

Behavior of gaseous radionuclides produced in the gold target at the J-PARC Hadron Experimental Facility

¹ J-PARC Center

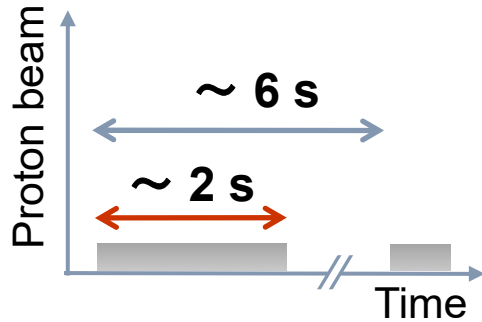
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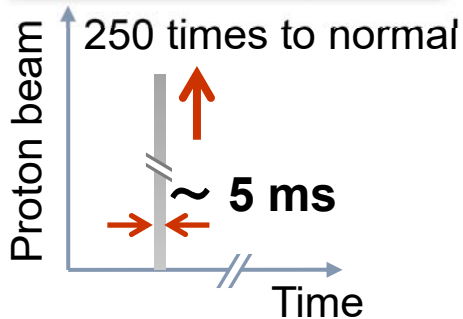
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K. Nishikawa^{1,2}, R. Kurasaki^{1,3}, R. Muto^{1,4},
Y. Kasugai¹, K. Saito^{1,2}, H. Yamazaki^{1,2}

Radioactive material leak incident at the Hadron Experimental Facility (May 2013)

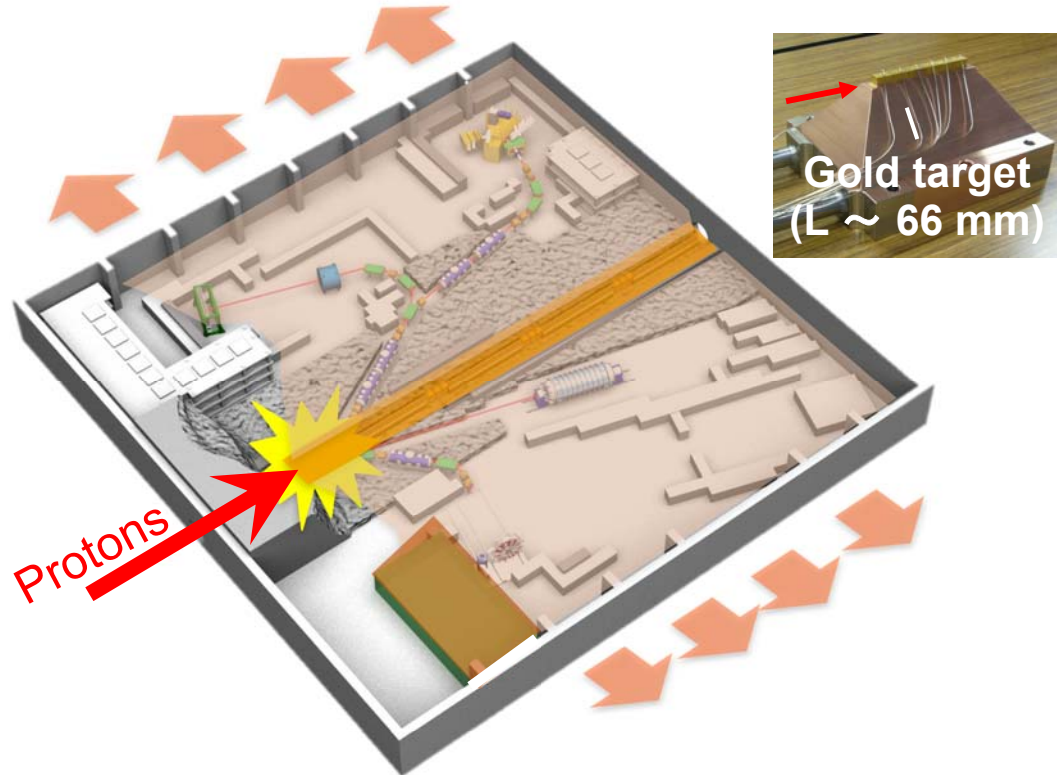


Slow-extraction
(Normal operation)

