Adsorption Interaction of ^{113m}In and $^{200\text{-}202}$Tl Isotopes with Quartz

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for collaboration of

Outline

1) Introduction
- Methods for studying chemical properties of superheavy elements (SHE)

2) Experiments with lighter analogues of SHE
- Preparation and purification of ^{113m}In and $^{200-202}$Tl
- Model experiments with ^{113m}In and $^{200-202}$Tl

3) Summary

4) Way to go

Methods for studying chemical properties of superheavy elements (SHE) and their lighter homologues

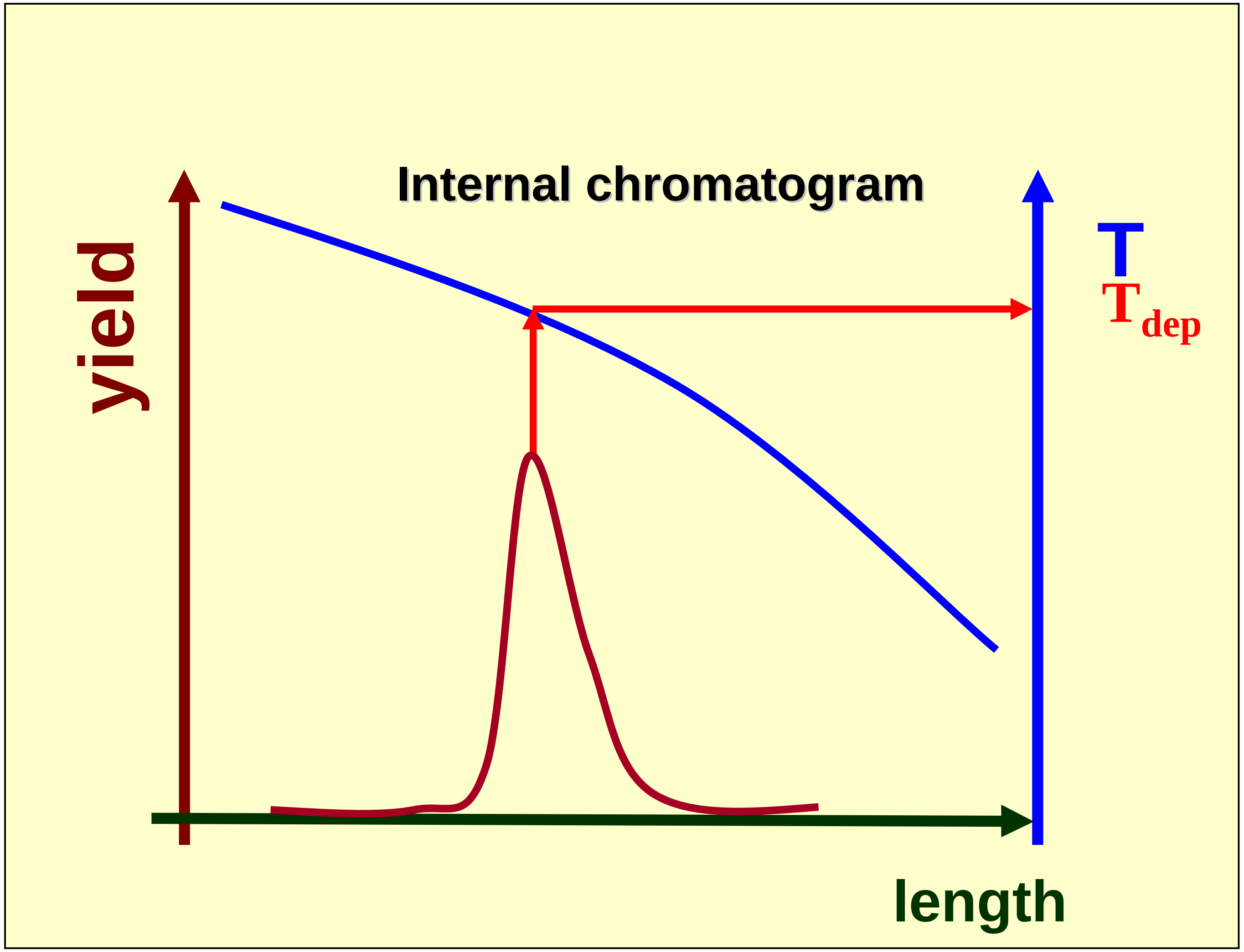
1) Gas phase methods: thermochromatography and isothermal chromatography
2) "Wet-chemical" methods
3) Electrochemical methods *etc*

Main methods used in the present research:

Isotope separation – "wet chemistry" and gas phase method

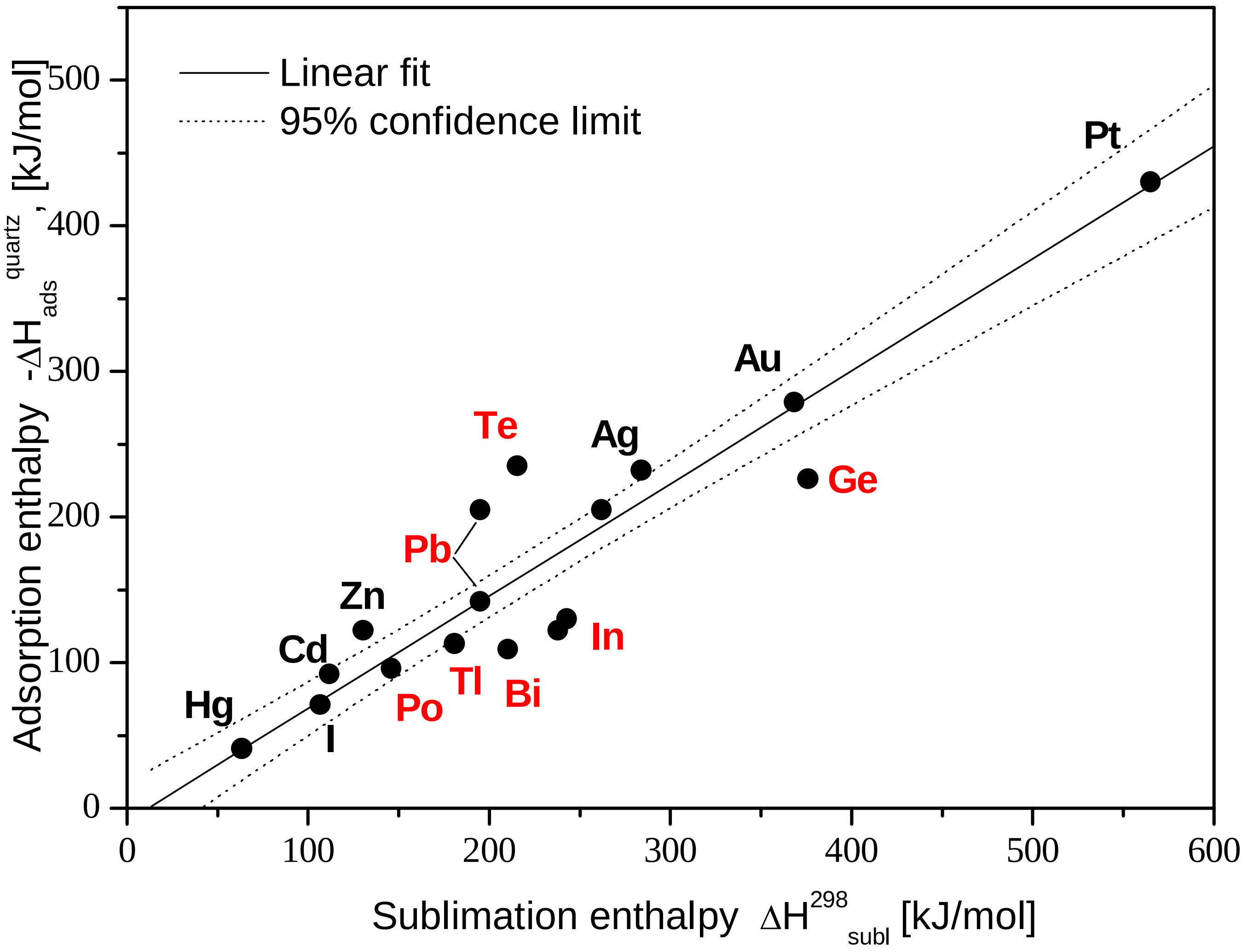
Chemical properties – thermochromatography

Thermochromatography



Result: $T_{dep} \rightarrow \Delta H_{ads}$

Motivation:
Unclear speciation of p- and s- elements (quartz)



Experiments with lighter analogues of SHE

Isotope production and purification

In-113m and Tl-200-202

1) Neutron activation method
"+" available at PSI upon request
"+" high specific activities of isotopes can be reached
"-" only long lived isotopes can be studied (exception - generators)
"-" good separation methods for specific isotopes are needed
"-" contaminations with carrier amounts

2) Cyclotron produced isotopes:
"+" practically no limitation in half-lives
"+" high isotopic purity and carrier free amounts
"+" variety of isotopes (combination target/beam)
"-" not available by request (scheduled beam-times)

3) Commercially available generator systems
"+" easy to work with
"-" price and shipment time
"-" available only for limited isotopes (radiopharmacy application)

