RIKEN GARIS as a Promising Interface for Superheavy Element Chemistry –Production of ^{261}Rf Using the GARIS/Gas-jet System–

RIKEN Nishina Center

H. Haba, D. Kaji, Y. Kasamatsu, Y. Kudou, K. Morimoto, K. Morita, K. Ozeki, T. Sumita, and A. Yoneda

Osaka Univ.

Y. Komori, K. Ooe, and A. Shinohara

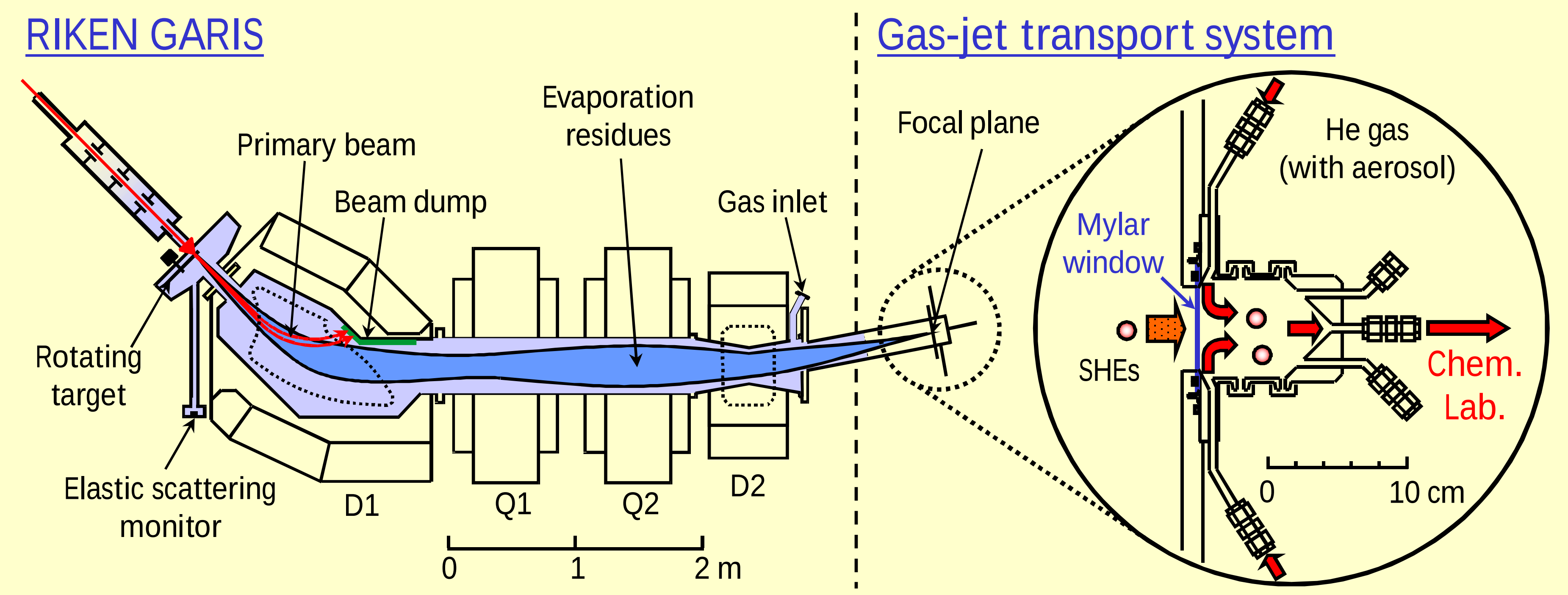
JAEA

N. Sato and K. Tsukada

1. Introduction

RIKEN Gas-filled Recoil Ion Separator GARIS as a pre-separator for superheavy element (SHE) chemistry

➡ Startup of SHE chemistry in RIKEN



Breakthroughs in SHE chemistry

Chemical experiments under low background condition

Stable and high gas-jet transport efficiency

New chemical systems

Commissioning with a prototype gas-jet transport system coupled to GARIS

Haba *et al.*, JNRS 8(2007)55.

$^{169}Tm(^{40}Ar,3n)^{206}Fr$ (Z=87)

$^{208}Pb(^{40}Ar,3n)^{245}Fm$ (Z=100)

Haba *et al.*, JNRS 9(2008)27.

$^{238}U(^{22}Ne,5n)^{255}No$ (Z=102)

→ *The GARIS/gas-jet system is a powerful tool for the next-generation SHE chemistry.*

Extremely low background condition

Beam-independent high gas-jet efficiency

For the SHE chemistry coupled to the recoil separator

- Berkeley Gas-filled Separator (BGS)@LBNL

Omtvedt *et al.*, JNRS 3(2002)121.

$^{208}Pb(^{50}Ti,n)^{257}Rf$ (4.7 s)

→ Identification of ^{257}Rf with a liquid scintillator after a liquid-liquid solvent extraction

- TransActinide Separator and Chemistry Apparatus (TASCA)@GSI

Even *et al.*, GSI Sci. Rep. 2008, p. 143 (2009).

$^{244}Pu(^{22}Ne,5n)^{261}Rf$ (68 s)

→ Anion-exchange chromatography of ^{261}Rf in diluted HF solution with ARCA

- GARIS@RIKEN

→ *^{248}Cm-based hot fusion reactions*

Z	Reaction	$T_{1/2}$ (s)	σ (pb)
104	^{248}Cm(^{18}O,5*n*)^{261}Rf$^{a/b}$	68/3	13000
105	^{248}Cm(^{19}F,5*n*)^{262}Db	34	1500
106	^{248}Cm(^{22}Ne,5*n*)^{265}Sg$^{a/b}$	9/16	240
107	^{248}Cm(^{23}Na,5/4*n*)$^{266/267}$Bh	1.7/17	50
108	^{248}Cm(^{26}Mg,5*n*)^{269}Hs	9	7

In this work

- Developments of a rotating ^{248}Cm target system and a gas-jet chamber for asymmetric hot fusion reactions
- Productions of ^{261}Rf$^{a/b}$ and its homologues ^{169}Hf and ^{85}Zr for chemical studies using the GARIS/gas-jet system

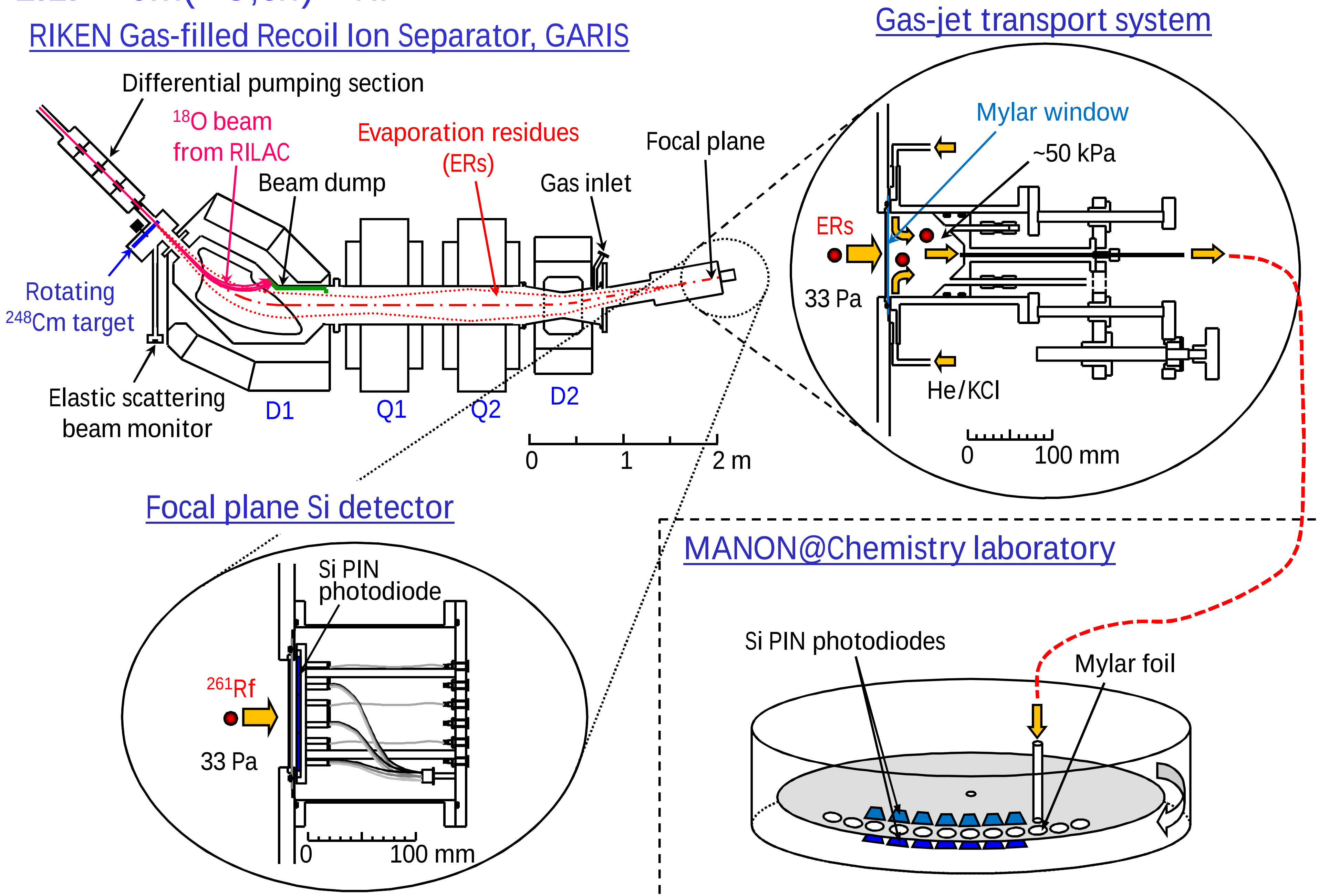
  ^{248}Cm(^{18}O,5*n*)^{261}Rf$^{a/b}$ (68 s/3 s)

