Sep. 26 (Thu.) 2019
SAF: Safety for Intensity Frontier
14:40-15:00

Behavior of Tritium Release from a Stainless Vessel of the Mercury Target as a Spallation Neutron Source

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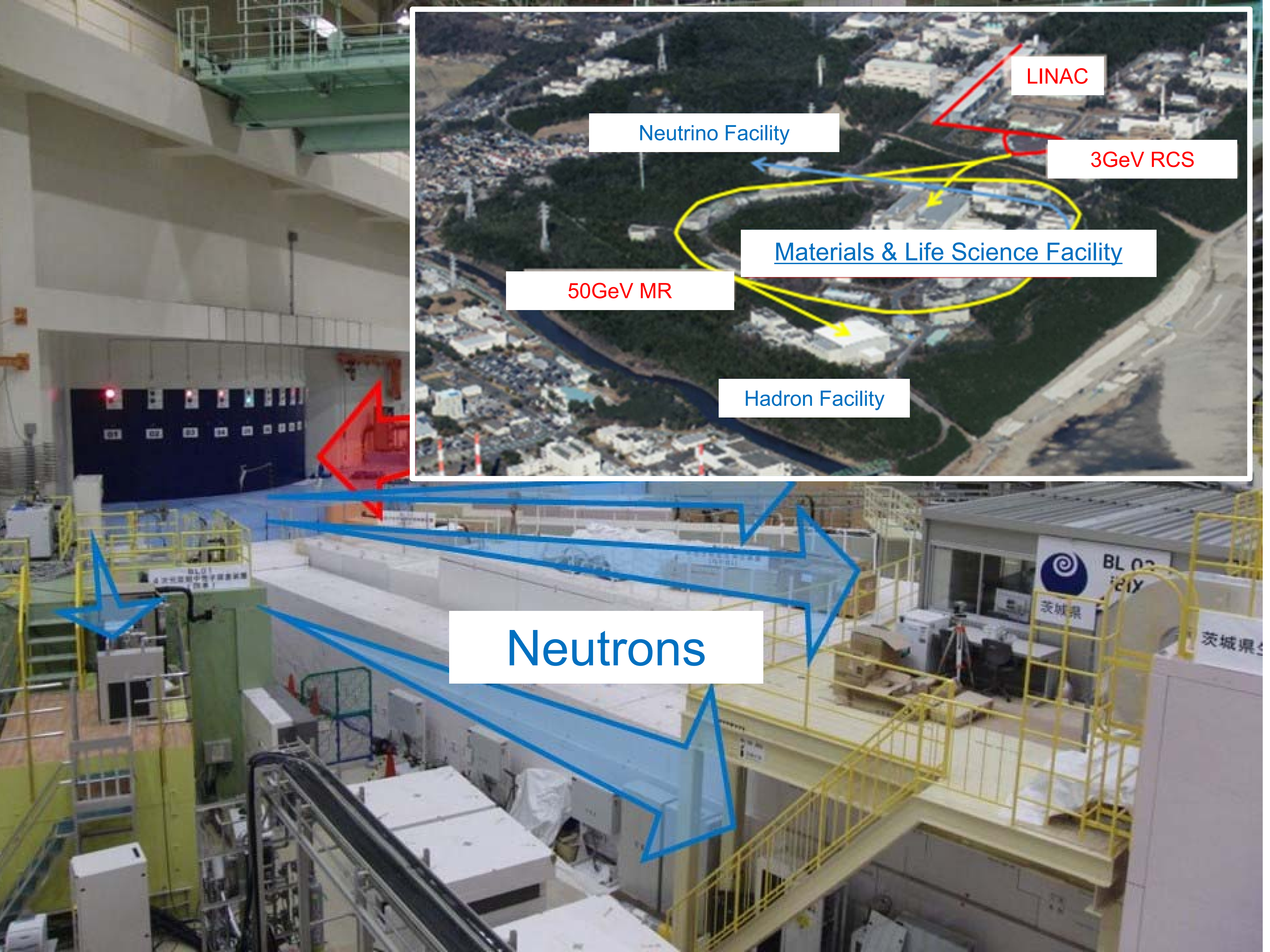
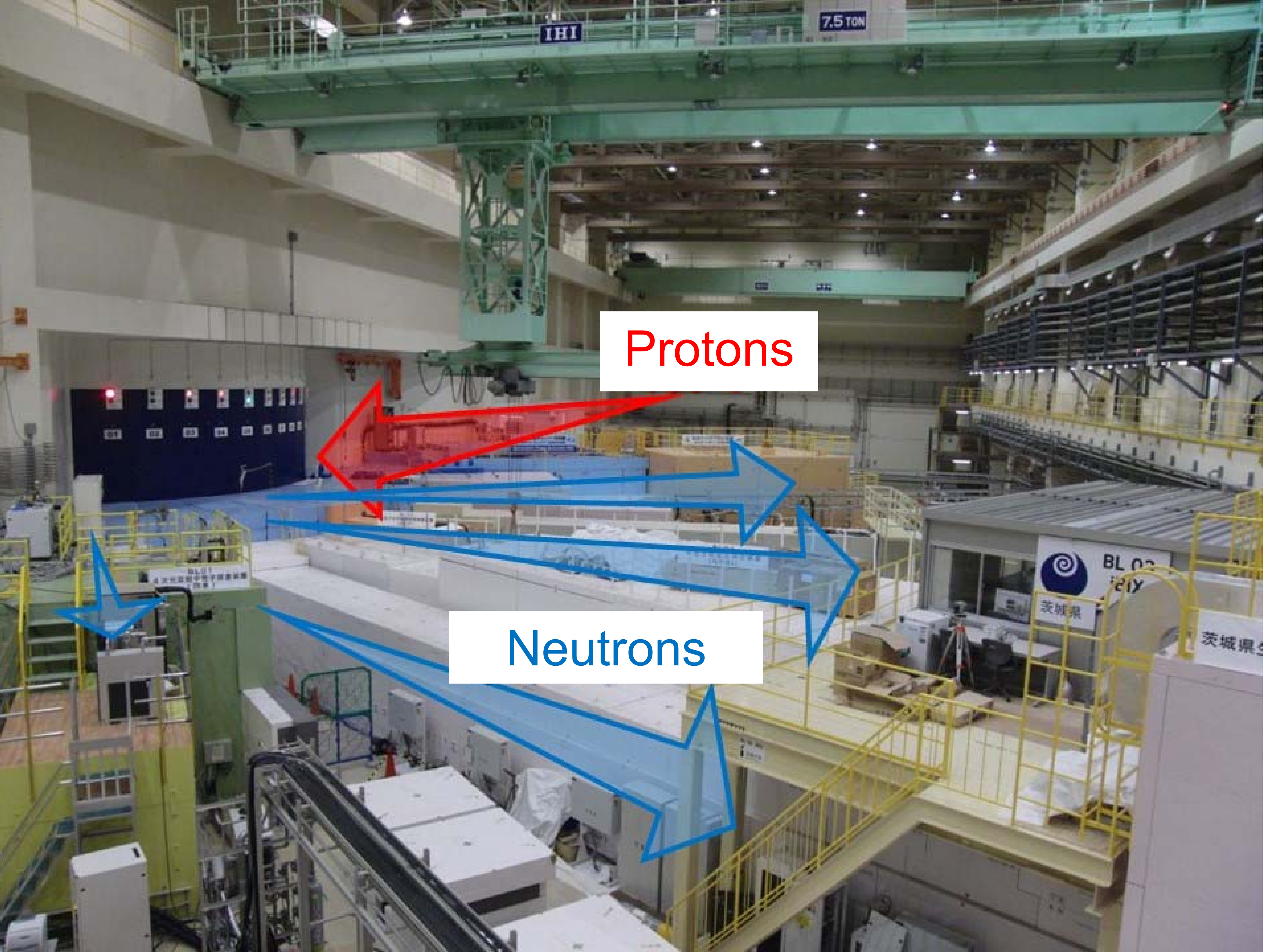
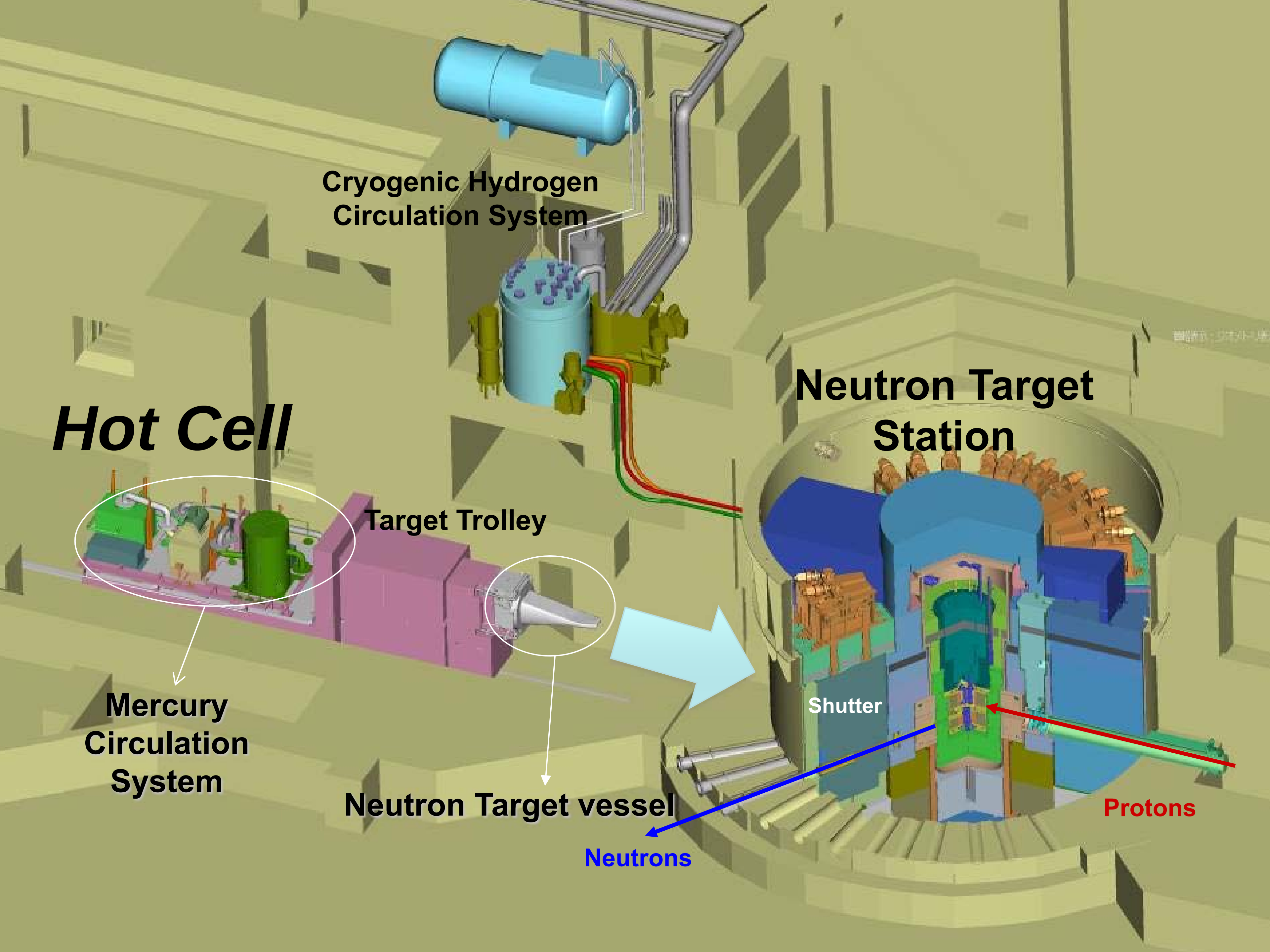
Introduction