11:55 on May 23rd



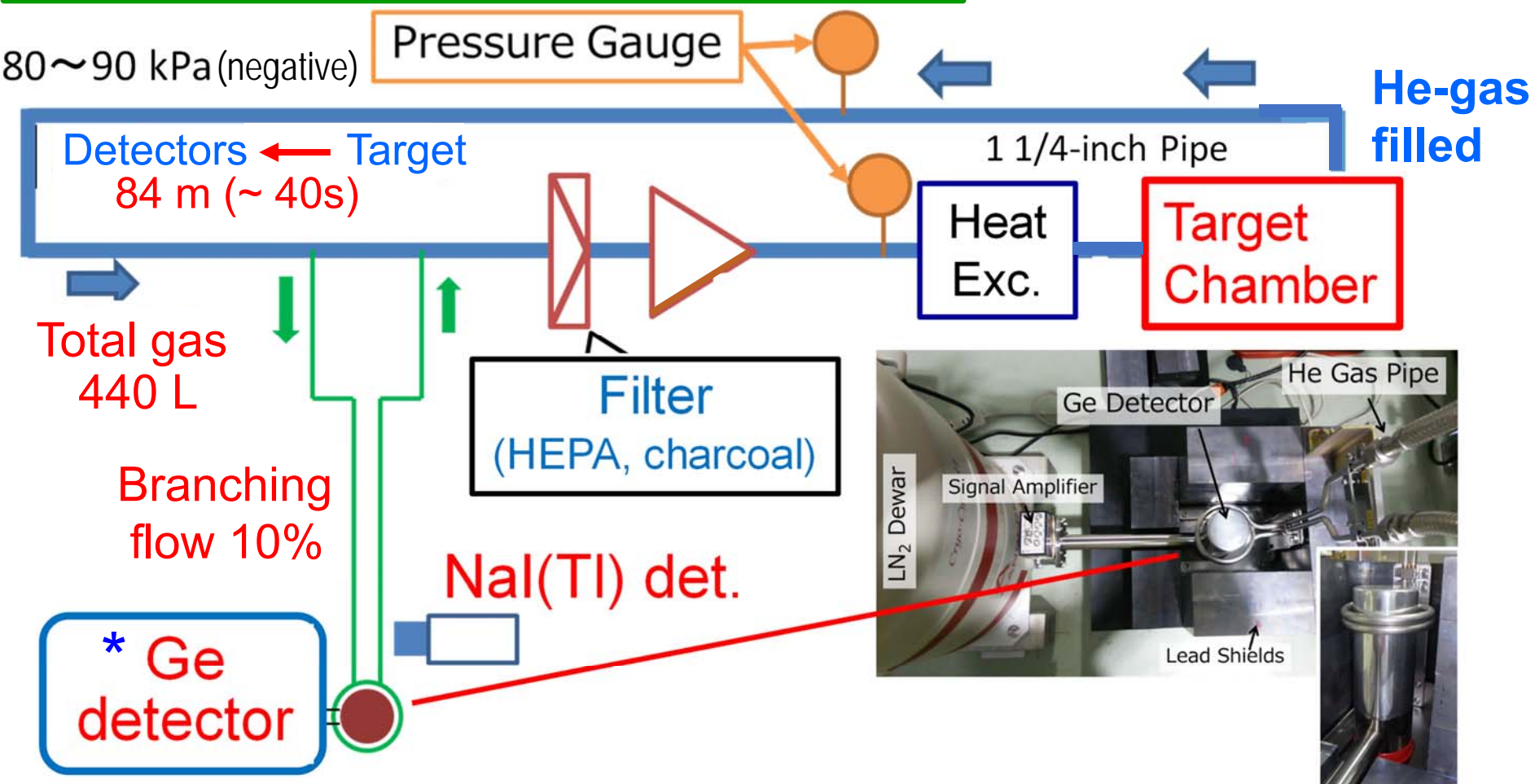
Abnormal beam due
to malfunction of the
instruments (1 shot)



Radionuclides in the gold target partially leaked
outside of the target.

Countermeasure against recurrence of similar incidents
□ Monitoring the soundness of the target

Countermeasure against recurrence of the incidents



Schematic view of the helium gas circulation system

(R. Muto *et al.*, EPJ Web of Conf. 153, 07004 (2017))

