Vacuum thermochromatography: Revival of a gas phase adsorption separation method to be coupled to a future „CHEMSEP“

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Method

Distinction noble gas ↔ metallic behaviour of E112 and E114 → **gas adsorption techniques on clean metal surfaces (e.g. no oxides, no ice)**

Adsorption properties → **gas chromatography**
→ ***thermochromatography***

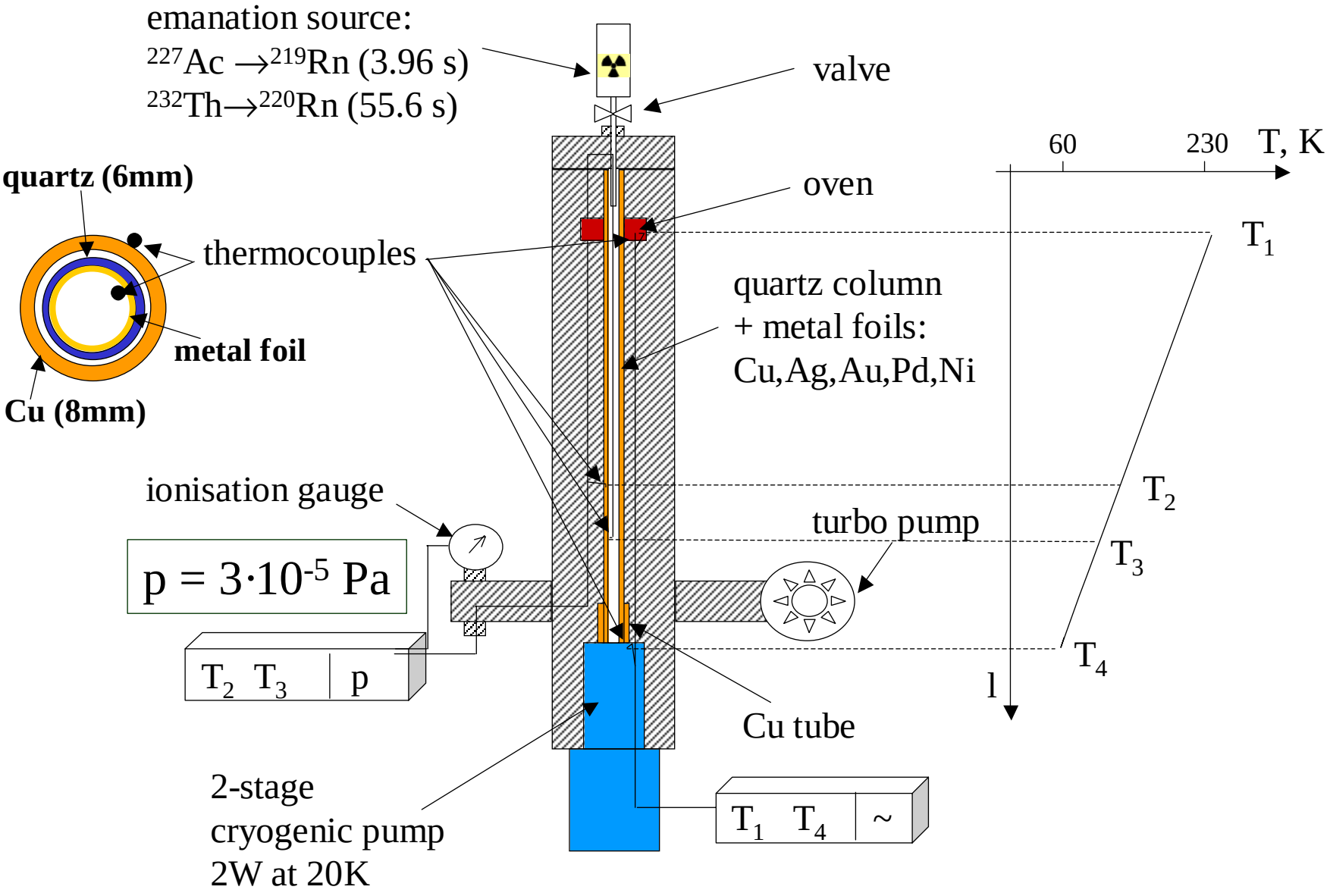
→ Vacuum thermochromatography

(G. Rudstam and B. Grapengiesser 1973)

Advantages:

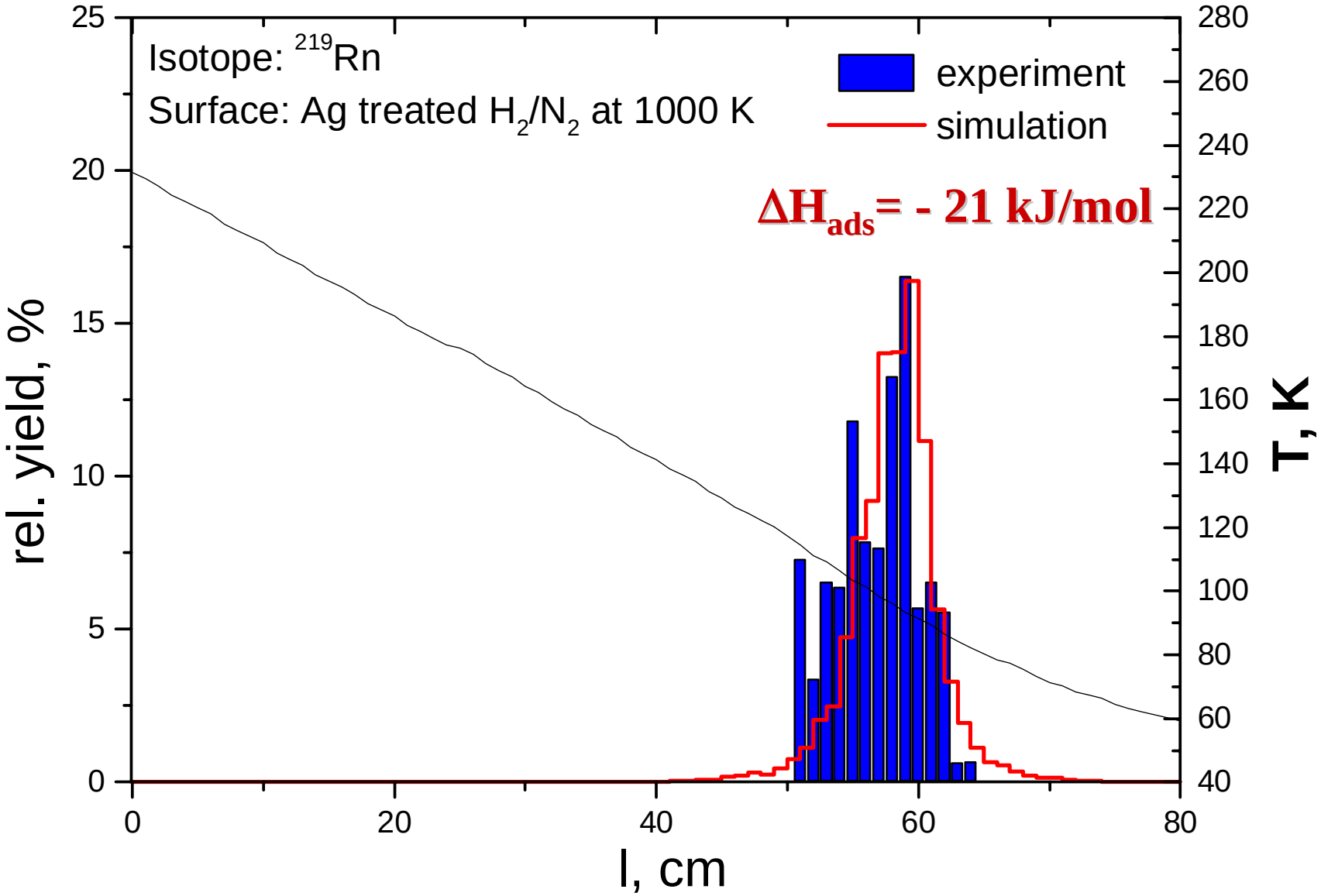
- + no carrier gas = less surface contamination
- + fast separation
- + good heat isolation at low temperatures
- + good α-resolution
- + comfortable coupling to physical separators