- J-PARC（Japan Proton Accelerator Research Complex）
  - Located in Takai-mura, Ibaraki, Japan
  - Consists of accelerator and experimental facilities for studying cutting edge science on particle physics, nuclear physics, material science, life science etc.
  - Run jointly by JAEA and KEK
- Materials and Life Science Experimental Facility
  - Spallation Neutron Source with mercury
- Mercury Target Vessel（Stainless）
  - Periodical exchange work => Observed tritium release
  - Report the release behavior and the analytical interpretation


LINAC
Neutrino Facility
3GeV RCS
Materials & Life Science Facility
50GeV MR
Hadron Facility
Neutrons



Protons
Neutrons
IHI
7.5 TON

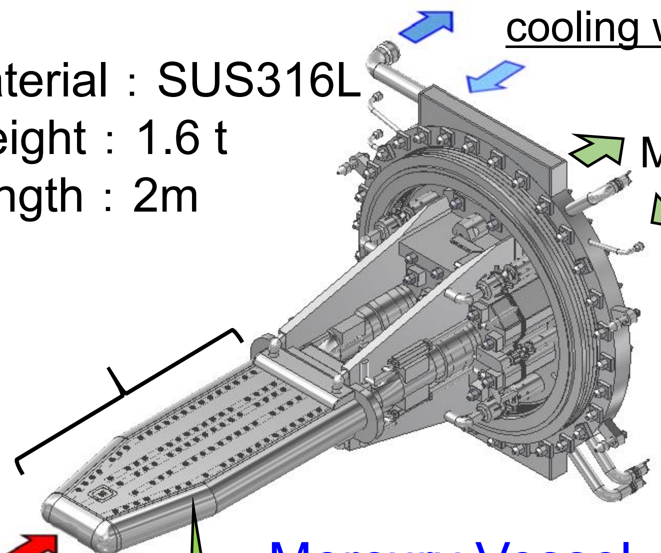


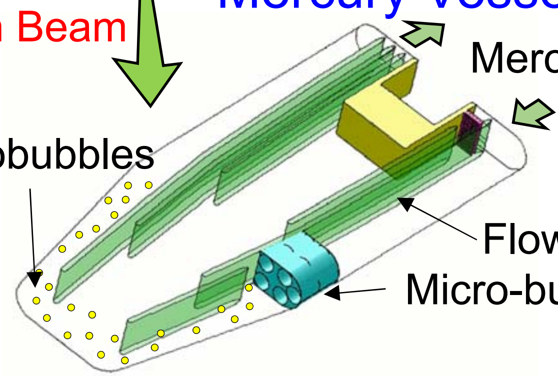
Cryogenic Hydrogen Circulation System
Hot Cell
Target Trolley
Neutron Target Station
Shutter
Mercury Circulation System
Neutron Target vessel
Protons
Neutrons

Outline of the Neutron Target

Neutron Target Vessel

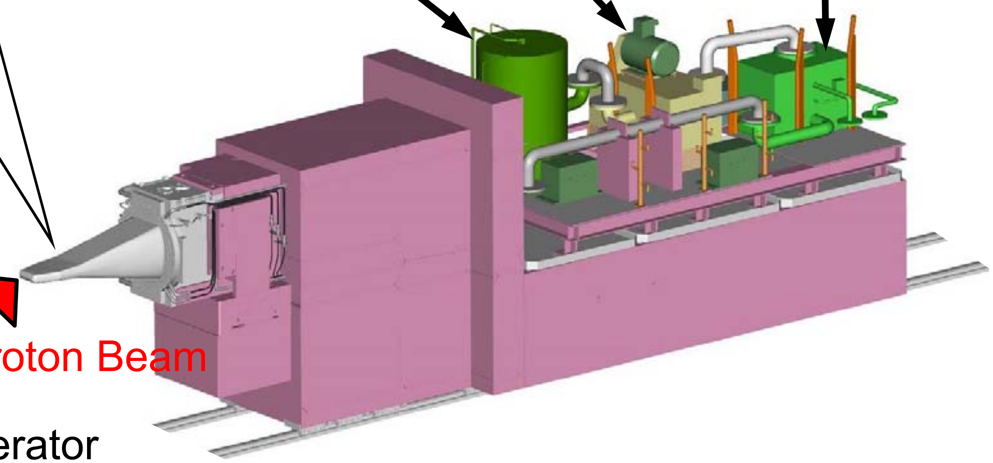
Material : SUS316L
Weight : 1.6 t
Length : 2m



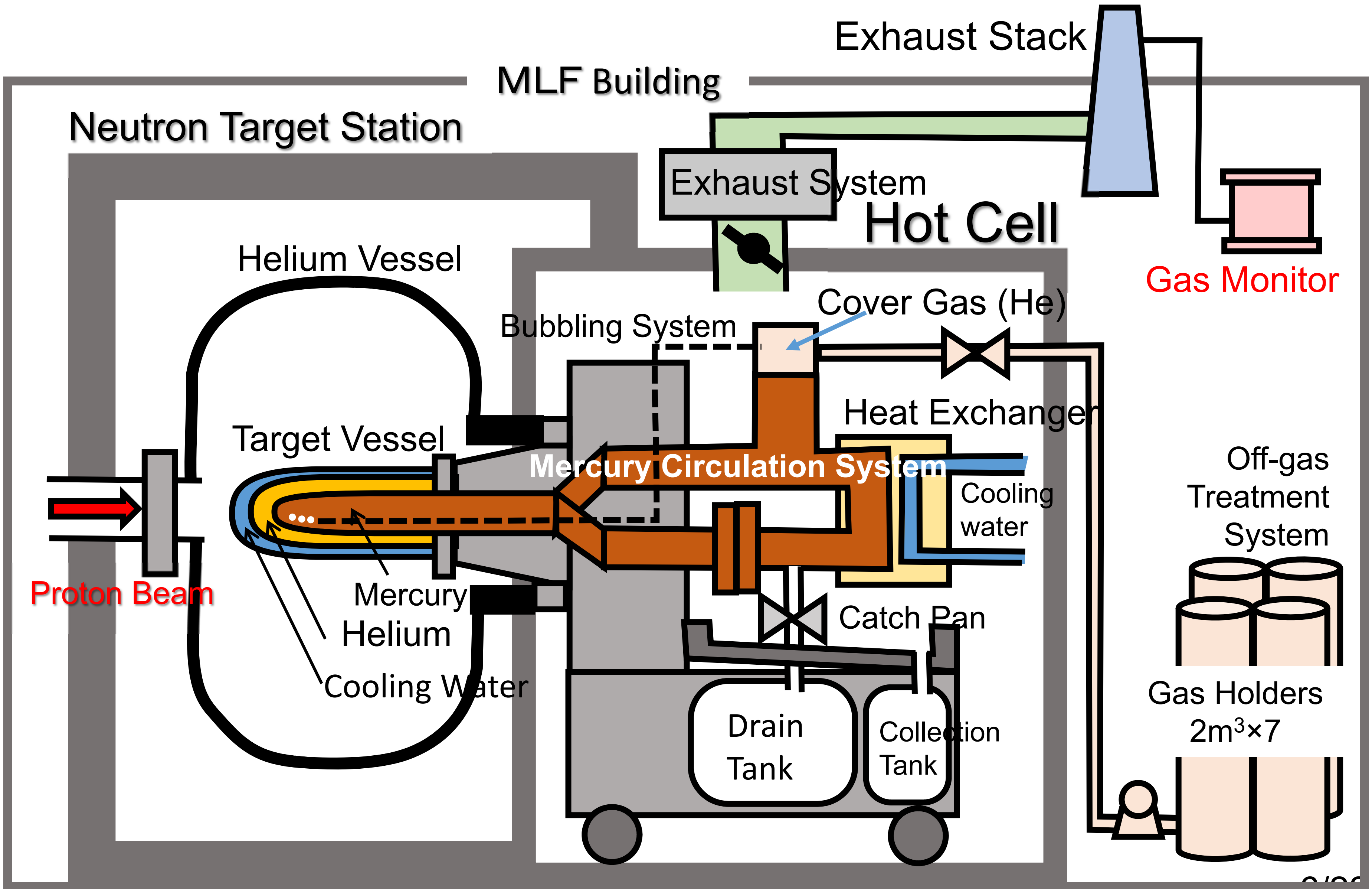


*Mitigation of cavitation damage by microbubbles

Length :	12 m
Weight :	315 ton
Mercury Volume :	1.5 m^3
Mercury Flow rate :	41m^3/hr