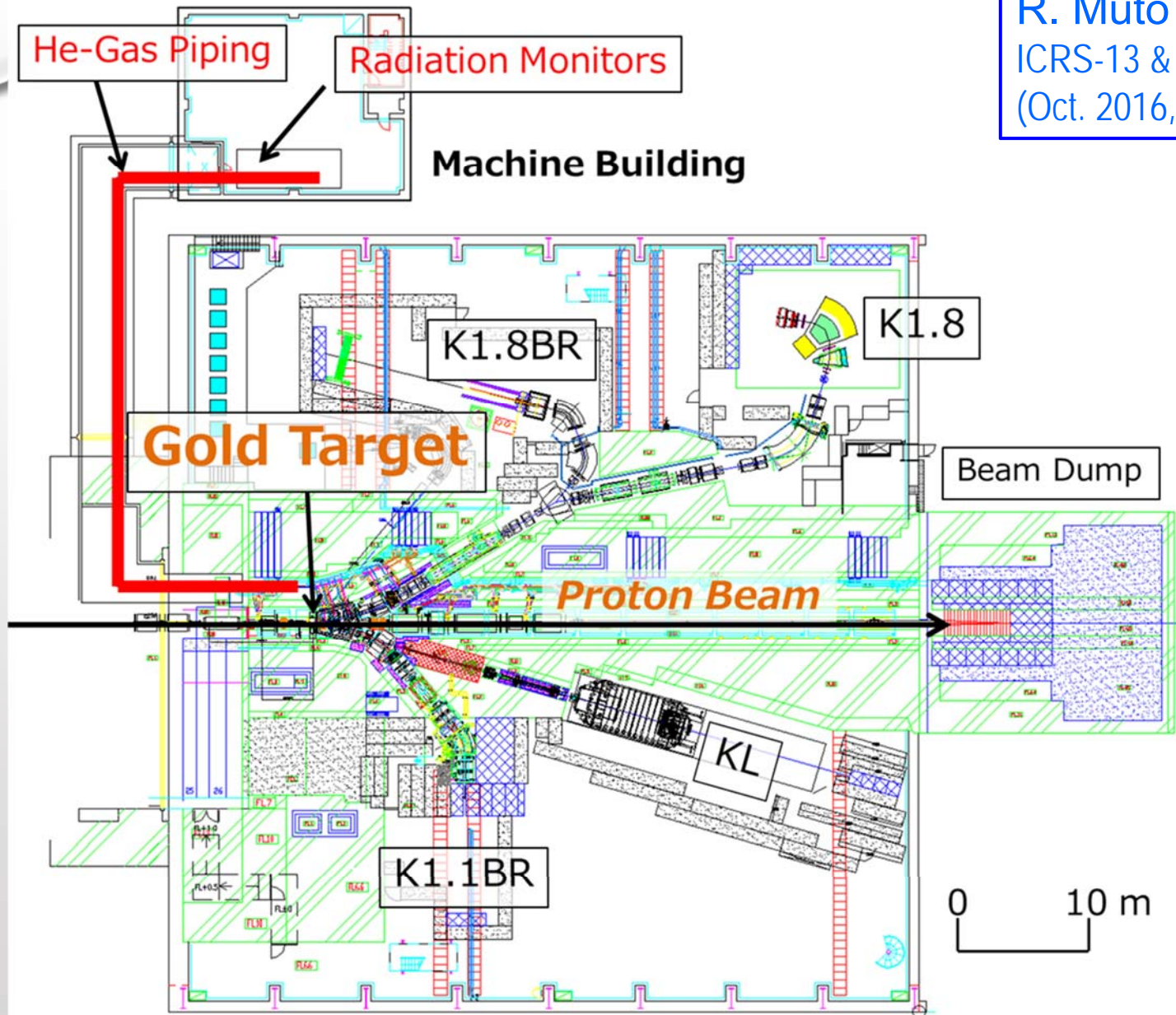
In operation
since April, 2015 ~

* Upgraded (April, 2017)

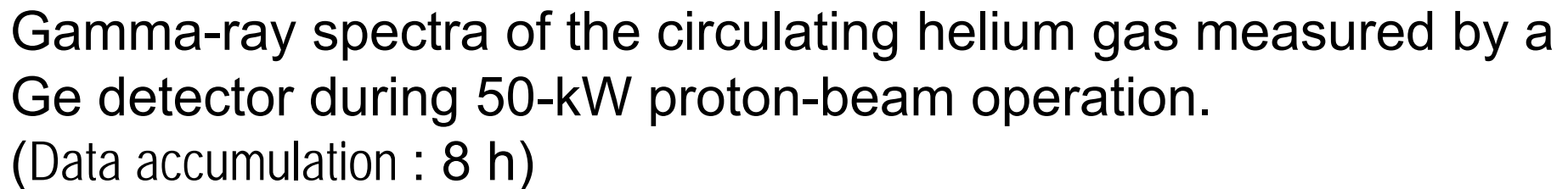
0.5 keV/ch x 16,384 ch, Spectrum meas. : 1,200 s (20 min)

Machine Building

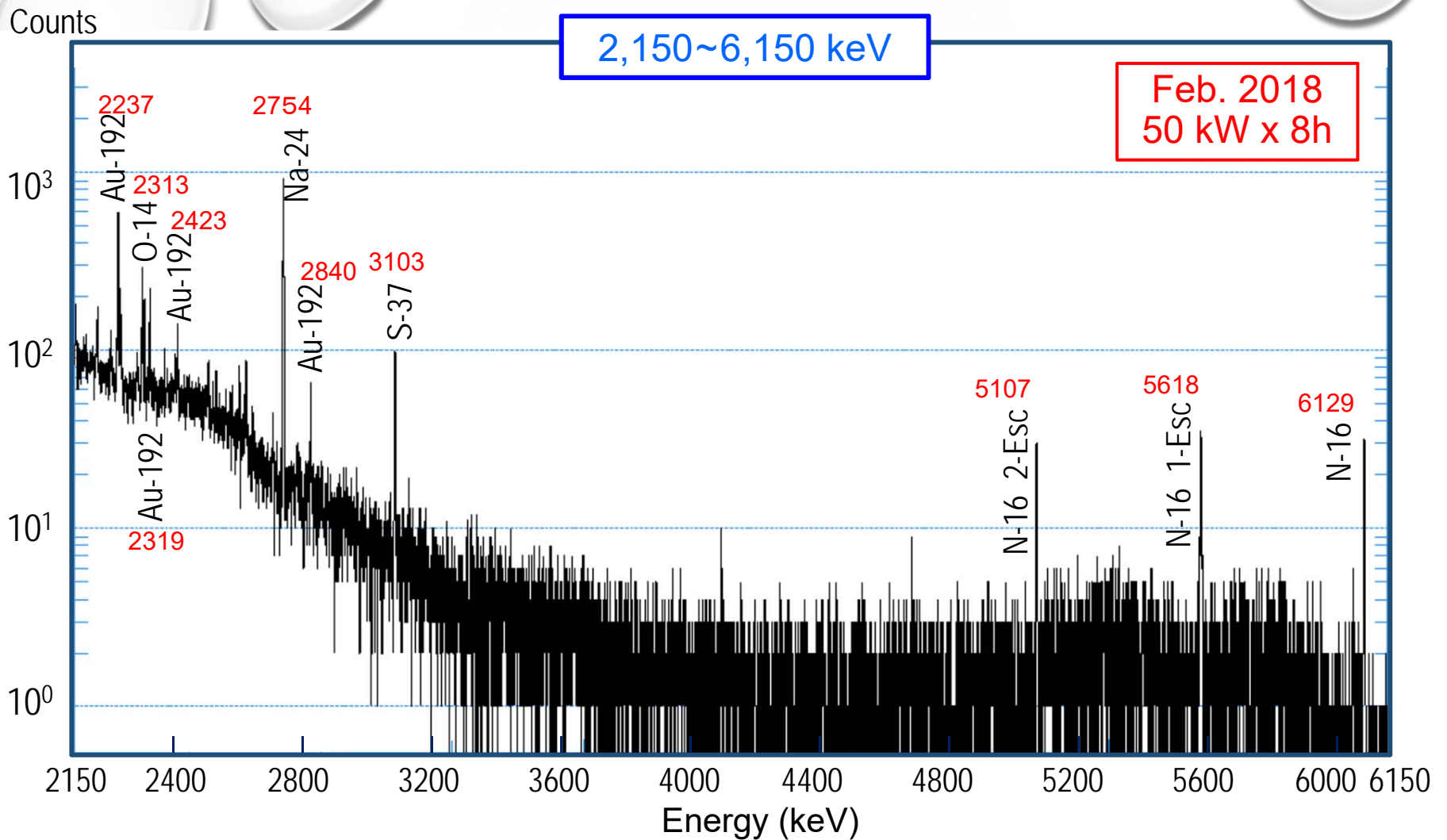
R. Muto *et al.*,
ICRS-13 & RPSD-2016
(Oct. 2016, Paris)



