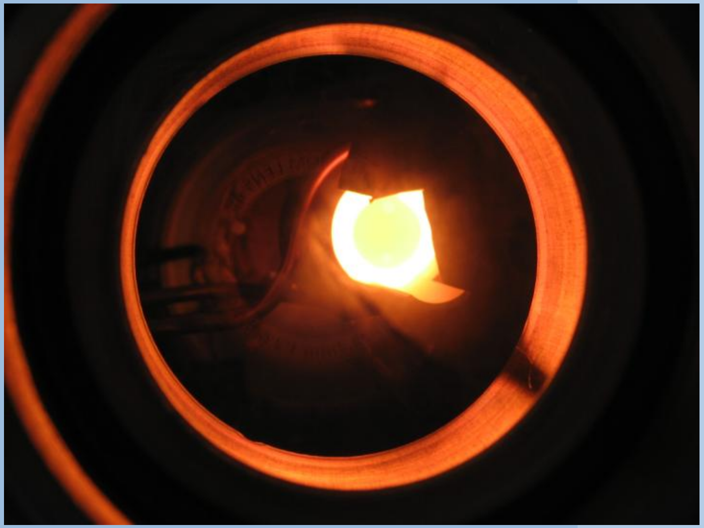
UNIVERSITÄT BERN

PAUL SCHERRER INSTITUT

TAN 2011 – Sochi, Russian Federation

10.09.2011

THERMAL RELEASE OF p-ELEMENTS FROM METAL MATRICES



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University of Bern &
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OUTLINE

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- Thermal Release

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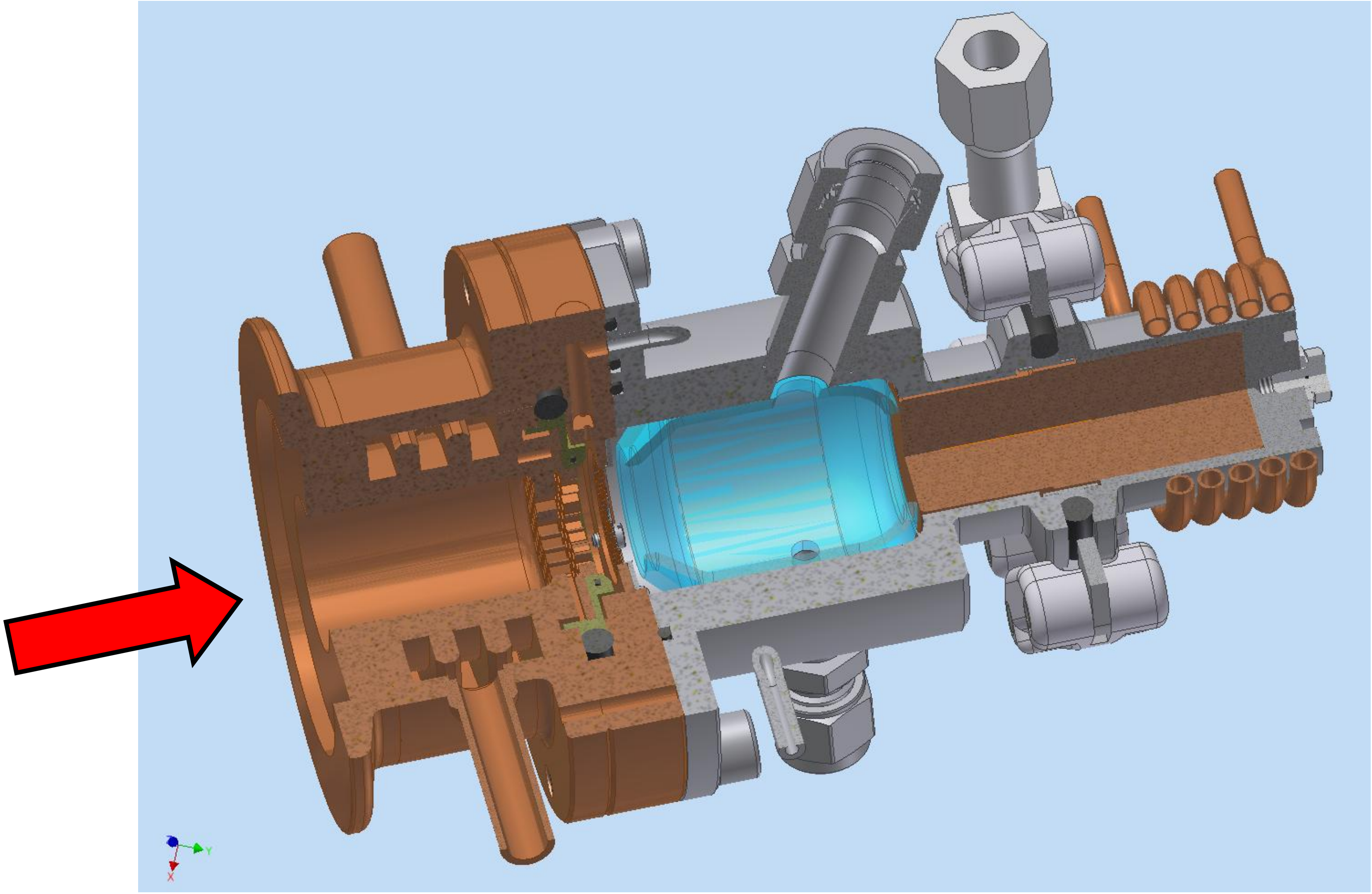
Introduction