  ^{nat}Gd(^{18}O,*xn*)^{169}Hf (3.24 min) and ^{nat}Ge(^{18}O,*xn*)^{85}Zr (7.86 min)
- Production of ^{265}Sg

  ^{248}Cm(^{22}Ne,5*n*)^{265}Sg$^{a/b}$ (9 s/16 s)

2. Experimental

2.1. $^{248}Cm(^{18}O,5n)^{261}Rf$



- $^{18}O^{5+}$ beam

  Energy: 95 MeV

  Intensity: 6 pμA
- ^{248}Cm target

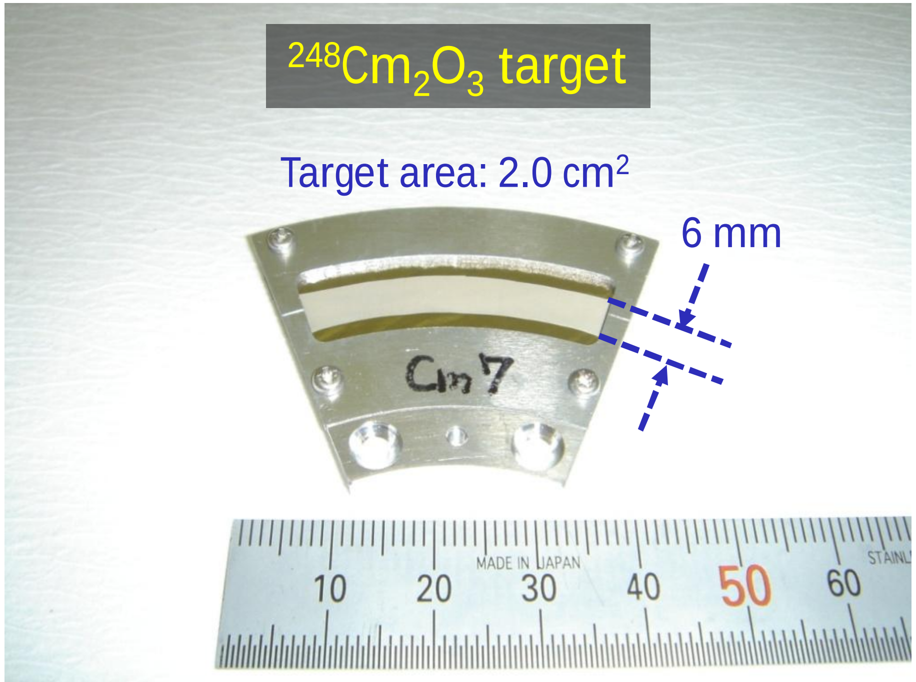
  280-μg/cm² $^{248}Cm_2O_3$ on 2.0-μm Ti×8