Hadron Experimental Facility



Gamma-ray spectra of the circulating helium gas measured by a Ge detector during 50-kW proton-beam operation.
(Data accumulation : 8 h)



Gamma-ray spectra of the circulating helium gas measured by a Ge detector during 50-kW proton-beam operation.
(Data accumulation : 8 h)

γ -emitting nuclides observed in the circulating He gas

Detected nuclides

C-10	19.3 s
N-16	7.1 s
O-14	70.6 s
O-19	26.9 s
O-20	13.6 s
F-20	11.0 s
Ne-23	37.2 s
Ne-24*	3.38 m
Na-24(* _{daut})	15.0 h
S-37	5.05 m
Ar-41	110 m
Hg-191m	50.8 m
Hg-192**	4.85 h
Au-192(** _{daut})	4.94 h
Hg-193m	11.8 h
Hg-195	9.9 h
Hg-195m	41.6 h

Calculations for nuclide production in the solid components

Nuclides produced in **beam windows** (Ti-6Al4V alloy)

Ti 90%, Al 6%, V 4 %

- Dominant nuclides **Sc-47, 44** etc. : **N.D.** in He gas
- C-10 ~ Ar-41 : Produced in solid, **Detected** in He

Nuclides produced in the **target** (Au) **Au 100%**

- Dominant nuclides **Au-196, 194, 198** etc. : **N.D.** in He
- C-10 ~ Hg-195m : Produced in solid, **Detected** in He

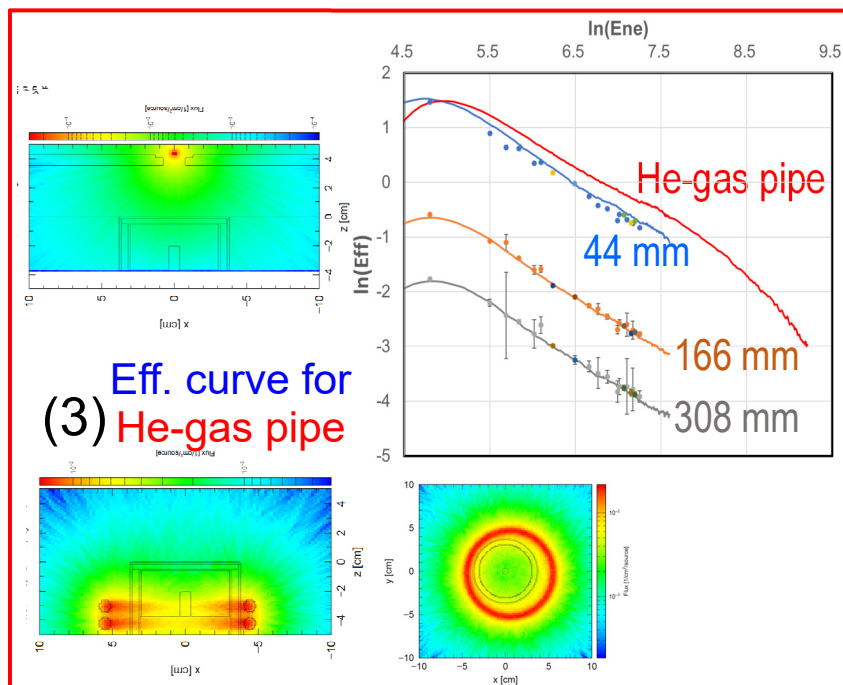
Nuclides forming **volatile chemical species** selectively transports to the gaseous phase.

C, N, O, F, Ne, S, Ar, Hg nuclides

Determining radioactivity from γ -ray spectra measurements

Efficiency calibration of the Ge detector

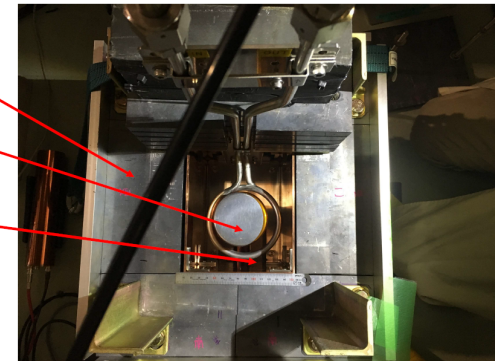
- (1) Measuring γ -ray efficiency curves for the Ge detector using standard point sources (Eu-152, Co-60, Cs-137, Na-22)
- (2) EGS5 simulations \rightarrow Optimizing the detector structure by reproducing the results (1)
- (3) Adding the He-gas pipe flowing around the Ge detector in the geometry \Rightarrow Calculating the efficiency curve for circulating He gas by EGS5



Pb shielding

Ge detector

He-gas pipe



Count rate for each γ -ray peak



Radioactivity (Bq) for each nuclide
can be determined.

