

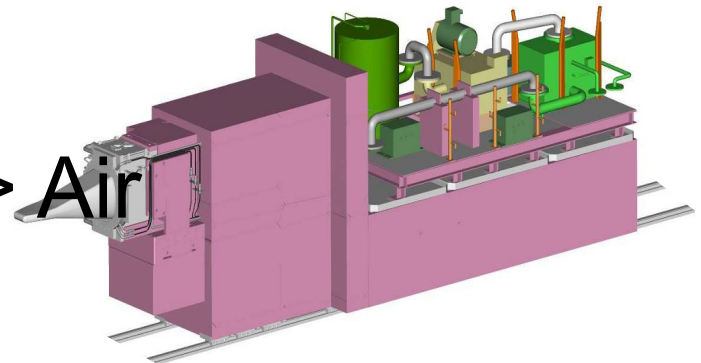
# Simplified Model for Simulating Tritium Behavior in the Spallation Neutron Target System

Yoshimi Kasugai

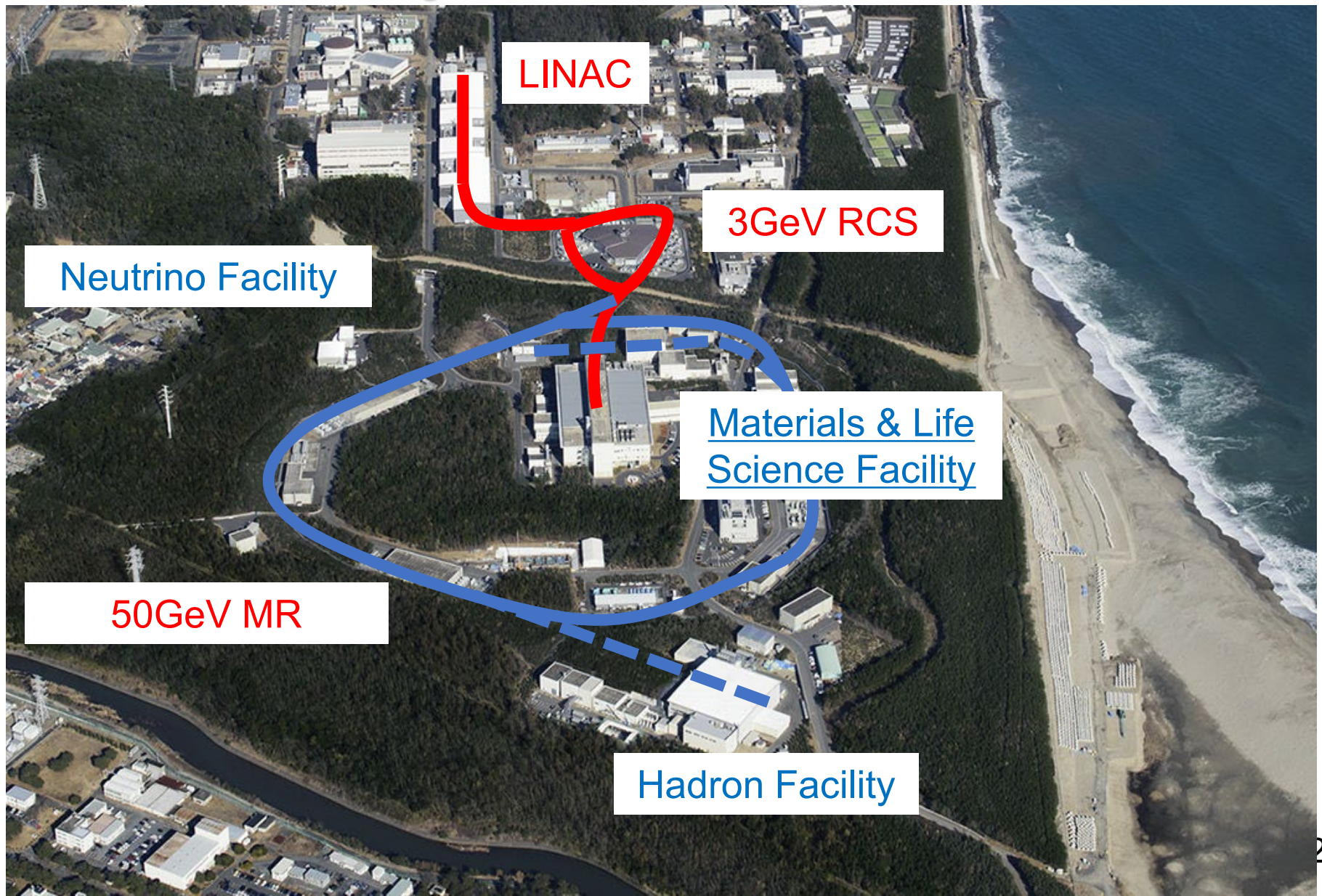
J-PARC (JAEA)