Vacuum thermochromatography setup

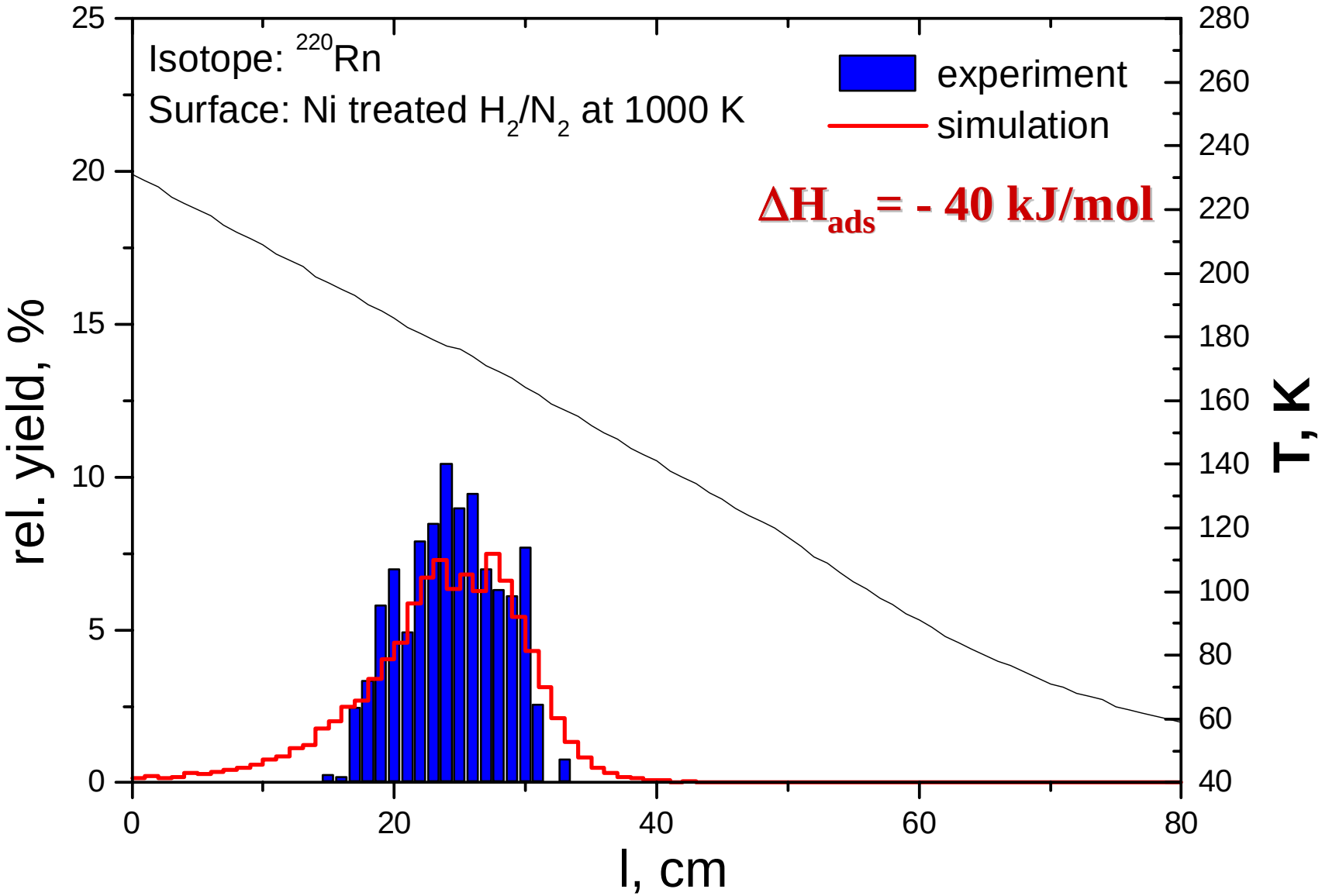


measurement of ^{212}Pb or ^{211}Bi: HPGe-γ-detector / collimator

Results


Isotope: ^{219}Rn
Surface: Ag treated H_2/N_2 at 1000 K
experiment
simulation
ΔH_{ads} = - 21 kJ/mol
rel. yield, %
T, K
l, cm
0
5
10
15
20
25
40
60
80
100
120
140
160
180
200
220
240
260
280

Results


Isotope: ^{220}Rn
Surface: Ni treated H_2/N_2 at 1000 K
experiment
simulation
ΔH_{ads}= - 40 kJ/mol
rel. yield, %
T, K
l, cm
0
5
10
15
20
25
0
20
40
60
80
40
60
80
100
120
140
160
180
200
220
240
260
280

Radon Adsorption Results

	$-\Delta H_{ads}$ kJ/mol
Ag	23±2
Au	29±2
Pd	35±3
Ni	38±3
Cu	39±3

Empirical model

A.R. Miedema, B.E. Nieuwenhuys Surf. Sci. 104, 491-509 (1981).