Calculation geometry for production of radionuclides in the gold target and the beam windows

Target $15_x \times 11_y \times L\ 66\ \text{mm}$

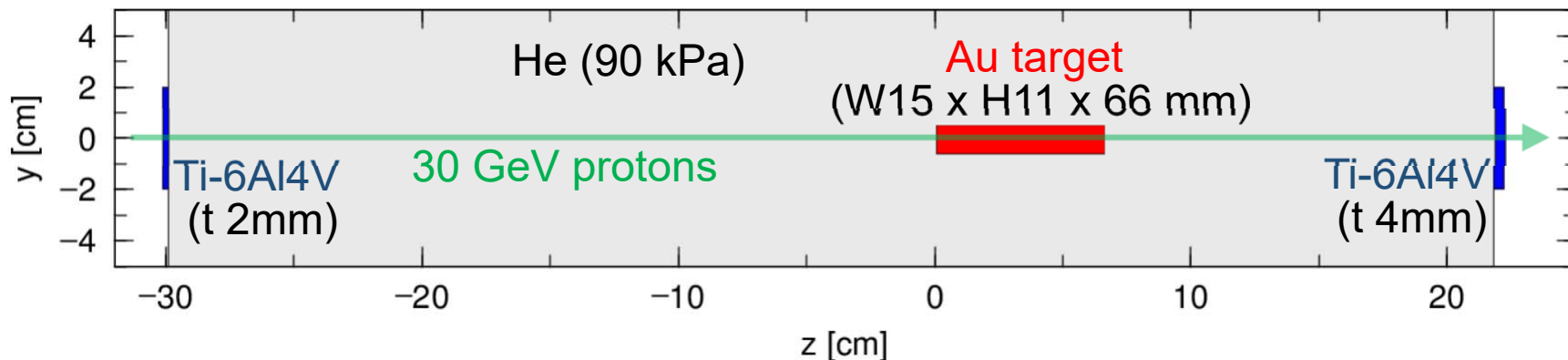
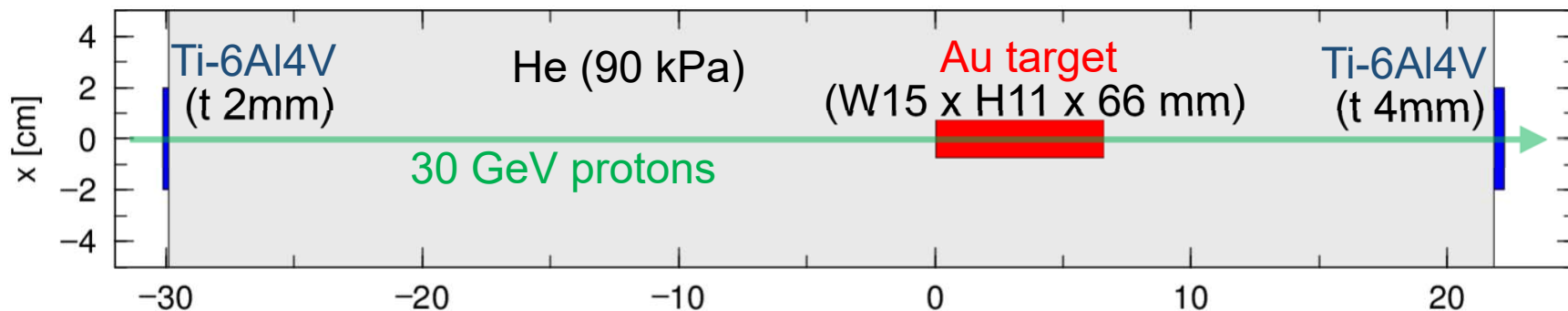
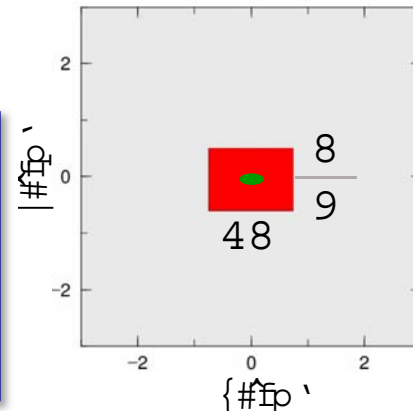
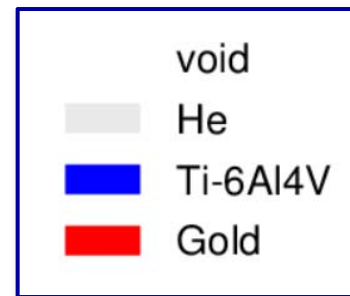
Au 100% ; $19.32\ \text{g/cm}^3$

Beam window $\Phi 40 \times t\ 2\ (\text{in}) / t\ 4\ (\text{out})\ \text{mm}$

Ti 90%, Al 6%, V 4 % ; $4.43\ \text{g/cm}^3$

He gas filled 90 kPa ; $0.16\ \text{mg/cm}^3$

30 GeV protons $\sigma_x\ 2.5\ \text{mm}, \sigma_y\ 1.0\ \text{mm}$



Calculation conditions for production of radionuclides in the gold target and the beam-windows