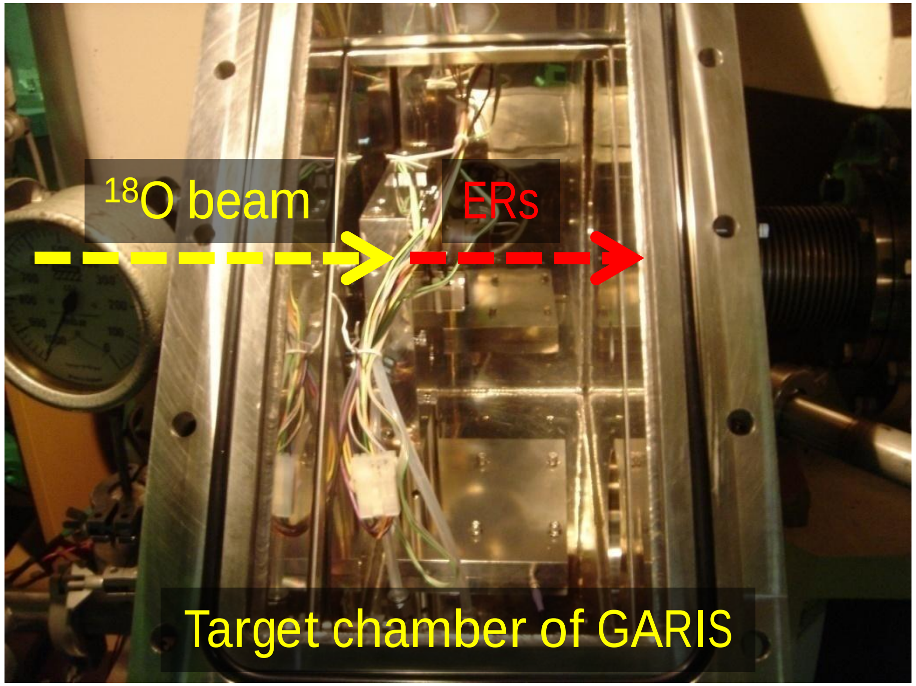
  1000 rpm

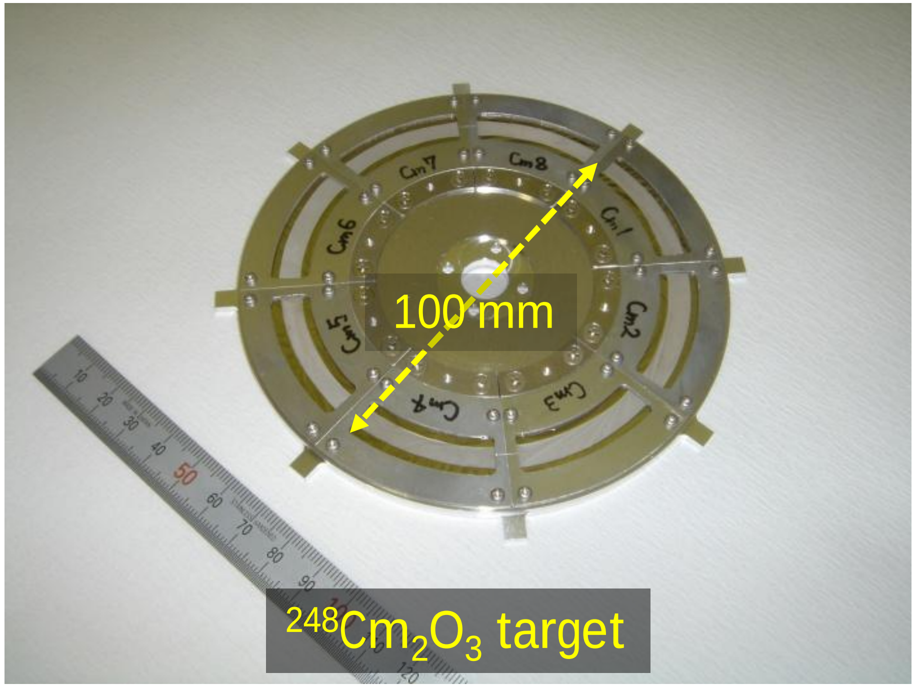
  He/water-cooled
- GARIS

  $B\rho$ = 1.58, 1.73, 1.86, and 2.01 Tm

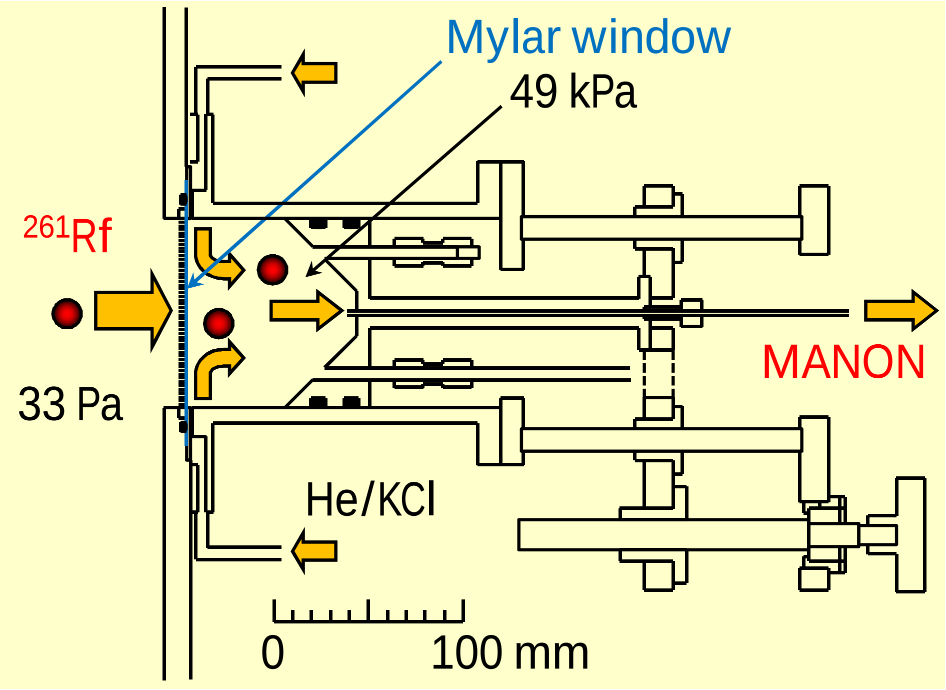
  He: 34 Pa

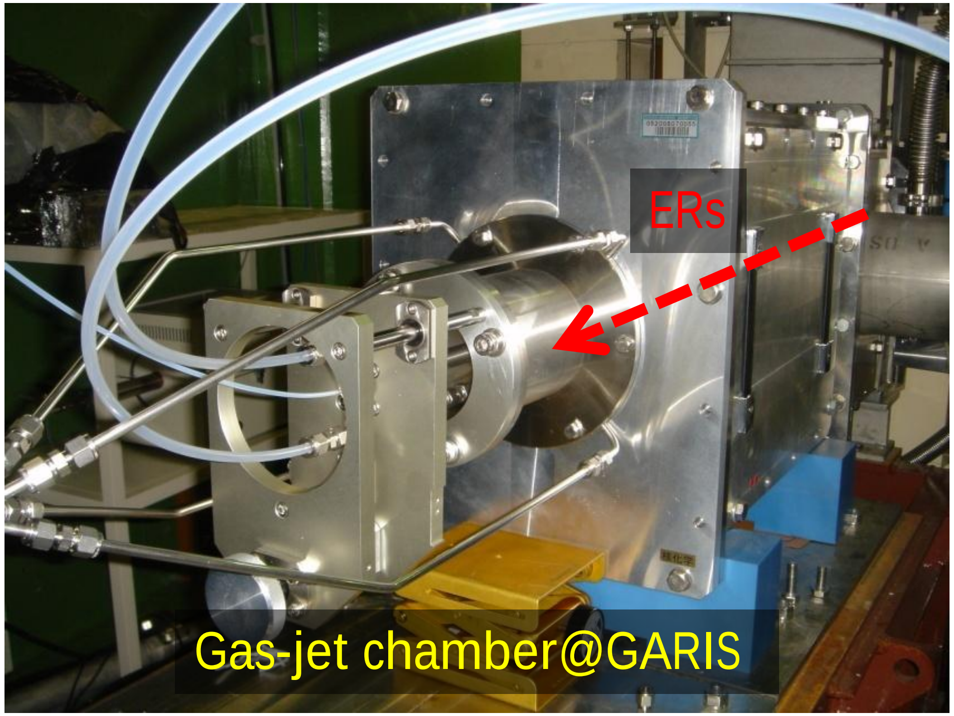


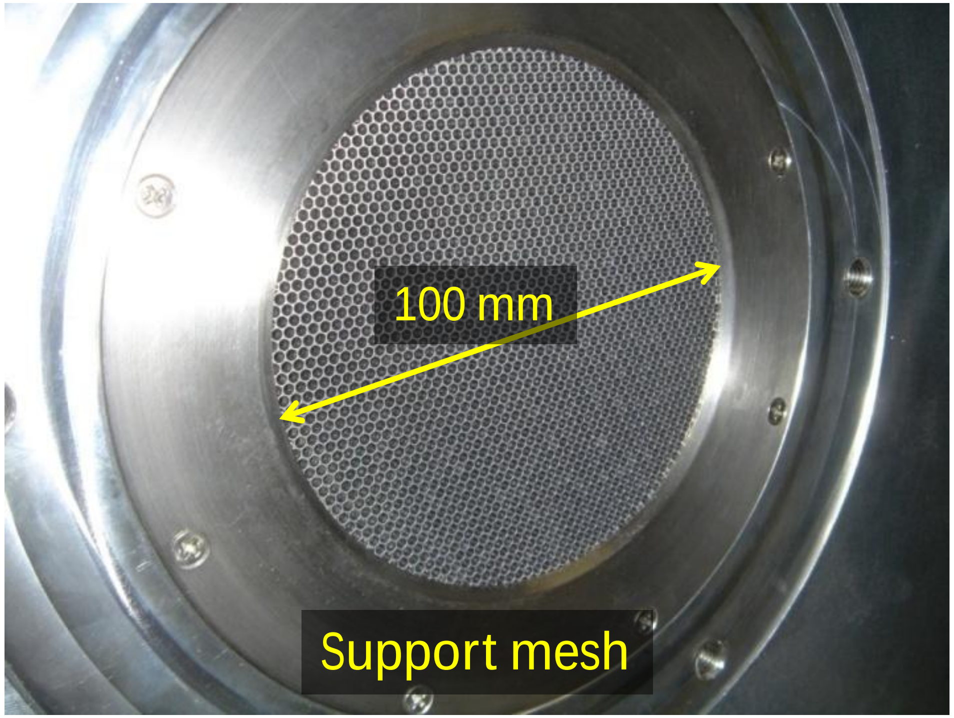




- Gas-jet chamber for hot fusion reactions
  - Size of the entrance window: Φ100 mm
  - Chamber depth: 20 mm
  - Window: 0.5-µm Mylar foil
  - Support mesh: 78% transparency
- He/KCl gas-jet
  - He: 2.0 L min^{-1}
  - Aerosol generator: 620 ^{o}C
  - Teflon capillary: 2 mm i.d. × 10 m







• MANON at the chemistry laboratory

Aerosol coll. on 0.5-μm Mylar foil

30 s for $^{261}Rf^{a}$ ($T_{1/2}$ = 68 s)

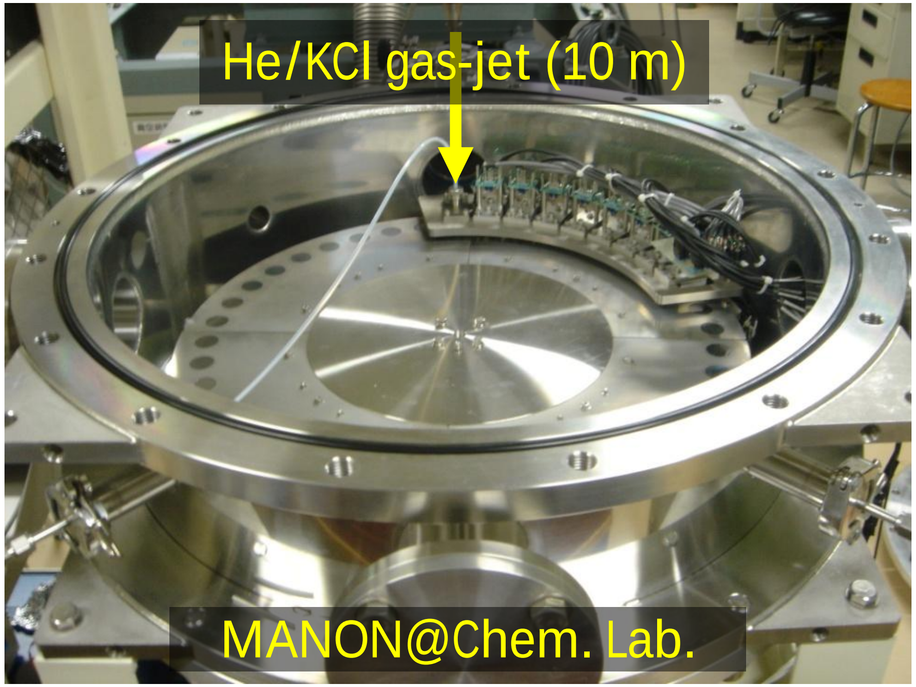
1.5 s for $^{261}Rf^{b}$ ($T_{1/2}$ = 3 s)

Hamamatsu Si PIN photodiode

S3204-09 (18×18 mm^2)×7 pairs

→ Counting eff.: 76%

DAQ: Iwatsu A3100 (LIST mode)