Model experiments with ^{113m}In and $^{200-202}$Tl

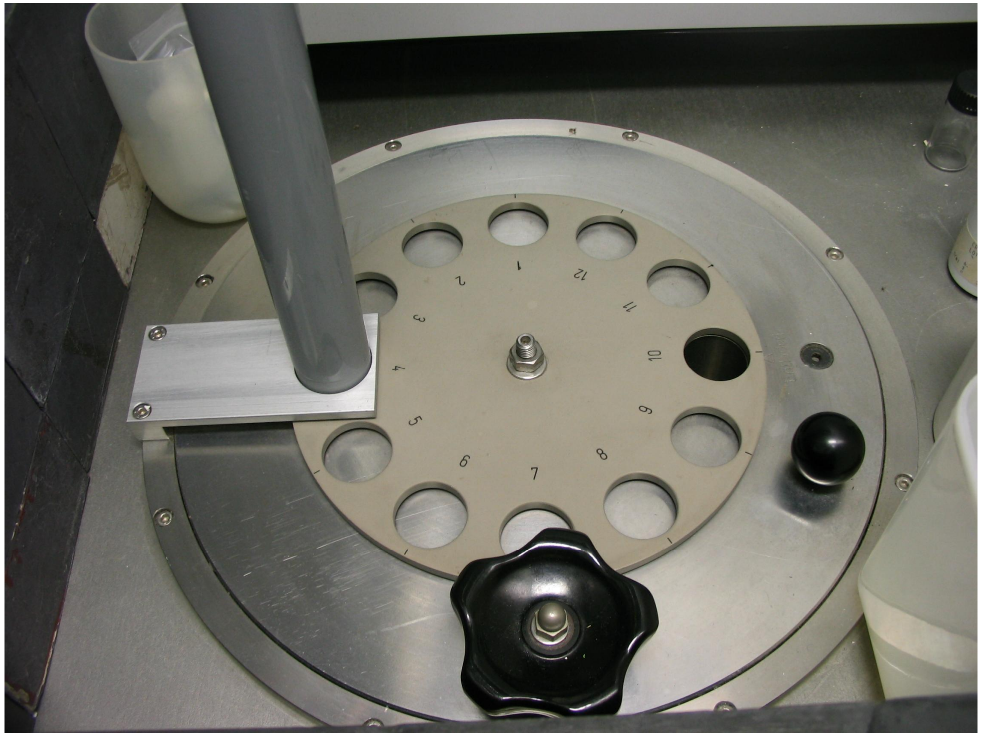
^{113m}In production and purification

1) ^{nat}Sn metal irradiation (0.5 g sample, 2 hrs)
2) Sample dissolution in $HCl_{(conc)}$
3) Deposition on anion-exchange column Dowex® 1X8
4) Elution with 1M HCl

^{113}Sn → ^{113m}In, $T_{1/2}$ = 115.1 d → 99.5min, IT 392 keV

Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

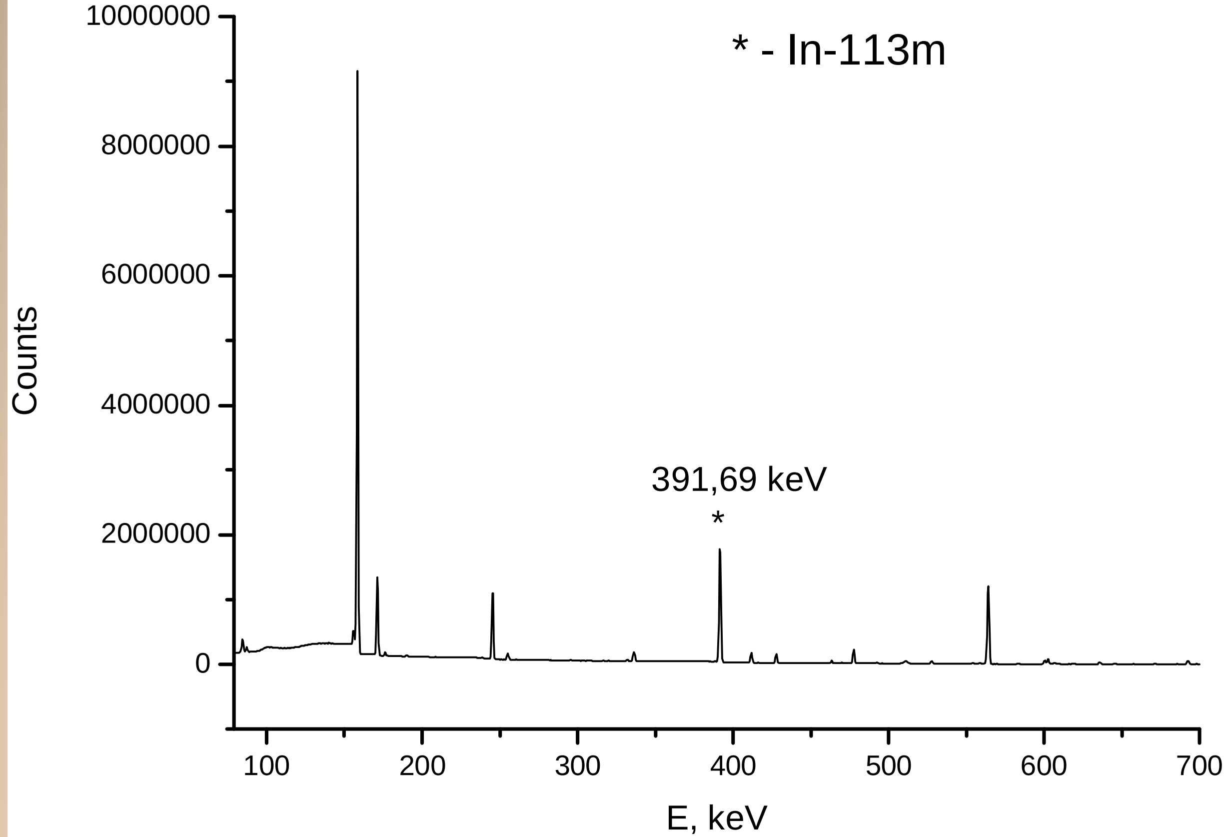
^{113m}In and $^{200\text{-}202}$Tl production by neutron irradiation



Pneumatic system for remote irradiation of samples by mediate neutron flux

Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

^{113m}In purification results

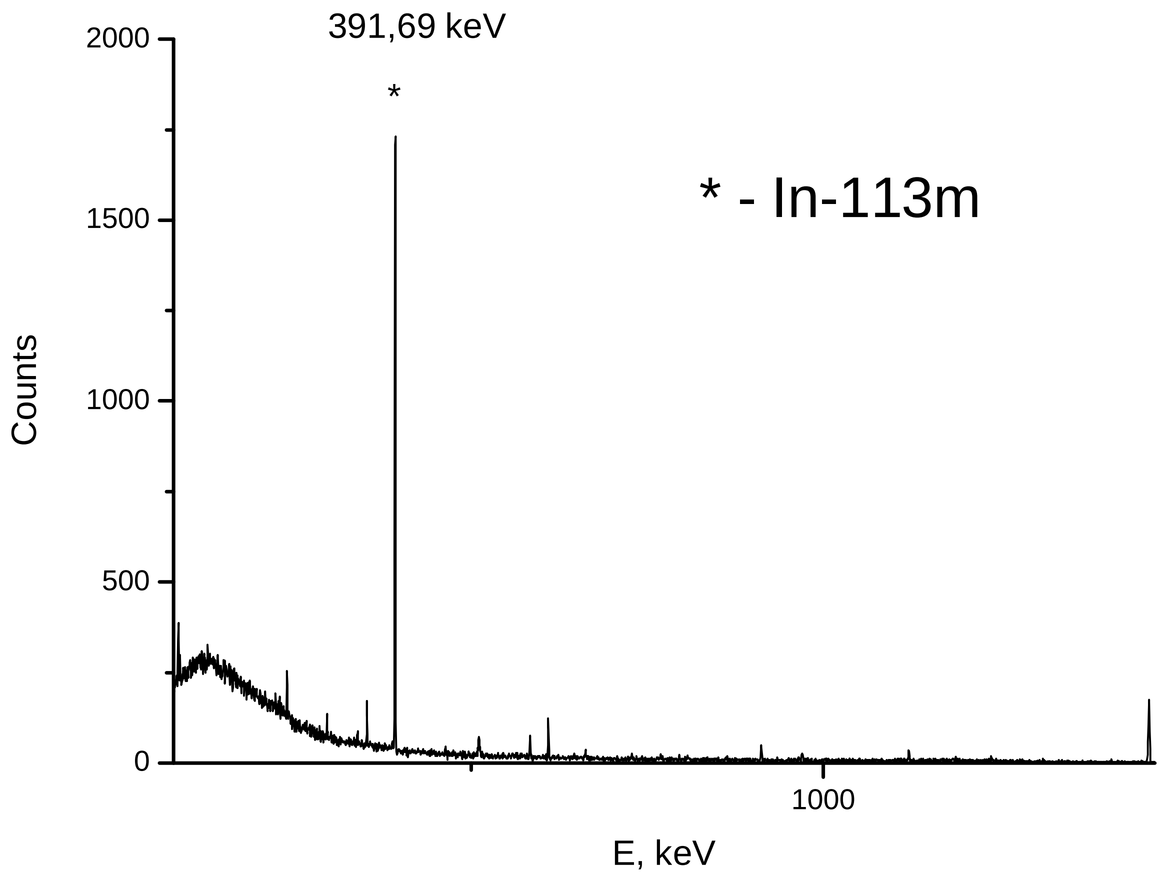


Main activity:
^{125}Sb (427, 87 keV)
^{117m}Sn (158, 56 keV)
^{113}Sn (391, 69 keV)
^{113m}In (391, 69 keV)