Description of ΔH_{ads} proportional to the enthalpy of adhesion:

$$\Delta\gamma^{ad}(A,B) = -2\,\Phi\,(\gamma^0(A)\,\gamma^0(B))^{1/2}$$

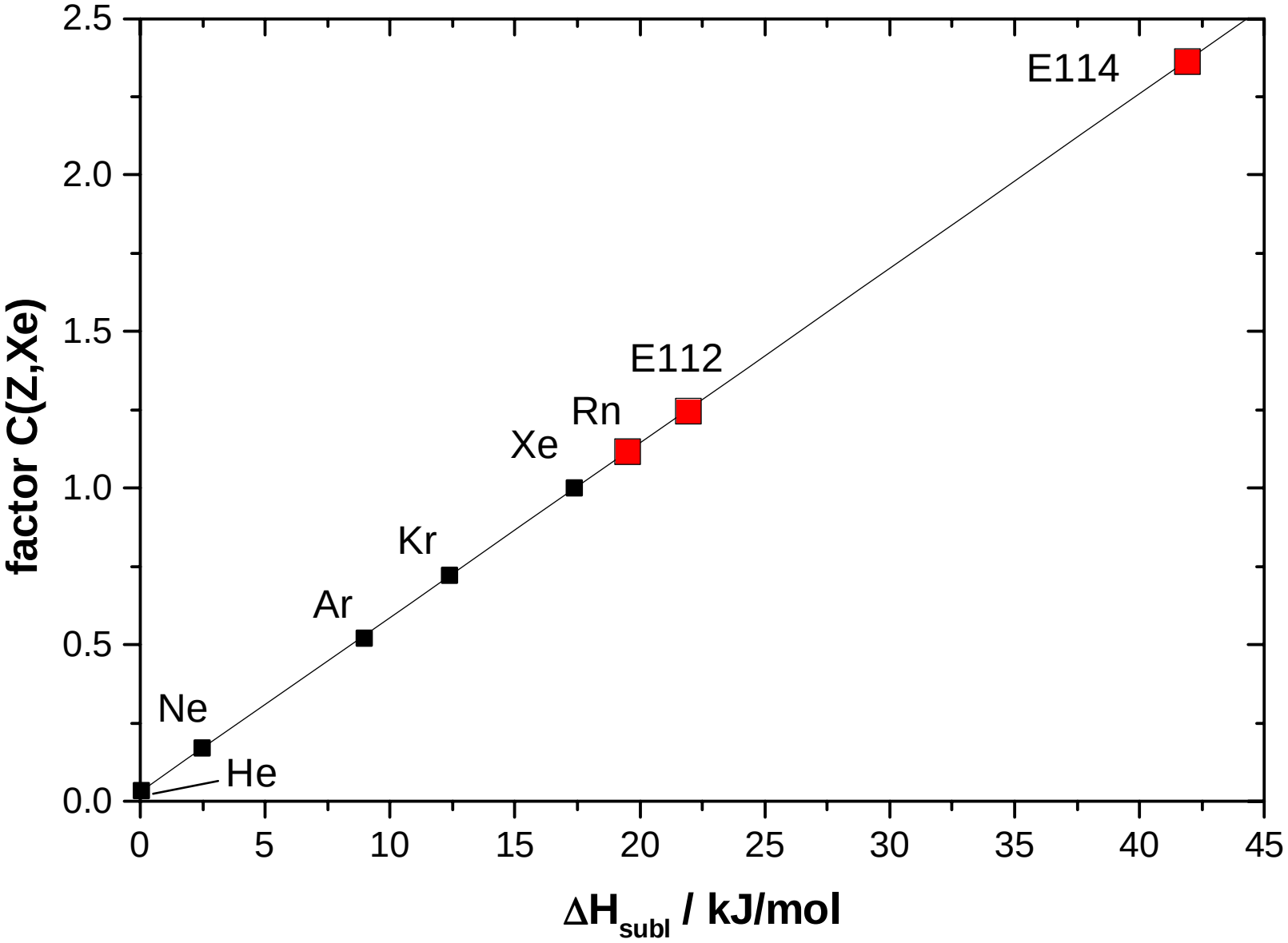
$$\Delta H_{ads} = 0.71*10^9\,F\,\Phi\,V_A^{2/3}\,(\gamma^0(A)\,\gamma^0(B))^{1/2}$$