Introduction

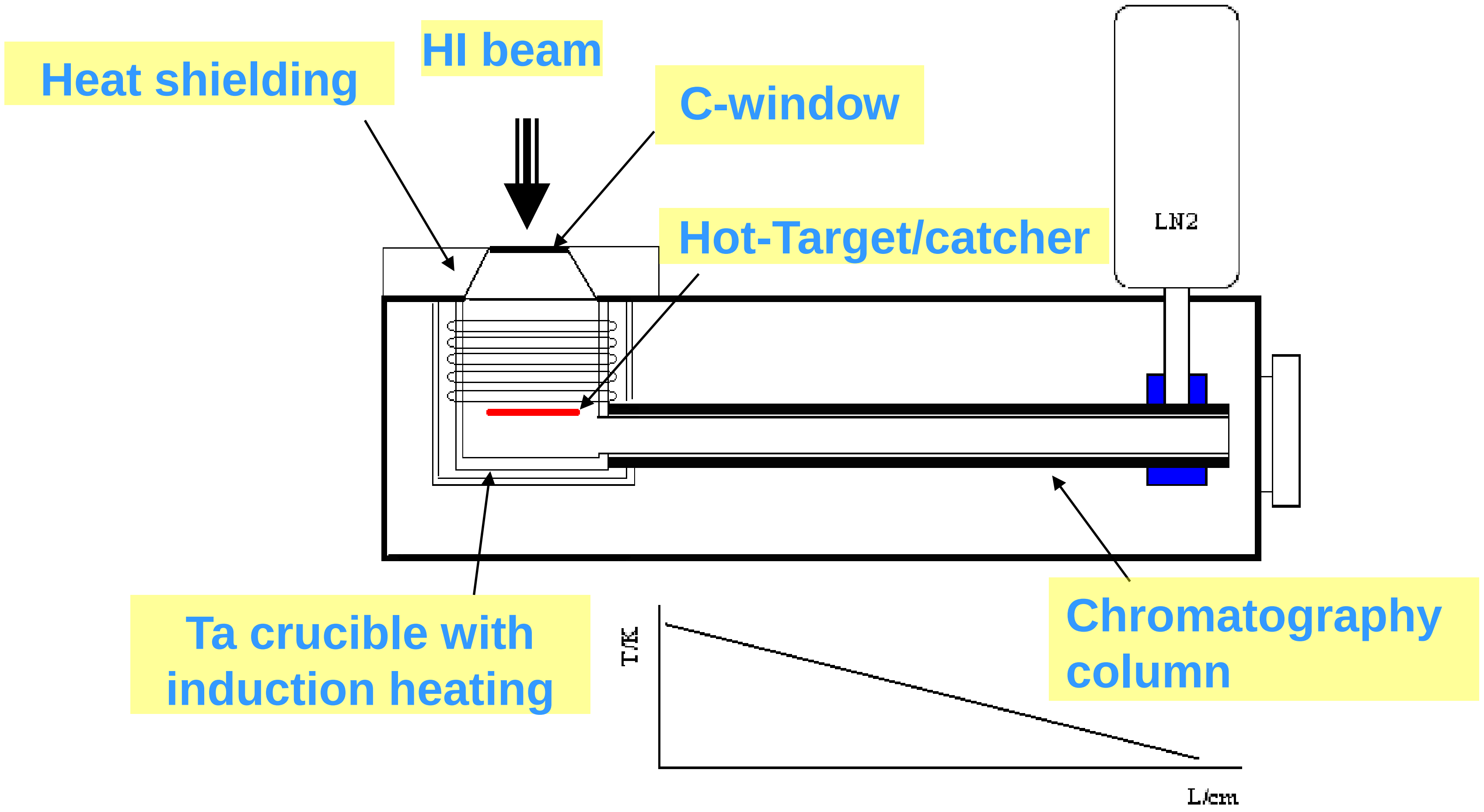
- Change to vacuum-chromatography

Pros:
- No aerosols
- Less pollution
- Better spectroscopic resolution

Cons:
- Target overheating
- No stopping gas anymore

- Idea: Solid catcher or (ISOLDE like) coupled target-catcher

Vision


Heat shielding
HI beam
C-window
Hot-Target/catcher
LN2
Ta crucible with induction heating
Chromatography column
T/K
L/cm

Thermal Release

-Release can be measured easily
-If the diffusion is the rate determining factor, diffusion coefficients can be calculated from the release rate

$$F = 1 - \frac{8}{\pi^2} \cdot \exp\left(- \frac{Dt}{d^2} \right)$$

-Further the activation energy can be deduced

$$\ln\left(\frac{\left(-\ln\left((1-F)\frac{\pi^2}{8} \right) \right) d^2}{t} \right) = -\frac{Q}{RT} + \ln(D^0)$$

F relative release

D is the diffusion coefficient or diffusivity in *m^2/s*

D_0 is the ***preexponential*** factor in *m^2/s*

t is the bake out time in ***s***

d thickness of the foil in ***cm***

Q activation energy in ***J/mol***

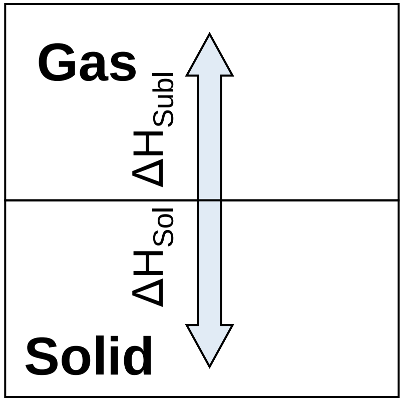
R is the IDEAL GAS constant in *$J/mol*K^{-1}$*