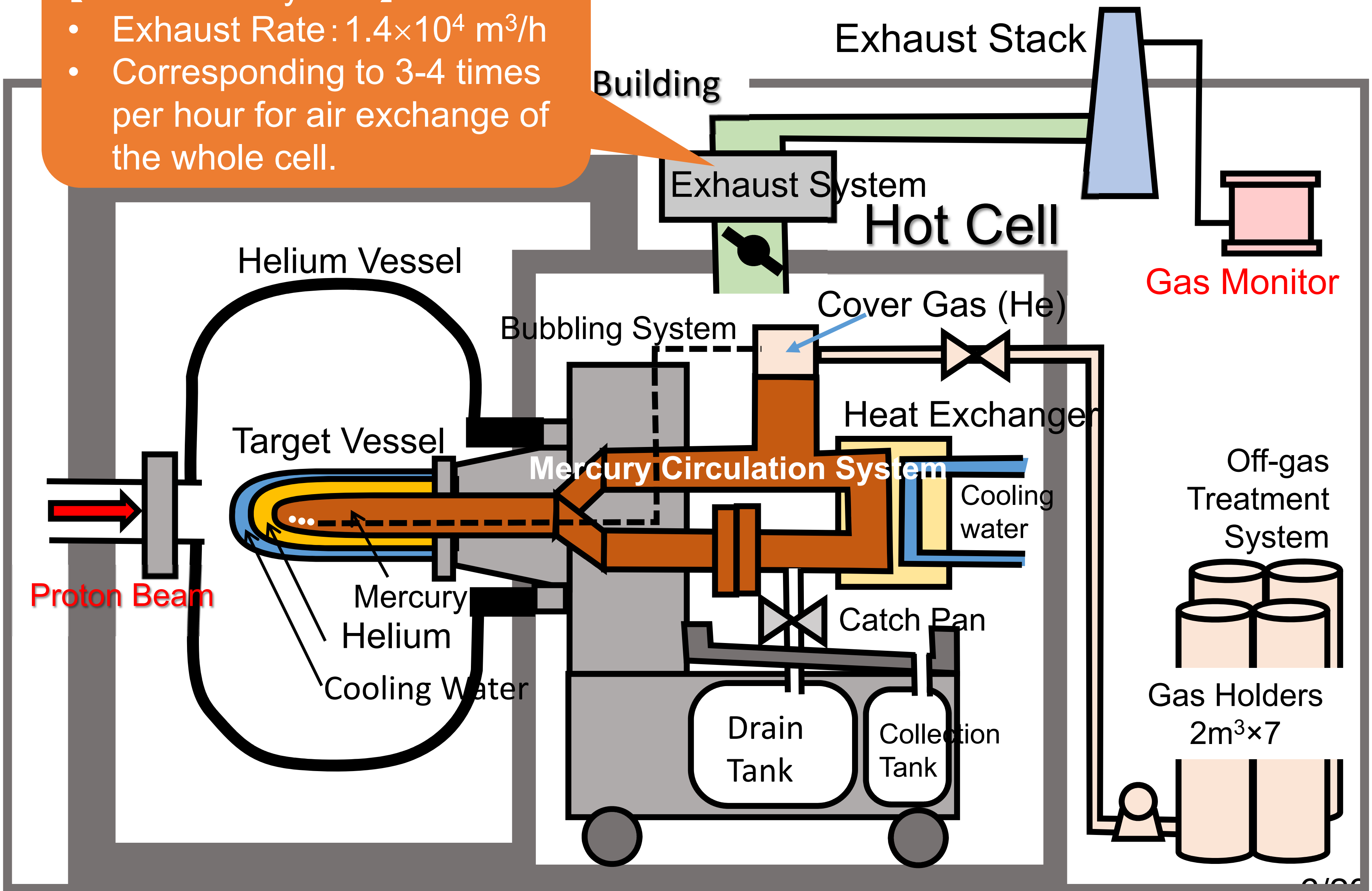
Concept of the Radioactivity Confinement



Concept of the Radioactivity Confinement

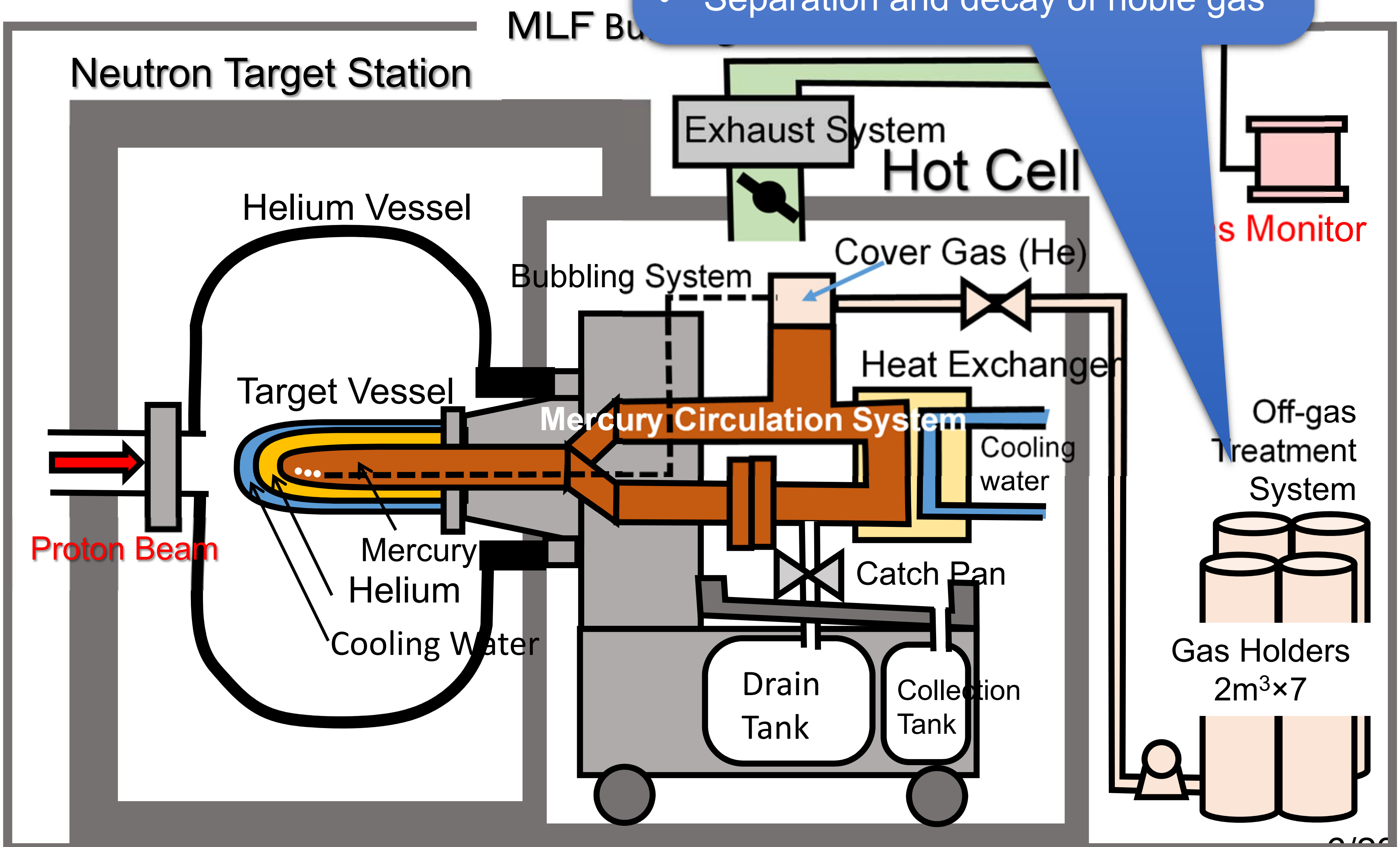
【Ventilation System】

- Exhaust Rate : 1.4×10^4 m^3/h
- Corresponding to 3-4 times per hour for air exchange of the whole cell.