• Focal plane Si detector

Hamamatsu Si PIN photodiode

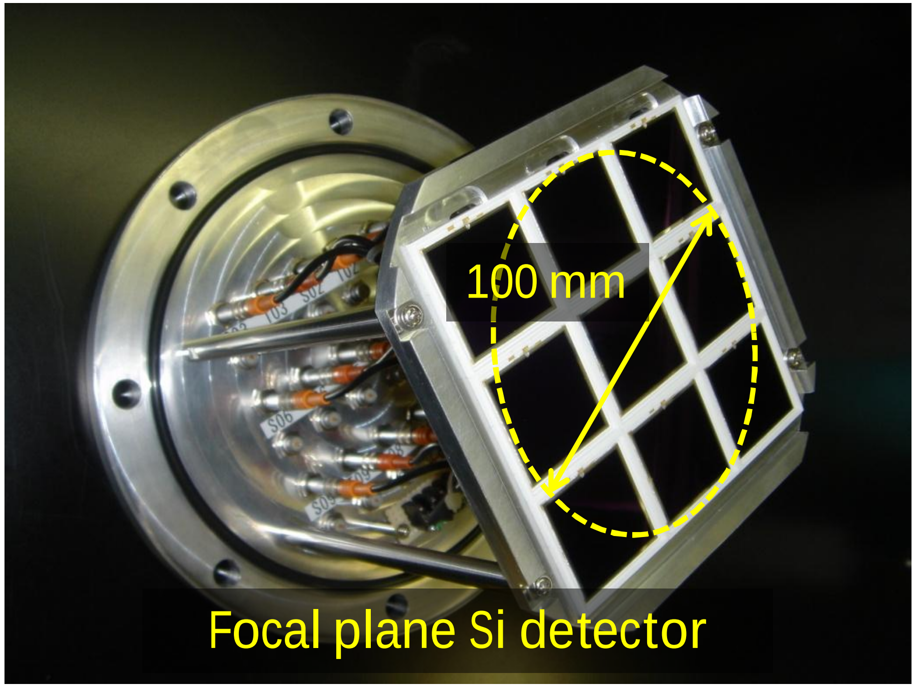
S3584 (28×28 mm^2)×9

→ Counting eff.: 50%

DAQ: Iwatsu A3100 (LIST mode)

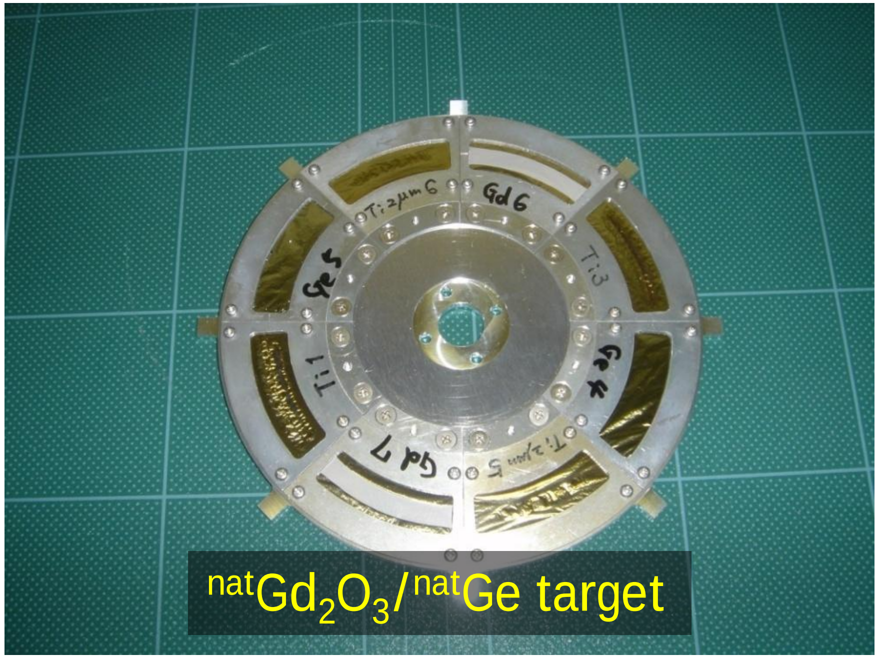
Cycle of beam ON (100 s) and OFF (100 s)

→ Evaluation of a gas-jet yield



2.2. $^{nat}Gd(^{18}O,xn)^{169}Hf$ and $^{nat}Ge(^{18}O,xn)^{85}Zr$

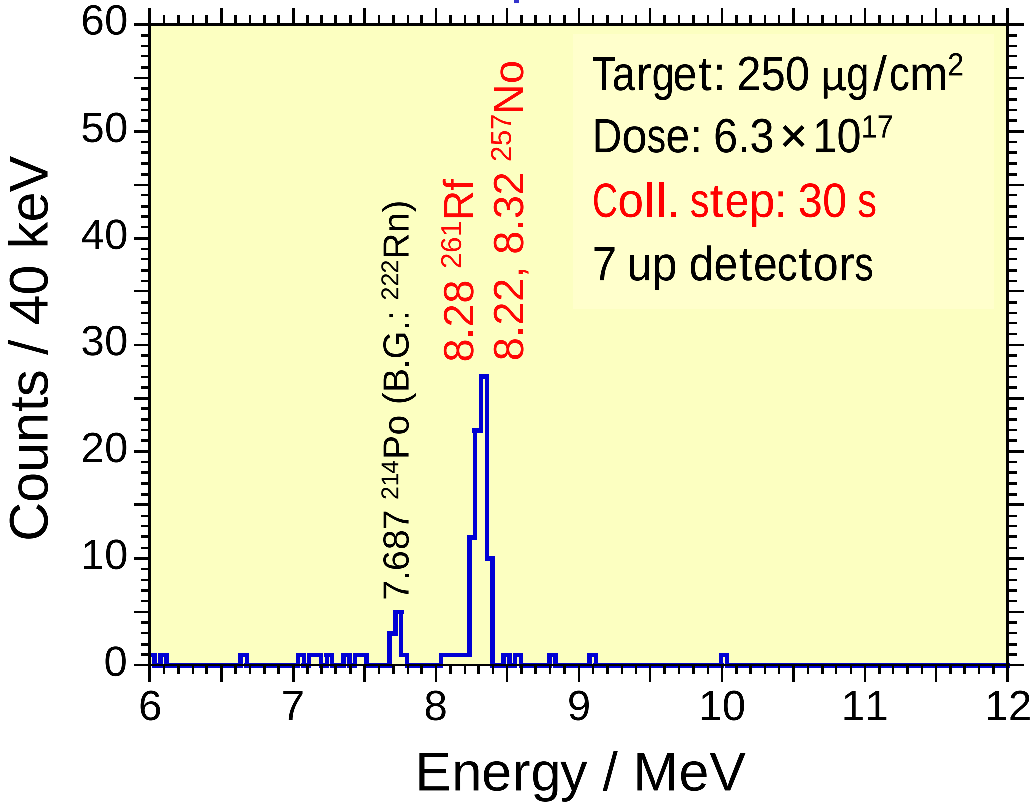
- ^{18}O beam ($^{169}Hf/^{85}Zr$)
  Energy: 95 MeV
  Intensity: 6/0.5 pμA
- $^{nat}Gd_2O_3/^{nat}Ge$ target
  300-μg/cm^2 $^{nat}Gd_2O_3$ on 2.0-μm Ti × 2
  290-μg/cm^2 ^{nat}Ge on 2.0-μm Ti × 2
- GARIS ($^{169}Hf/^{85}Zr$)
  $B\rho$ = 1.48–1.63/0.88–1.04 Tm
  He: 34 Pa
- He/KCl gas-jet
  See the $^{248}Cm(^{18}O,5n)^{261}Rf$ exp.
- **γ**-ray spectrometry with Ge detector
  Glass filter ADVANTEC GB-100R
    ^{169}Hf: 60-s coll., 60-s cool., 60-s meas.
    ^{85}Zr: 300-s coll., 60-s cool., 300-s meas.
  Direct catch of ERs with 20-μm Al
    → Evaluation of a gas-jet yield