J. Crank, Mathematics of Diffusion, Oxford Univ. Press London (1975)

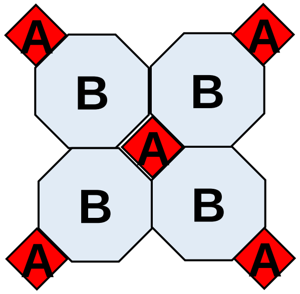
Thermal Release

Release Enthalpy

$$\Delta H_f = \Delta H_{Subl} - \Delta H_{Sol}$$



Miedema model: Intermetallic solid solution



$$\Delta H_{sol} = \frac{2 \cdot V_{Asol}^{2/3}}{n_{WSA}^{-1/3} + n_{WSB}^{-1/3}} \cdot \left(Q \cdot \left(n_{WSA}^{1/3} + n_{WSB}^{1/3} \right)^2 - P \left(\Phi_A^* - \Phi_B^* \right)^2 - R_m \right)$$

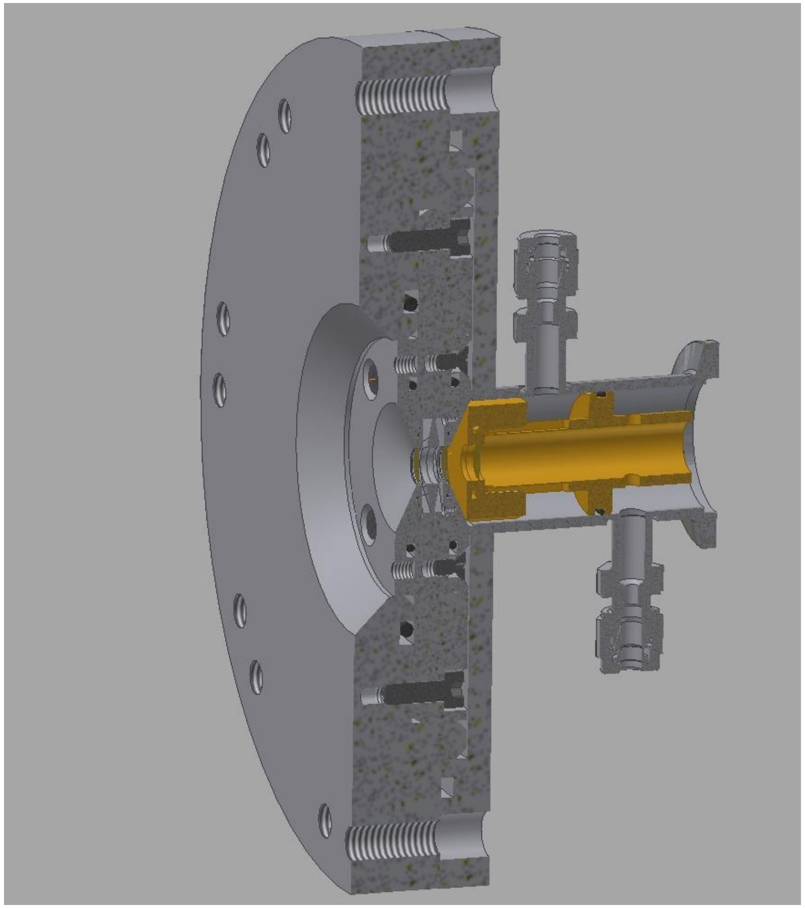
$$V_{Asol} = V_A \cdot \left(1 + a \cdot \left(\Phi_A^* - \Phi_B^* \right) \right)^{3/2}$$

n_{WS} = electron density at the boundary
V_{Asol}= molar volume of the species in solution
Φ^* = chemical potential of electrons
$P/Q/R_m$ = proportionality factors (empirically derived)

Semi empirical model adjusted to hundreds of binary compounds

A.R. Miedema, J. Less-Comm. Met. 46, 67 (1975)

Thermal Release - Experimental



- Si, Ti, Steel, Ni, Ge, Y, Zr, Nb, Mo, Rh, Hf, Ta, W, Re, and Pt matrices irradiated