Φ ... dissimilarity parameter (calculated)
γ^0... surface energy at T=0 K
F ... geometrical factor (empirically 0.31)
V_A ... Volume of the spherical adsorbate atom

$$\Rightarrow \Delta H^M_{ads}(Z) = C(Z,Xe)*\,\Delta H^M_{ads}\,(Xe)$$

Z	Ne	Ar	Kr	Xe
C(Z,Xe)	0.17	0.52	0.72	1

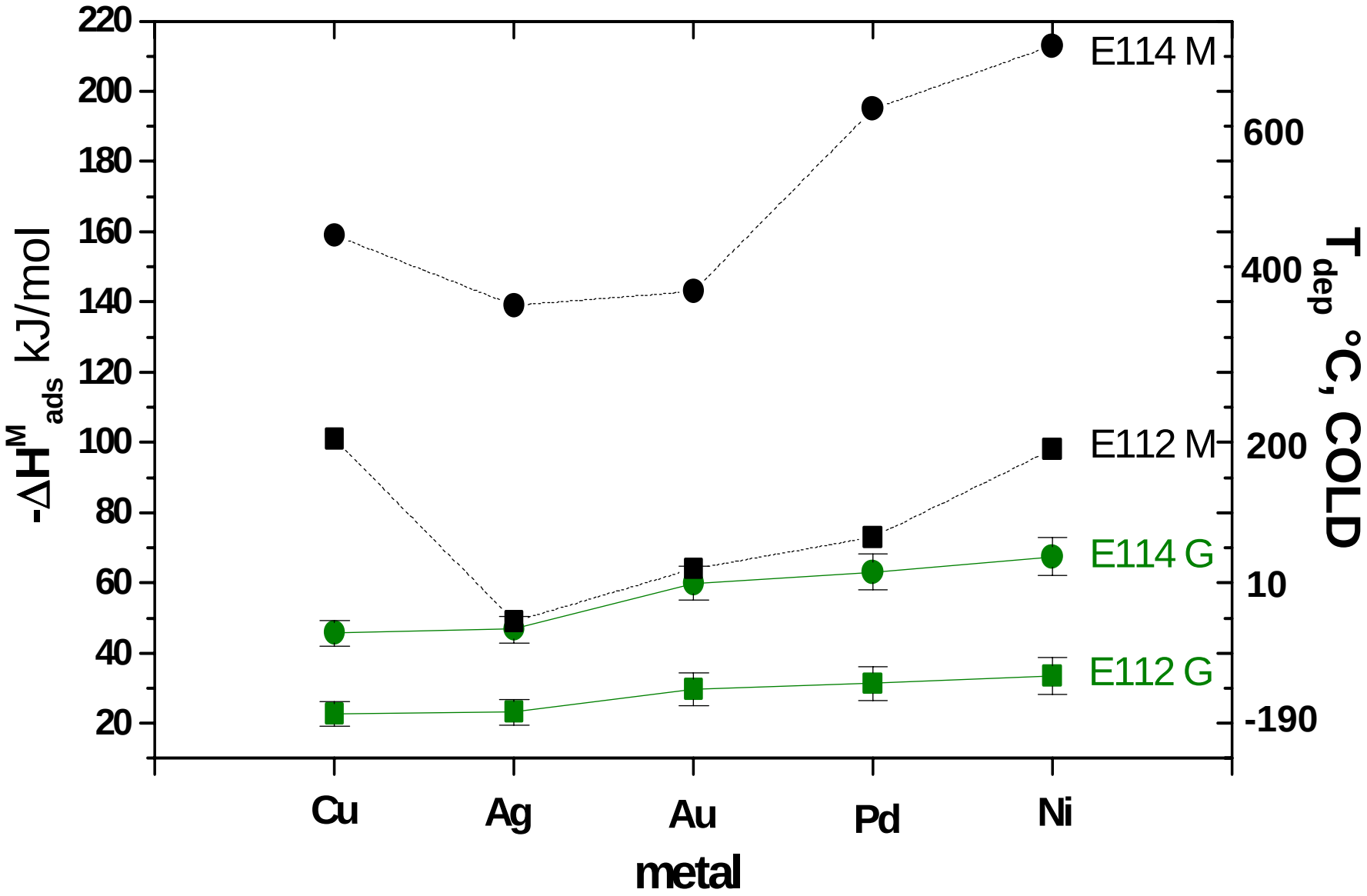
Empirical correlation of C(Z, Xe)



Radon Adsorption
Experimental Results⇔Model

	$-\Delta H_{ads}$	$-\Delta H^{M}_{ads}$
	kJ/mol	
Ag	23±2	26±2
Au	29±2	33±3
Pd	35±3	35±3
Ni	38±3	37±3
(Cu	*39±3*	*25±2)*

Prediction of the adsorption behavior of E112 and E114



On-line Vacuum thermochromatography

Advantages:

+ no carrier gas = less surface contamination
+ fast separation
+ good heat isolation at low temperatures
+ good α-resolution

