3$ and AtC_2H_5.

17 18

He

F Ne

Cl Ar

Br Kr

I Xe

Cs Ba La* Hf Ta W Re Os Ir Pt Au Hg Tl Pb Bi Po At Rn

Fr Ra Ac** Rf Db Sg Bh Hs Mt Ds Rg Cn 113 Fl 115 Lv 117 118

119 120

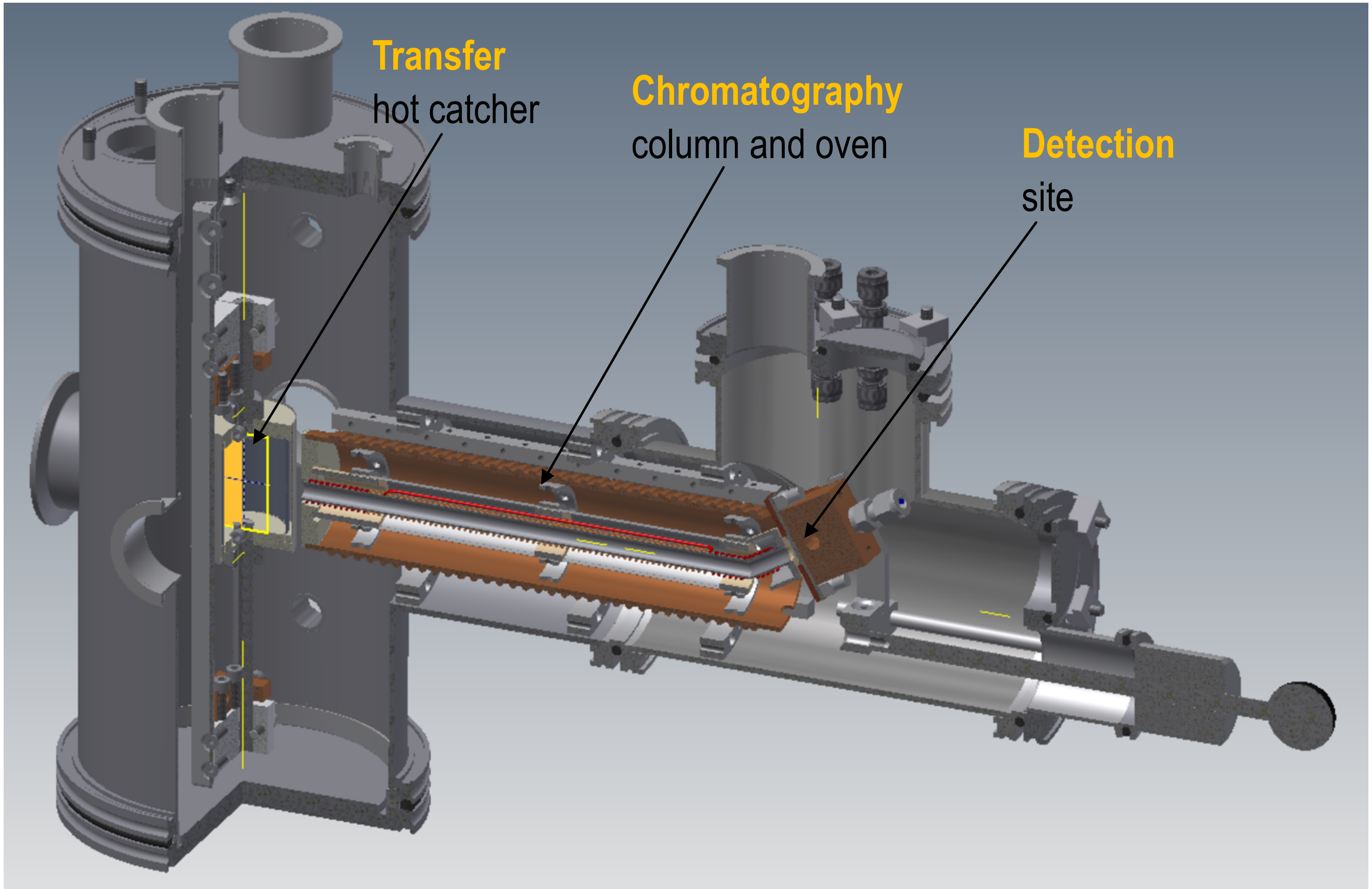
* Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu

** Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr

Design of a Vacuum Experiment

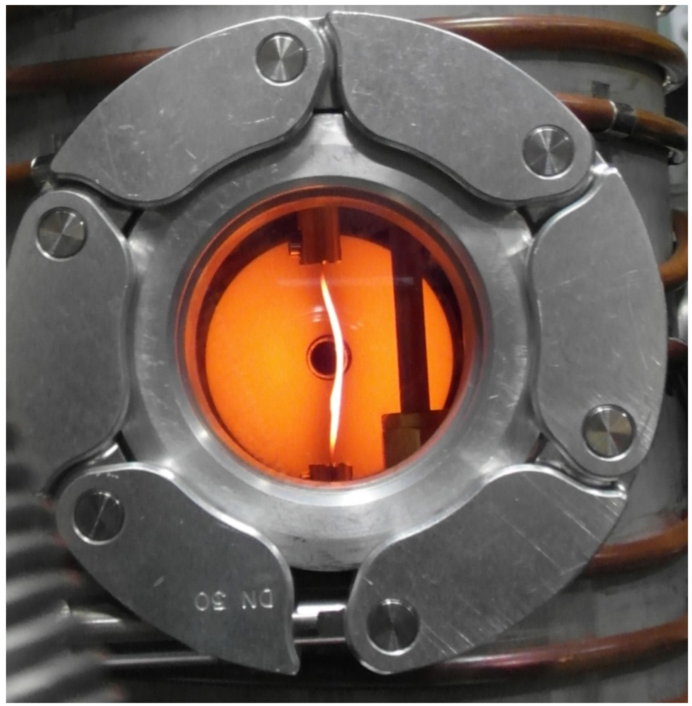
Isothermal Vacuum Chromatography IVAC

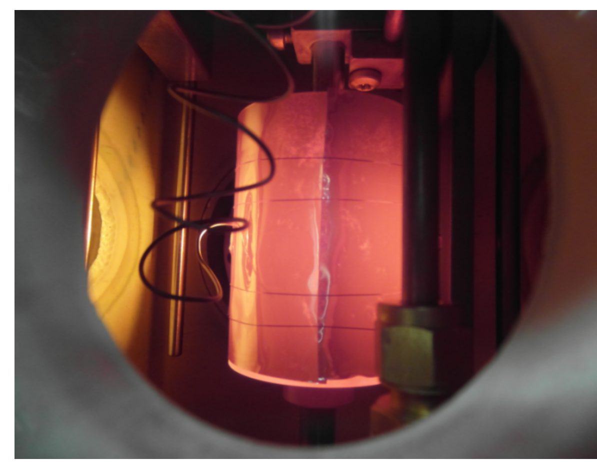
Patrick Steinegger
Dave Piguet



Successful Proof-of-principle experiment in 2014 collaboration with ASRC @ JAEA!