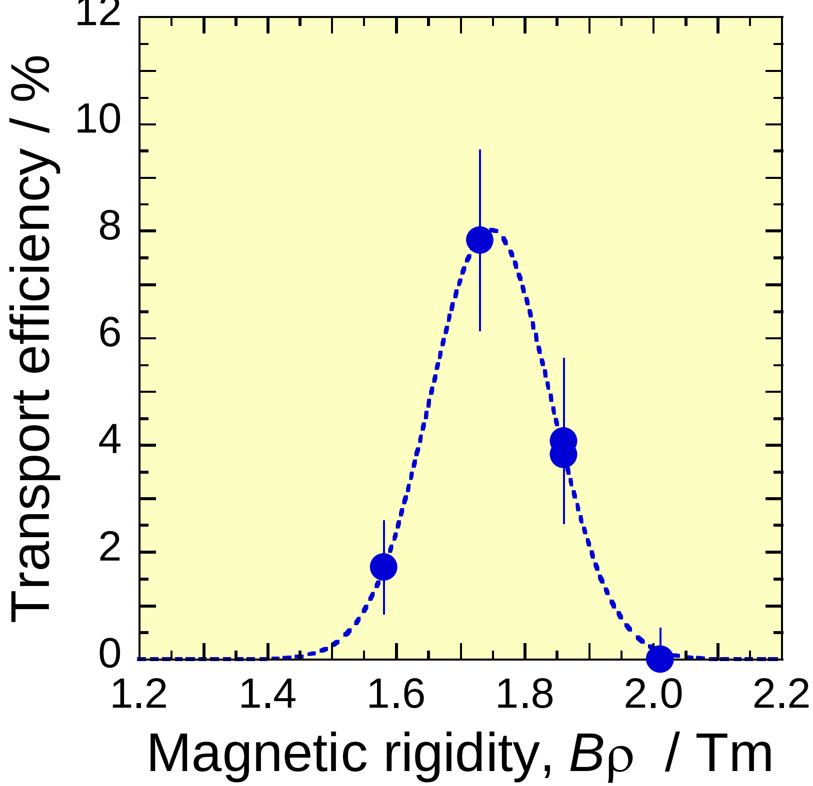
3. Results and Discussion

3.1. $^{248}Cm(^{18}O,5n)^{261}Rf^{a}$

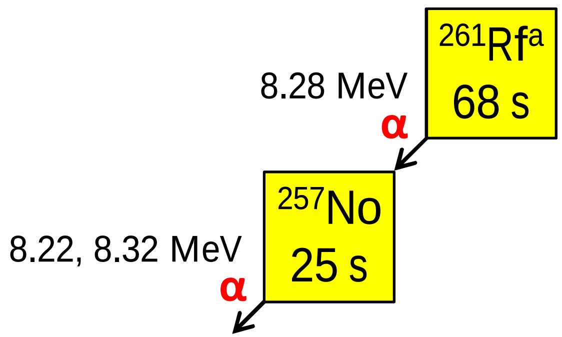
α spectrum



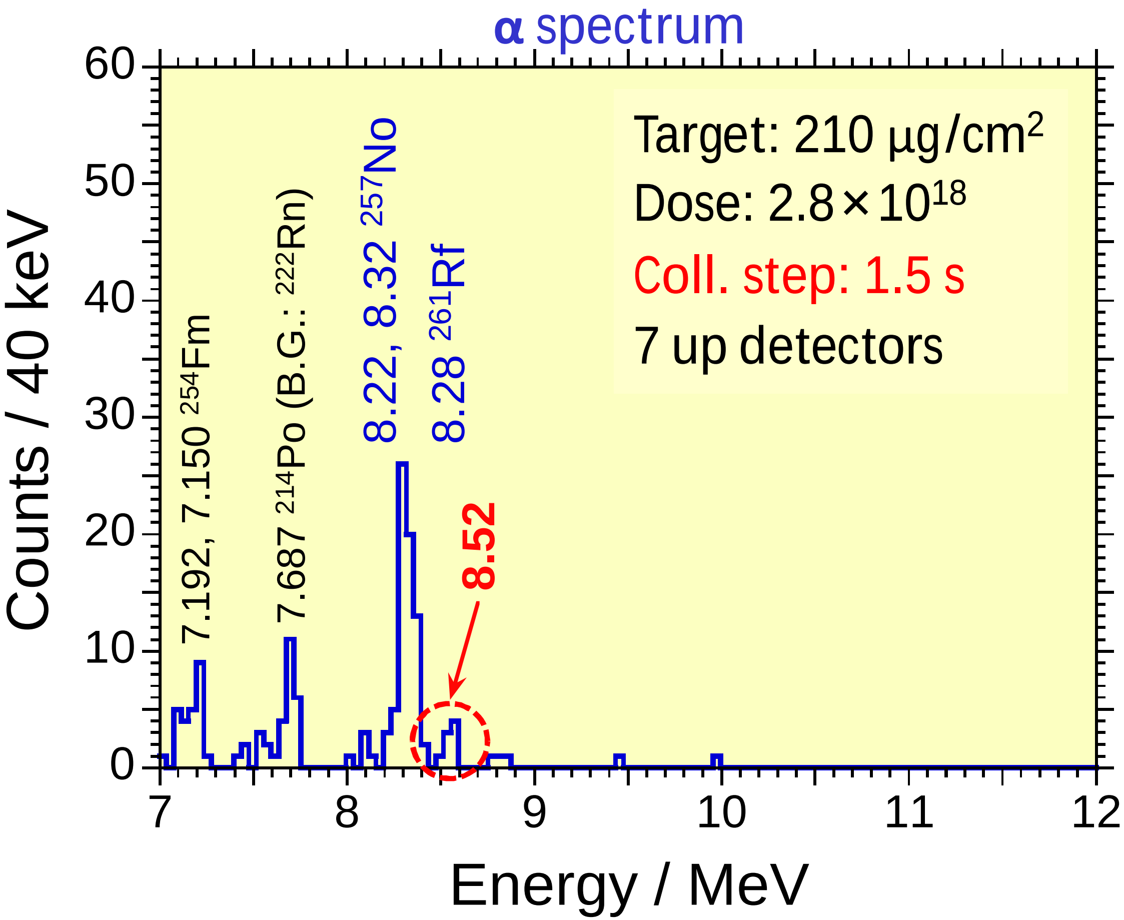
GARIS efficiency vs. *B*ρ

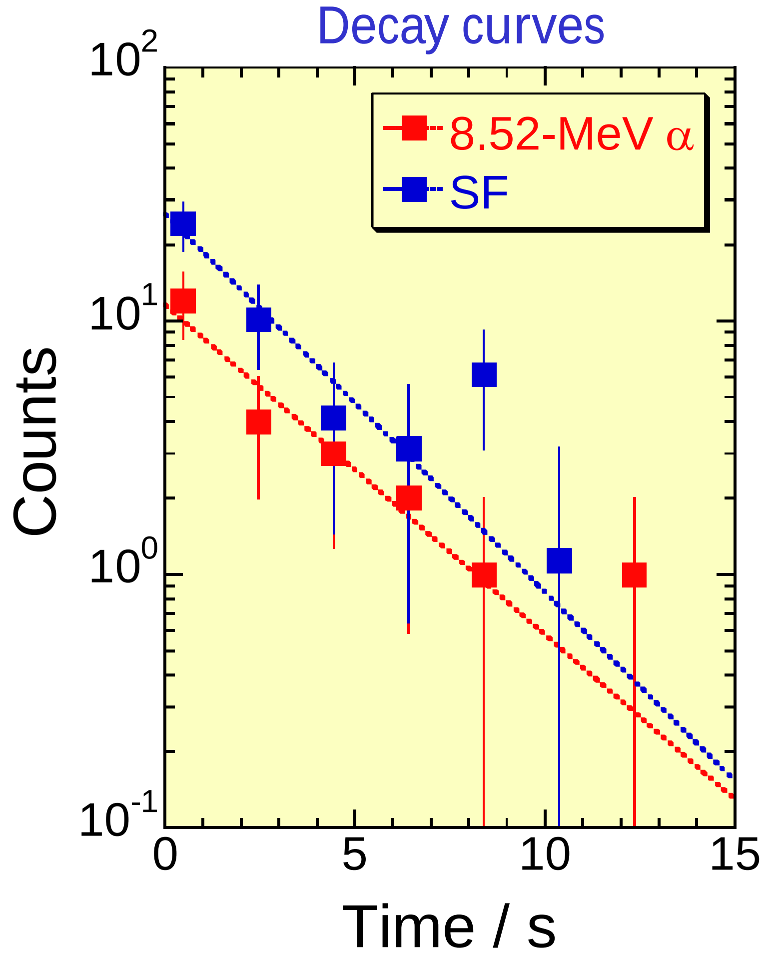


- 161 **α** (58 **α-α**) / 8.2-h meas.
- GARIS eff.: 7.8±1.7% for **Φ**100 mm (σ = 13 nb@JAEA)
- ***B*ρ** = 1.75±0.02 Tm, Δ*B*ρ/*B*ρ = 12.7±1.9%
- Gas-jet eff.: 52±12%



3.2. ^{248}Cm(^{18}O,5*n*)^{261}Rf^{b}





Numbers of 8.52-MeV α and SF

Step [s]	No. of events 8.52-α	No. of events SF	Dose [10^{18}]
30	0	5	0.57
1.5	20	66	2.8

$T_{1/2}$ of 8.52-MeV α and SF

	$T_{1/2}$ [s]
8.52-MeV α	2.3 ± 0.8
SF	2.0 ± 0.6
Average	2.1 ± 0.5

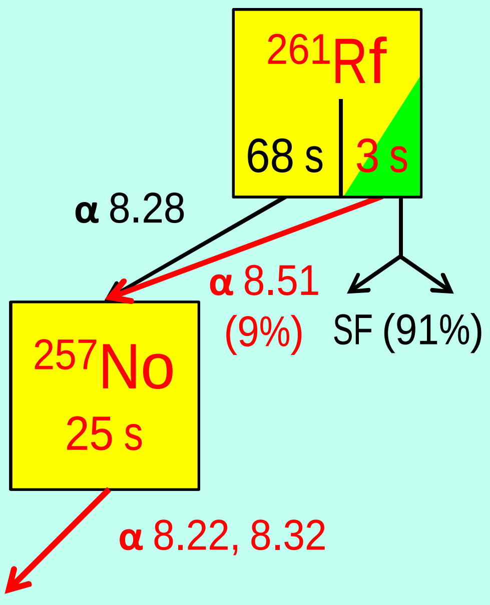
Correlated events on 8.52-MeV α

No.	1st α (8.52-MeV)		2nd α	
	E_α (keV)	ΔT (s)	E_α (keV)	ΔT (s)
1	8467	4.6	8289	0.6
2	8538	1.2	8306	34.6
3	8455	2.5	8233	34.0
4	8541	6.4	8223	8.3
5	8439	0.4	8178	32.0
6	8511	4.0	8225	77.3

$E_{\alpha 2}$ = 7900–8600 MeV; E_{SF} > 20 MeV
ΔT_2 = 250 s

2nd α

E_α = 8.18–8.31 MeV
$T_{1/2} = 22^{+14}_{-6}$ s



Düllmann and Türler:
PRC **77**, 064320 (2008).