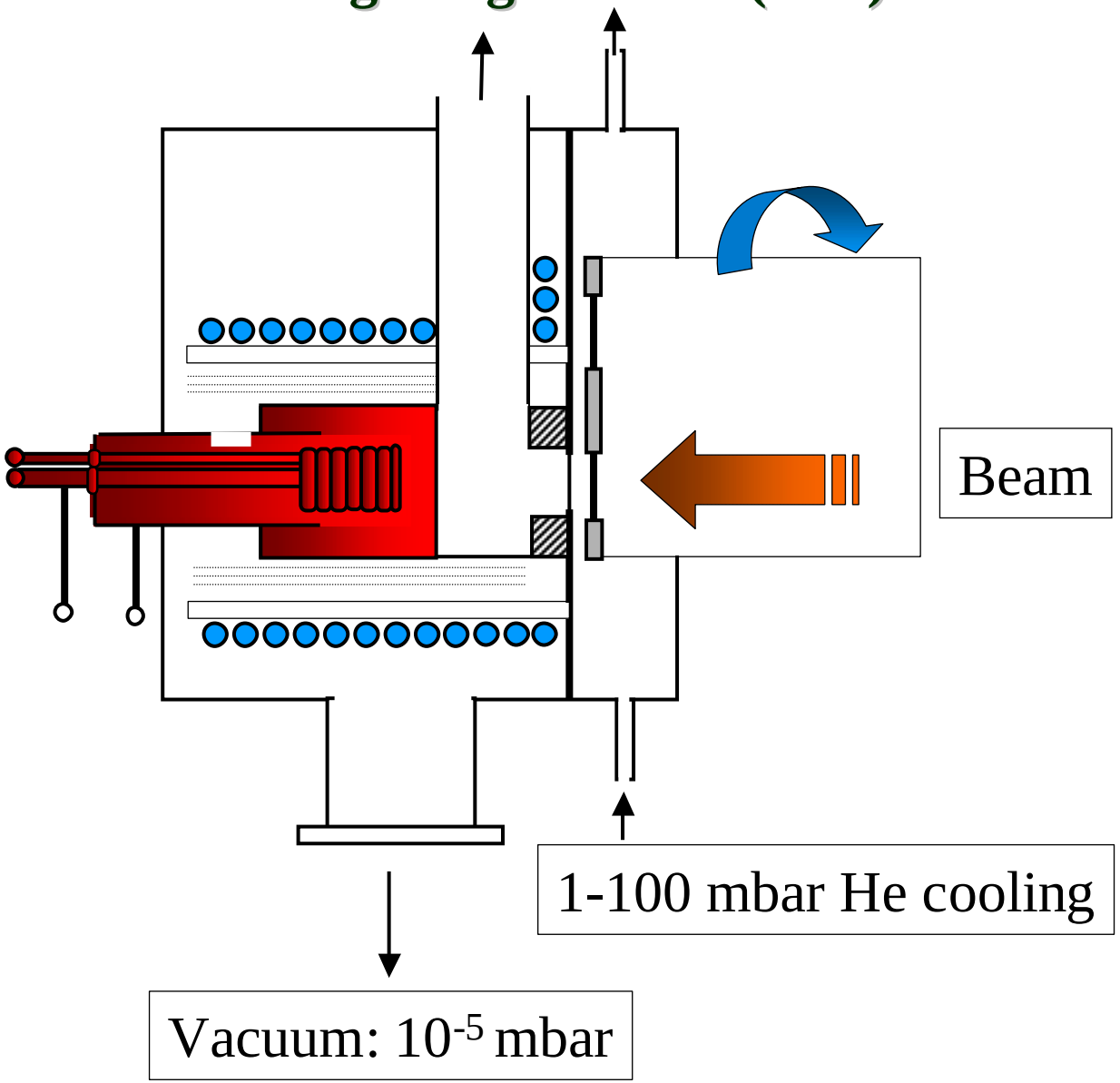
Problem:

- transfer of recoils

Solutions:

A hot catcher system
B coupling to physical separator
C adsorber system (batch wise)