# Introduction

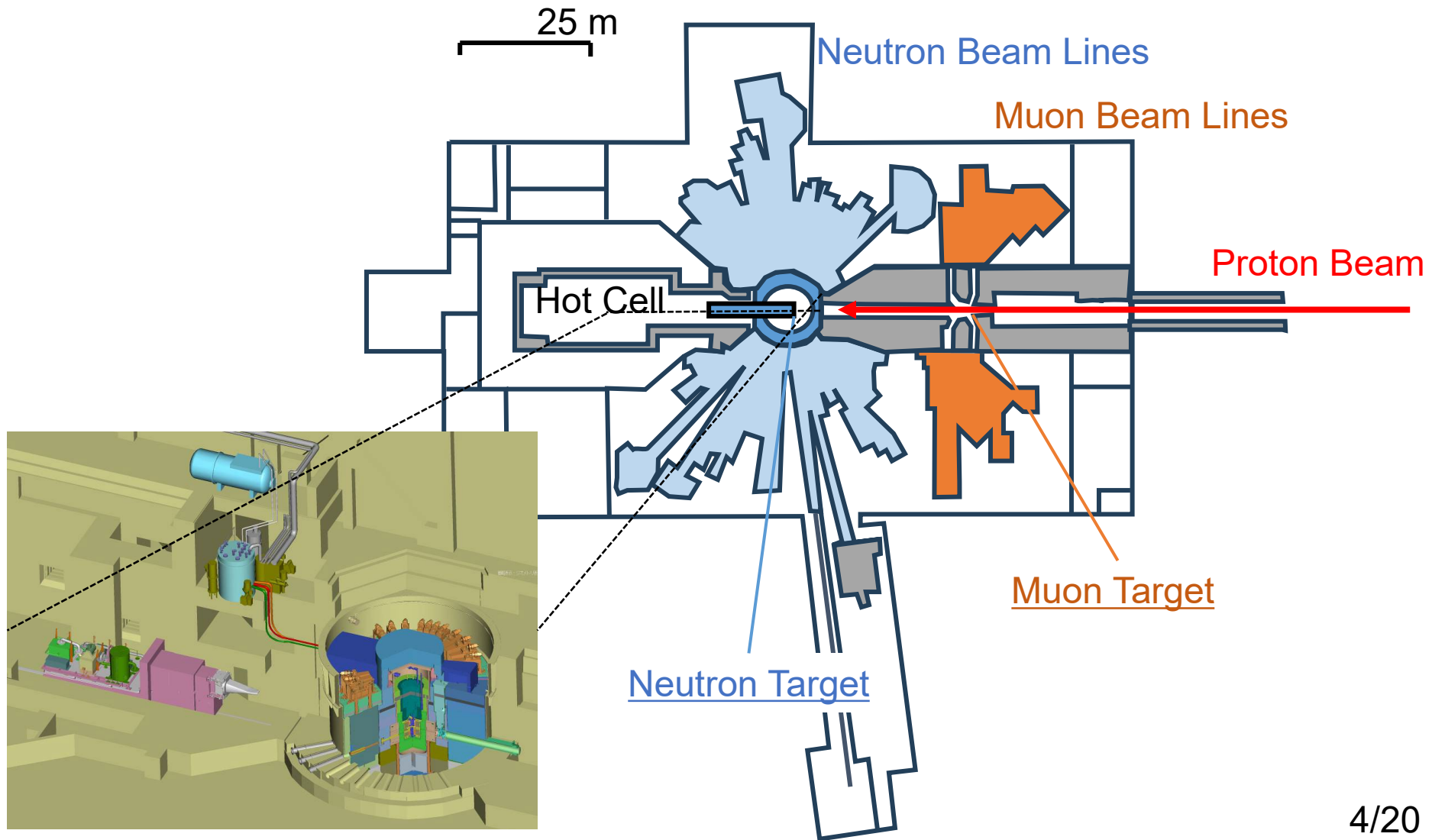
- Spallation Neutron Source using mercury
  - Materials and Life Science Experimental Facility (MLF) of J-PARC
- Mercury Target Vessel
  - Stainless Steel (SS)
  - Radiation and Pitting Damage
  - Periodical exchange work => Tritium release
- Behavior of tritium
  - Mercury => Stainless => Air