Hot Catcher - Release

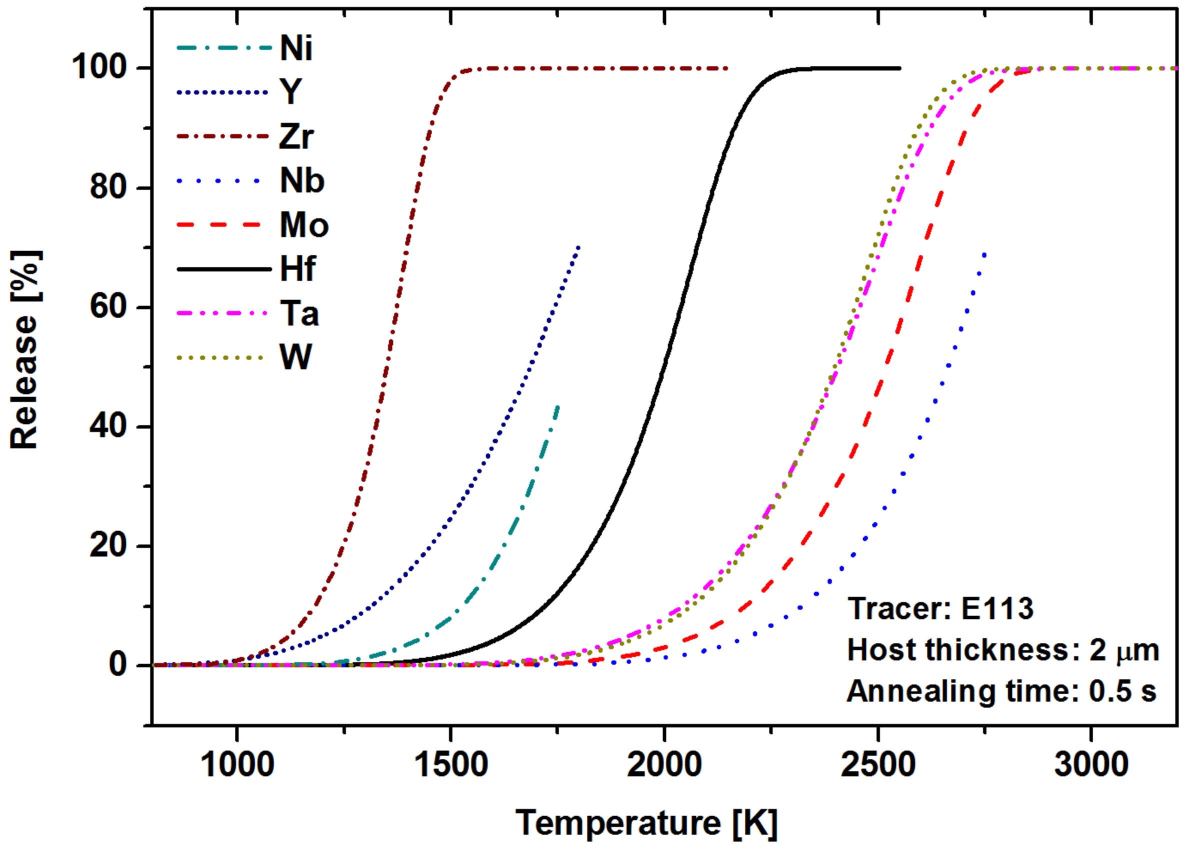




Release Kinetics E113

Prediction for the thermal release of E113 from various metal matrices acting as **hot catcher**.

$$F[\%] = 100 - \frac{800}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \times \exp\left[- \frac{(2n+1)^2 \pi^2 D t}{d^2} \right]$$



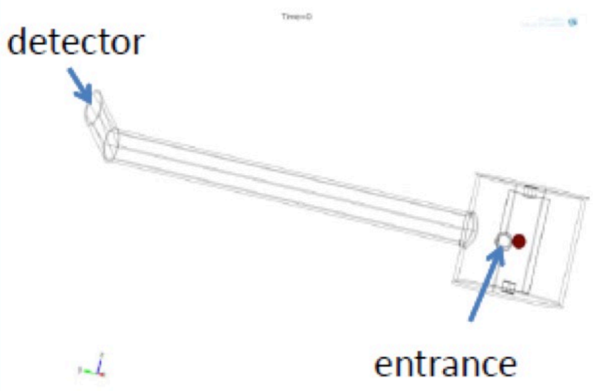
[1]