Confirmation of $^{261}Rf^b$ by $^{248}Cm(^{18}O,5n)^{261}Rf^b$

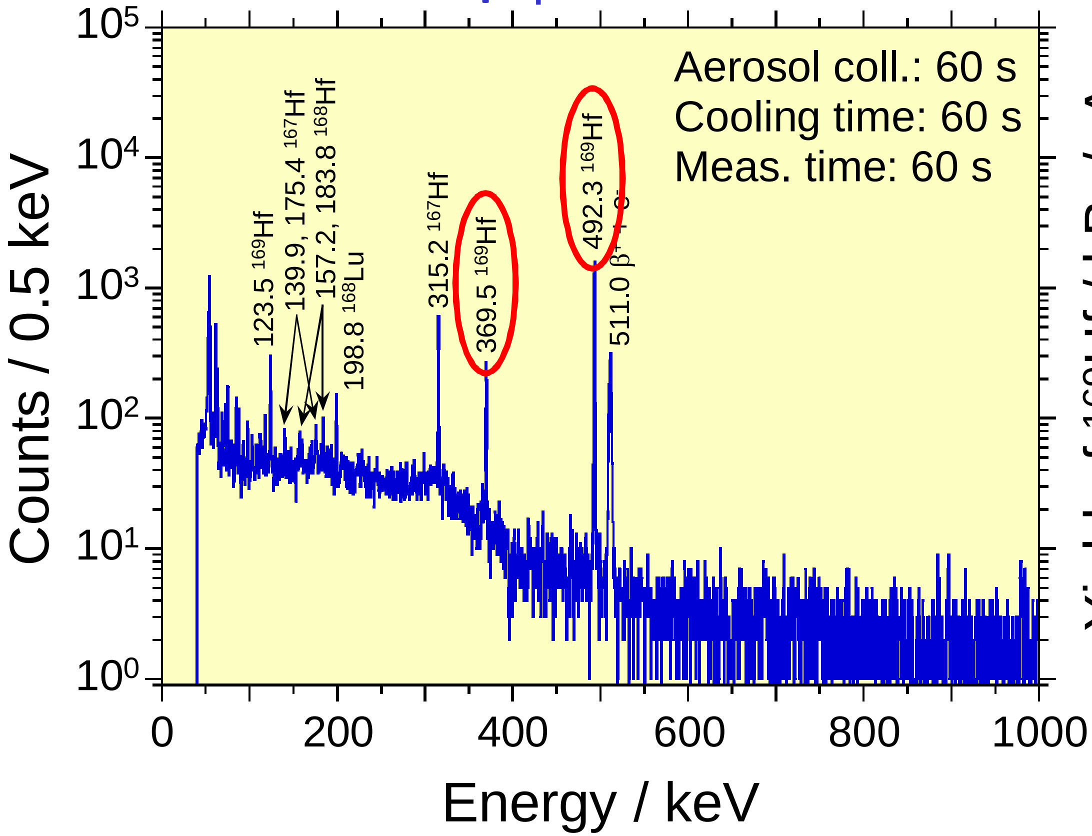
- $E_\alpha = 8.52 \pm 0.03$ MeV
- $T_{1/2} = 2.1 \pm 0.5$ s
- Branching ratio (SF/α) = 72 ± 7%/28 ± 7%
- $\sigma = 6.8 \pm 1.5$ nb

assumptions: $\sigma(^{261}Rf^a)$ = 12 nb, transport time of 2.2 s

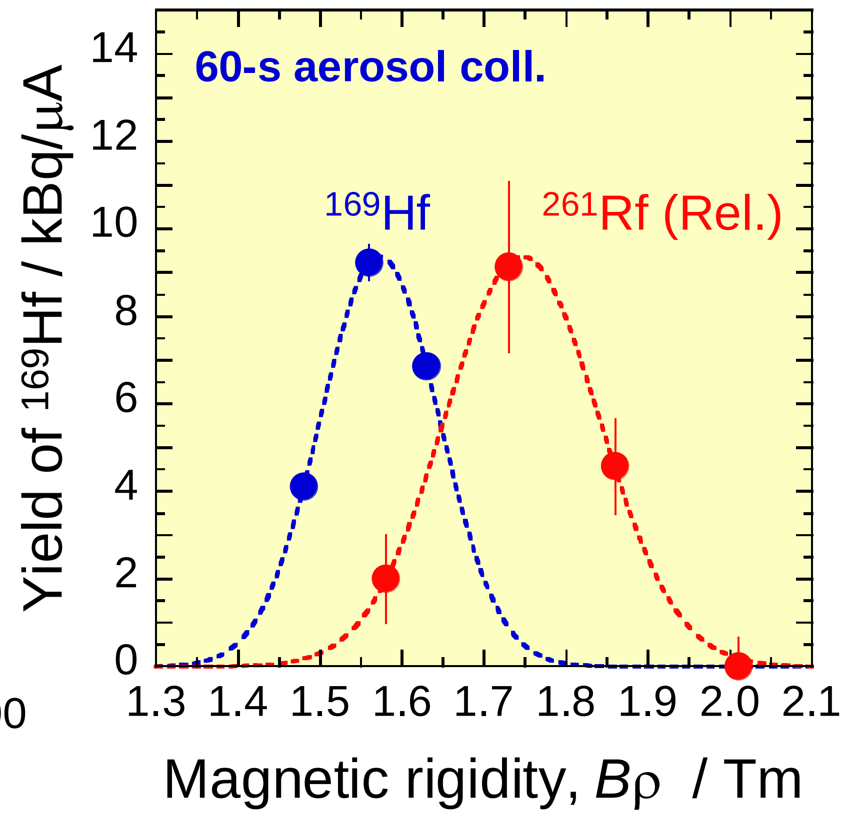
→ σ ratio($^{261}Rf^a$/$^{261}Rf^b$) = 1.8 ± 0.4

3.3. ^{nat}Gd(^{18}O,*xn*)^{169}Hf

γ spectrum



Yield of ^{169}Hf vs. *B*ρ

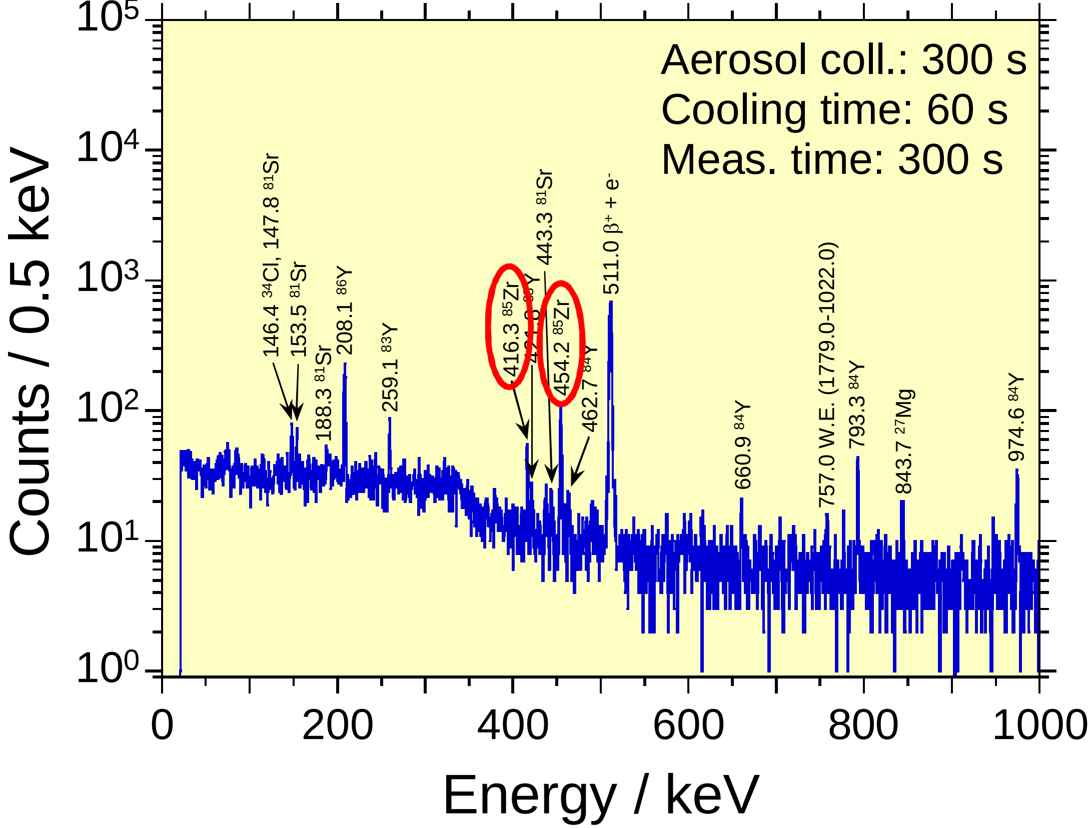


- ^{167}Hf ($T_{1/2}$ = 2.05 min), ^{168}Hf (9.458 min), ^{169}Hf (3.24 min)
  ^{168}Lu (6.7 min)
- *B*ρ = 1.57 ± 0.01 Tm, Δ*B*ρ/*B*ρ = 11.0 ± 0.5%
- Gas-jet eff.: 85 ± 2%