Gamma spectrum for irradiated ^{nat}Sn (before purification)

Model experiments with ^{113m}In and $^{200-202}$Tl

^{113m}In purification results



Main activity:
^{113m}In (391, 69 keV)

Gamma spectrum for eluated ^{113m}In

Model experiments with ^{113m}In and $^{200-202}$Tl

$^{200-202}$Tl production

1) $^{nat}PbO_2$ irradiation (0.5g sample, 2hrs)

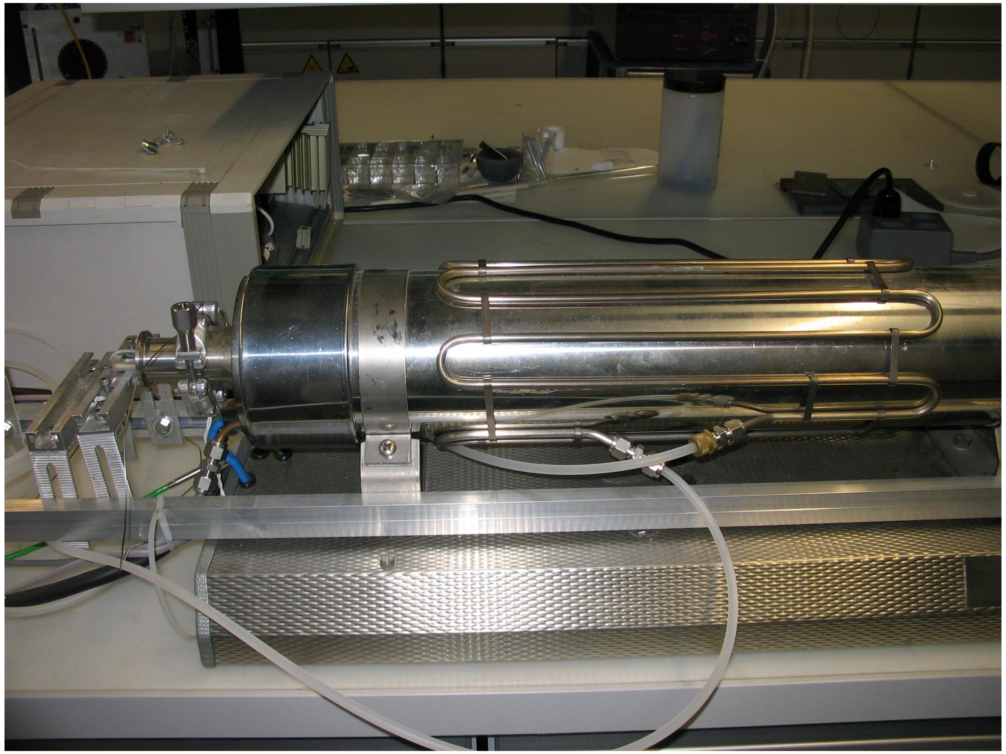
2) Solid $PbO_{2(irradiated)}$ can be used as the source of $^{200-202}$Tl

3) ^{200}Tl ($t_{1/2}$=26.1 h), ^{201}Tl ($t_{1/2}$=73.1 h) and ^{202}Tl ($t_{1/2}$=12.23 d),

4) Carrier-free Tl can be investigated

Model experiments with ^{113m}In and $^{200-202}$Tl

^{113m}In and $^{200-202}$Tl thermochromatography experiment on quartz



Gradient: 1000/1300 – 30/-140 ^{0}C

Column material: quartz

Column length: 110 cm

Carrier gas: Different

Thermochromatography oven set-up

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In-113m and Tl-200-202

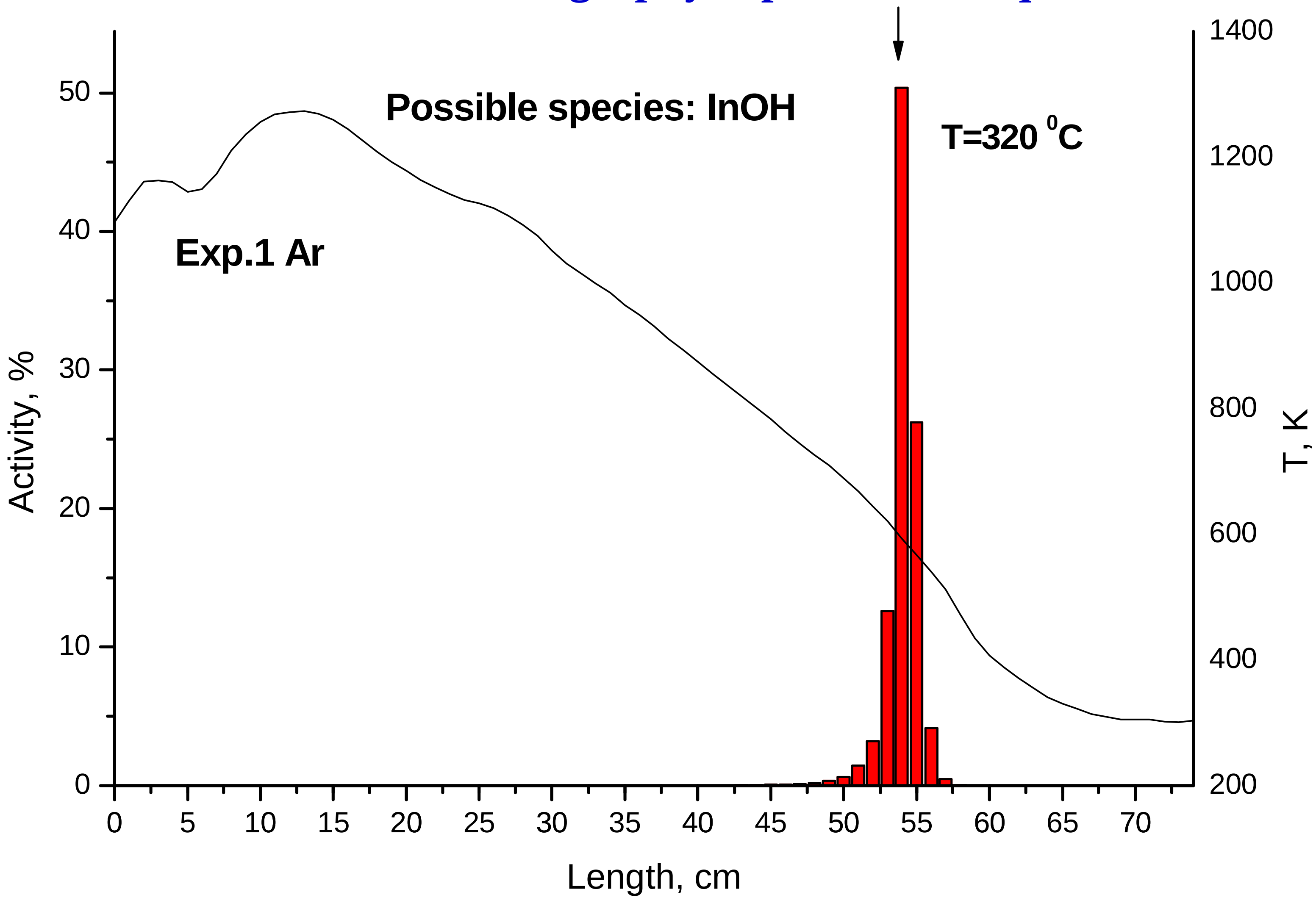
Model experiments with ^{113m}In and $^{200\text{-}202}Tl$

Details of experiments

Starting material	Carrying gas/mixture	Expected species
In-eluated/Ta	Ar 50ml/min	In-metal
Sn-irr./Al_2O_3 , PbO_2-irr./Ta	H_2 25ml/min	In and Tl metals
Sn-irr./Al_2O_3, PbO_2-irr./Ta	O_2 25ml/min	In_2O_3 and Tl_2O_3
Sn-irr./Al_2O_3	O_2/H_2O 25ml/min	$In(OH)_3$ and TlOH
Sn-irr./Al_2O_3	H_2/H_2O 25ml/min	InOH, In_2O, Tl_2O TlOH
Sn-irr./Al_2O_3	He/H_2O 25ml/min	InO, InOH, TlOH, Tl_2O