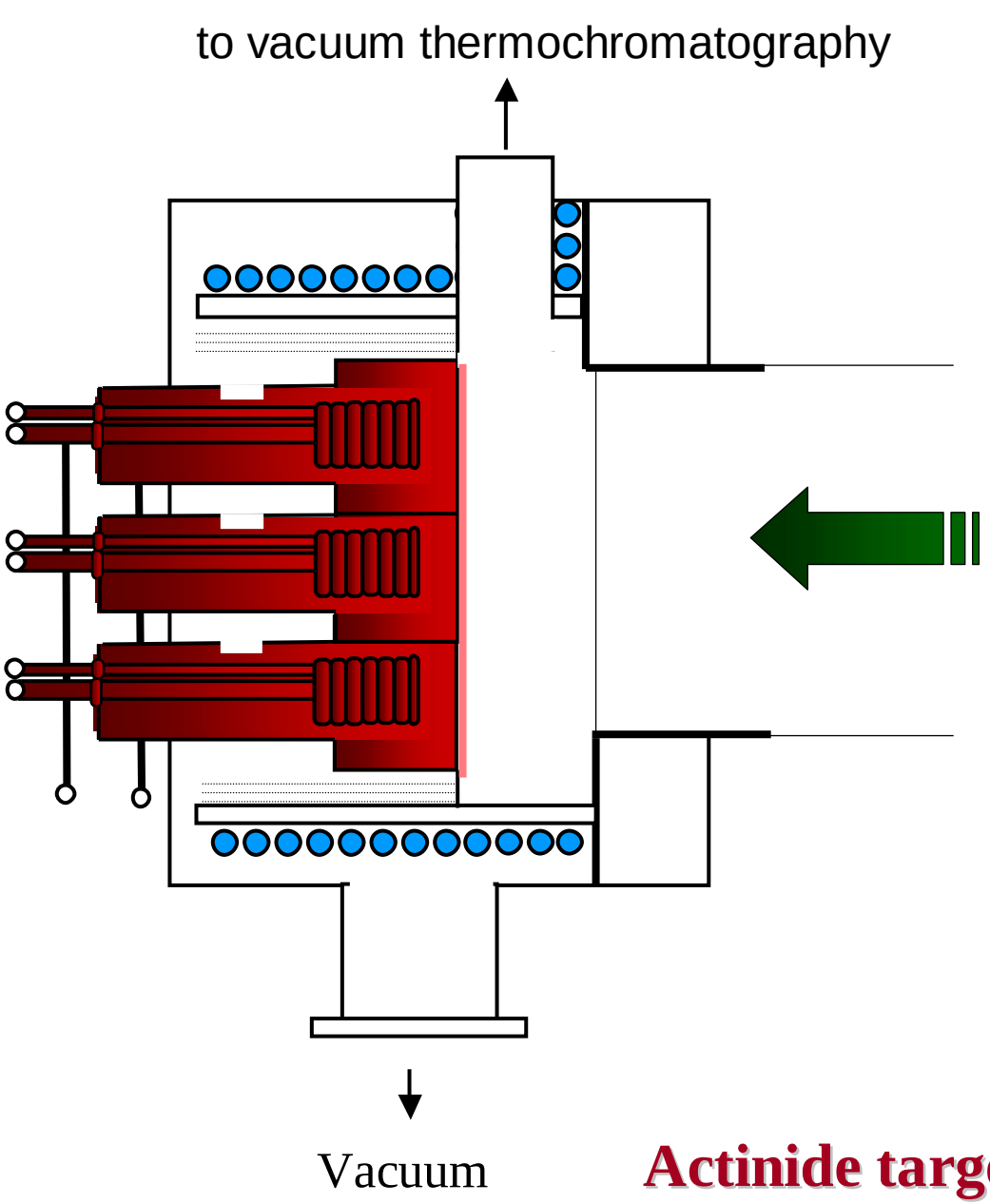
Vacuum Thermochromatography without Rn-Suppression: Rotating Target Wheel (GSI)

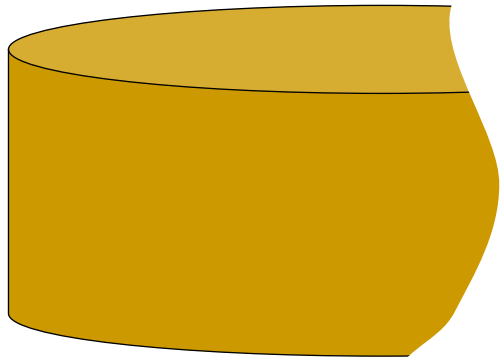


1h 400 pnA ^{40}Ca: vacuum $2 \cdot 10^{-5}$ mbar

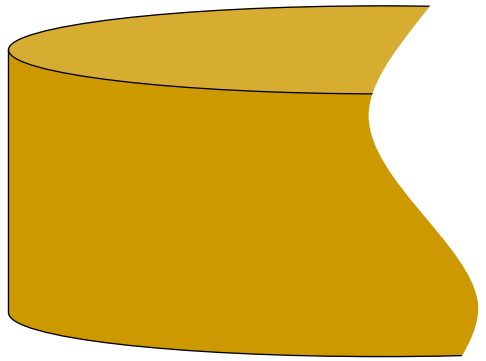
800 μg/cm^2 lanthanide targets: 2.2 mg/cm^2 (5μm) Ti backing