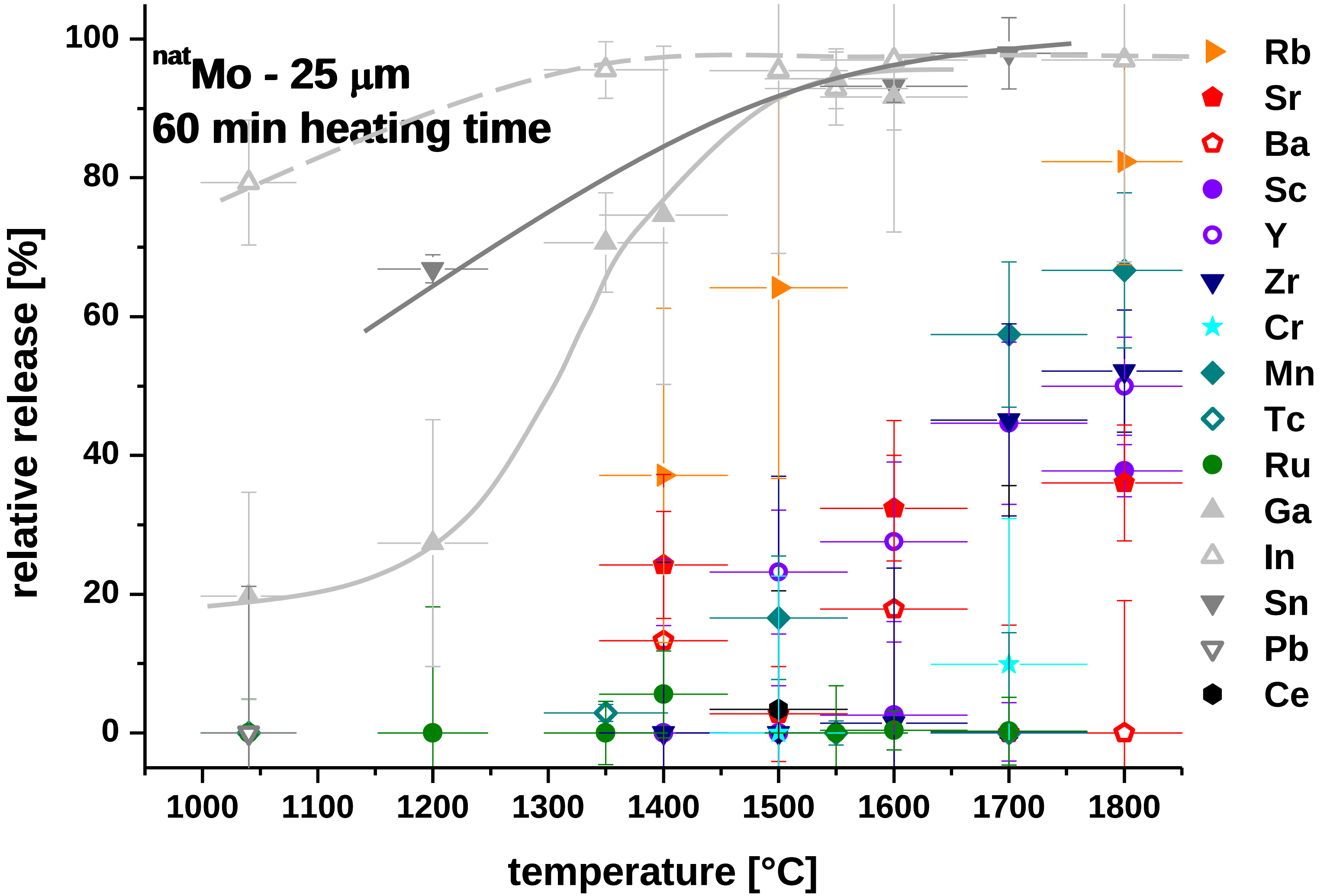
- Irradiation of metal foils with ^{40}Ar or ^{4}He from the Philips Cyclotron @ PSI

- E_{Beam} ≈ 305 MeV (^{40}Ar)
  80 MeV (^{4}He)

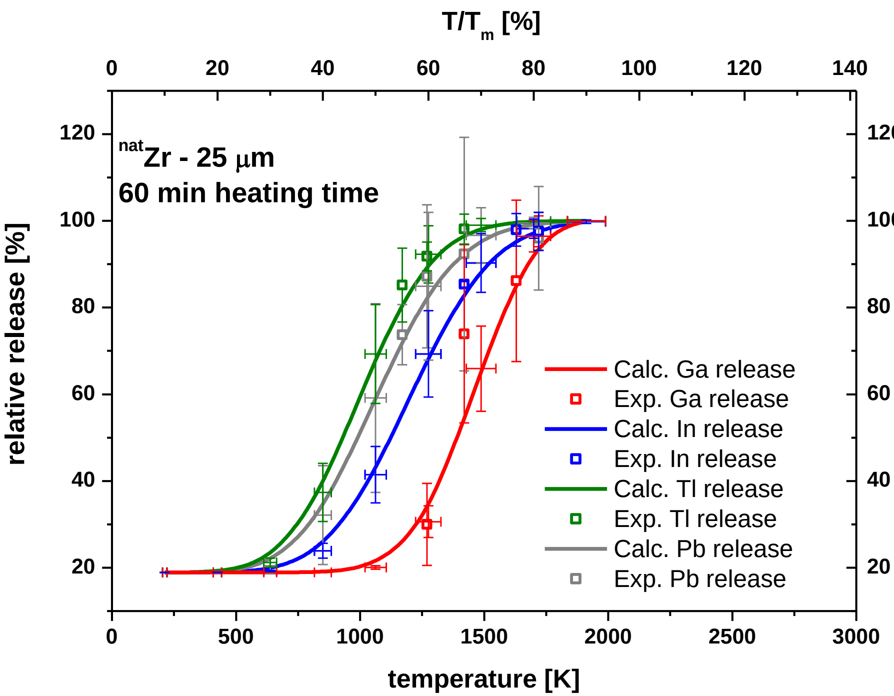
- I_{Beam} ≈ 500 nA (^{40}Ar)
  125 nA (^{4}He), both electrical

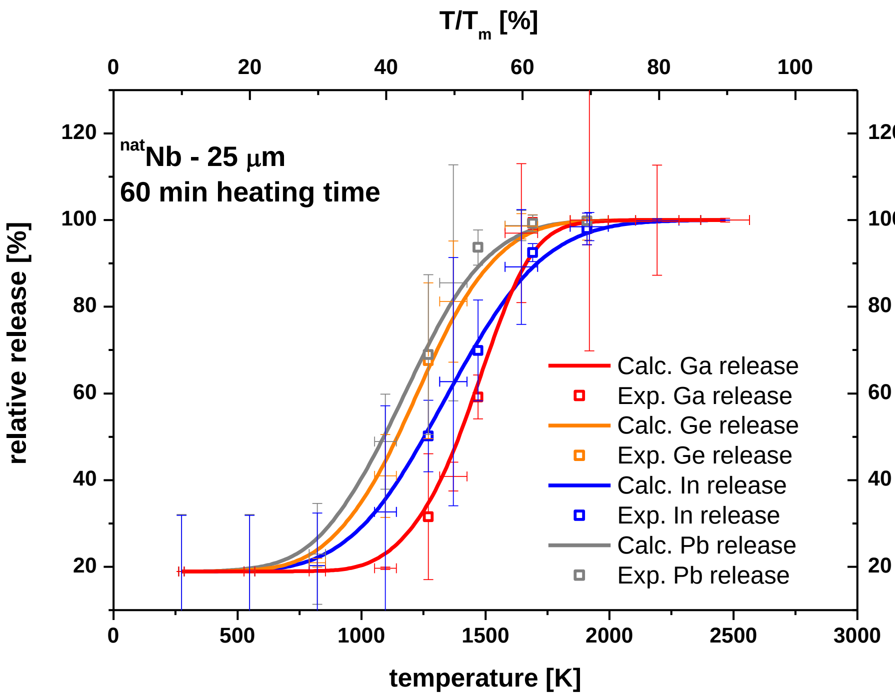
- Zn, Cd, and Hg alloy target used
  Ga/Ge, In/Sn, and Tl/Pb isotopes and transfer products from the beam and the matrices