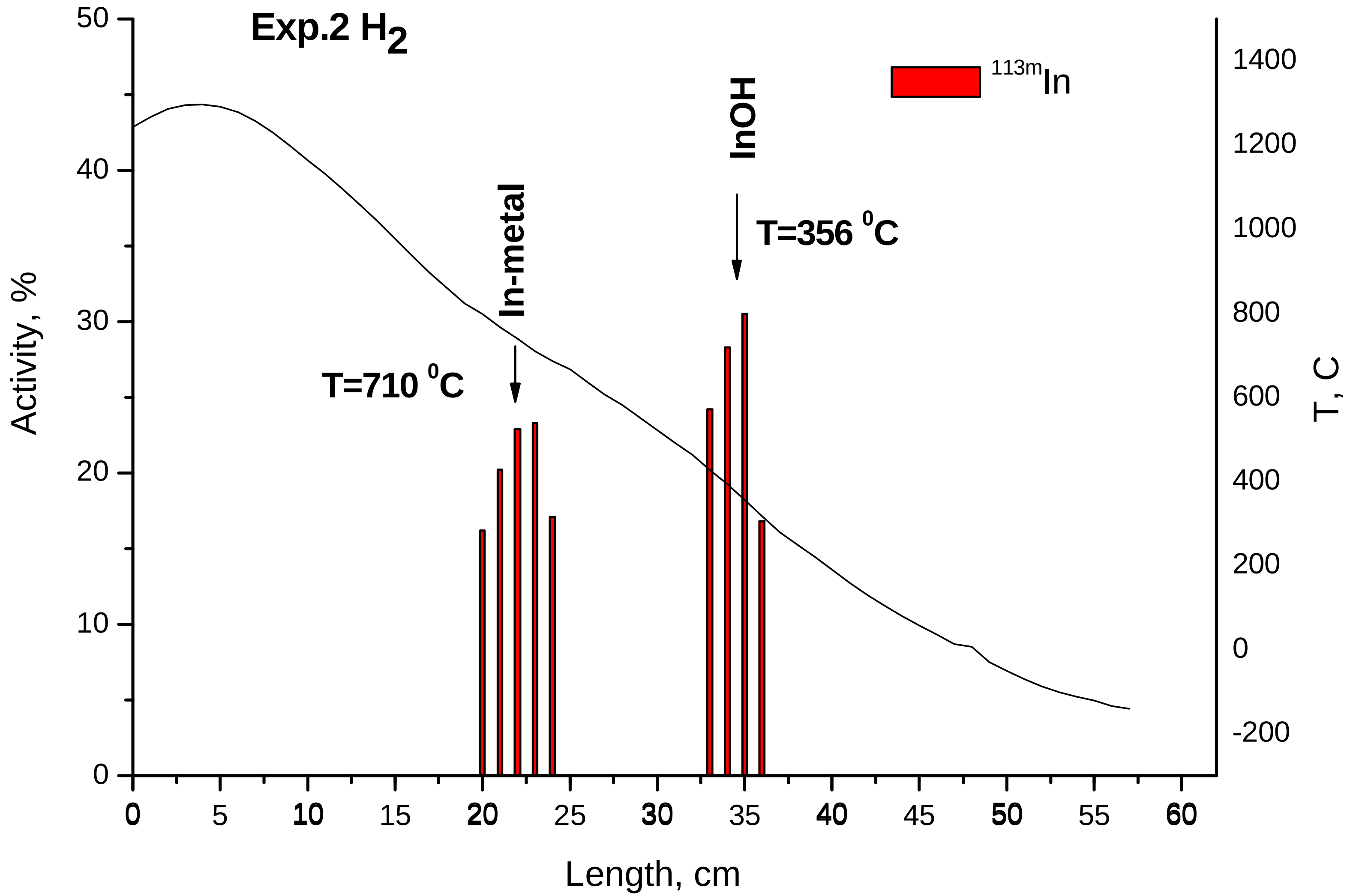
Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

^{113m}In thermochromatography experiment on quartz



Model experiments with ^{113m}In and $^{200-202}$Tl

^{113m}In thermosublimation experiment on quartz



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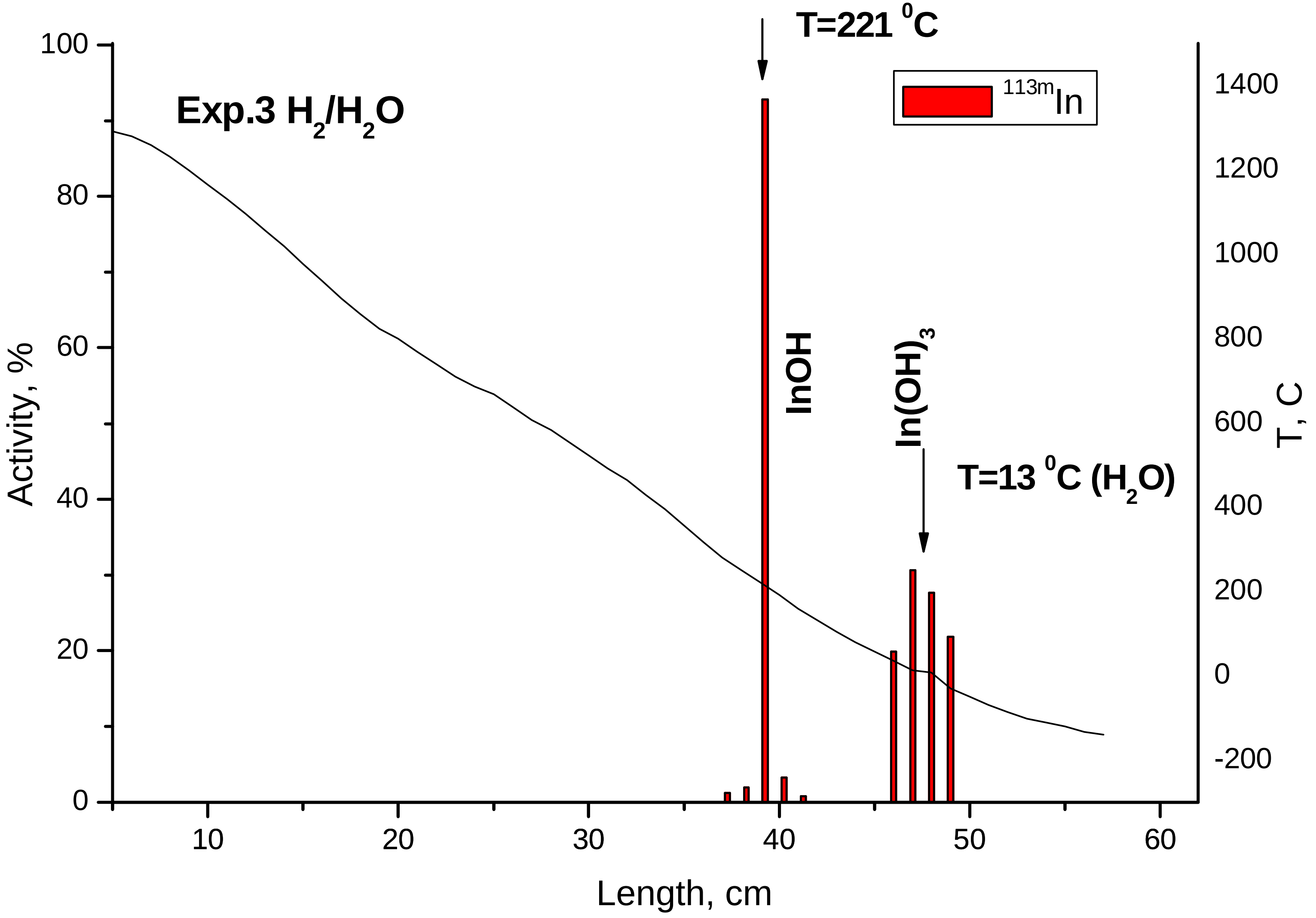
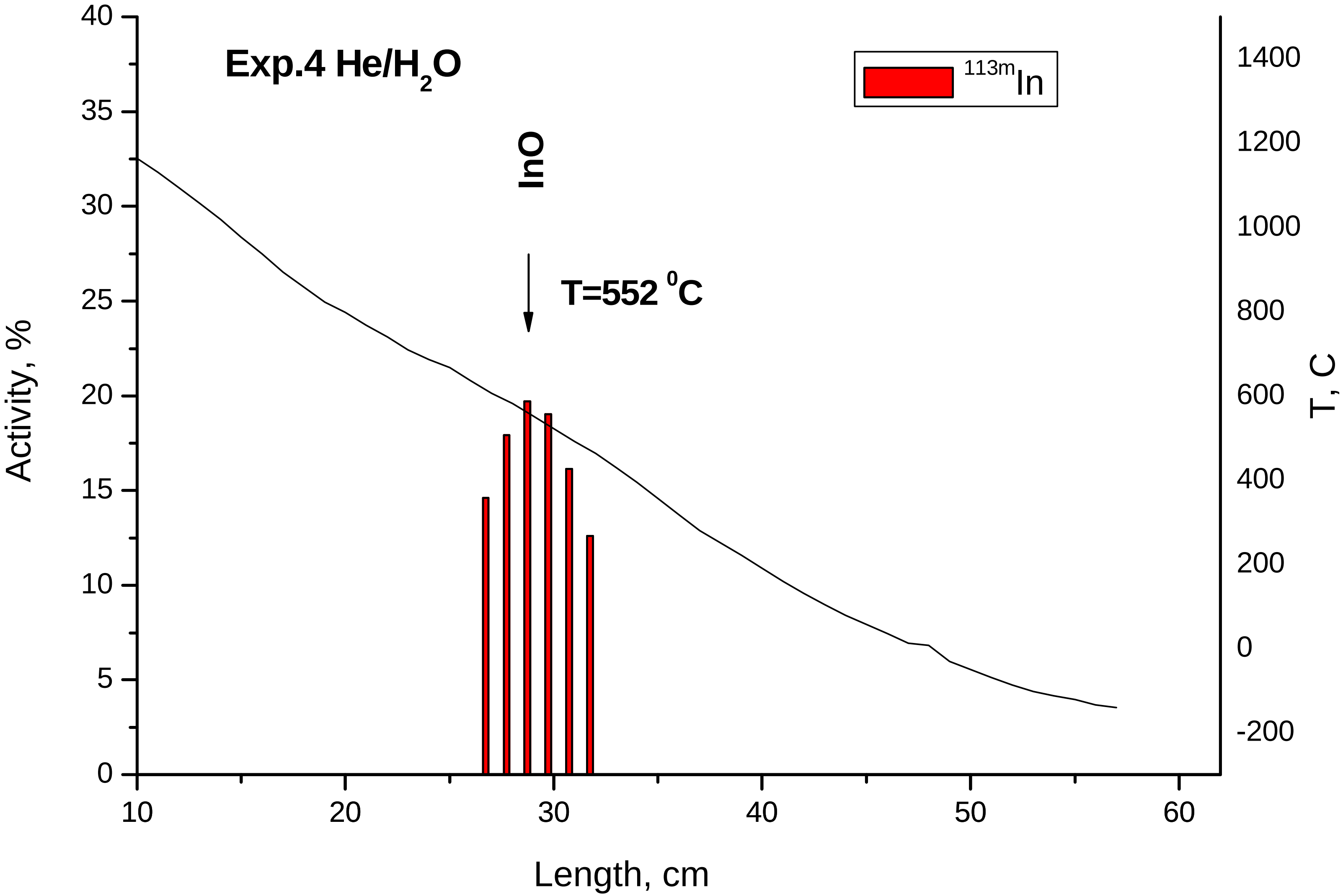
Model experiments with ^{113m}In and $^{200-202}$Tl