*D. Wittwer et al., Radiochimica Acta **101** (2013) 4*

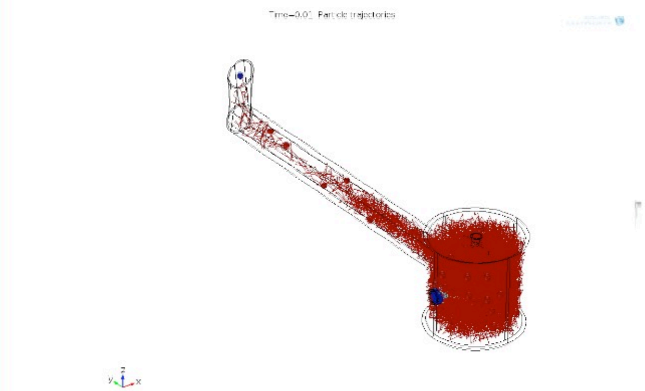
COMSOL Multiphysics®

IVAC time simulation w/o adsorption retention

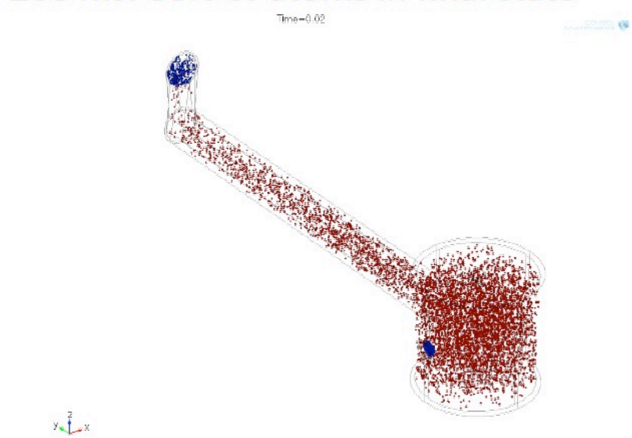
0 ms: Start of release from 3 mm spot

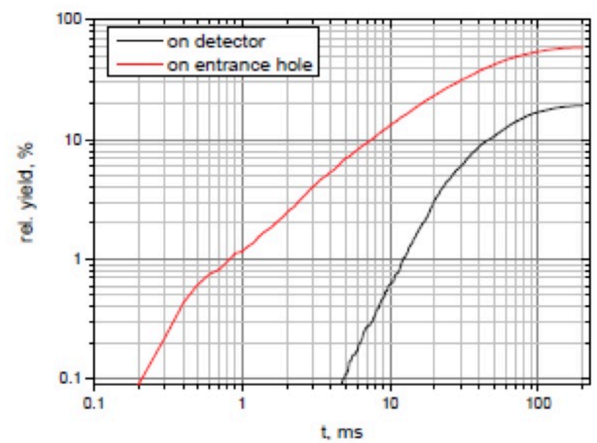


19 ms: First atom arrives @ detector

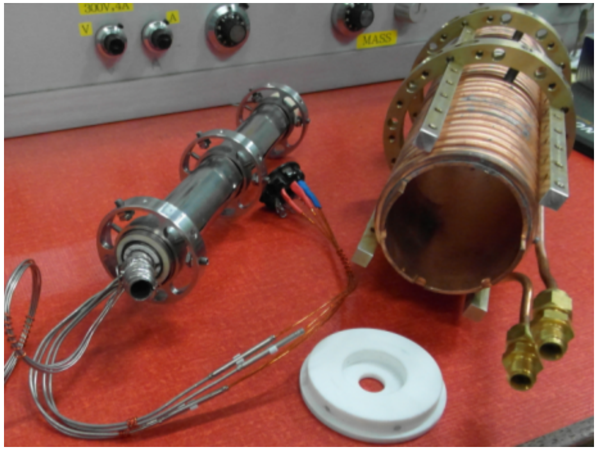


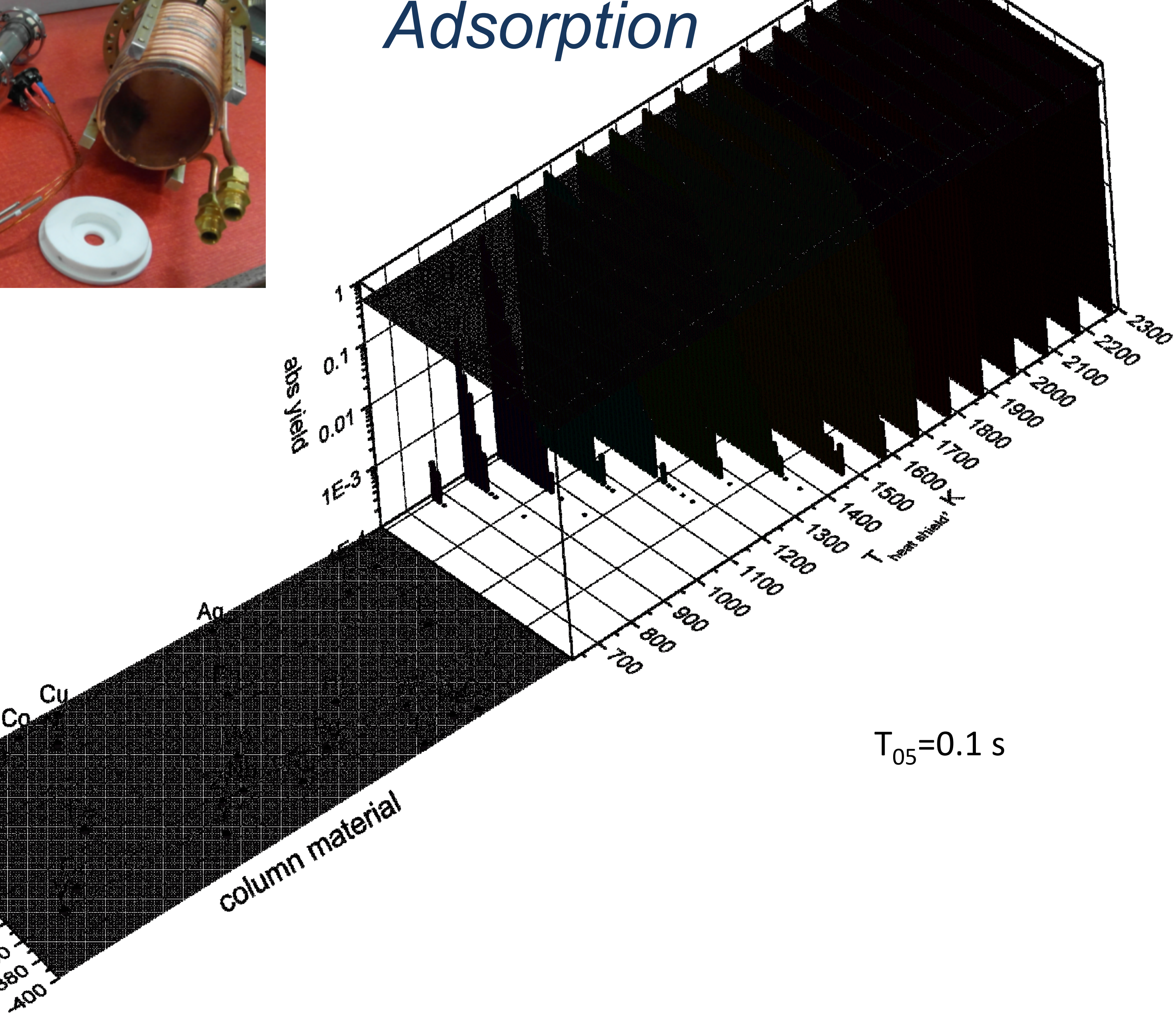
200 ms: 80% of atoms in final state



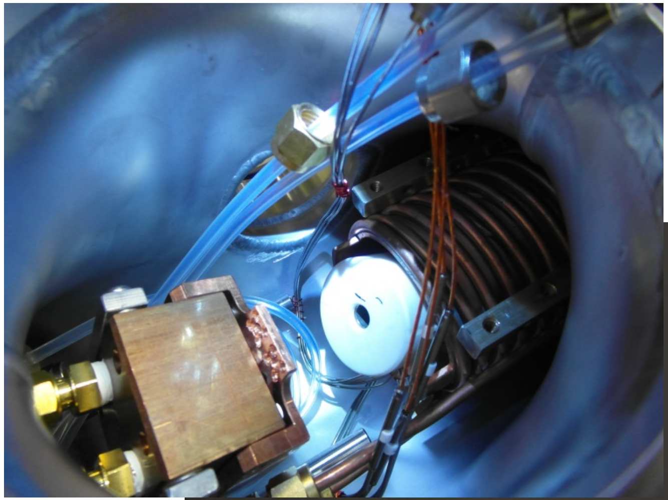


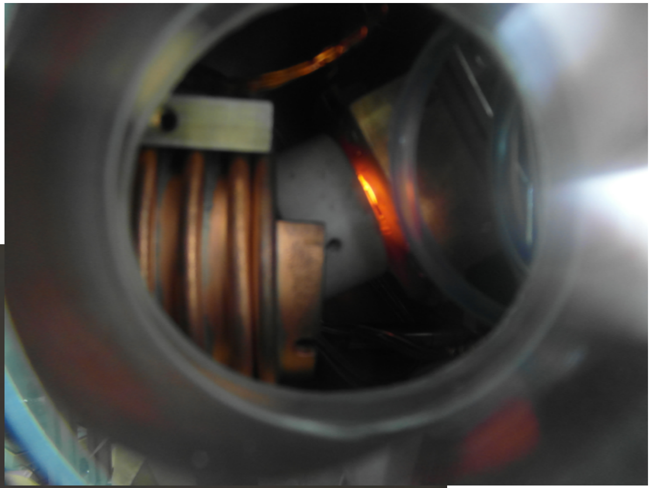
Adsorption

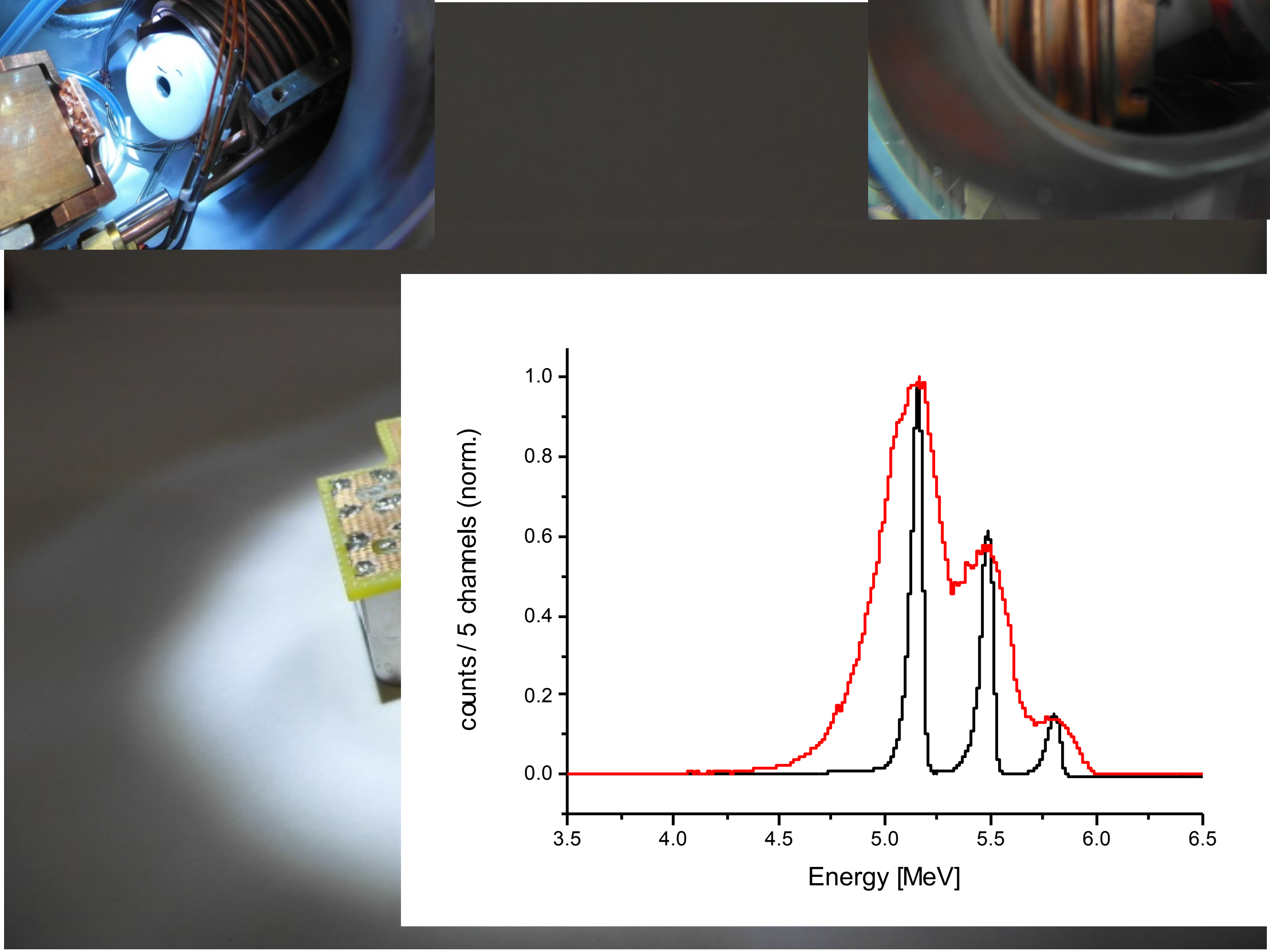




T_{05}=0.1 s

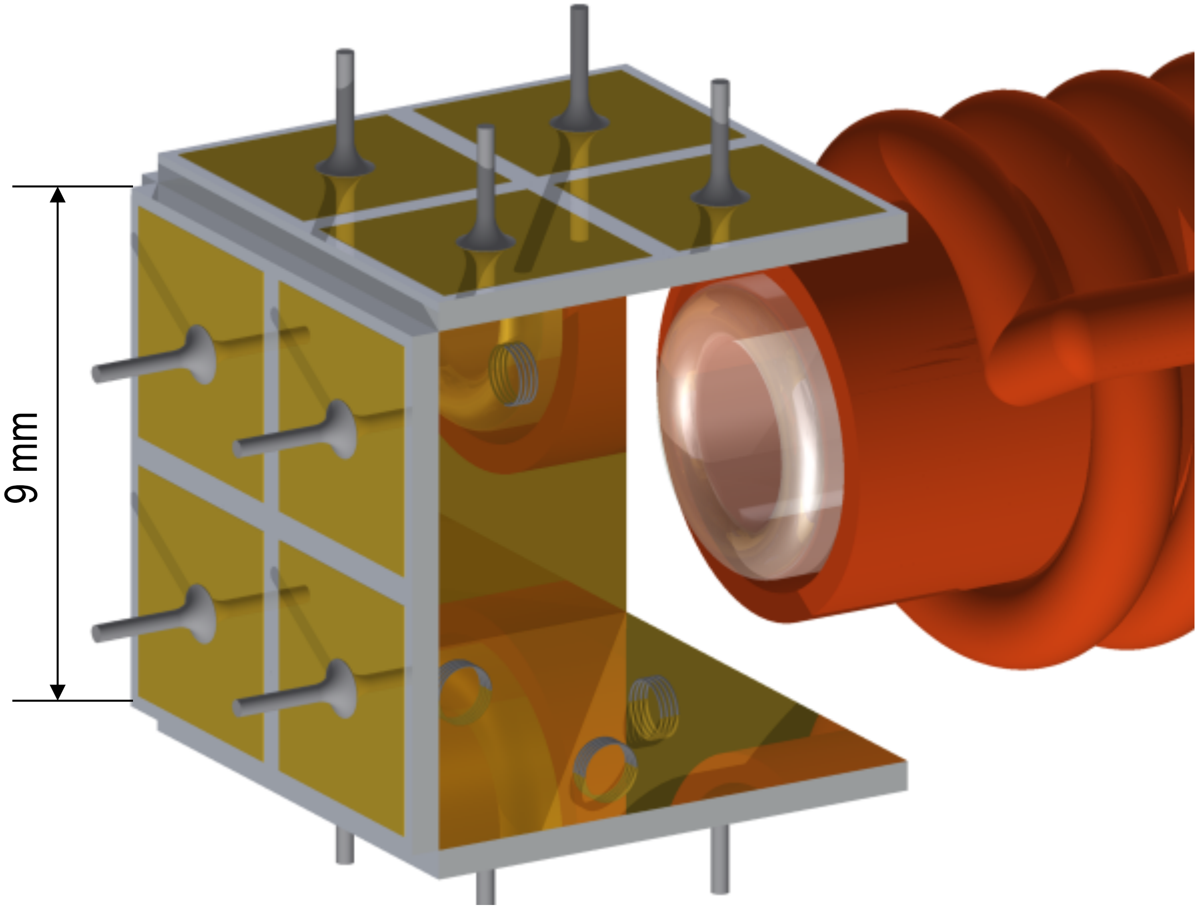






P. Steinegger (2014)

Diamond detector assembly