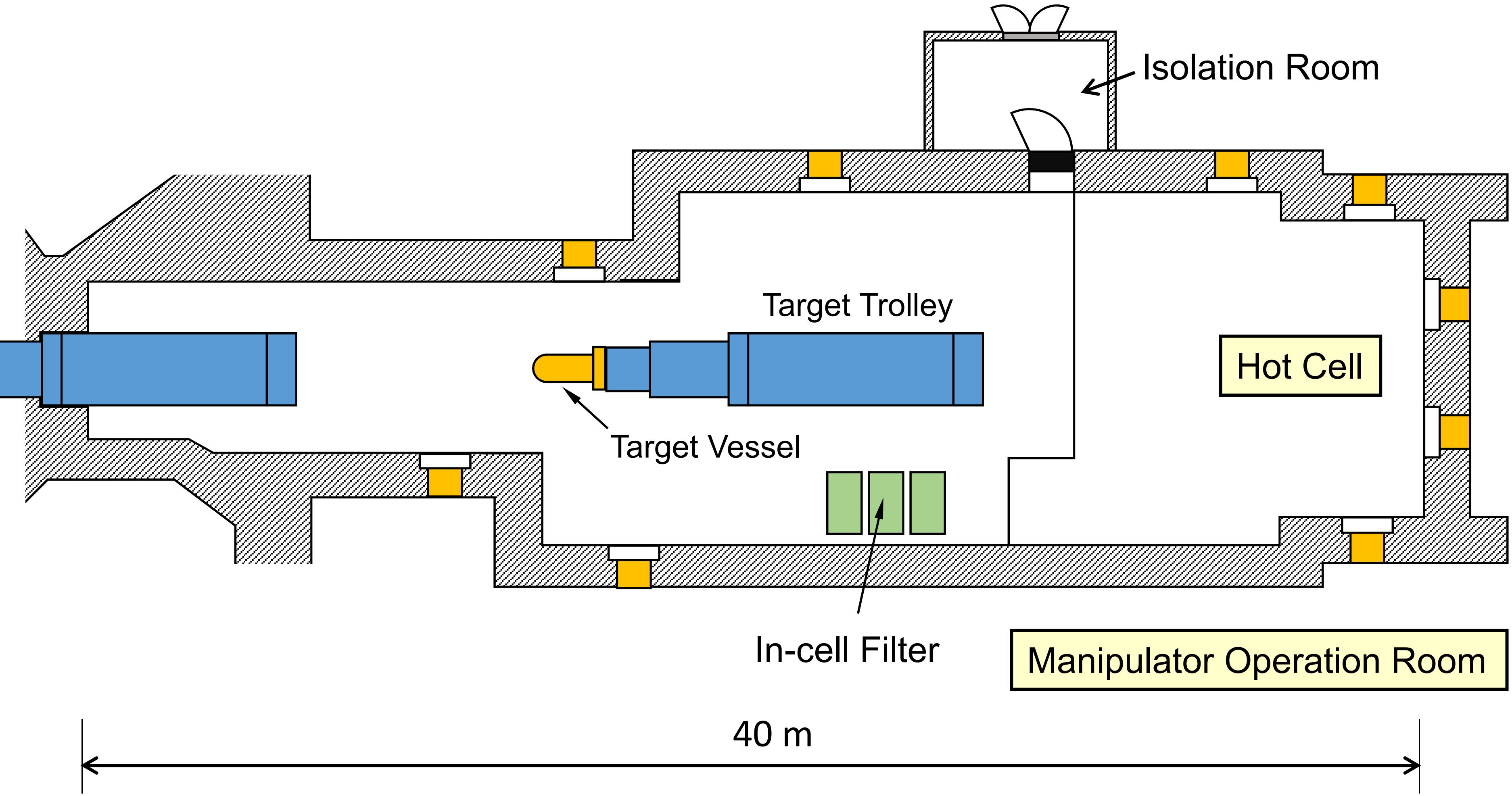


Concept of the Radio

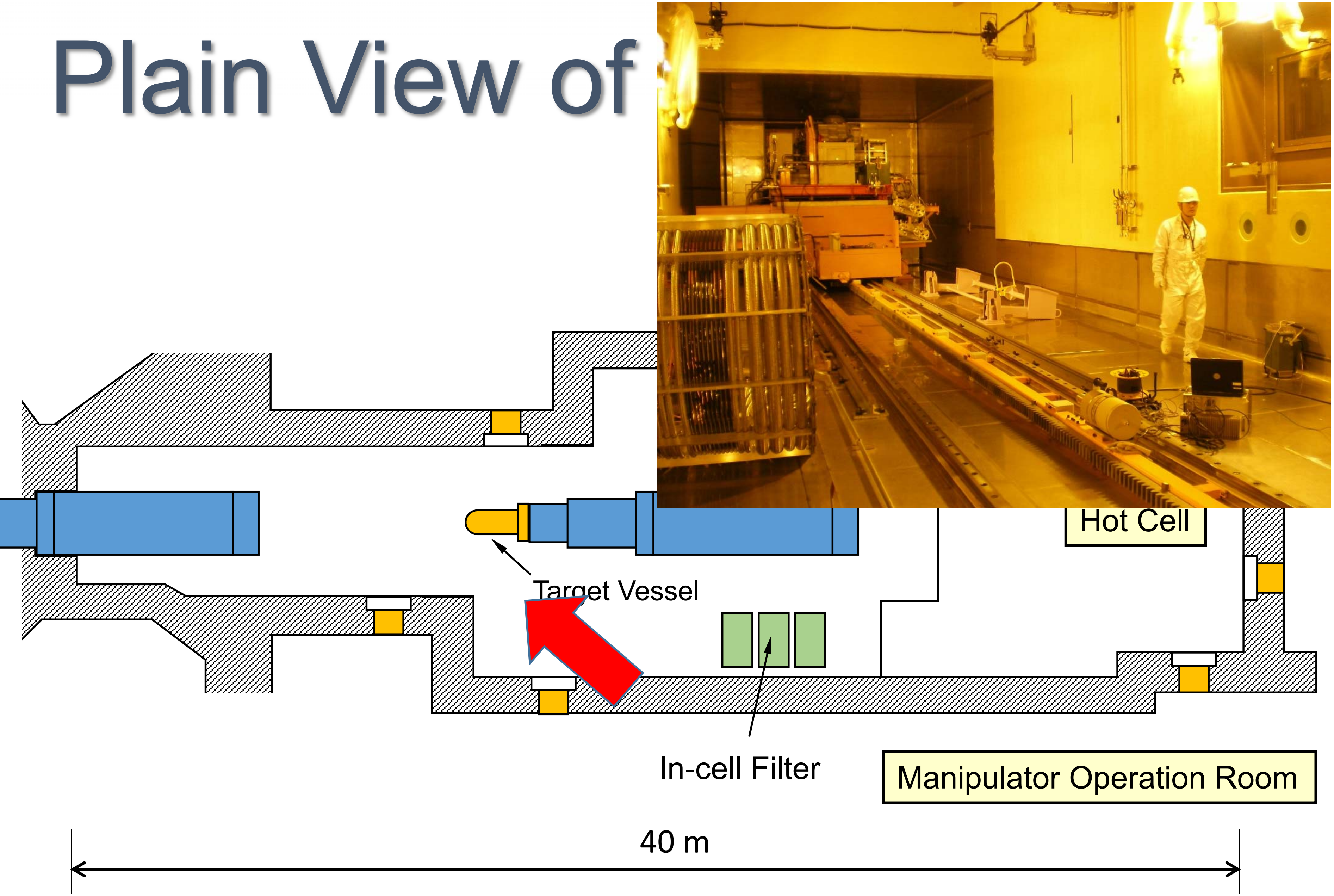
【Off-gas treatment System】
- Removal of mercury
- Removal of tritium
- Separation and decay of noble gas



Plain View of the Hot Cell



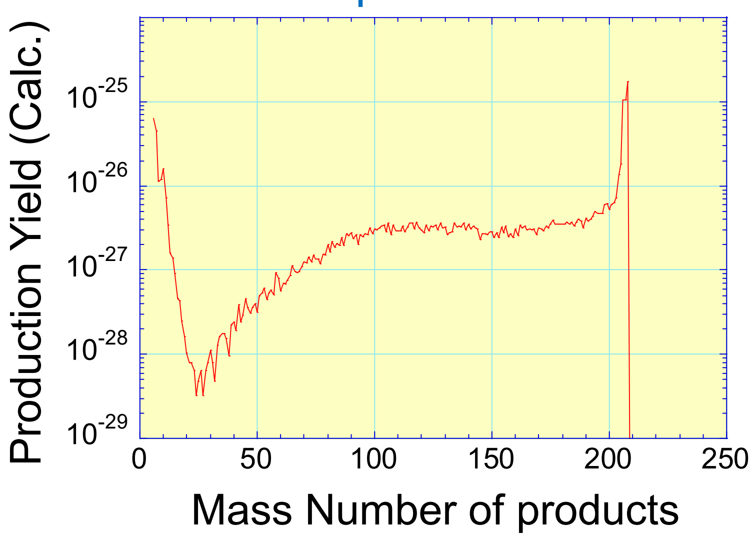
Plain View of

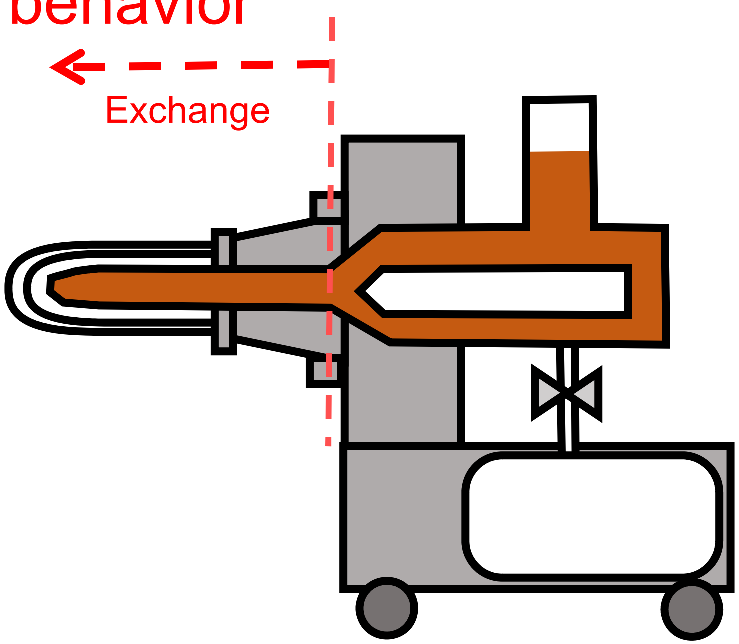


Why We need to Understand the Tritium Behavior?

- Various kinds of radioactive nuclides produced
- Tritium, : ^{3}H, T
  - ~10^{14}Bq for 1MW-1 year operation.
- Suppression of tritium release during the target exchange work
  - Need understanding the T(^{3}H) behavior

Mass Yield for spallation reactions





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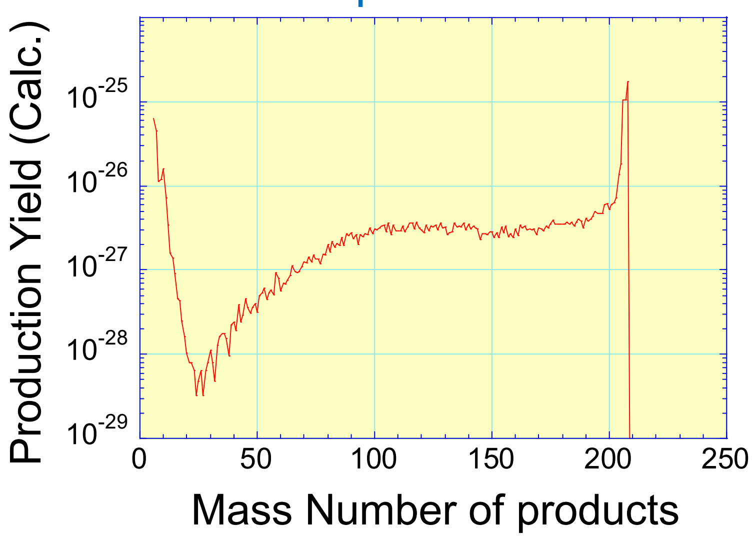