^{113m}In thermosublimation experiment on quartz



Model experiments with ^{113m}In and $^{200-202}$Tl

^{113m}In thermosublimation experiment on quartz



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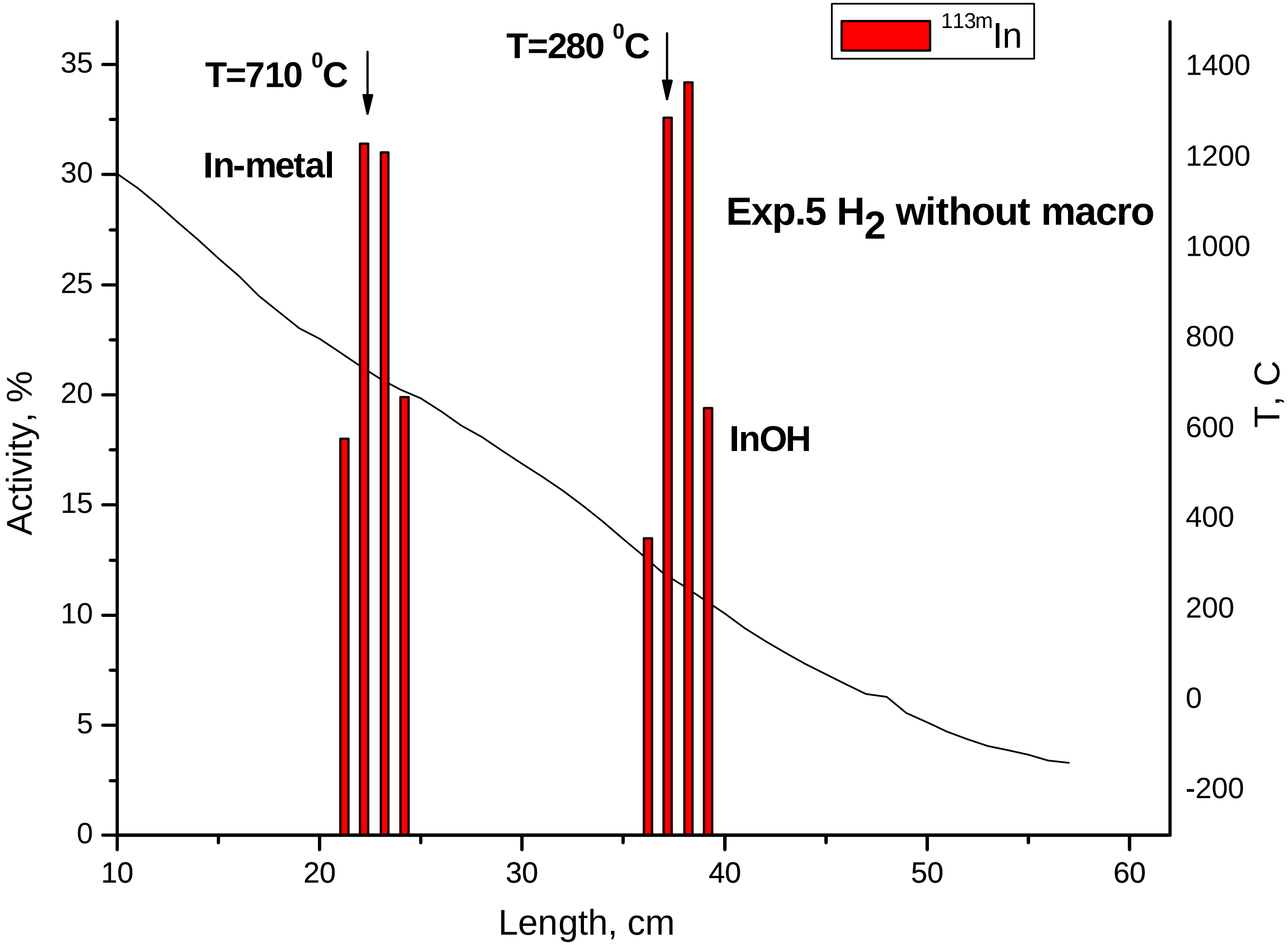
In-113m and Tl-200-202

Model experiments with ^{113m}In and $^{200-202}$Tl

^{113m}In thermochromatography on quartz



Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

^{113m}In thermosublimation experiment on quartz

Exp. 7 Dry Sn-irradiated $+O_2$

Exp. 8 Dry Sn-irradiated $+O_2/H_2O$

↓

No volatile In species – because of low volatility

of oxides and hydroxides in high oxidation states for In

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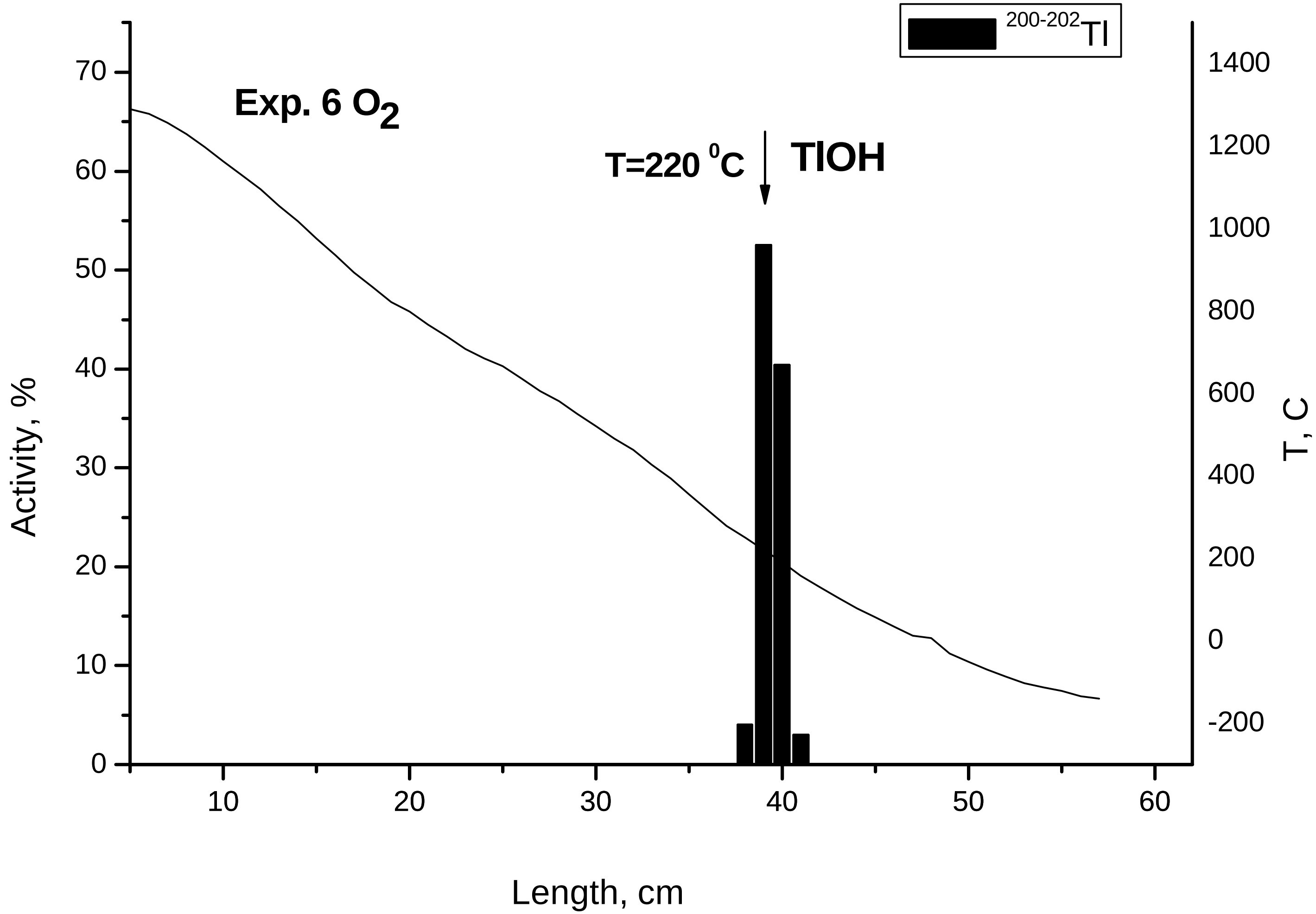
In-113m and Tl-200-202