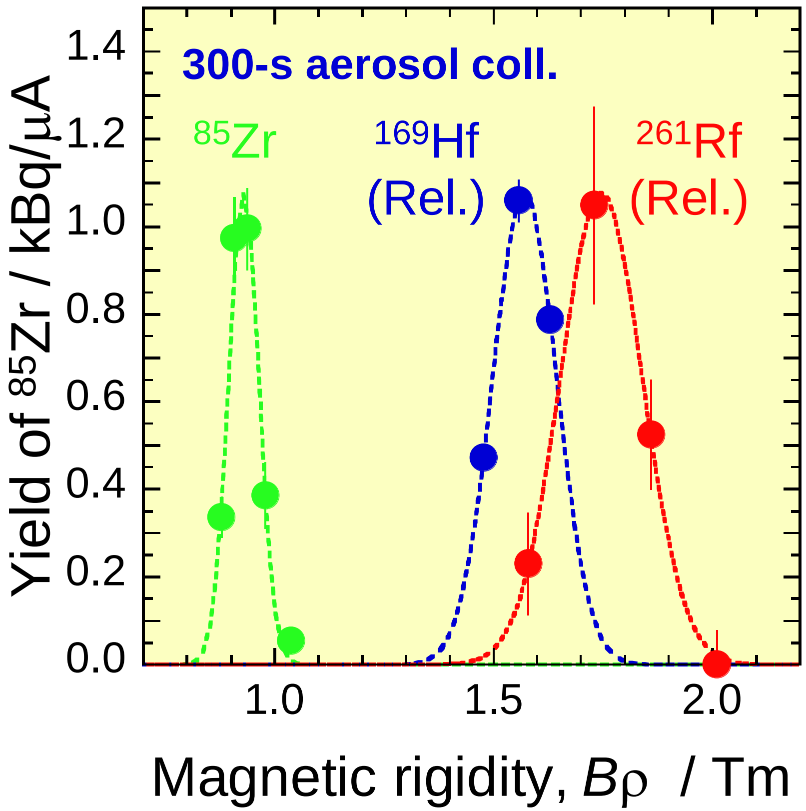
  Ranges of ^{169}Hf: 21–34 mm ≈ 100-mm chamber depth

3.4. $^{nat}Ge(^{18}O,xn)^{85}Zr$

γ spectrum



Yield of ^{85}Zr vs. $B\rho$



- ^{85}Zr (7.86 min)
  ^{27}Mg (9.458 min), ^{28}Al (2.2414 min), ^{29}Al (6.56 min), ^{34}Cl (32.00 min), ^{81}Sr (22.3 min), ^{83}Y (2.85 min), ^{84}Y (40 min), ^{86}Y (48 min)
- $B\rho = 0.930 \pm 0.002$ Tm, $\Delta B\rho / B\rho = 8.75 \pm 0.57\%$
- Gas-jet eff.: $36 \pm 4\%$

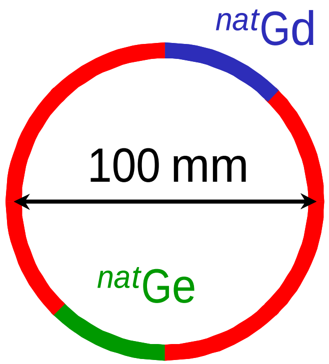
  Ranges of ^{85}Zr: 103–114 mm ≈ 100-mm chamber depth

3.5. Simultaneous chemical experiments with $^{261}Rf^{a}$, ^{169}Hf, and ^{85}Zr

Mixed $^{248}Cm/^{nat}Gd/^{nat}Ge/$ targets

→ Ready for chemistry experiments of Rf together with its homologues

Change of the magnet setting of GARIS: <1 min



Yields of $^{261}Rf^{a}$, ^{169}Hf, and ^{85}Zr at the chemistry laboratory

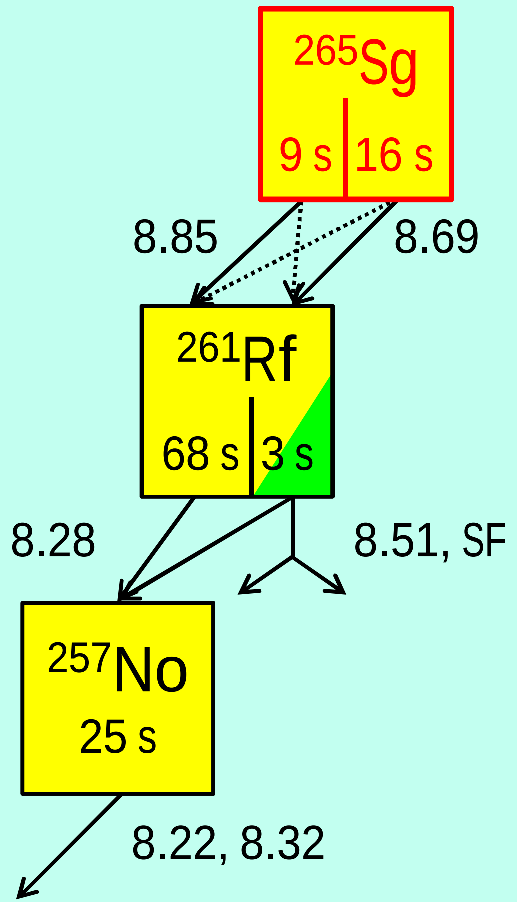
	$^{248}Cm(^{18}O,5n)^{261}Rf^{a}$	$^{nat}Gd(^{18}O,xn)^{169}Hf$	$^{nat}Ge(^{18}O,xn)^{85}Zr$
Beam energy (MeV)	95	←	←
Beam intensity (pμA)	6	1	1
Target thickness (μg/cm^2)	250	300	300
Magnetic rigidity (Tm)	1.75	1.57	0.93
GARIS He (Pa)	32	←	←
Mylar window (μm)	0.5	←	←
Support mesh (%)	78	←	←
Gas-jet He (kPa)	49	←	←
He flow rate (L/min)	2	←	←
KCl generator (°C)	620	←	←
Gas-jet eff. (%)	52 ± 12	85 ± 2	36 ± 4
Yield at Chem Lab.	0.5 atoms/min	9.4 kBq/60-s coll.	1.1 kBq/300-s coll.

4. Production of $^{265}Sg^{a/b}$ using the GARIS/gas-jet system

4.1. Experimental conditions

Reaction	$^{248}Cm(^{22}Ne,5n)^{265}Sg$
Cross section	200-300 pb*
Beam energy (MeV)	118
Beam intensity (pμA)	3
$^{248}Cm_2O_3$ thickness (μg/cm^2)	250
Recoil energy (MeV)	9.4
Magnetic rigidity (Tm)	1.73, 1.94, 2.05, 2.16
GARIS He (Pa)	32
Mylar window (μm)	0.65
Support grid (%)	78
Gas-jet He (kPa)	48
He flow rate (L/min)	2
KCl generator (^{o}C)	600