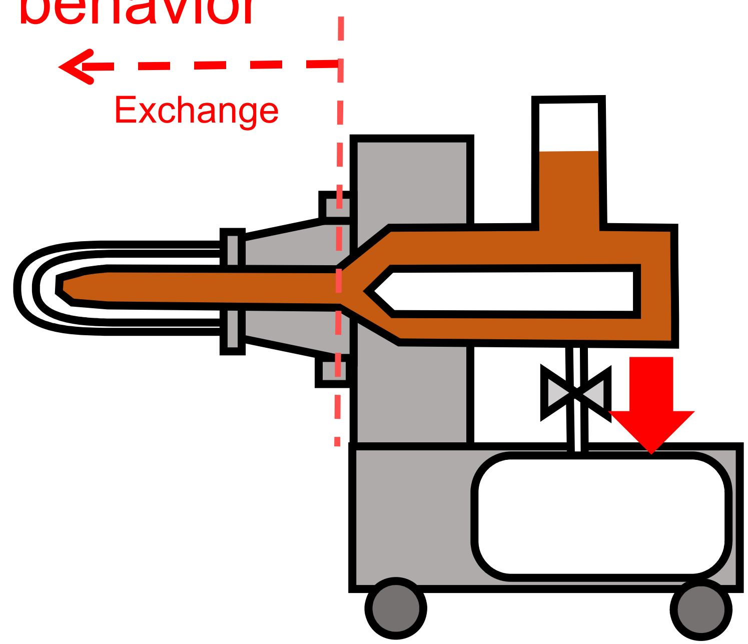

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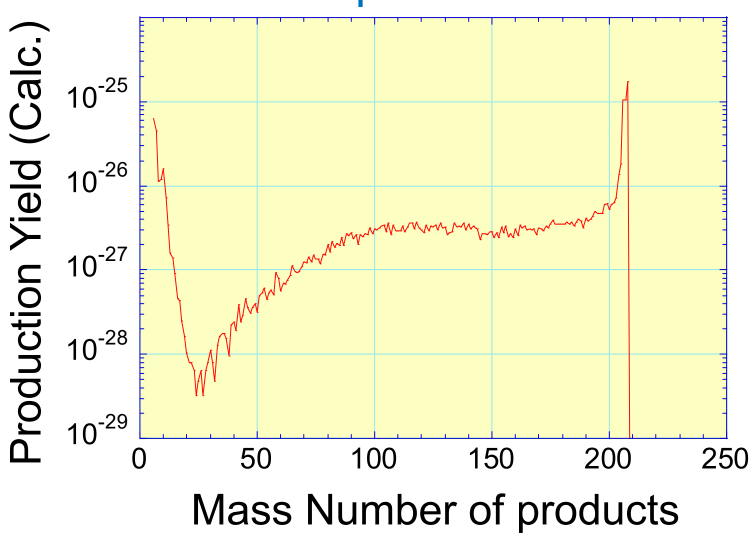
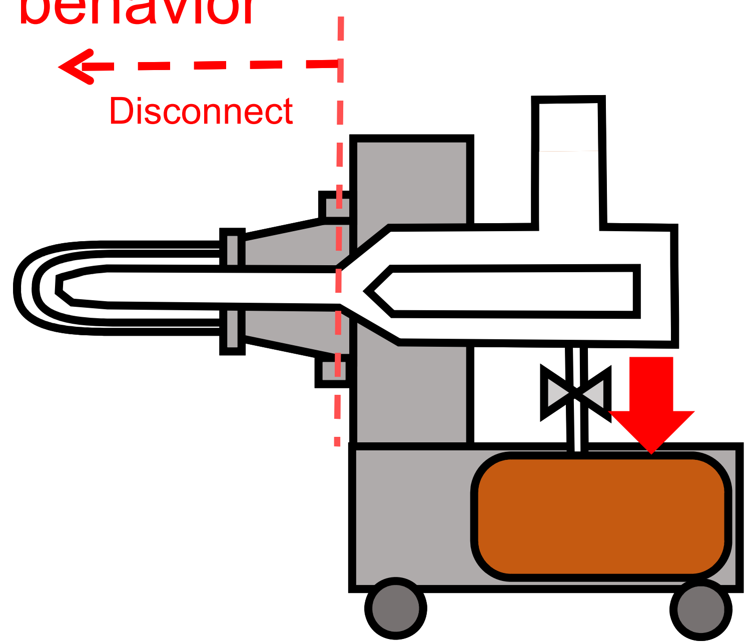
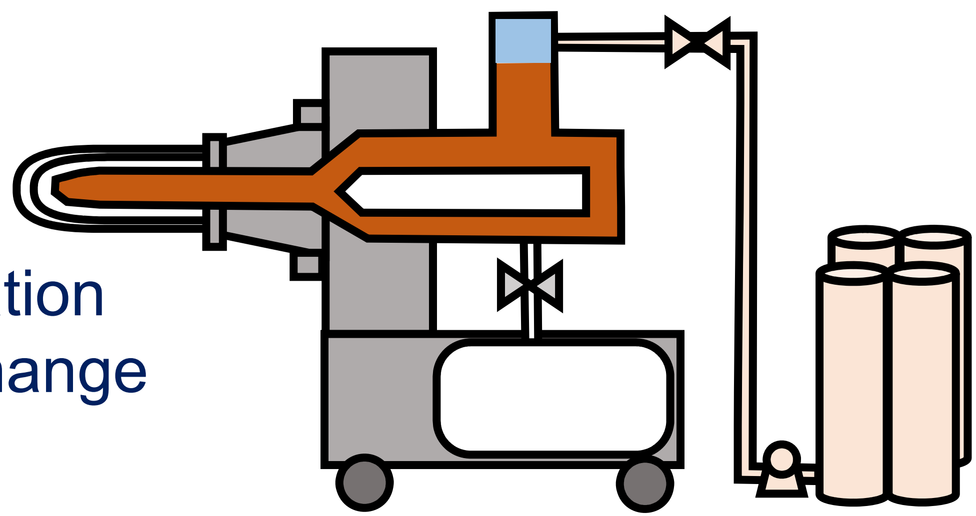




Prediction of the Tritium Behavior

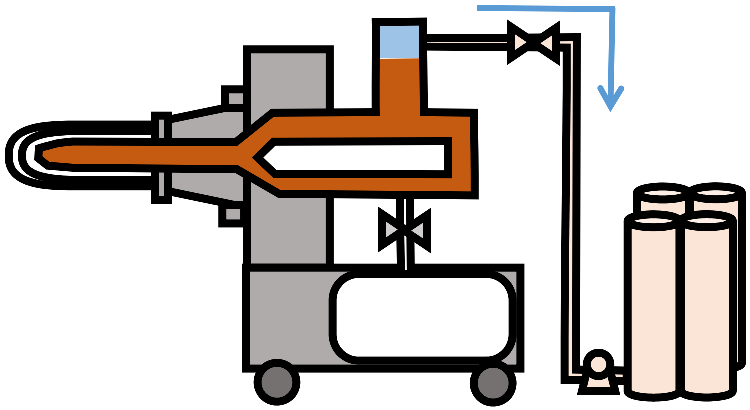
- (1) : Contained in the helium cover gas
  - =>Transfer to the off-gas system
  - =>Release after the treatment
- (2) : Contained in the mercury
  - =>Mercury drain

The predictions were confirmed in the preparation process for the first exchange work (Nov. 2011).

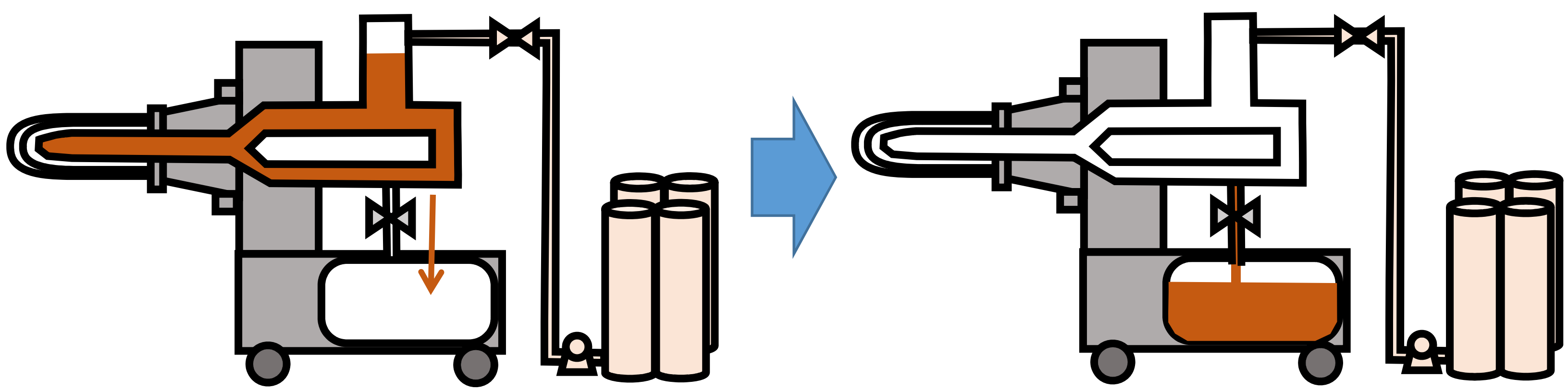


The Preparation Process for the First Exchange Work

(1) Transfer the cover gas to the off-gas system

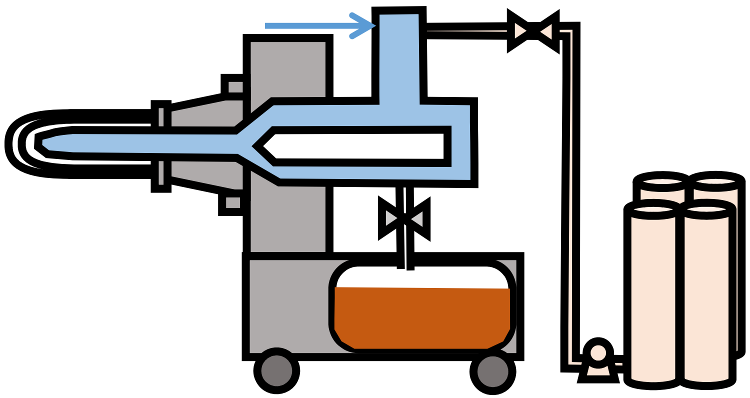


(2) Drain the mercury to the drain tank



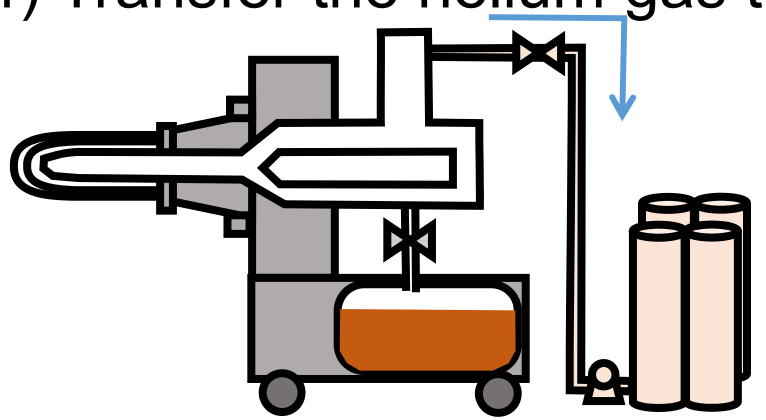
The Preparation Process for the First Exchange Work – Flushing -

(3) Inject helium gas in the circulation system



Repeated Several times

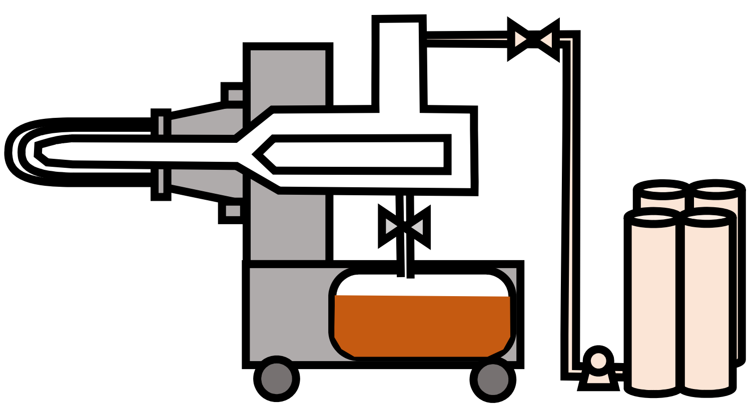
(4) Transfer the helium gas to the off-gas system



We checked the whole amounts of radioactive gases transferred the off-gas system.