Future Transactinide Chemistry

Summary s- and p- elements

- Gas phase chemical systems:
  - * classical gas chromatography
    - reactions with H_2, H_2O, O_2, S, Se, F_2, Cl_2
    - organometallic chemistry (groups 12-14)
  - * vacuum chromatography
    - element-metal/element-quartz interaction
    - high temperature detectors

1 17 18

H He

Li F Ne

Na Cl Ar

K Br Kr

Rb Sr Y Zr Nb Mo Tc Ru Rh Pd Ag Sb Te I Xe

Cs Ba La* Hf Ta W Re Os Ir Pt Au Hg Tl Bi Po At Rn

Fr Ra Ac** Rf Db Sg Bh Hs Mt Ds Rg Cn 113 Fl 115 Lv 117 118

119 120

* Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu

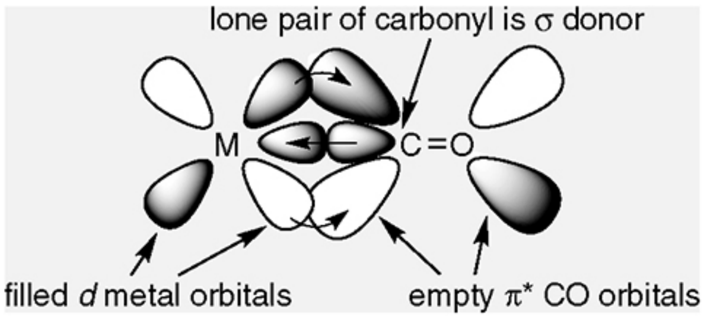
** Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr

Future Transactinide Research

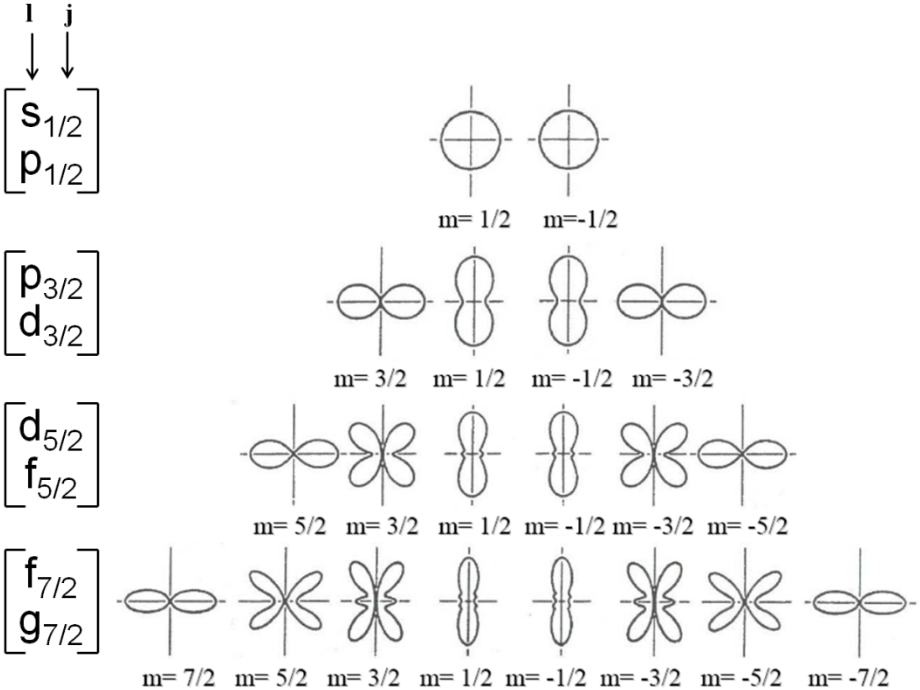
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	113	Fl	115	Lv	117	118
119	*120*																

*	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
**	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Carbonyl Chemistry with Transactinides

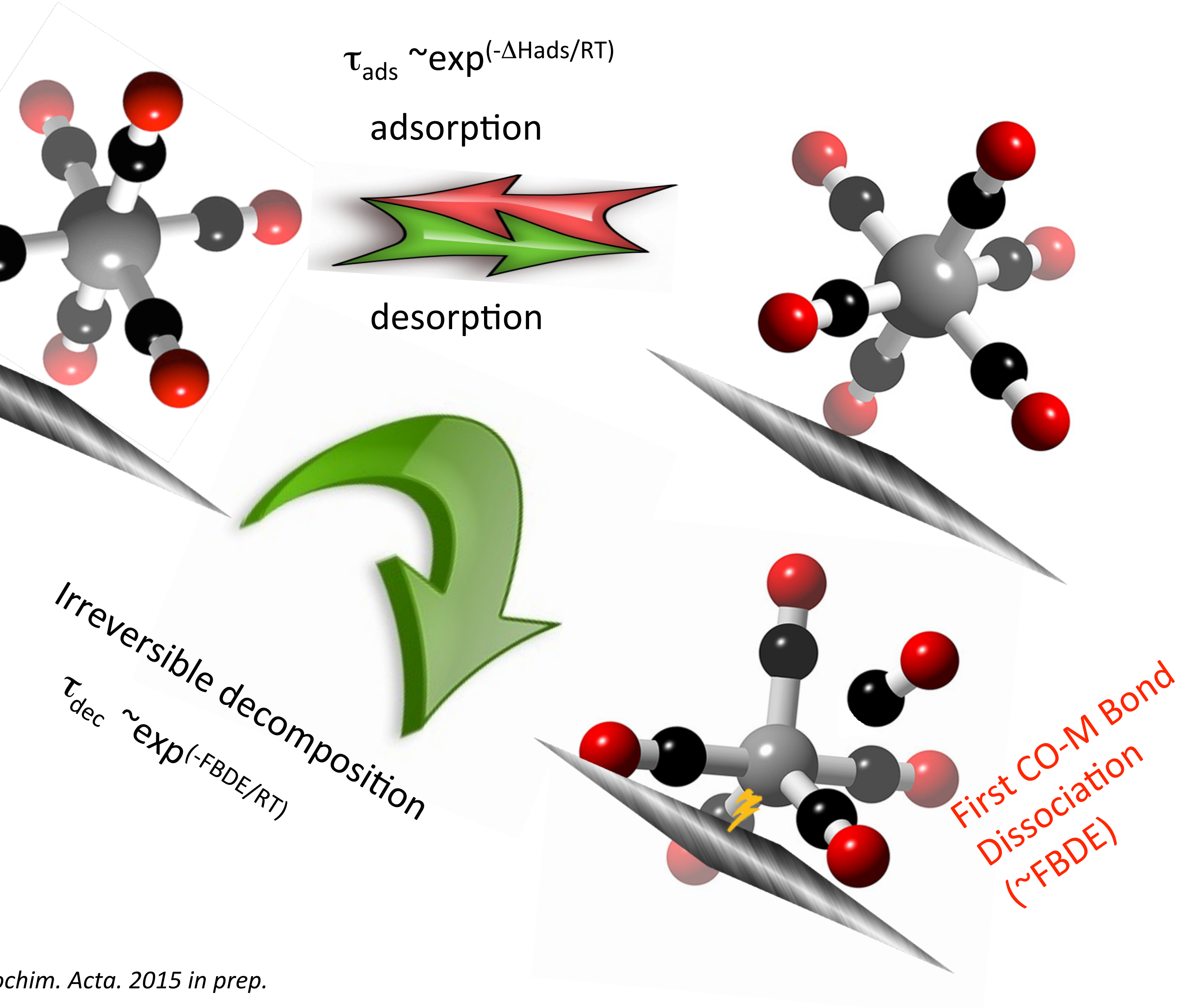


Dirac Orbitals
relativistic



O.L. Keller, Radiochim. Acta 37 (1984) 169; adapted from H. White, Phys. Rev. 38 (1931) 513

2-Step Decomposition for $M(CO)_6$



I.Usoltsev et al., Radiochim. Acta. 2015 in prep.

The CO Collaboration

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J.V. Kratz
N. Wiehl

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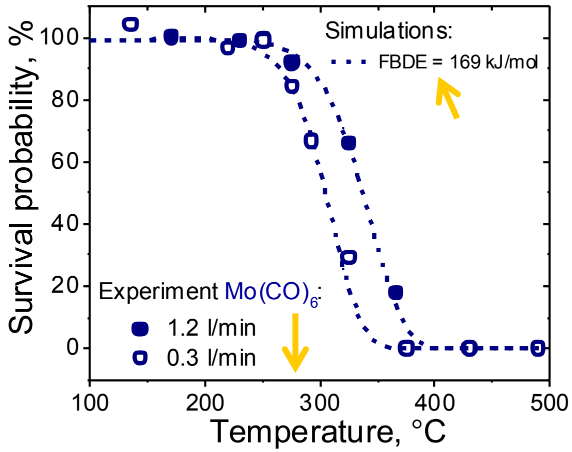
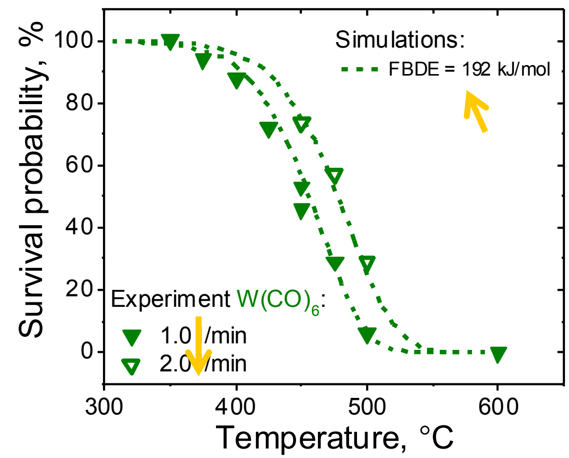
PSI Villigen (CH)
U. Bern (CH)
R. Eichler
N. Chiera
D. Piguet
P. Steinegger
A. Türler