Vacuum Thermochromatography at a Recoil Separator



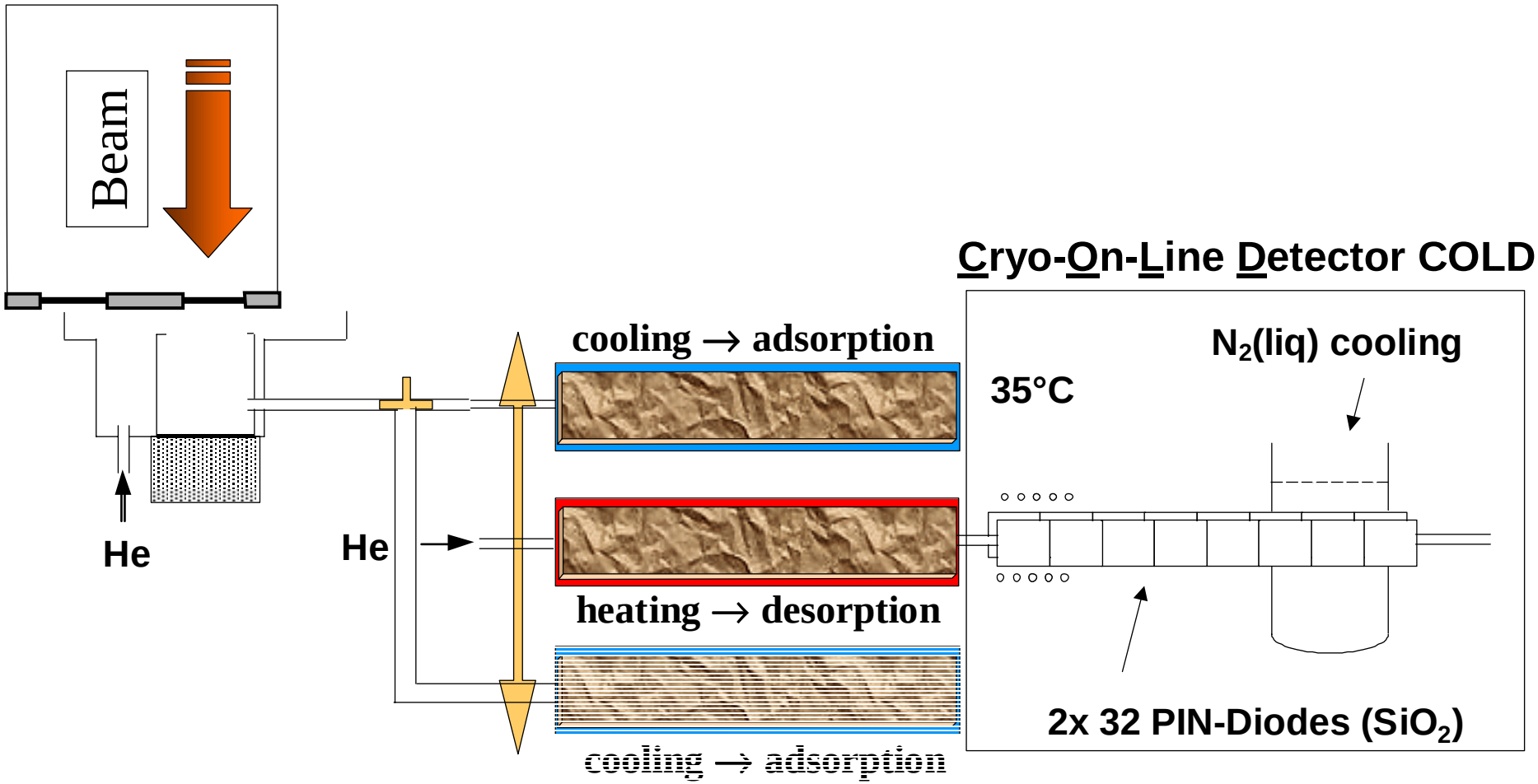


CHEMSEP



Actinide target technology development!!!

Adsorber System (114-Chemistry)



112/114-Chemistry