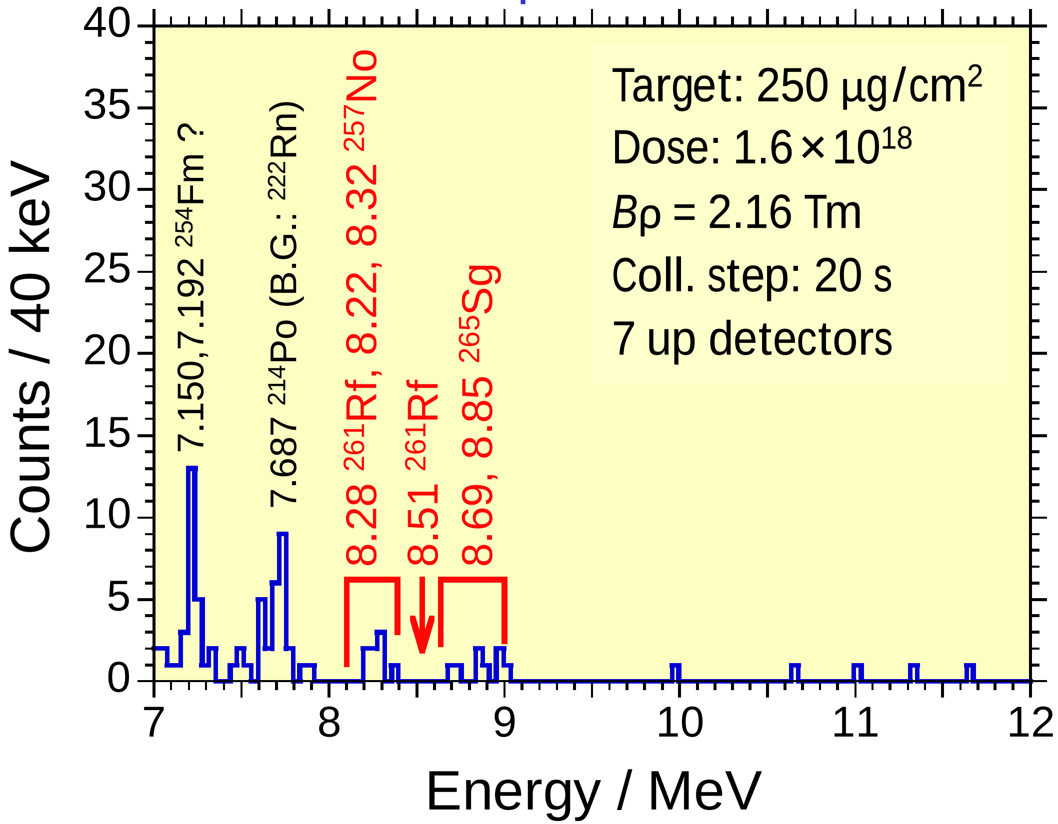
*Düllmann and Tüler: Phys. Rev. C 77, 064320 (2008).



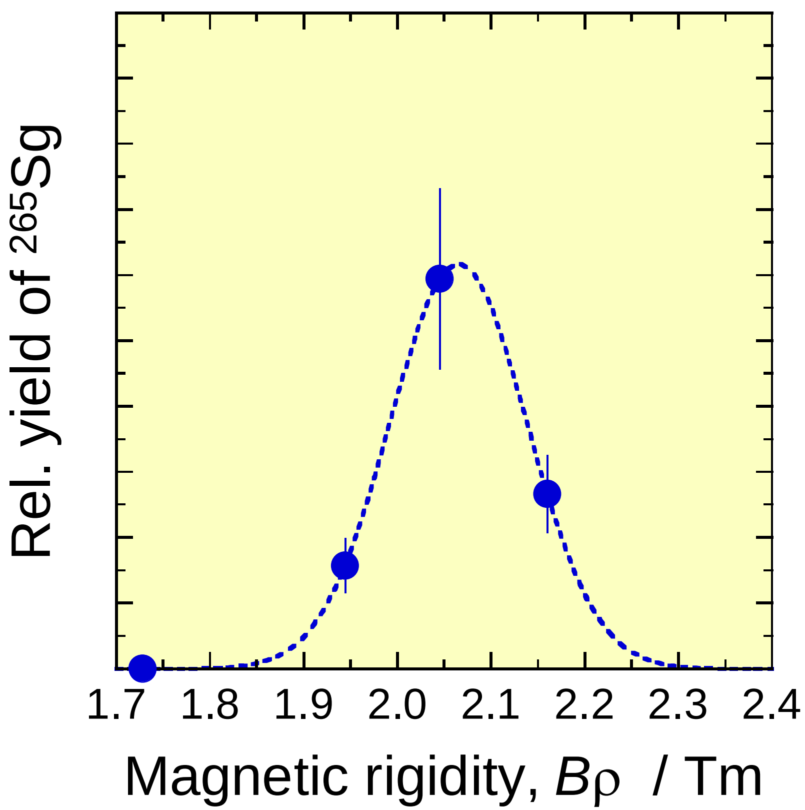
Düllmann and Türler:
PRC 77, 064320 (2008).

4.2. Results of ^{248}Cm(^{22}Ne,5*n*)^{265}Sg

α spectrum

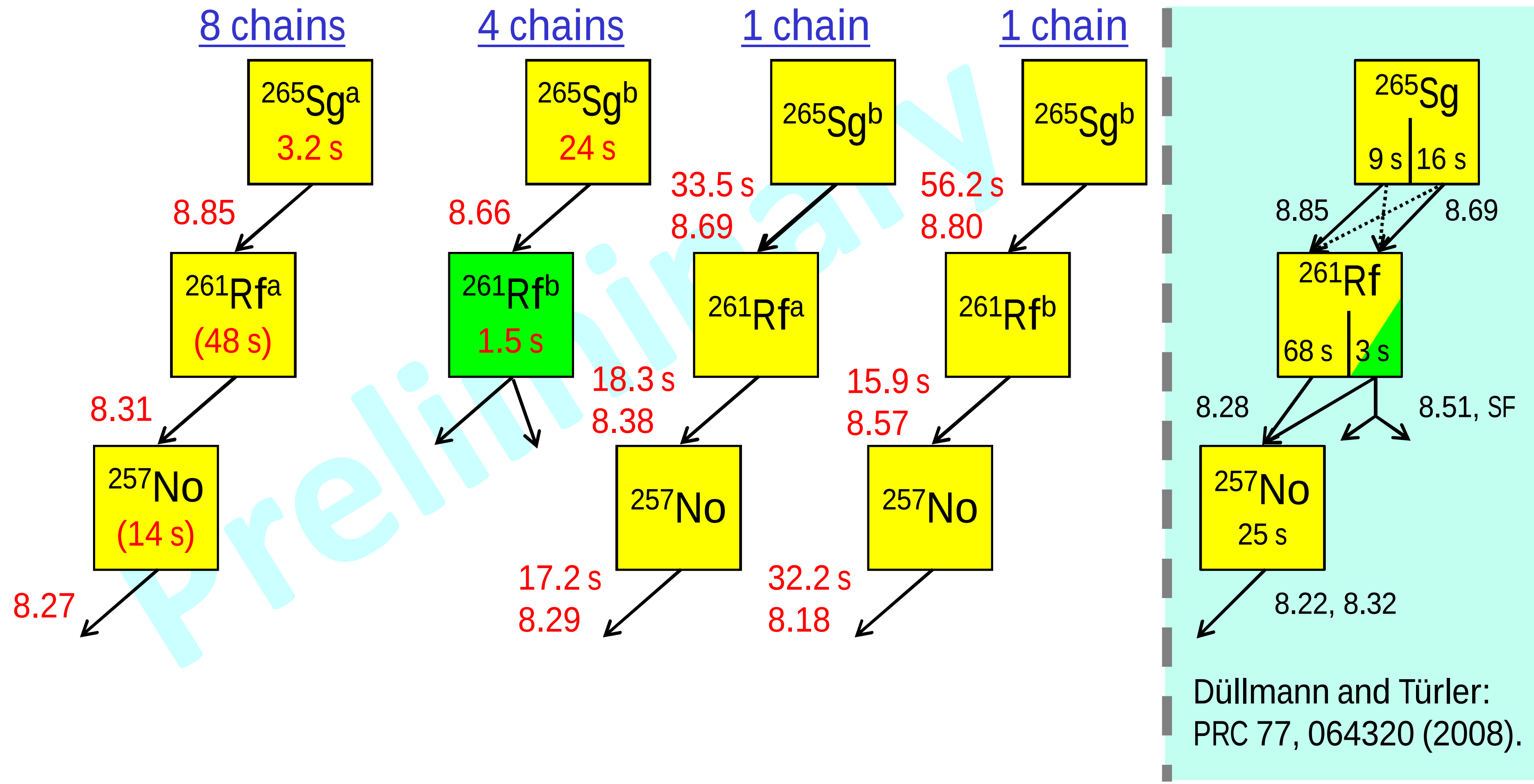


Rel. yield of ^{265}Sg



- No background peaks above 8 MeV
- 14 correlations on ^{265}Sg$^{a/b}$
- $B\boldsymbol{\rho}$ = 2.07 ± 0.01 Tm, $\Delta B\rho/B\rho$ = 8.4 ± 1.1%

Observed decay chains on ^{265}Sg



- $\sigma(^{265}Sg^{a}/^{265}Sg^{b})$ = ~400 pb/~200 pb at 118 MeV
- Yield at the chemistry laboratory: ~4 atoms/h

5. Summary

- The target and gas-jet transport systems for asymmetric ^{248}Cm-based hot fusion reactions have been installed on GARIS to start SHE chemistry at RIKEN.

- The group-4 isotopes of ^{261}Rf, ^{169}Hf, and ^{85}Zr for chemical studies were produced in the ^{18}O-induced reactions on ^{248}Cm, ^{nat}Gd, and ^{nat}Ge targets, and were successfully extracted by the gas-jet to the chemistry laboratory after the physical preseparation by GARIS.

- The present result demonstrates that the GARIS/gas-jet system is promising to explore new frontiers in SHE chemistry.
  - (i) The background radioactivities originating from unwanted reaction products are strongly suppressed.
  - (ii) The intense primary heavy-ion beam is absent in the gas-jet chamber and hence high gas-jet transport efficiency is achieved.
  - (iii) The beam-free conditions make it possible to investigate new chemical systems that were not accessible before.