Transferred Radioactive Gases

- Noble gas（^{127}Xe）
  - Whole products：~10^{12}Bq
  - The whole of them were transferred.
- Tritium
  - Whole products : ~10^{13}Bq
  - Only ~10^{11}Bq were transferred.
  - The chemical form is all HT.



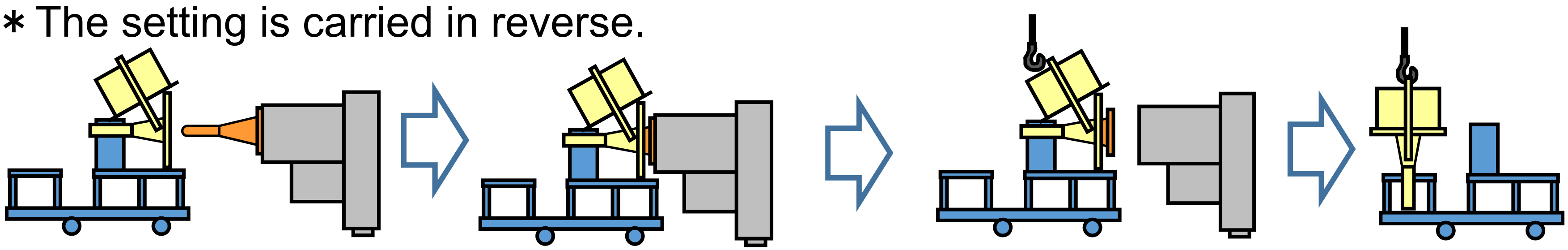
Finally the radioactive gas concentration of the flushing gas decreased sufficiently.

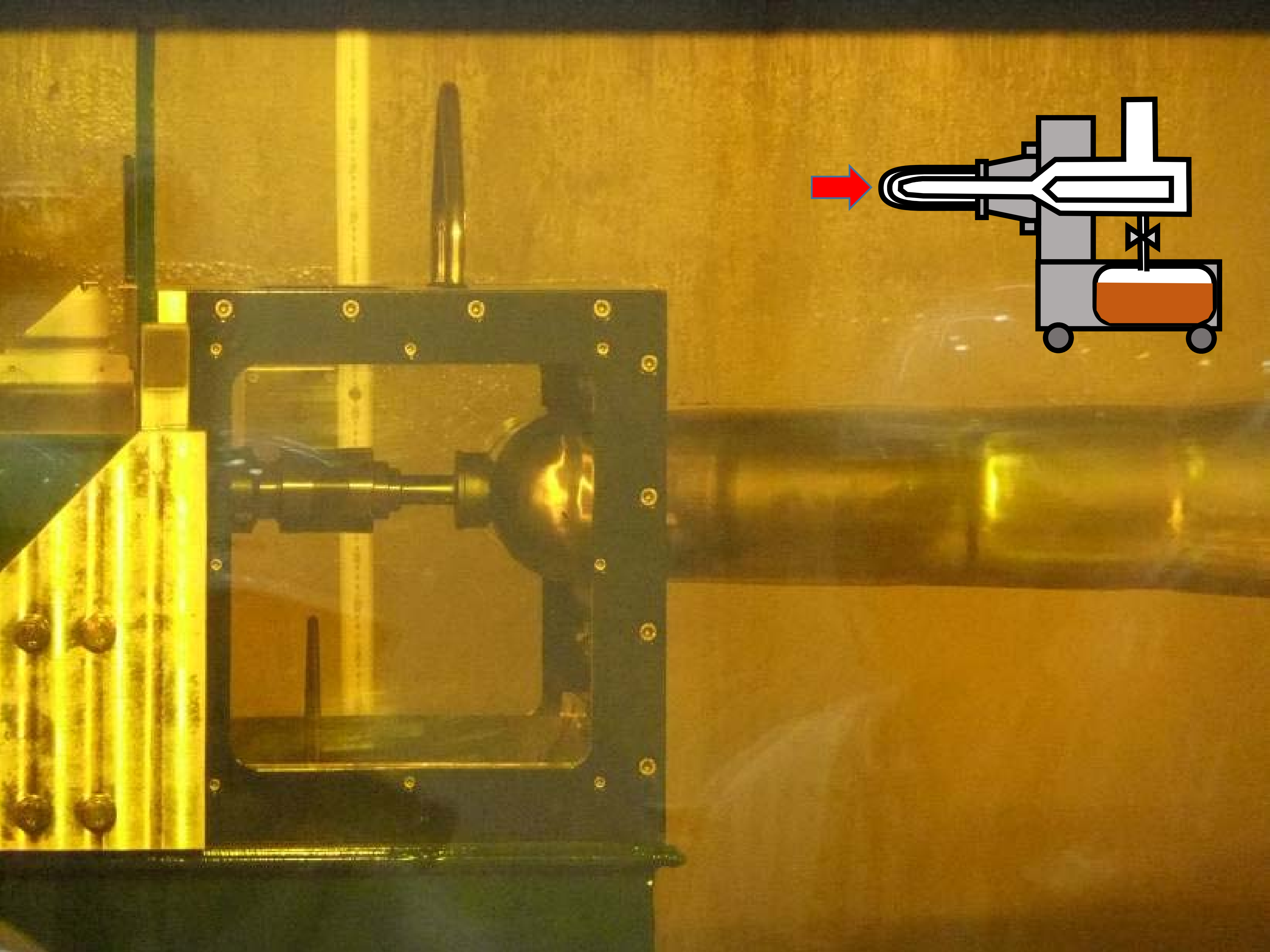
Process of the Exchange Work（Nov. 2011）

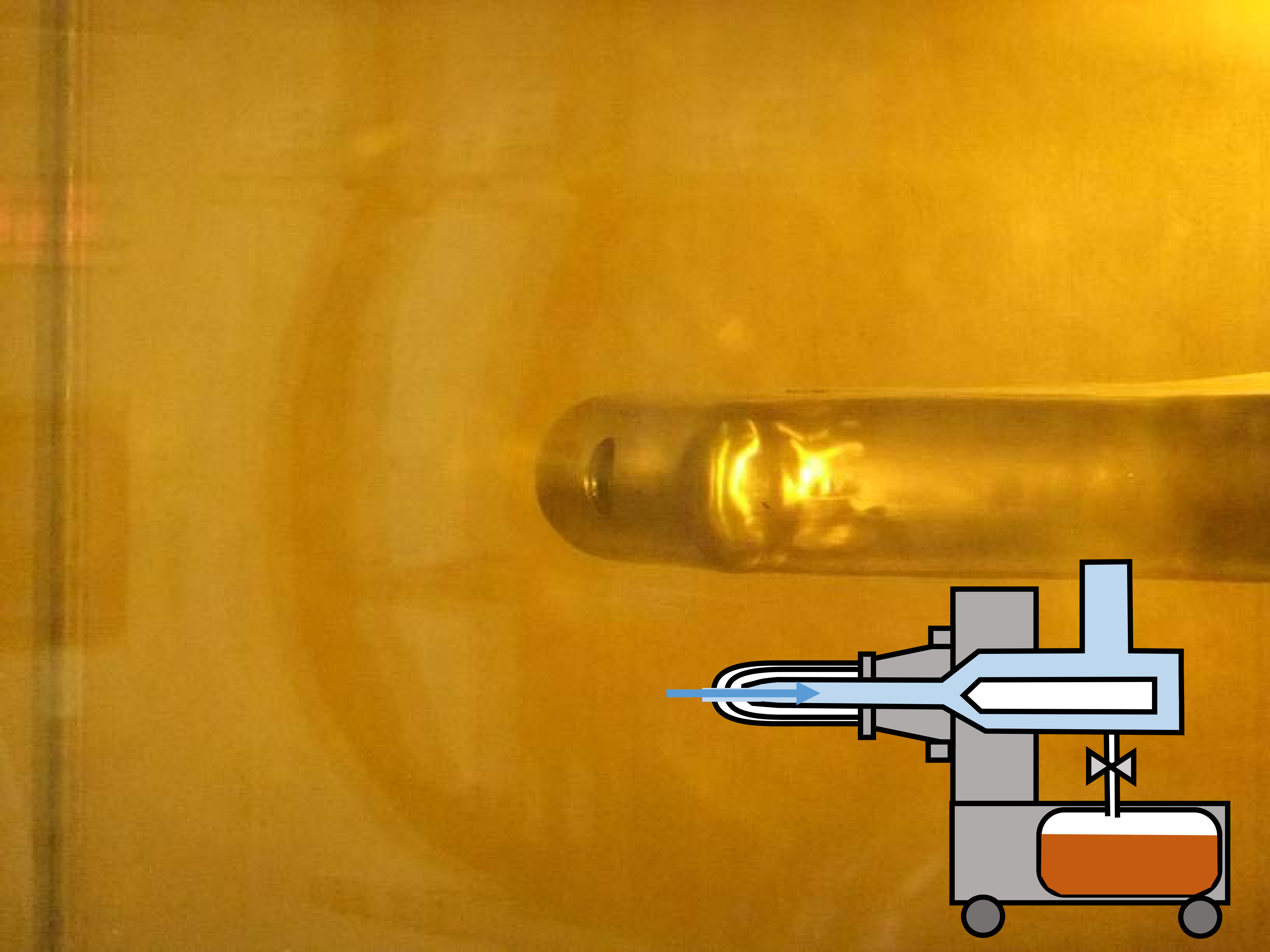
1. Cutting the specimen from the top.
2. Removal of the used target.
3. Setting of the new target.