Niigata U (J)
K. Ooe

Decomposition studies: $Mo(CO)_6$ + $W(CO)_6$

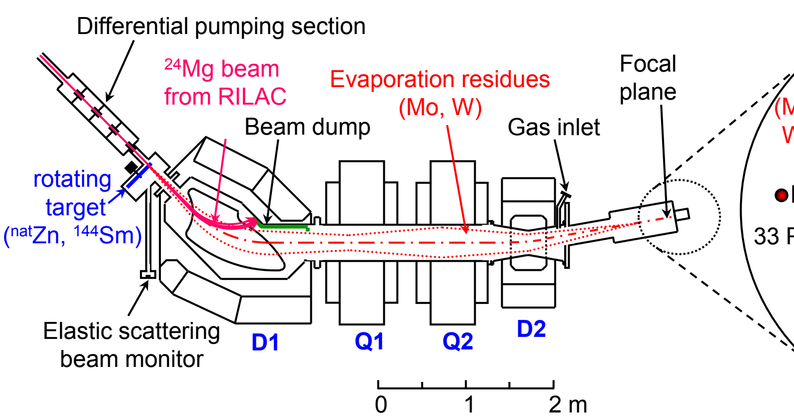


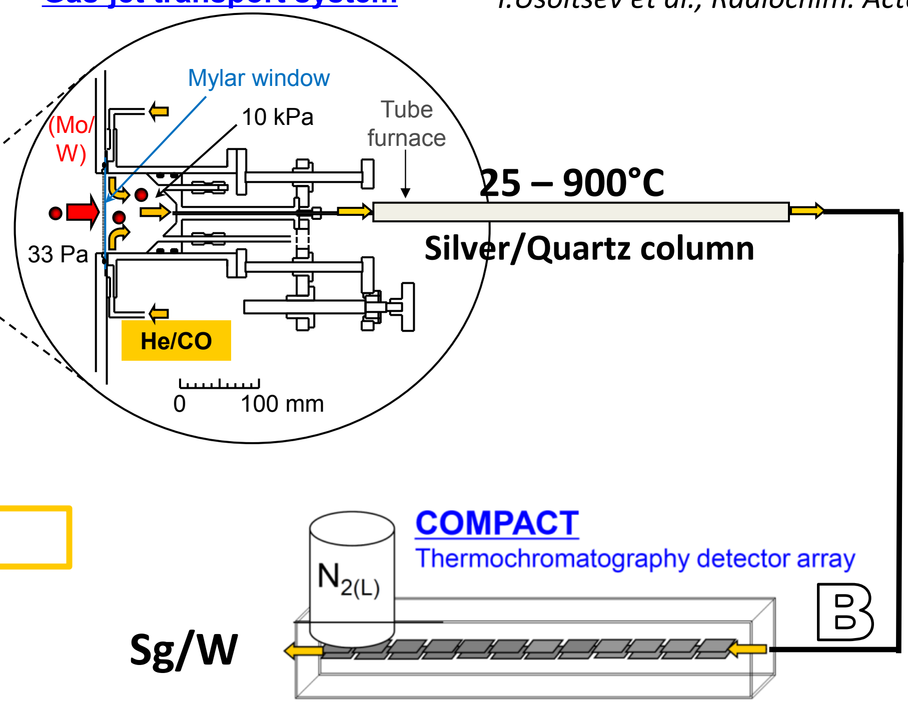


RIKEN GARIS

Gas-jet transport system

I.Usoltsev et al., Radiochim. Acta. 2015 in prep.



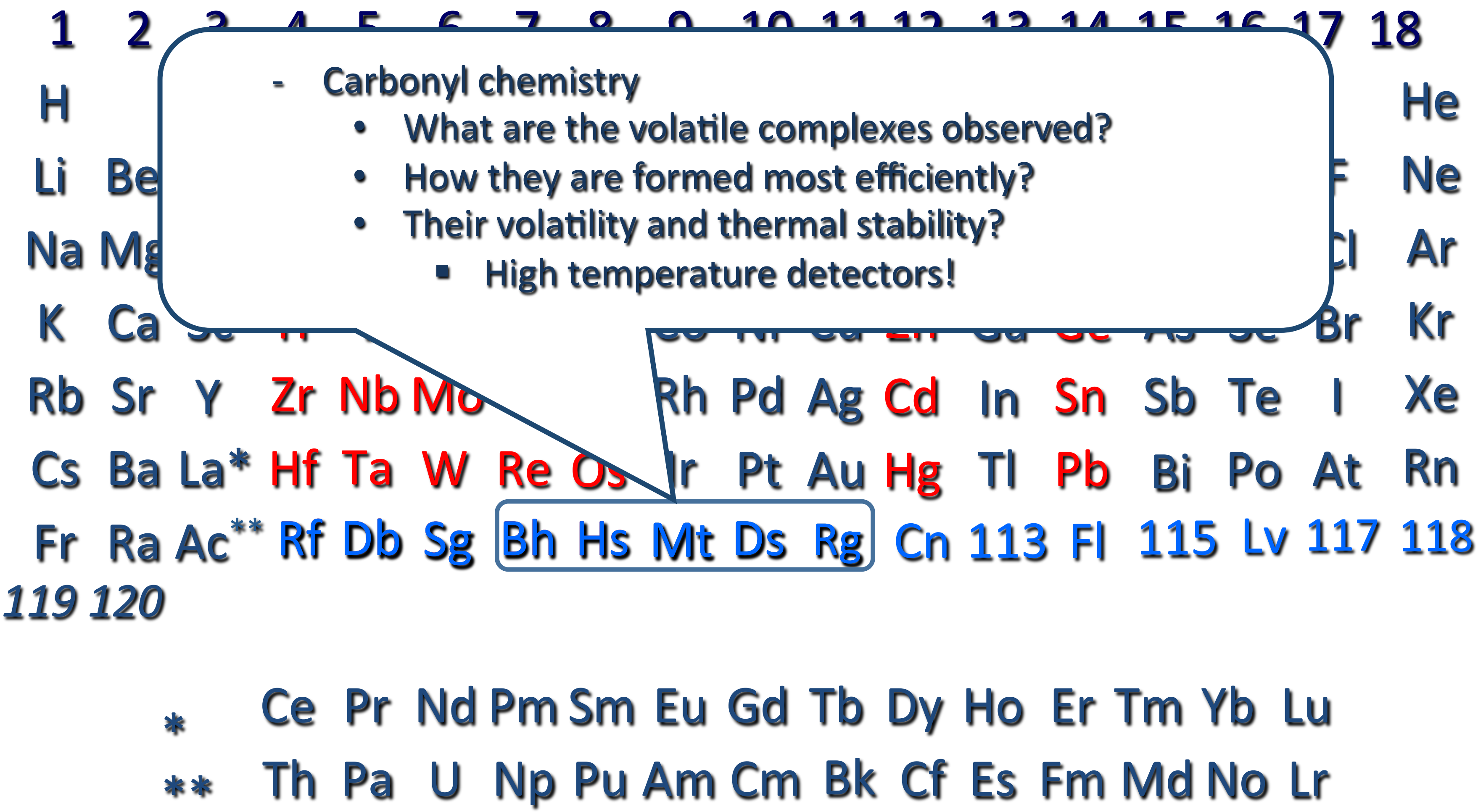


K.E. Lewis et al., J. Am. Chem. Soc. 106 (1984) 3905

Beam time proposal of the «CO collaboration» accepted at RIKEN ML-PAC Jan. 2015

Exciting Future Carbonyl Chemistry

- Carbonyl chemistry
  - What are the volatile complexes observed?
  - How they are formed most efficiently?
  - Their volatility and thermal stability?
    - High temperature detectors!