Code : PHITS 3.02 + DCHAIN-SP 2014

n : 1 meV~20 MeV : JENDL-4.0

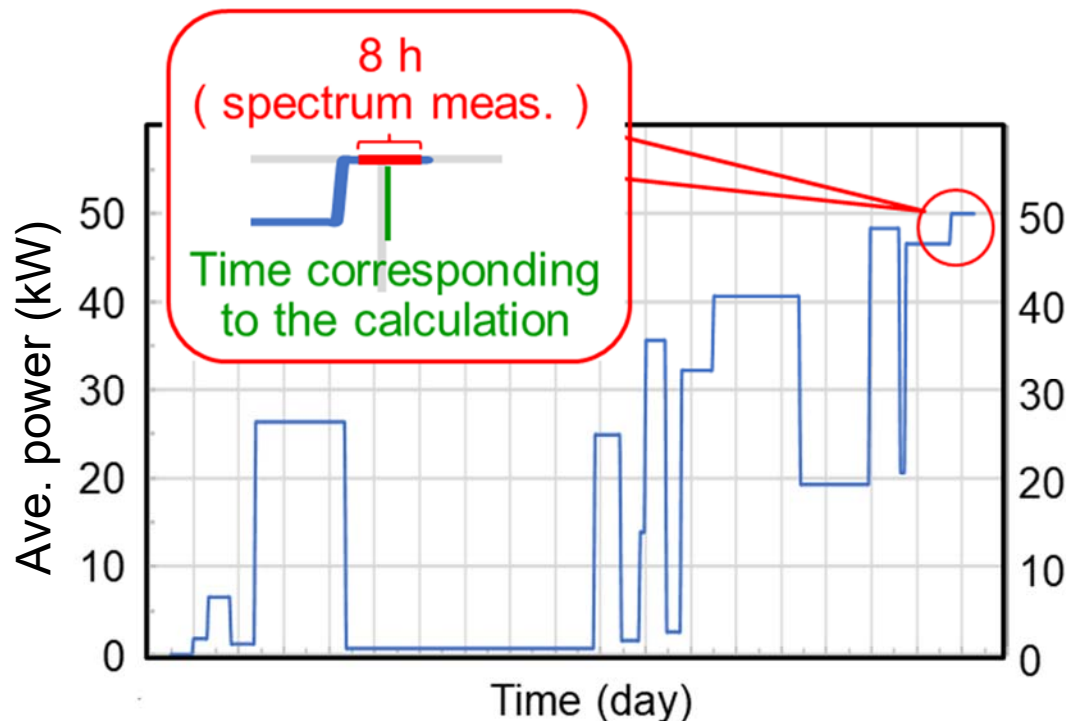
20 MeV~3 GeV : INCL-SMM-GEM

3 GeV~30 GeV : JAM-SMM-GEM

p, π^\pm : 1 eV~1 MeV : Bertini-GEM

1 MeV~3 GeV : INCL-SMM-GEM

3 GeV~30 GeV : JAM-SMM-GEM



Simplified time profiles for proton-beam operation during Jan.-Feb., 2018 at the J-PARC HEF.

Activity of radionuclides

evaluated in 40 s* after the reaction by the proton-beam

*40 s

Transfer time for He circulation from the target chamber to the Ge detector

Radioactive decay before arriving at the Ge detector

Calculation results (Summary)

- Dominant nuclides produced in **solid**
- Nuclides **detected** in **He gas**

Radionuclides produced in beam windows

	Nuclide	Act [Bq]	HL	keV	Br(%)
1	Sc 47	5.7E+09	81.6h	159	68
2	Sc 44	5.8E+09	3.9h	1,157	100
5	Sc 44m	2.5E+09	58.6h	271	87
6	Al 28	2.5E+09	2.2m	1,779	100
Det	C 10	2.4E+07	19.3s	718	99
Det	N 16	3.5E+06	7.1s	6,129	67
Det	O 14	1.9E+07	70.6s	2,313	99
Det	O 19	1.4E+07	26.9s	1,357	50
Det	O 20	2.1E+06	13.6s	1,057	100
Det	F 20	9.0E+07	11.0s	1,634	100
Det	Ne 23	1.8E+08	37.2s	440	33
Det	Ne 24	1.1E+08	203s	472	100
Det	Na 24	1.8E+09	15.0h	1,369	100
Det	S 37	1.2E+08	303s	3,103	94
Det	Ar 41	5.4E+08	110m	1,294	99