Model experiments with ^{113m}In and $^{200-202}$Tl

$^{200-202}$Tl thermosublimation experiment on quartz *



*** S. König results (PSI Annual Reports)**

Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

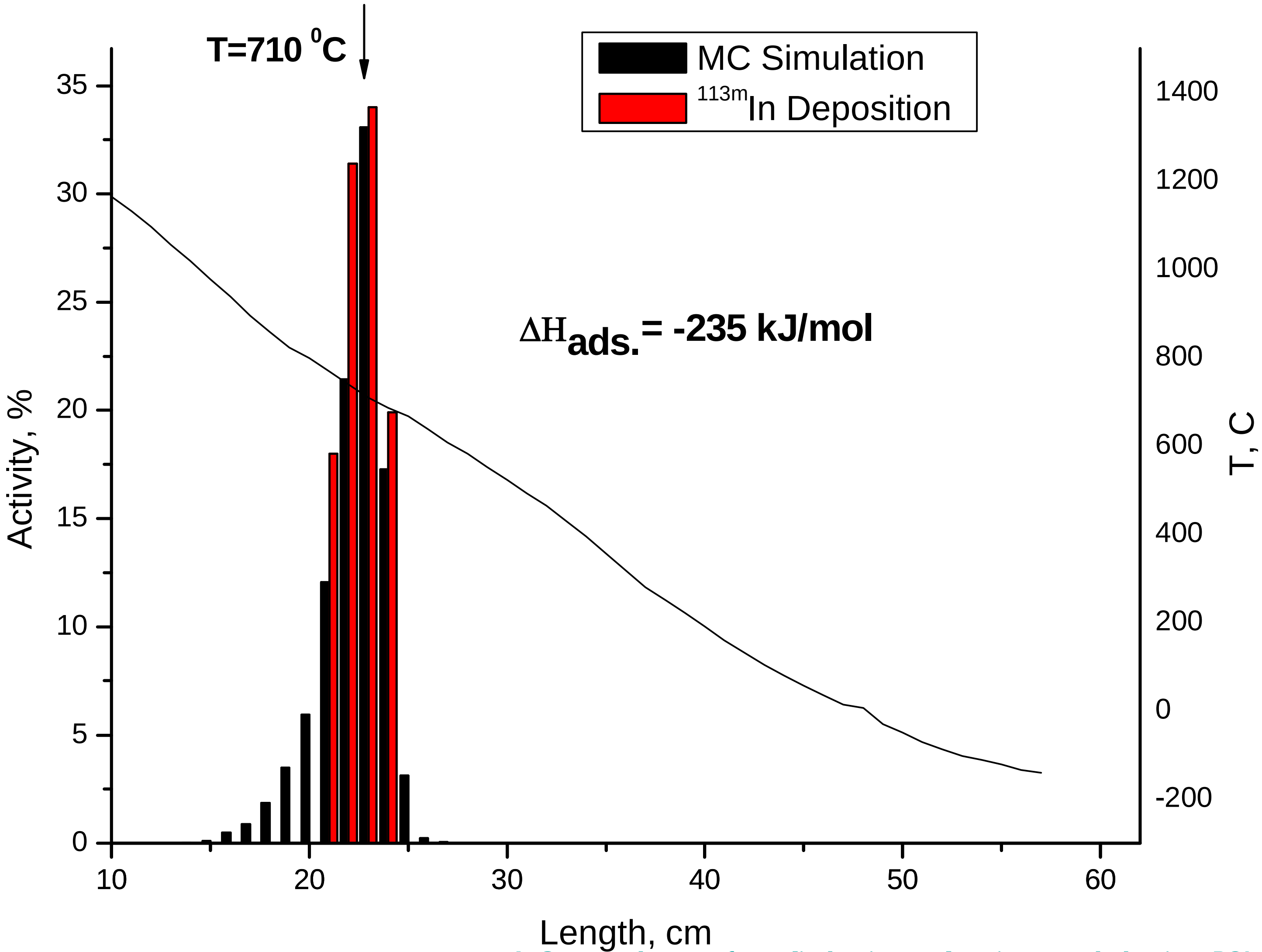
Summary of deposition temperatures on quartz

Isotope	Gas/Mixture	T_{dep} (exp), ^0C	$T_{dep.}$(Lit.)*, ^0C	Possible species
^{113m}In	H_2	320, 356 250,300,280 710(2)	330±20	InOH (?) In
^{113m}In	H_2/H_2O	221 13	-	InOH (?) $In(OH)_3$ (?)
^{113m}In	He/H_2O	552	-	InO (?)
$^{200\text{-}202}$Tl	H_2	202		TlOH
$^{200\text{-}202}$Tl	O_2	220	-	TlOH