Future Transactinide Research

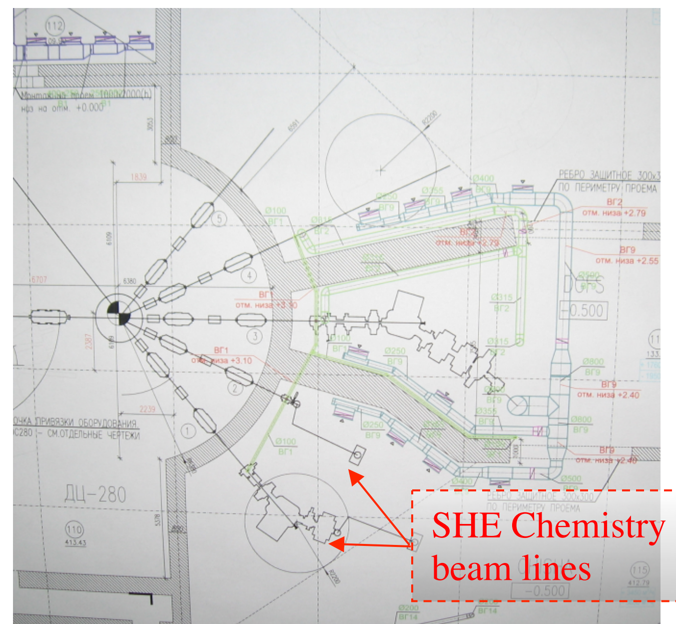
Summary Transition Metals

- Further gas phase chemical investigations
  - * low oxidation states oxides, hydroxides, chlorides ...
  - * volatile organo-metallic compounds

1	2																18
H																	He
Li	Be																Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V			Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb			Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*	Hf	Ta		Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	113	Fl	115	Lv	117	118
119	*120*																

*	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
**	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

SHE-Factory @ FLNR Dubna



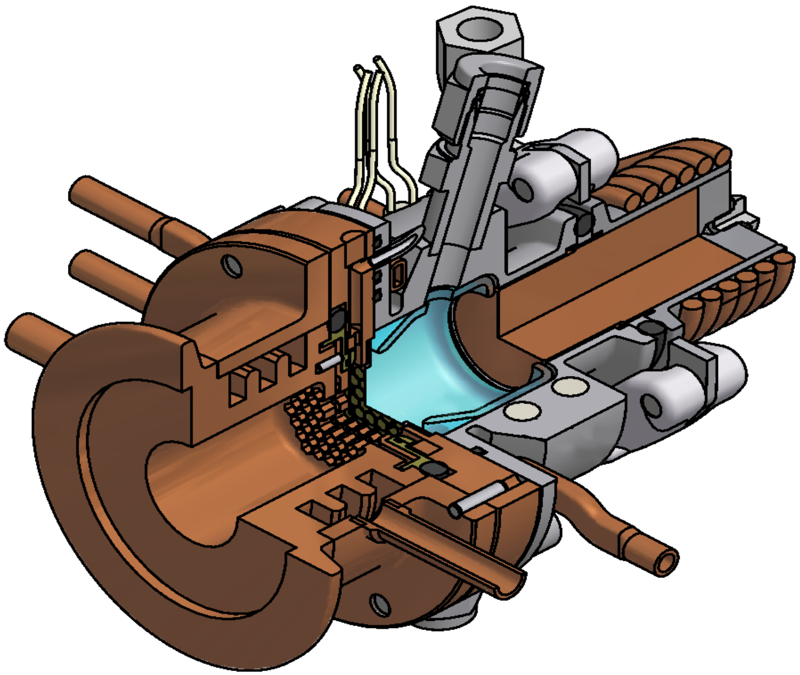
Requirements for Future Chemistry with SHE

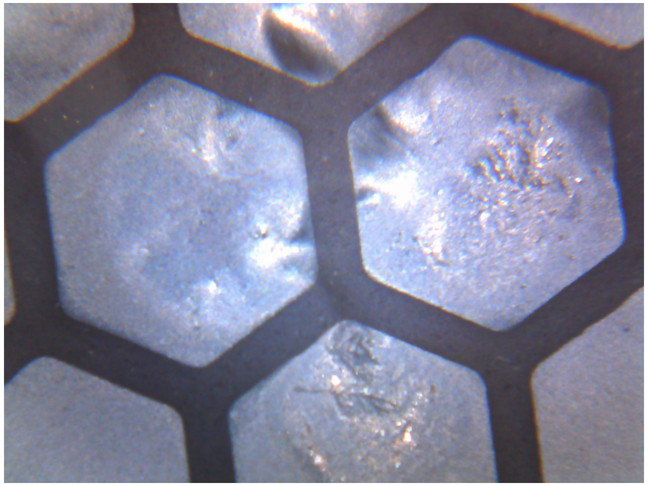
Preseparation for Chemistry:

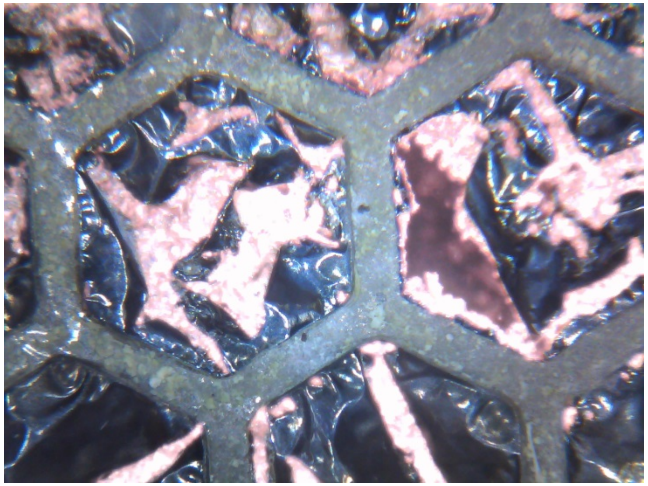
- transmission >50% with thick targets 0.5-1 mg/cm^2
- small focal plane image 2x2 cm^2
- primary beam separation
- ideal for chemistry would be mass < 240 separation from SHE

→ We need to discuss design options and start the real project soon!

10-fold beam intensity = target problems !