Thermal Release - Experimental



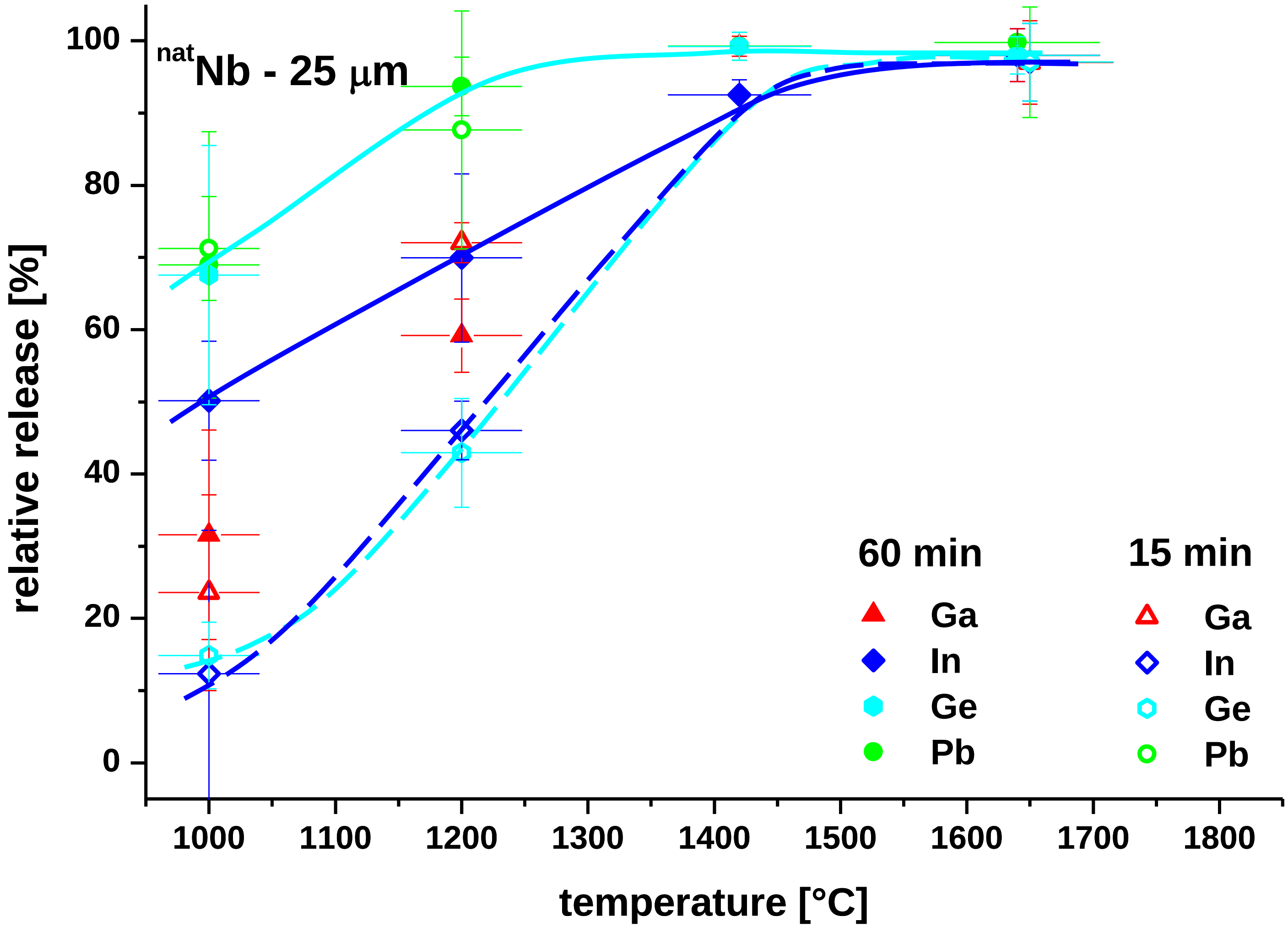
Thermal Release - Experimental



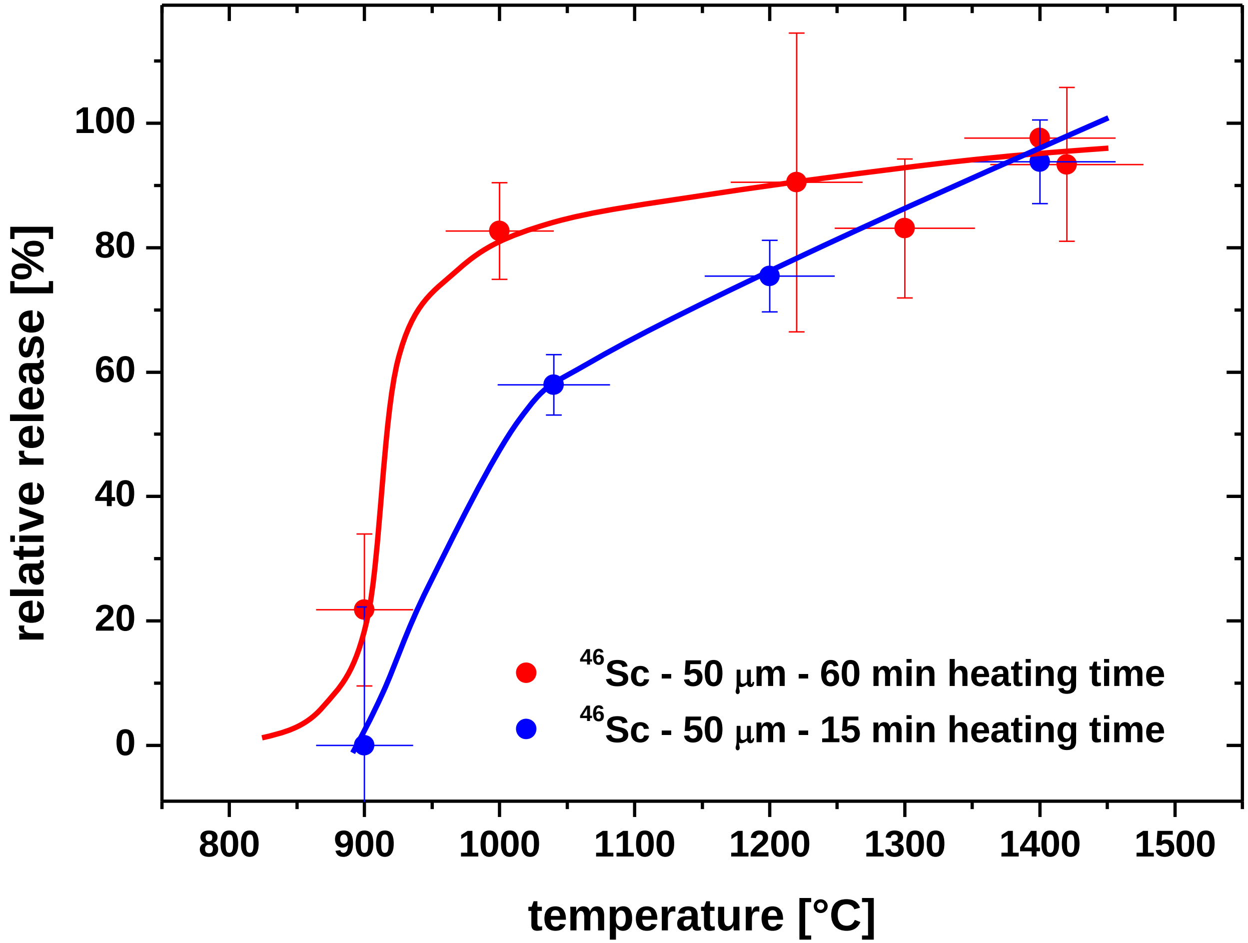


High melting metals as W, Re, Mo, and Nb are best suited