* B. Eichler JINR Communication, 1972

Model experiments with ^{113m}In and $^{200-202}$Tl

^{113m}In thermochromatography and Monte-Carlo simulation

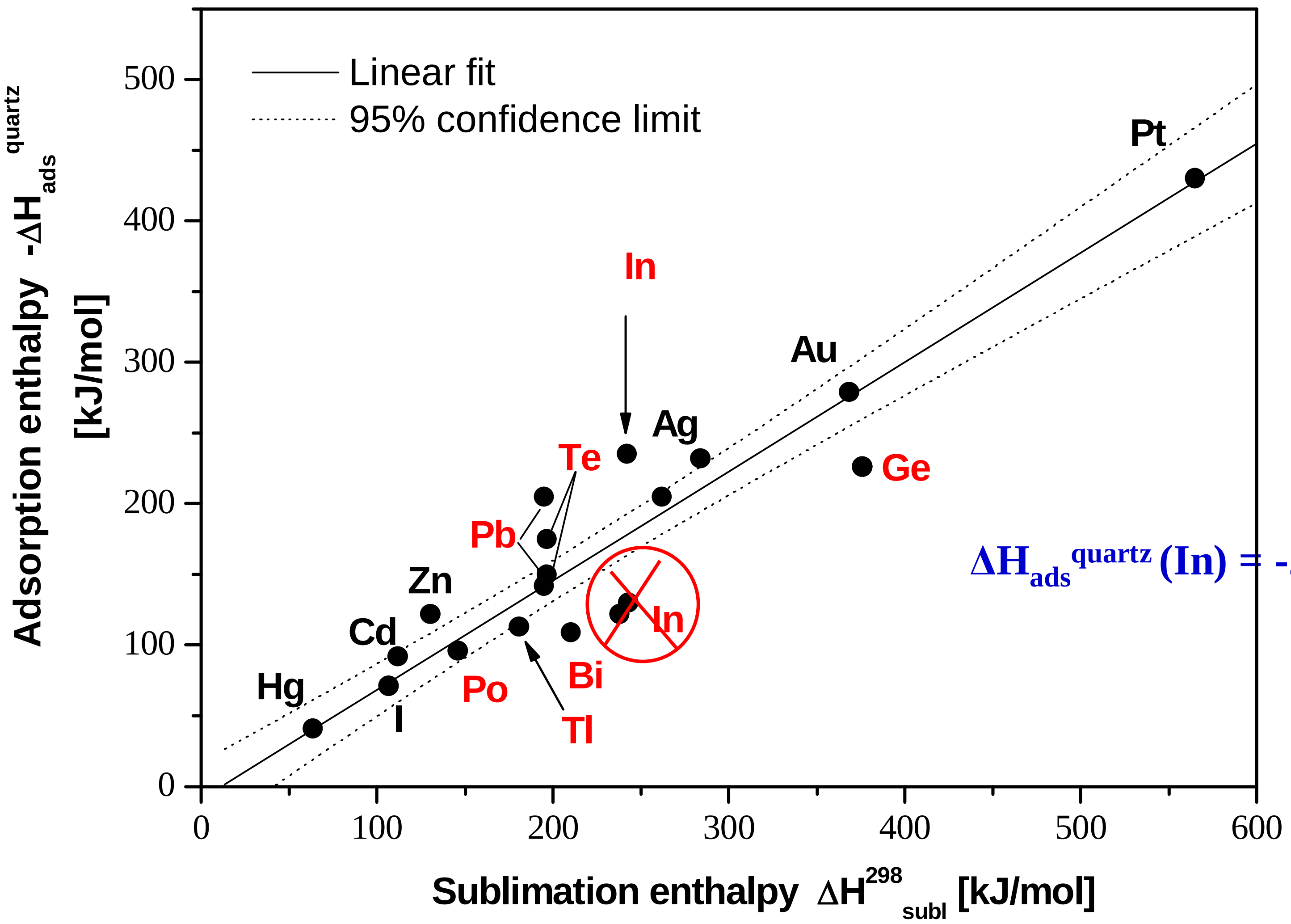


In-113m and Tl-200-202

In-113m and Tl-200-202

Model experiments with ^{113m}In and $^{200-202}$Tl

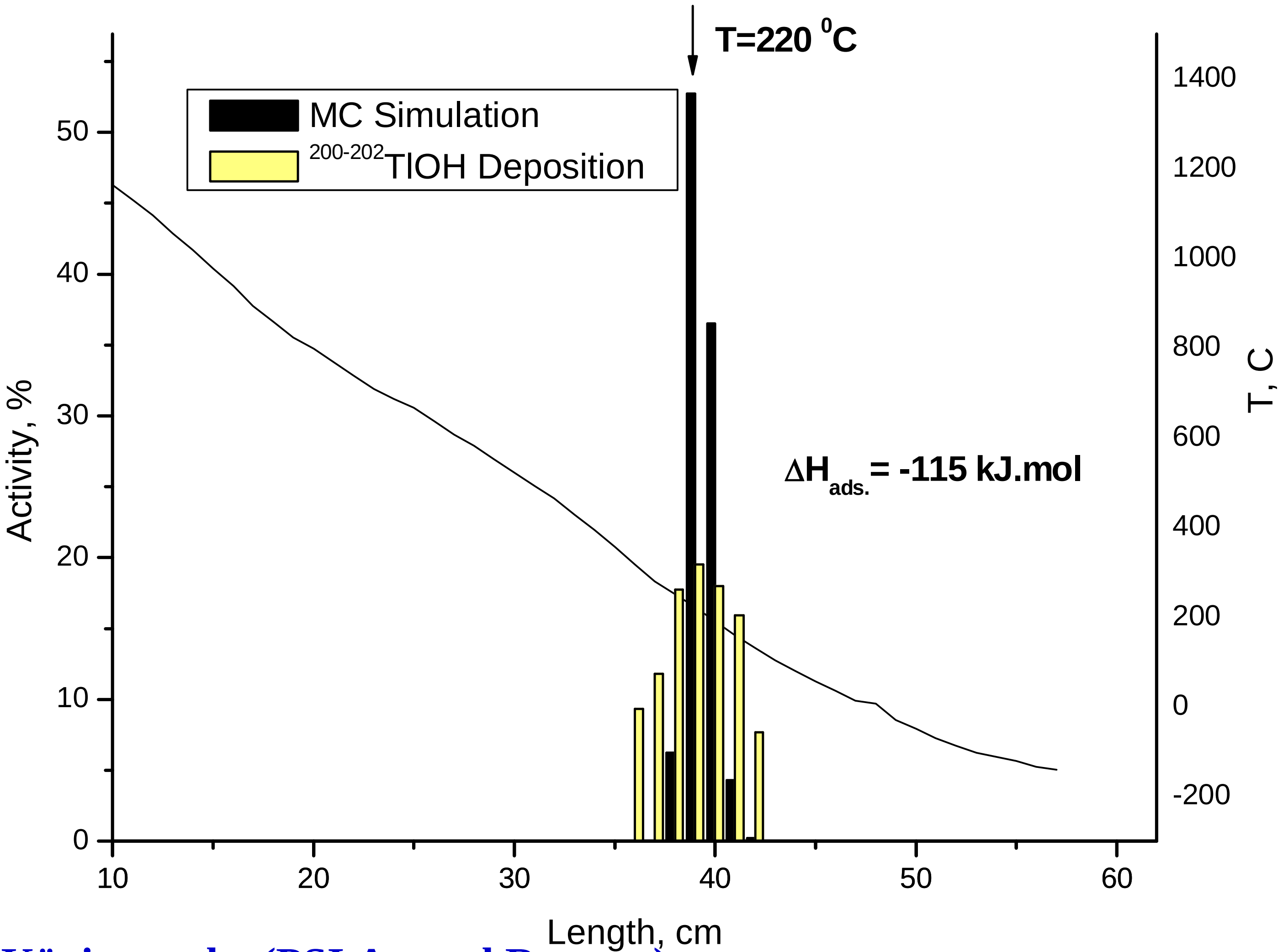
^{113m}In adsorption on quartz



ΔH_{ads}^{quartz} (In) = -235 kJ/mol

Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

$^{200\text{-}202}$TlOH thermochromatography and Monte-Carlo simulation *

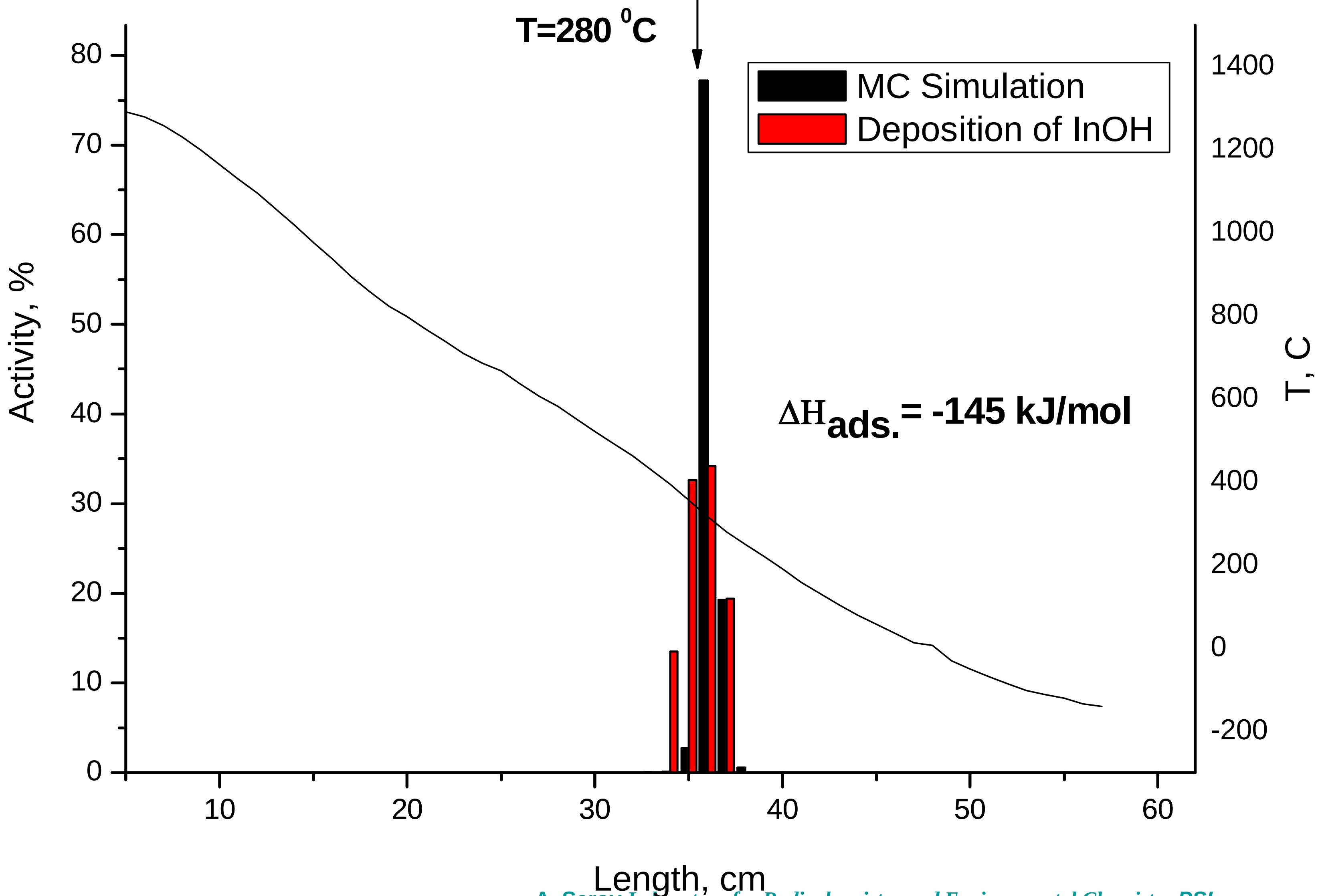


* S. König results (PSI Annual Reports)

In-113m and Tl-200-202

Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

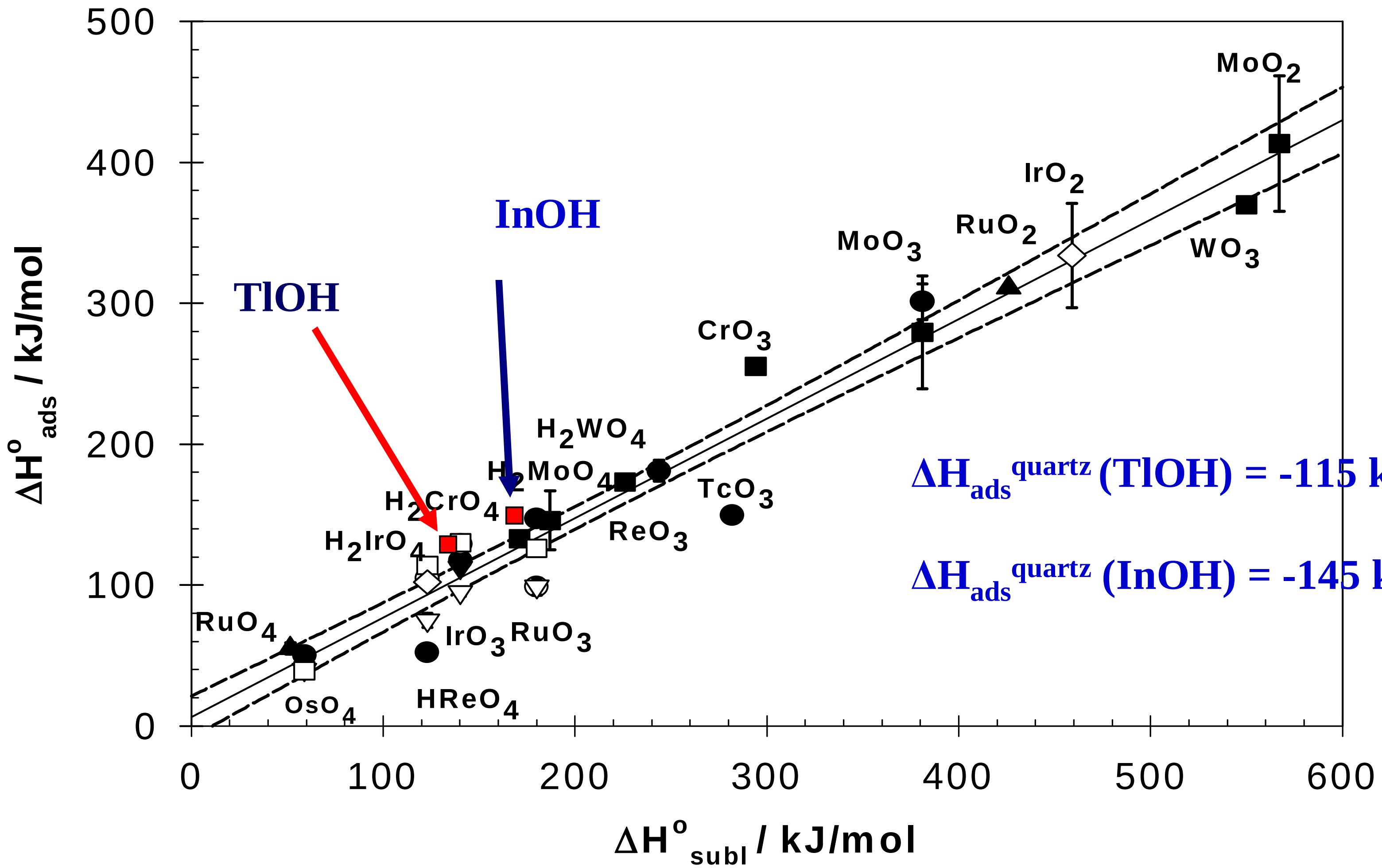
^{113m}InOH thermochromatography and Monte-Carlo simulation