daughter

daughter

Radionuclides produced in Au target

	Nuclide	Act [Bq]	HL	keV	Br(%)
1	Au 196	2.2E+12	6.2d	356	87
3	Au 194	8.9E+11	38.0h	328	61
5	Au 196n	6.7E+11	9.6h	188	37
6	Au 198	6.6E+11	64.7h	412	96
Det	Hg 191m	2.9E+10	50.8m	253	58
Det	Hg 192	1.1E+11	4.85h	275	50
Det	Hg 193m	5.4E+10	11.8h	408	33
Det	Hg 195	1.4E+11	9.9h	780	7
Det	Hg 195m	8.1E+10	41.6h	560	7
Det	Au 192	5.8E+11	4.94h	317	58
Det	C 10	6.8E+08	19.3s	718	99
Det	N 16	3.6E+08	7.1s	6,129	67
Det	O 14	3.8E+08	70.6s	2,313	99
Det	O 19	1.9E+09	26.9s	1,357	50
Det	O 20	2.5E+08	13.6s	1,057	100
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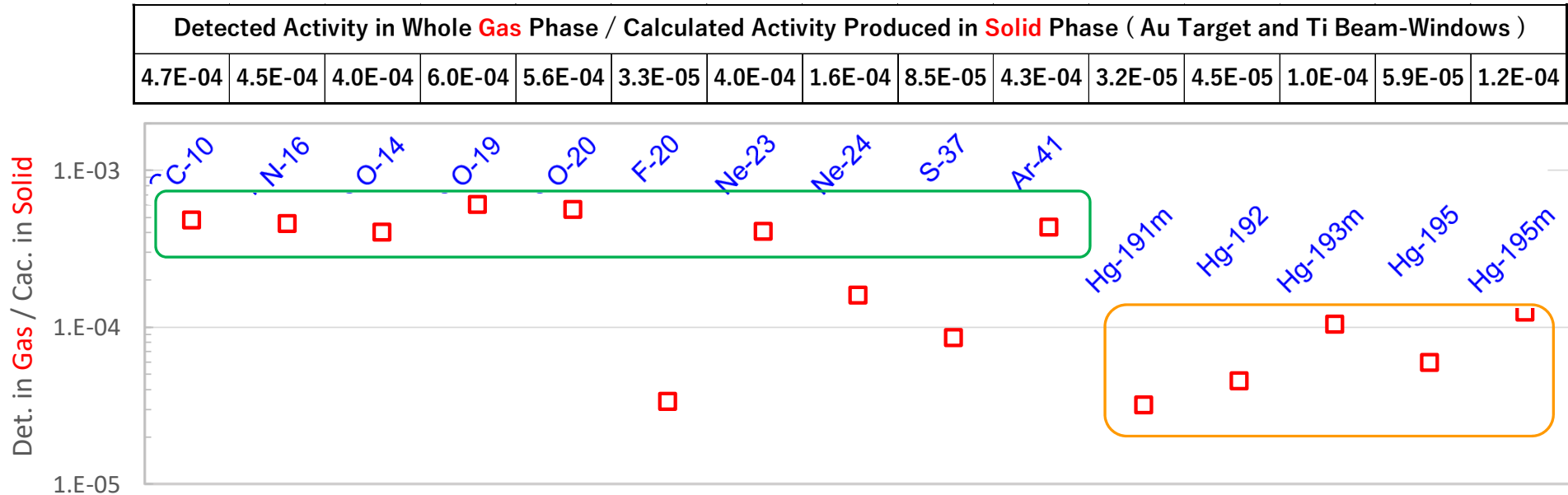
Comparison between detected activity in He gas and calculated activity in the solid components

	C-10	N-16	O-14	O-19	O-20	F-20	Ne-23	Ne-24	S-37	Ar-41	Hg-191m	Hg-192	Hg-193m	Hg-195	Hg-195m
Half-life (s)	19.25	7.12	70.61	26.91	13.57	11.03	37.2	202.8	303	6,577	3,048	17,460	42,480	35,640	149,760
γ -ray (keV)	718	6,129	2,313	1,357	1,057	1,634	440	472	3,103	1,294	253	275	408	780	560
Br (%)	98.5	67.0	99.4	50.4	100.0	100.0	33.0	100.0	94.0	99.1	57.6	50.4	32.5	7.0	7.0
Det. Eff.	1.2E-02	1.3E-03	4.6E-03	7.3E-03	8.8E-03	6.4E-03	1.9E-02	1.8E-02	3.3E-03	7.4E-03	3.4E-02	3.1E-02	2.1E-02	1.1E-02	1.5E-02
Det. Act. in Gas Phase	Detected Peak Counts [cps]														
	0.26	0.0089	0.046	0.26	0.078	0.056	1.01	0.79	0.020	0.77	1.1	4.7	2.4	0.41	0.68
	Detected Activity Measured by the Ge detector ($2.8 \times 10^{-5} \text{ m}^3$) [Bq]														
	2.1E+01	1.2E+01	1.2E+01	7.3E+01	8.8E+00	8.8E+00	1.6E+02	4.3E+01	6.3E+00	1.1E+02	5.9E+01	3.0E+02	3.6E+02	5.1E+02	6.3E+02
	Activity in Whole Gas Phase (0.44 m^3) [Bq]														
	3.4E+05	1.7E+05	1.6E+05	1.1E+06	1.4E+05	1.4E+05	2.5E+06	6.9E+05	1.0E+05	1.7E+06	9.3E+05	4.7E+06	5.6E+06	8.1E+06	1.0E+07
Calc. Act. in	Calculated Activity Produced in the Au Target and the Ti Beam-Windows [Bq] (Cooling Time : 40 s)														
Au target	6.8E+08						1E+09	6.0E+09	4.2E+09	1.0E+09	3.3E+09	2.9E+10	1.1E+11	5.4E+10	1.4E+11
Ti windows	2.6E+07						4E+07	1.9E+08	1.2E+08	1.3E+08	5.6E+08	—	—	—	—
Ratios	Detected Activity in Whole Gas Phase / Calculated Activity Produced in Solid Phase (Au Target and Ti Beam-Windows)														
	4.7E-04	4.5E-04	4.0E-04	6.0E-04	5.6E-04	3.3E-05	4.0E-04	1.6E-04	8.5E-05	4.3E-04	3.2E-05	4.5E-05	1.0E-04	5.9E-05	1.2E-04

Det. in Gas / Cac. in Solid



Rates released to gaseous phase for detected radionuclides



Characteristics of detected radionuclides

- ◆ C-10, N-16, O-14, O-19, O-20, Ne-23, Ar-41 : $4 \sim 6 \times 10^{-4}$

Nuclides produced in near-surface region fully release to gaseous phase.