Thermal Release - Experimental



Thermal Release - Application



D. Wittwer et al, Radiochim. Acta 99, 193 (2011)

Thermal Release - Experimental

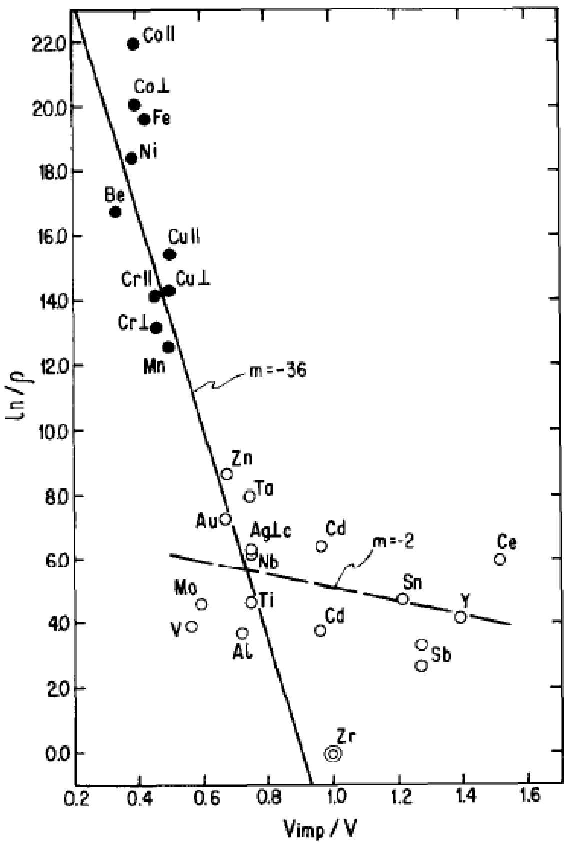
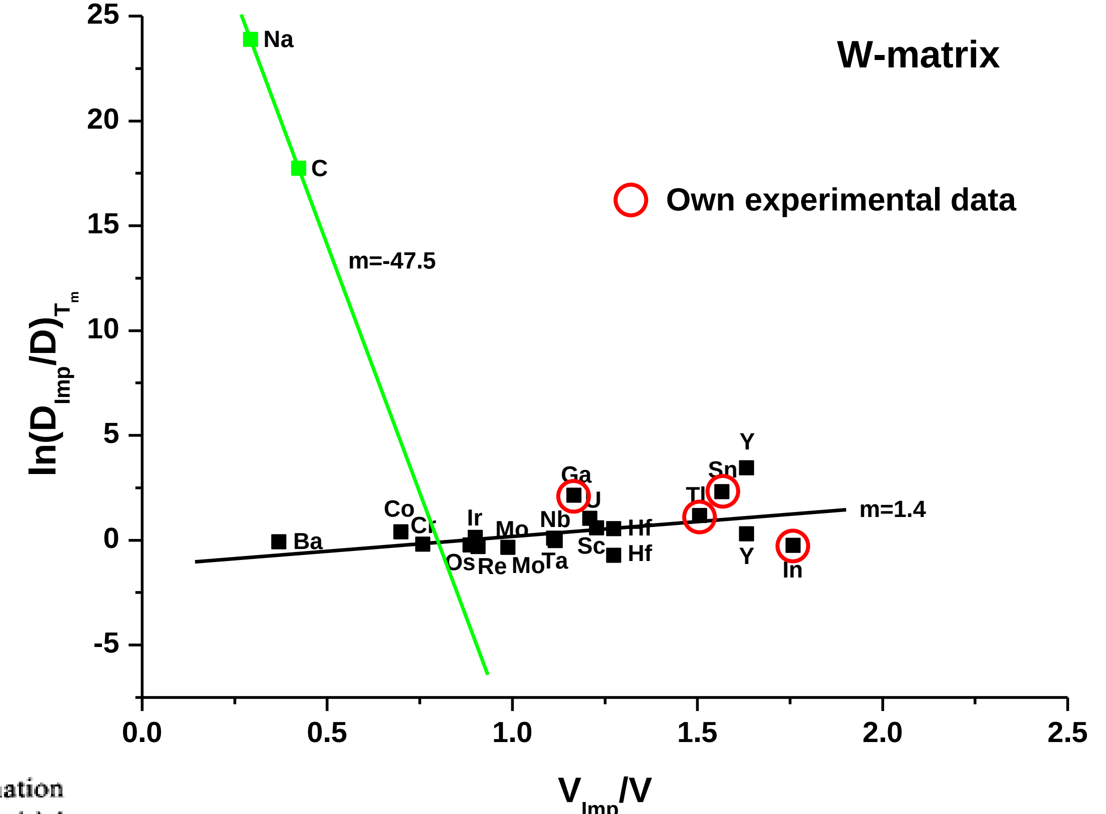