In-113m and Tl-200-202

Model experiments with ^{113m}In and $^{200\text{-}202}$Tl

$^{200\text{-}202}$TlOH and ^{113m}InOH enthalpy of adsorption on quartz



ΔH_{ads}^{quartz} (TlOH) = -115 kJ/mol

ΔH_{ads}^{quartz} (InOH) = -145 kJ/mol

Model experiments with ^{113m}In and $^{200-202}$Tl

Problem of pure metals

Even 99,9998 Sn – contains In and Sb admixtures

Possible solutions of the problem

1) **Ion-exchange chromatography**
- **Works well for In, hard to achieve high activity for Sb**
2) **Gas phase purification**
- **Perfect for In, very poor for Sb**
3) **Tin-organic compounds with Sn-C bonds (tetra-phenyl Tin)**
- **Experiments on the way**

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Summary and Future

Results and way to go

1) Prepared generator system for continuous obtaining ^{113m}In
2) Chemical interaction of ^{113m}In and $^{200-202}$Tl with quartz surface was investigated
3) First time adsorption temperature for In-metal was detected

To do

1) Investigation of In and Tl elemental state, oxides, and hydroxides interaction with Au, Pt

2) Other SHE analogues (Sb, Te etc)

3) Experiments with In, Tl and Pb species in vacuum

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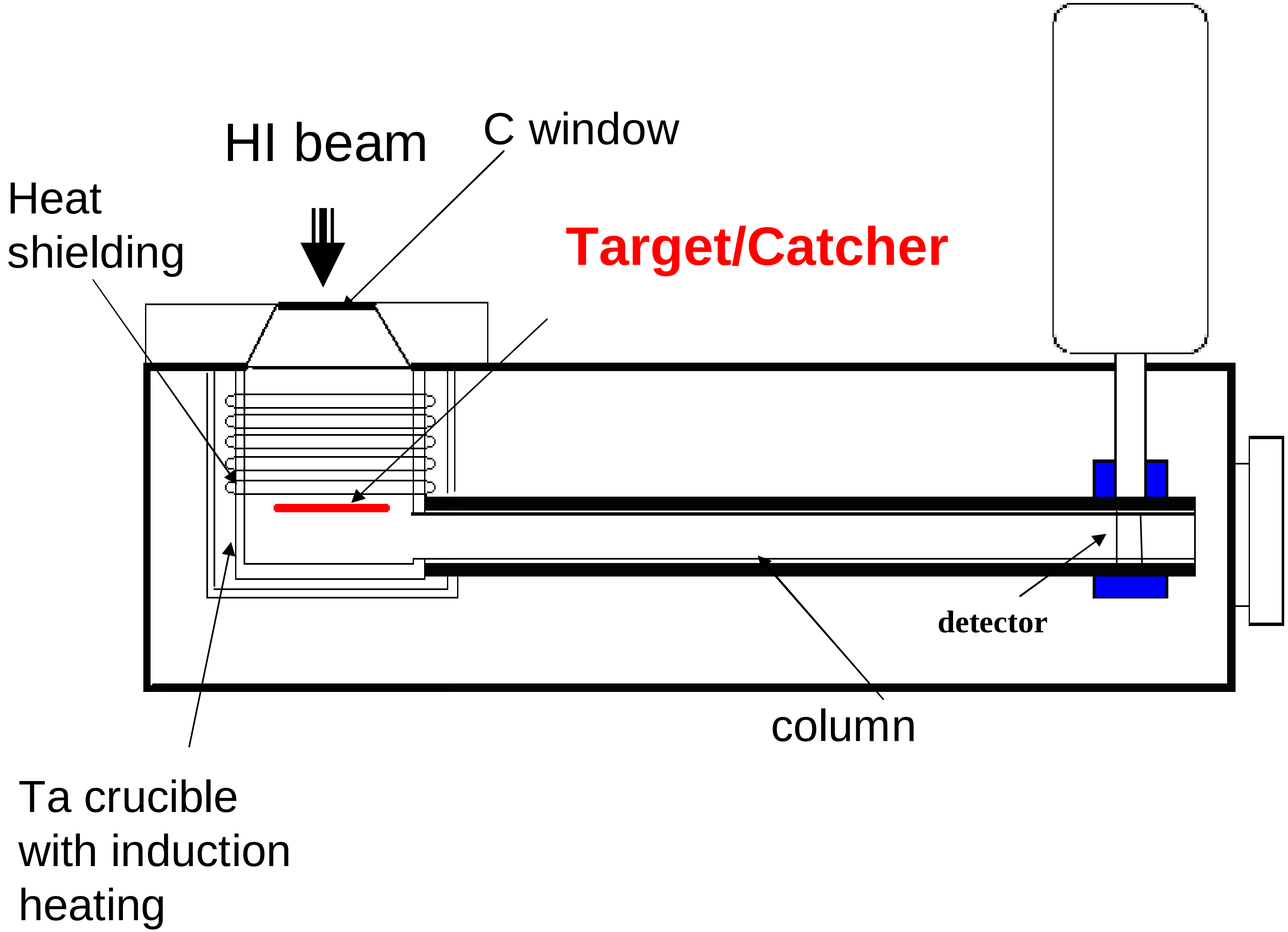
Acknowledgements:
To all members of our lab!

SNF for $$$

Thank you for your attention!

HOTCAT


HI beam
C window
Heat
shielding
Target/Catcher
detector
column
Ta crucible
with induction
heating

Kinetic model of linear

gas adsorption chromatography

Condition :
* Simple reversible single step adsorption process
  i.e. No change of the chemical state during the process and no irreversible reaction with the surface or diffusion into the surface
* zero surface coverage / carrier free amounts = single atoms

ments

Frenkel-type adsorption kinetics:
$\tau_a = \tau_o * \exp(-\Delta H_{ads}/RT)$
phonon frequency of the surface material : τ_o
-> sticking probability if needed

gas transport through tubes:
laminar flow

ΔH_{ads}

diffusion in the carrier gas
Gilliland's eqn.

for short-lived isotopes
radioactive decay: $t_{1/2}$
else: t_{exp}

Monte Carlo Simulation

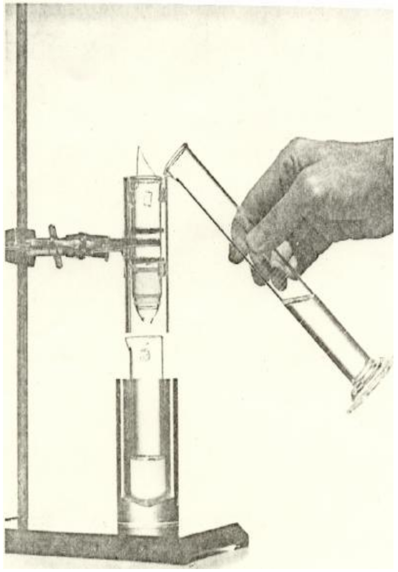
Zvara, I., Radiochim. Acta 38, 95 (1985).

Model experiments with ^{113m}In, ^{125}Sb and ^{125m}Te

^{113}Sn/^{113m}In generator

Q: What is the radionuclide generator?

A: A radionuclide generator is a system which permits separation of a daughter radionuclide from its parent. Parent has a relatively long half-life compared with the daughter.



The Original ^{99m}Tc/^{99}Mo Generator Without Shielding

- Parent: ^{99}Mo as molybdate ($^{99}MO_4^{-2}$)
- Daughter: ^{99m}Tc as pertechnetate($^{99m}TcO_4^-$)
- Adsorbent Material: Alumina (aluminum oxide, Al_2O_3)
- Eluent: saline (0.9% NaCl)
- Eluate: $^{99m}TcO_4^-$

(c) Reproduced from http://www.nucmedicine.com