- ◆ Ne-24 : 1.6×10^{-4}

The reason for discrepancy with Ne-23 (factor~2.5) : Not clarified.

- ◆ F-20 : 3.3×10^{-5} , S-37 : 8.5×10^{-5}

They are effected by chemical states of F and S nuclides on solid surface.

- ◆ Hg-191m, 192, 193m, 195, 195m : $0.3 \sim 1.2 \times 10^{-4}$

Behavior of Hg nuclides

Temp. of the gold target during beam operations

: 70~360°C (50-kW)

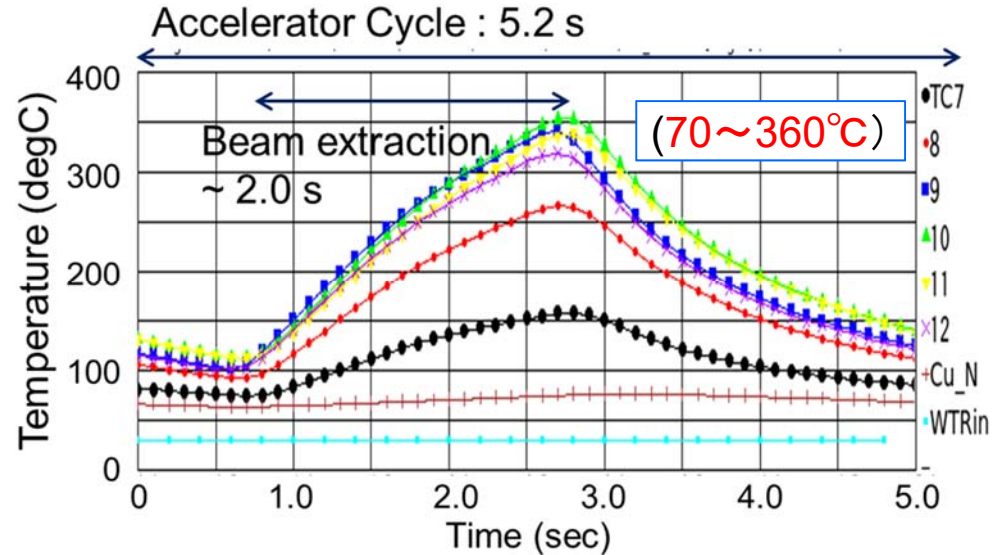


Hg nuclides partially present as a gaseous species during the beam operations.

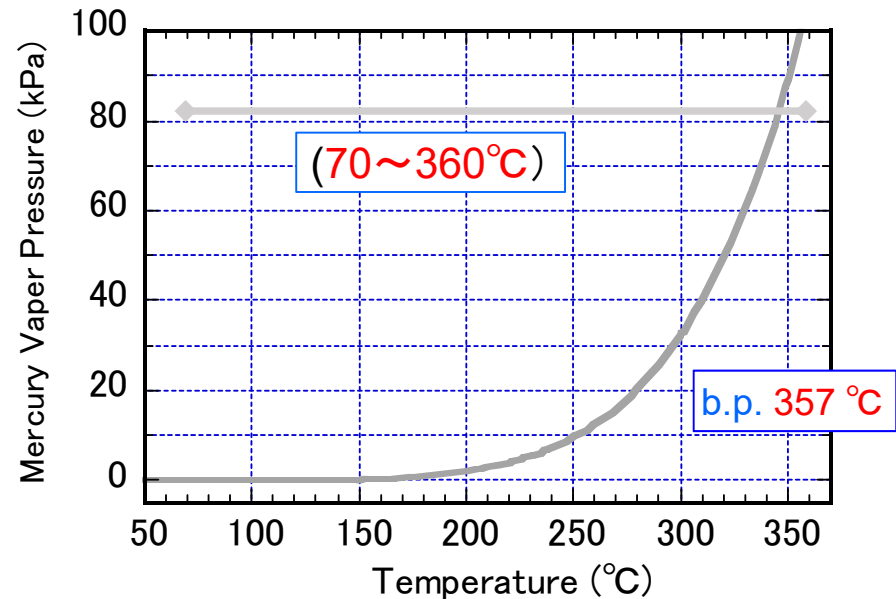
◆ Hg nuclides : $0.3 \sim 1.2 \times 10^{-4}$

◆ C, N, O, Ne, Ar nuclides : $4 \sim 6 \times 10^{-4}$

51 kW beam operation



Time profiles for the temperature of the target



Vapor pressure of metallic mercury (Hg)

Behavior of gaseous radionuclides in the He gas circulating system at the Hadron Experimental Facility

- ◆ Measuring γ -ray spectrum of the He gas
- ◆ Calculating the radionuclide production in solid components

Comparison
Discussion

Findings

- C, N, O, F, Ne, S, Ar and Hg radionuclides are selectively released to gas phase compared to other dominant radionuclides produced in the solid components.
- Selectivity in transporting to gas phase is related with elemental volatility of nuclides.

More systematic analyses are in progress to elucidate the transportation processes of various nuclides from solid to gas phase.