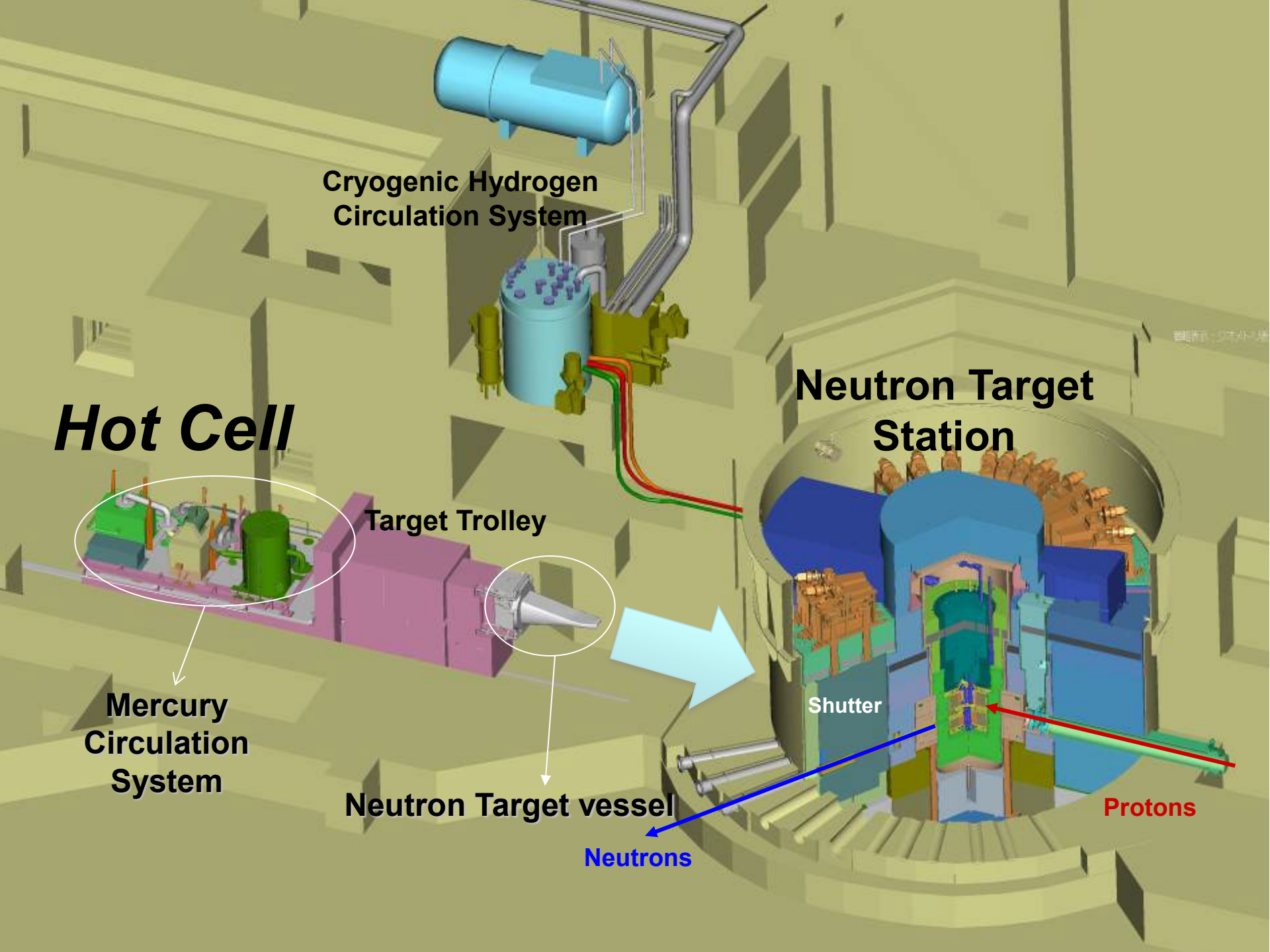


# Layout of J-PARC



# Materials & Life Science Experimental Facility (MLF)





**Cryogenic Hydrogen  
Circulation System**

**Hot Cell**

**Neutron Target  
Station**

**Target Trolley**

**Mercury  
Circulation  
System**