Using remote-handling system

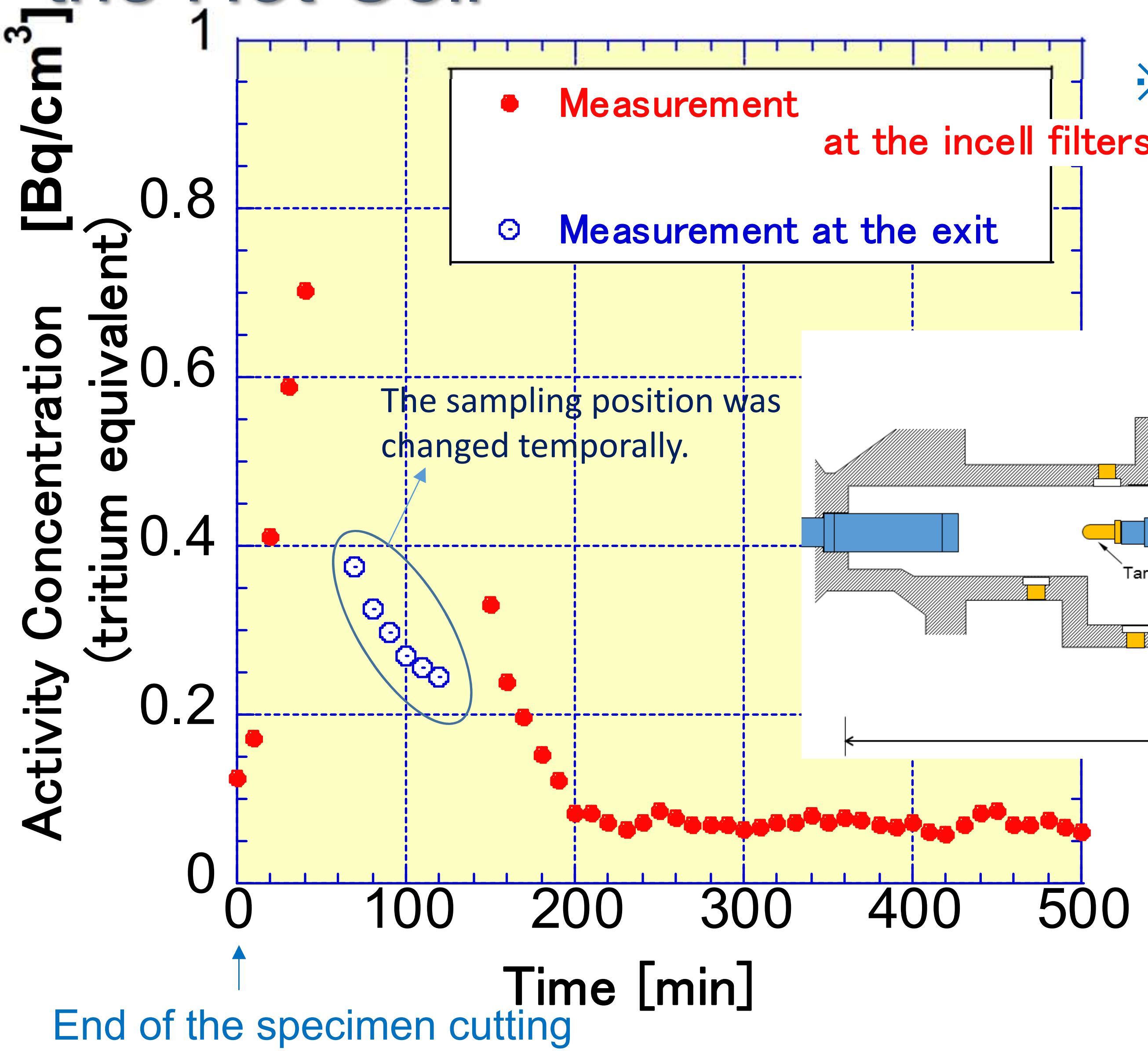
【Removal process of the target vessel】
＊The setting is carried in reverse.

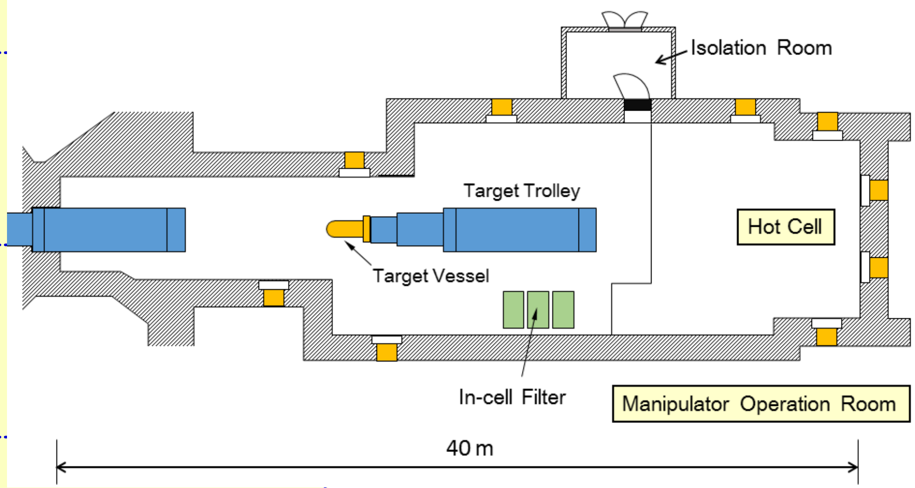






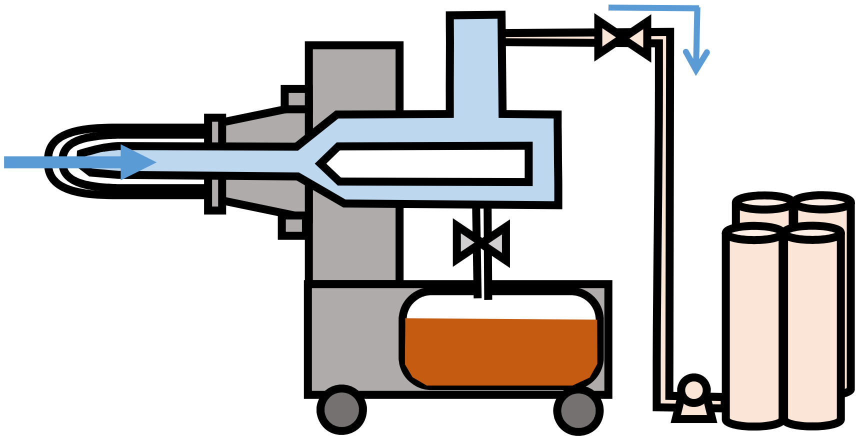
Variation of the Tritium Concentration in the Hot Cell





※ The chemical form of the released tritium was almost HTO.

After the Cutting (before the Exchange Work)



The inside surface of the system behaved as "a Unlimited Tritium Source."

For the system flushing, the gas(air) transfer carried out 6 times.

- 10^{11}Bq of tritium was transferred every time.
- No flushing effect!

We had to take mitigation measures for the tritium release on the premise of 10^{13}Bq of tritium remained in the target vessel and the circulation pipes.

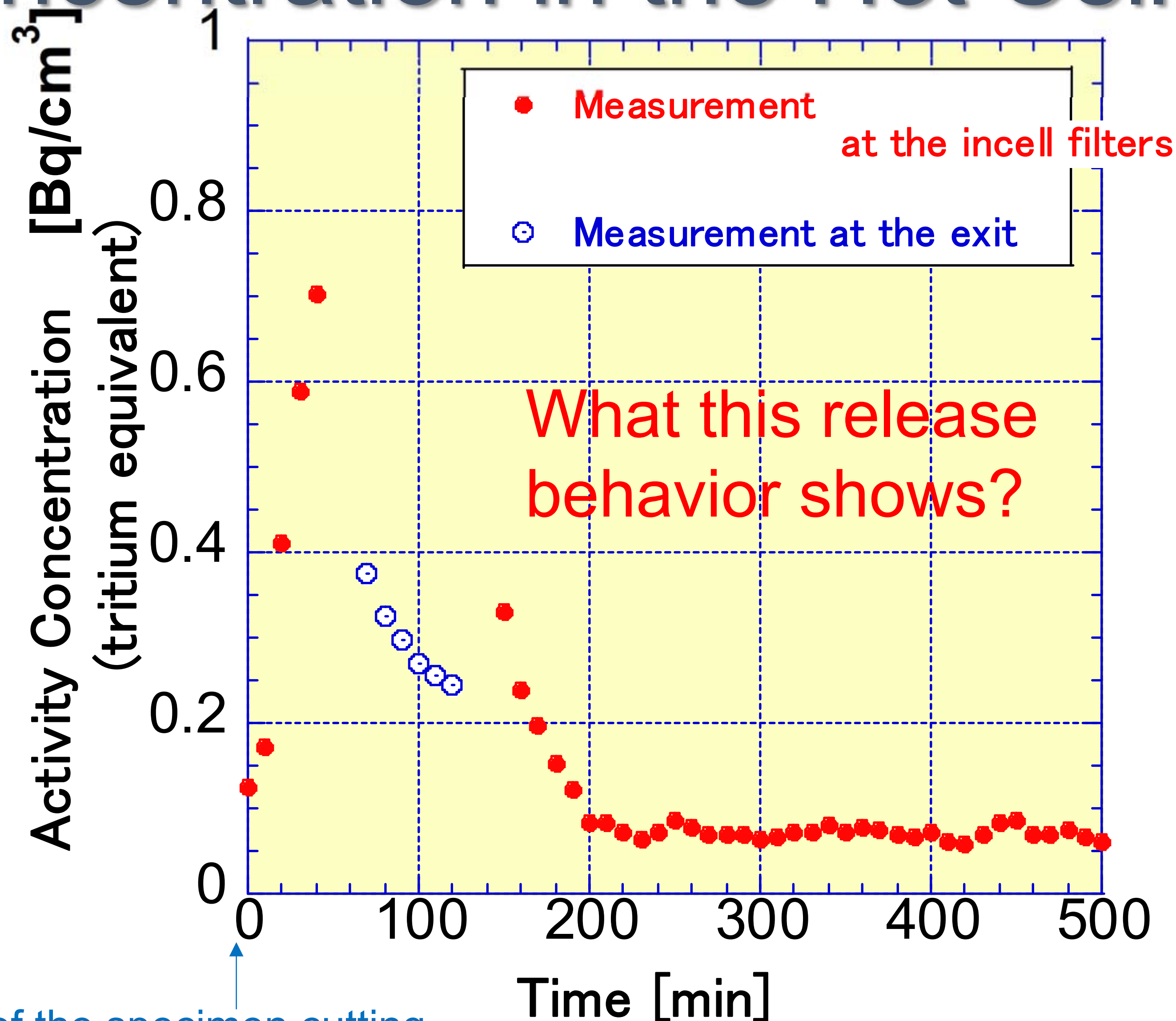
What We Recognize on the Tritium Behavior

- Tritium produced in mercury
  - ~10^{13}Bq tritium produced by March, 2011 by the beam operation.
  - The whole amount were absorbed to the stainless steel of the target vessel and the circulation pipes.
- The small part of the tritium was released as HT in helium-gas environment.
  - Decreased the HT release due to consumption of "H" or "H_2." in the system?
- In air environment, HTO was released.
  - The tritium was absorbed as hydrogen, and released via isotope exchange with water molecular.
  - $T + H_2O \rightarrow H + HTO$

- Since then and up to now, the HT-release condition has not been appeared even if we replaced air by helium gas in the circulation system.
- The inner surface may be fully contaminated by water?