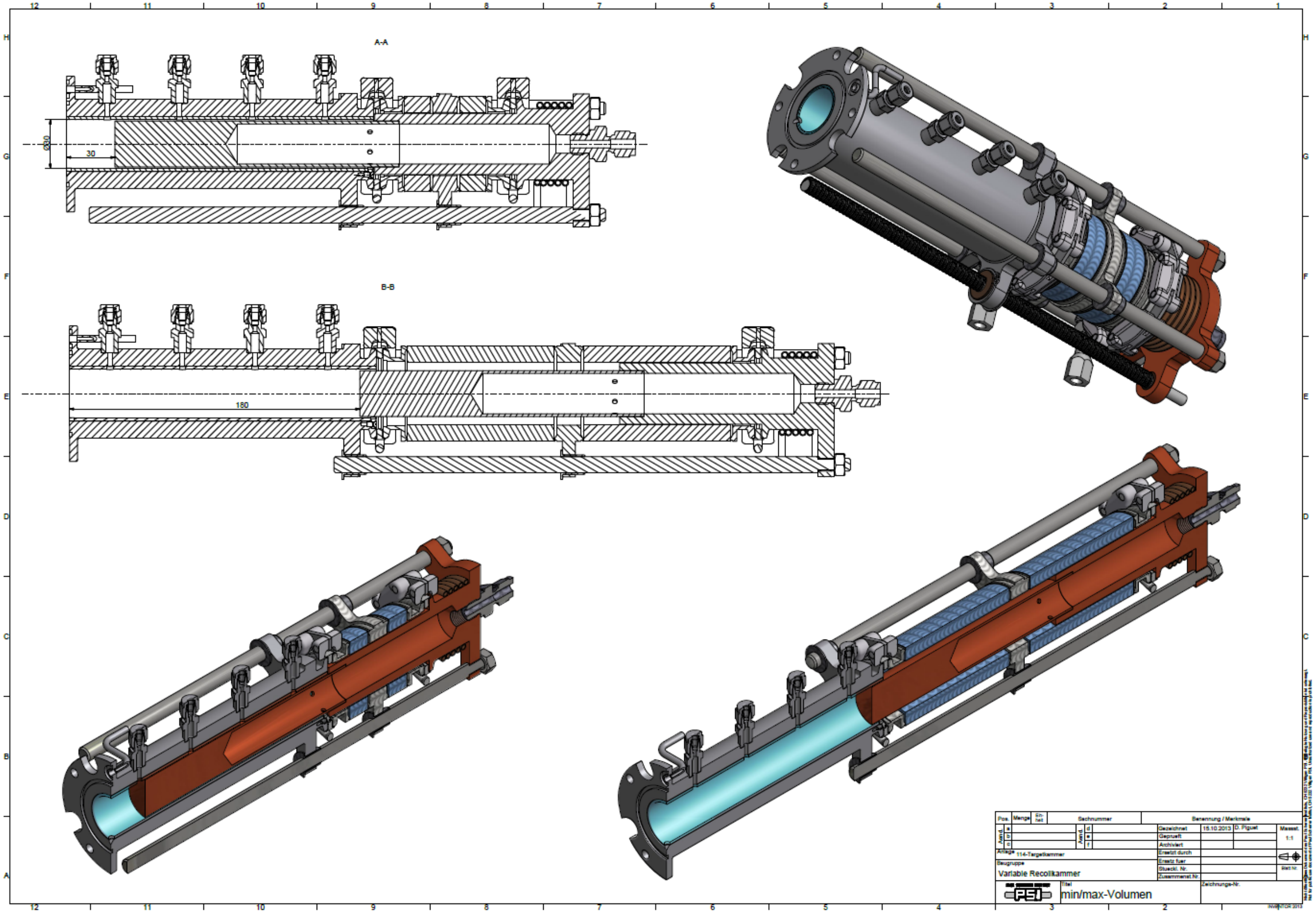




Problems of destruction:
- production (physics and chemistry)
- beam scattering/no-stripping (physics)
- stopping (chemistry)

👍 improved the target techniques needed
→ intermetallic targets (first step)

Recoil range of SHE in plasma???

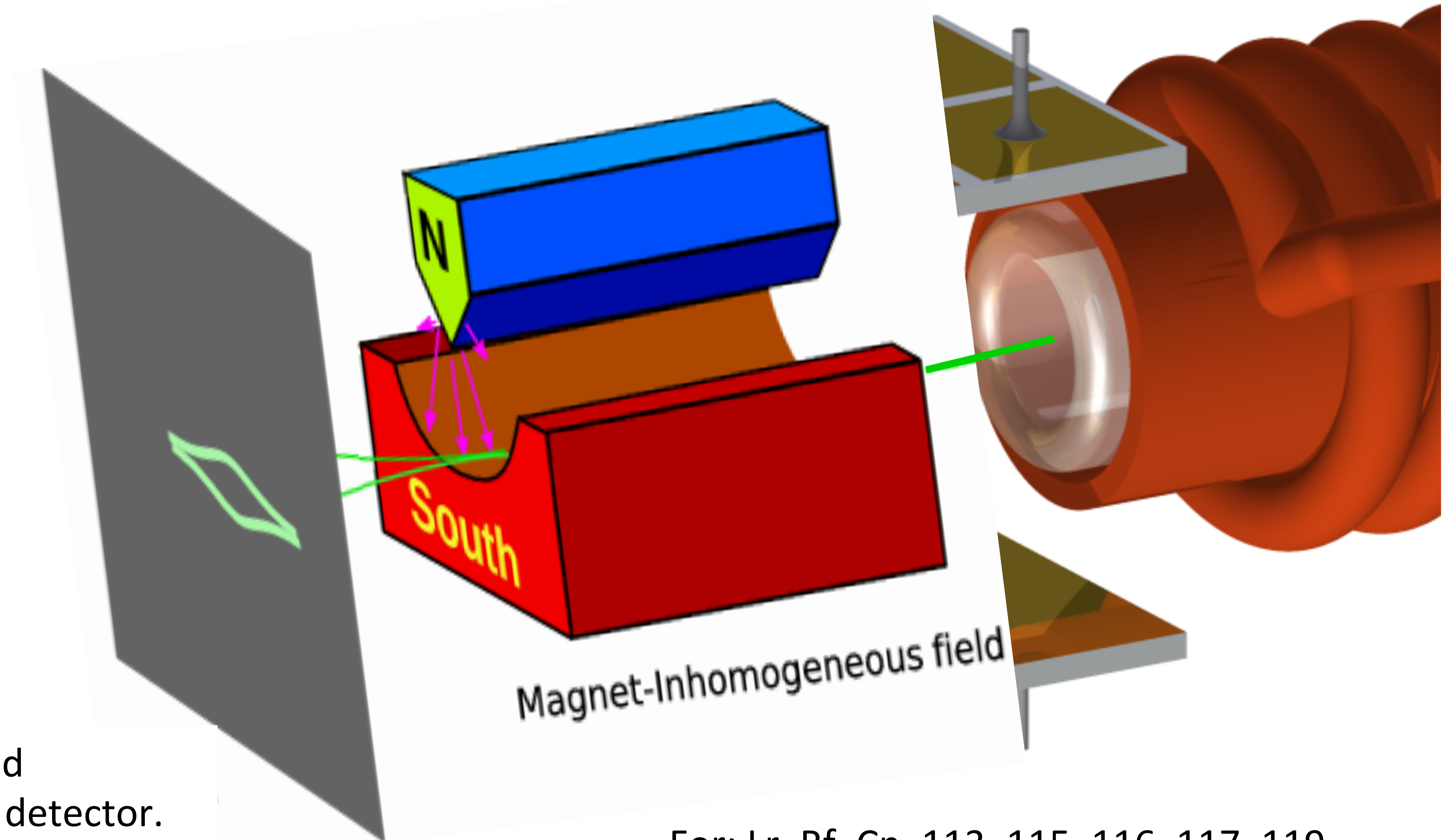


Experiment in collaboration with FLNR scheduled April 13.-26.2015 @FLNR

Future Transactinide Research

Stern Gerlach - «fast chemistry»

IVAC as atomic beam source



Pixelated
particle detector.
e.g. diamond array.

For: Lr, Rf, Cn, 113, 115, 116, 117, 119

Summary

Constraints are:
- short half-lives 1 min-1 s and below
- low production rates

Theory:
- **ask the right questions**

SHE Experiment:
- **data analysis, results**
- **thermodynamic & kinetic models**

Adequate technique for single atoms:
- **mulitstep processes**
- **separation, detection**
- **model experiments**
- **kinetic & thermodynamic model description**

Acknowledgements

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  - the Reimei Research Program (Japan Atomic Energy Agency);
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  - the Office of Science, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences, and Biosciences, Heavy Element Chemistry Program of the U.S. Department of Energy at Lawrence Berkeley National Laboratory under contract DE-AC02-05CH11231;
  - the National Natural Science Foundation of China (grant 11079006).