Fig. 1. ln ρ(840 °C) versus V_{imp}/V for hcp-Zr. Approximation by two straight lines; m: slope of the lines; (●) interstitial diffusers, (○) substitutional diffusers, (◎) hcp-Zr self-diffusion.

R. Tendler et al, J. of Nucl. Mat. 150, 251 (1987)



Data from:
G. Neumann, Self Diffusion and Impurity Diffusion in Pure Metals, Pergamon Materials Series

Thermal Release - Conclusion

- Volatile p-elements can be quantitatively released from metal matrices by heating

- High melting metals are more appropriate (under investigated conditions) due to lower release to melting temperature ratio (lower exposure)

- Phase changing metals as titanium, zirconium, or hafnium do not necessarily promote a fast release

- Time dependence irrelevant above certain temperatures ~60% T_m (minutes time scale)

- Possibilities are available to calculate an hypothetical release of SHE form metal matrices

Acknowledgements

People:

- Accelerator and ECR crew:
  PSI: Philips cyclotron
- Tech-shops @ University Bern, and PSI

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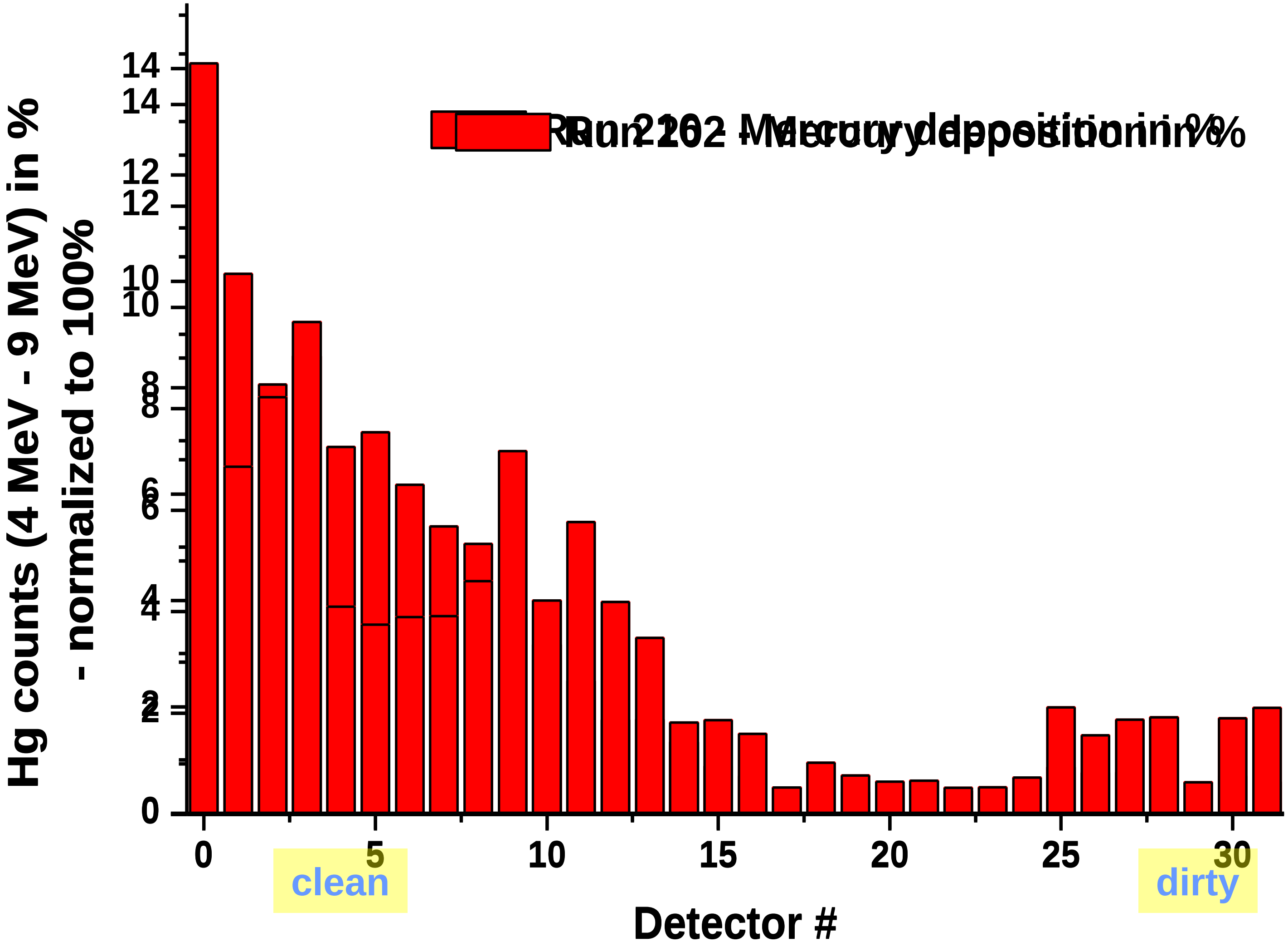
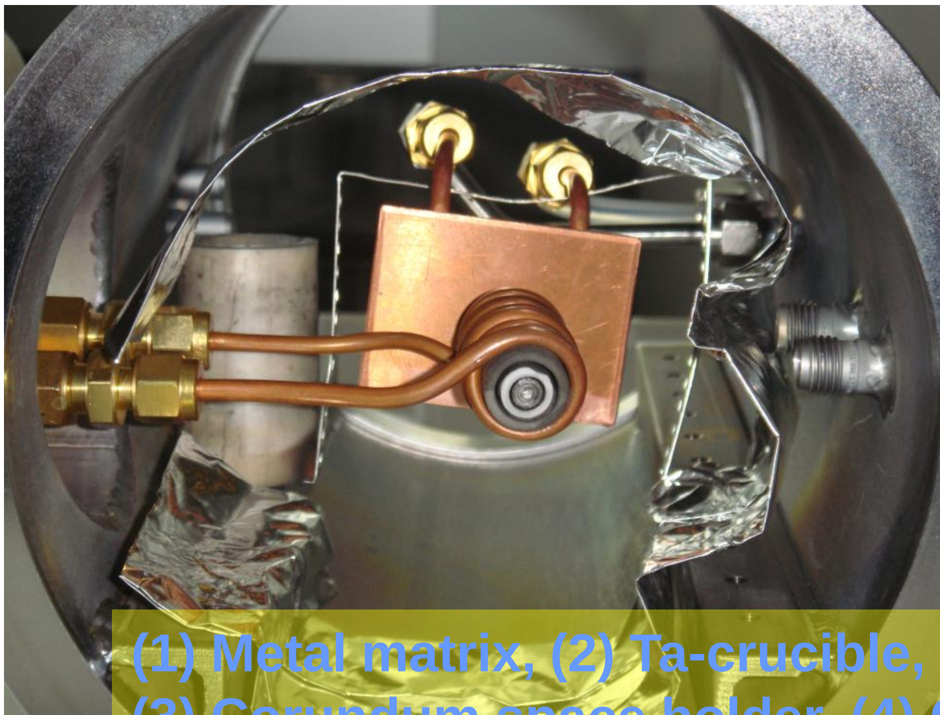
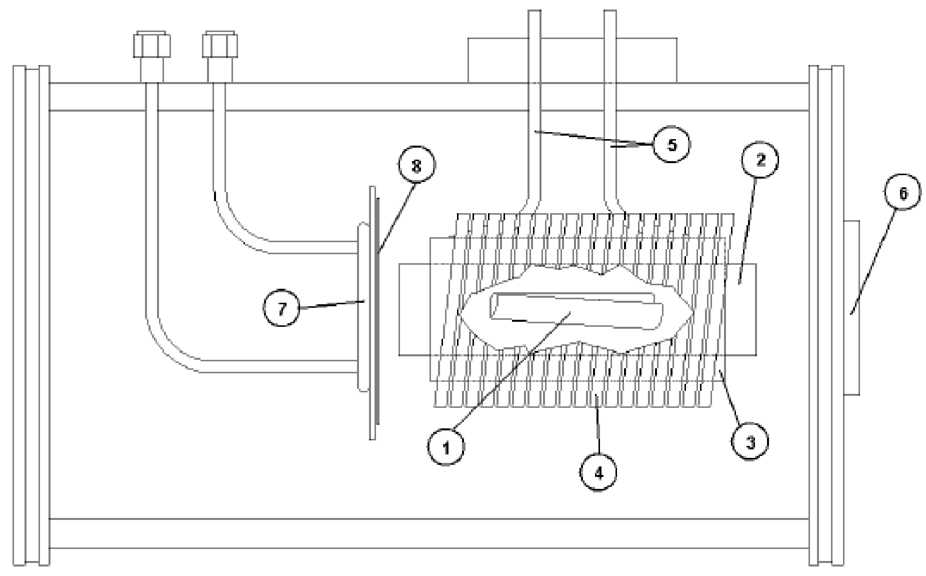
-Swiss National Science Foundation

Introduction



Thermal Release - Experimental





(1) Metal matrix, (2) Ta-crucible,
(3) Corundum space holder, (4) Copper coil for induction,
(5) Cooling for the furnace, (6) Vacuum window,
(7) Cooled carrier holding, (8) Foil to collect released products

Thermal Release