Variation of the Tritium Concentration in the Hot Cell

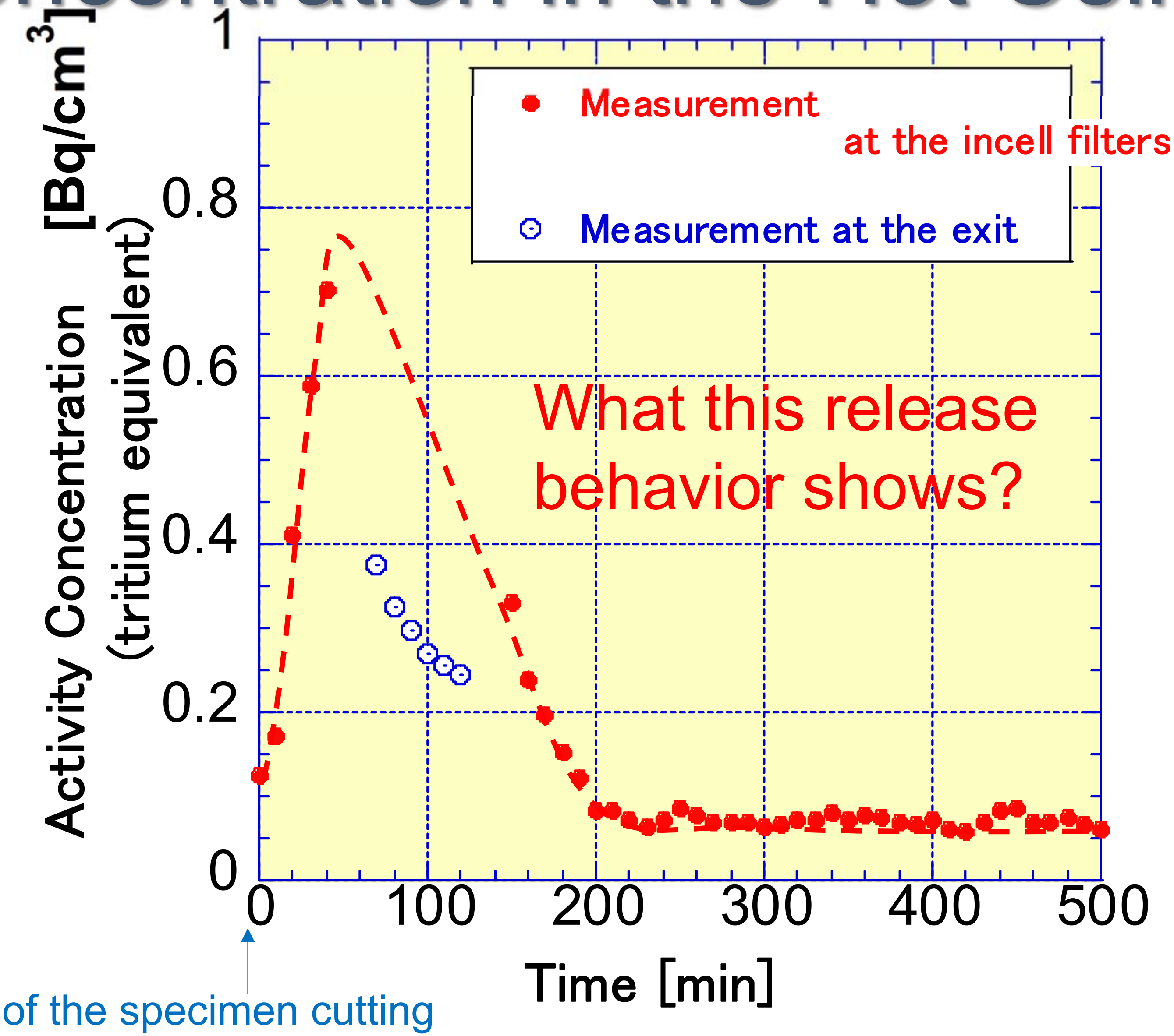
Again!



End of the specimen cutting

Variation of the Tritium Concentration in the Hot Cell

Again!



Gas Release by Diffusion from a Solid Surface

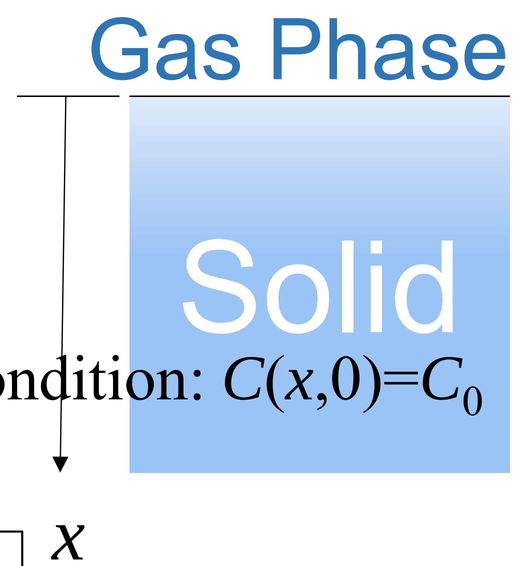
$$\frac{\partial C(x,t)}{\partial t} = D\frac{\partial^2 C(x,t)}{\partial x^2}$$

$C(x, t)$: Tritium concentration in a solid

D: Diffusion coefficient of tritium in a solid.

Boundary Condition

$C(0,t) = 0$



Initial Condition: $C(x,0)=C_0$

$C(\infty,t) = C_0$

$$\text{Release Rate} = C_0\sqrt{\frac{D}{\pi t}}$$

per unit time and per unit surface area at a solid surface

$$\frac{1}{m^2 s} = \frac{1}{m^3}\sqrt{\frac{m^2/s}{s}}$$

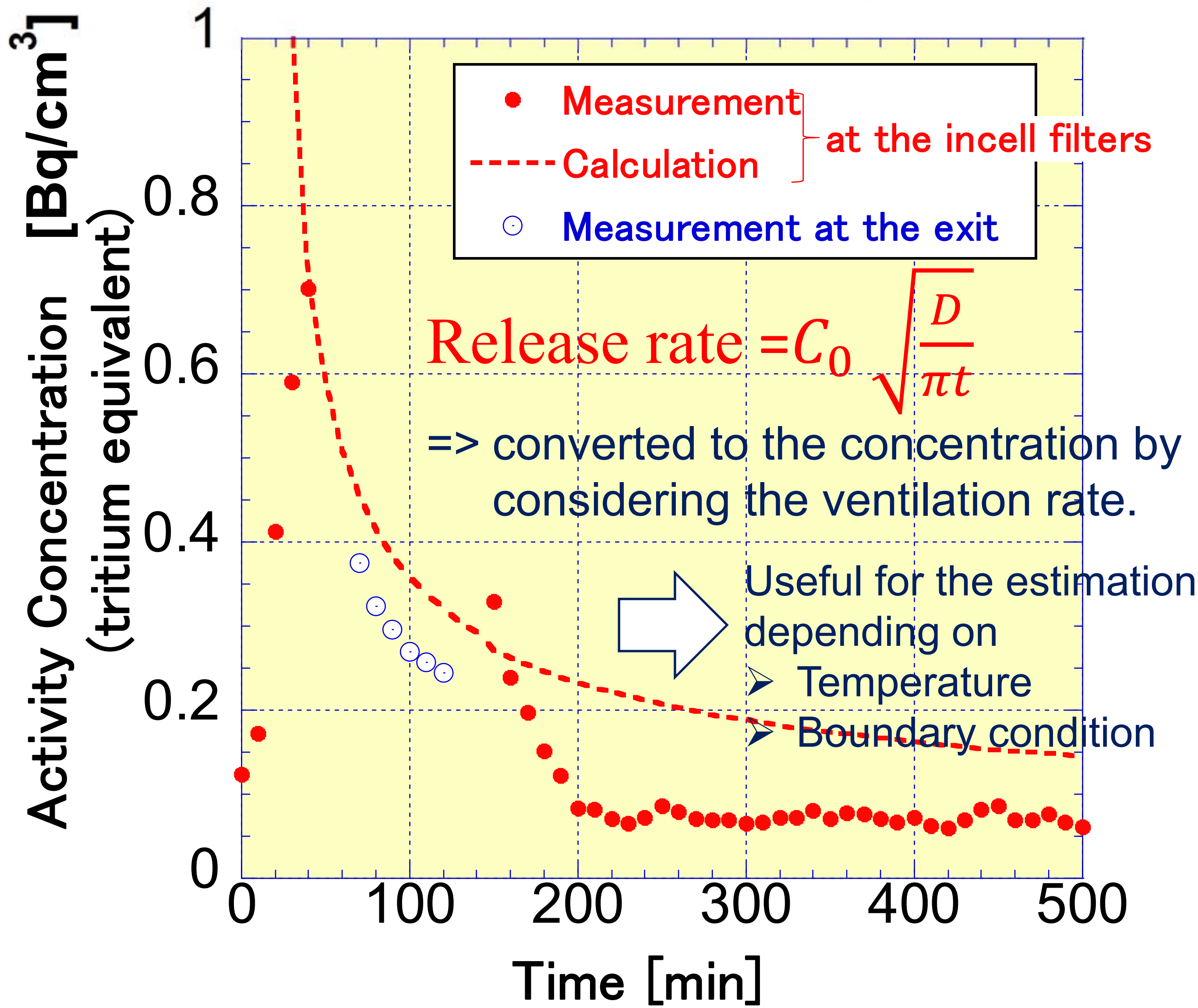
Estimation of the Diffusion Coefficient: *D* and the Concentration: *C*

- Diffusion coefficient of tritium in stainless steel : D
  - $$D[\mathrm{m^2/s}] = 5.9 \times 10^{-7} \exp\left(-\frac{51.9\ \mathrm{kJ{\cdot}mol^{-1}}}{RT}\right)$$
  - $D = 6\times10^{-16}$ m²/s (T=300 K)

Large deviations, by more than a factor of 10, among references

- Diffusion length: $\sigma = \sqrt{2Dt}$
  - t =3 years =9.5×10^{7} second
  - $\sigma = 3.5 \times 10^{-4}\mathrm{m} = 350\ \mu\mathrm{m} \rightarrow \ 2\sigma = 700\ \mu\mathrm{m}$
- Tritium concentration in stainless steel: C_0
  - Effective area：~20 m^2
  - C_0=7×10^{8} Bq/cm³ by supposing that 1×10^{13} Bq of tritium spread to the depth of 2σ.

Tritium Concentration【Analytical Result】



Summary and Unsolved Issues

- Tritium produced in the mercury is almost absorbed with the stainless steel
- After the mercury was removed, the absorbed tritium is released from the stainless surface by gas diffusion process in solid.
  - The chemical form of the released tritium depends largely on the environment covering the surface.

The issue is

- What is the mechanism of the mercury absorption with the stainless steel