**Neutron Target vessel**

**Shutter**

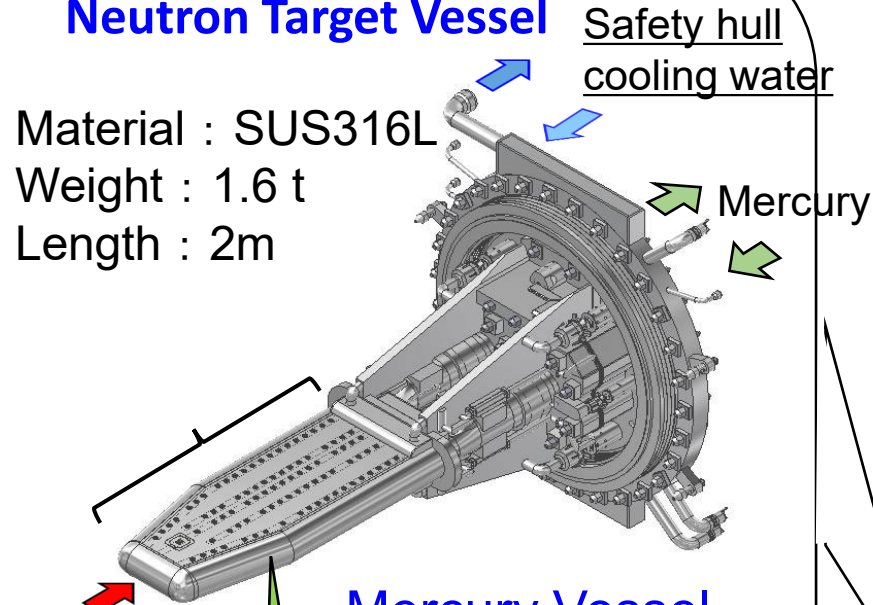
**Neutrons**

**Protons**

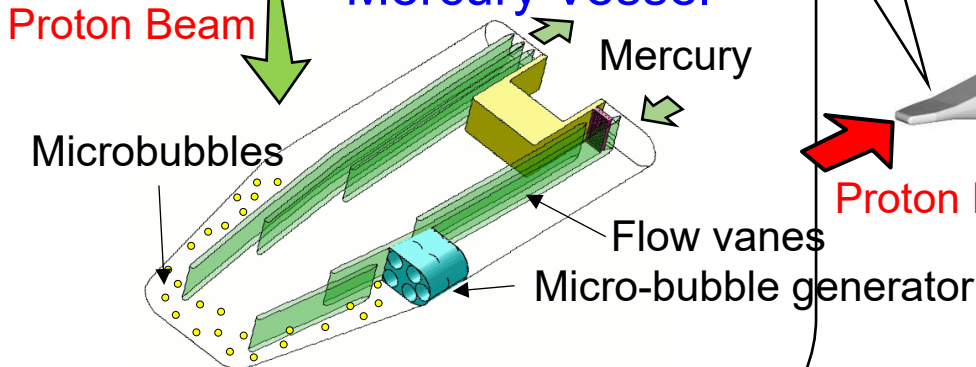
# Outline of the Spallation Neutron Target

## Neutron Target Vessel

Material : SUS316L  
Weight : 1.6 t  
Length : 2m



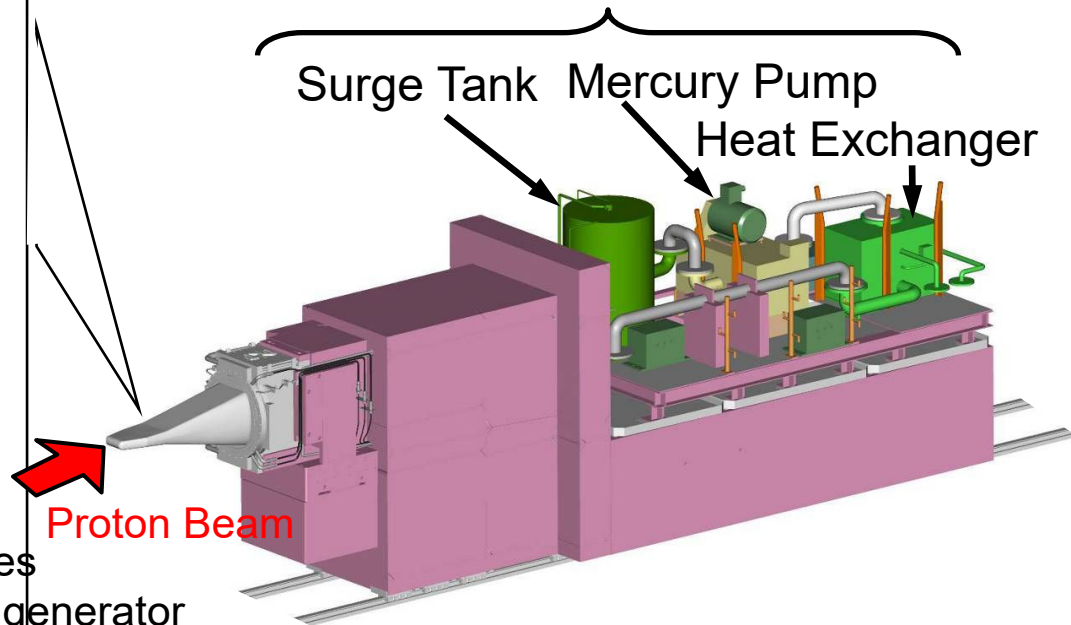
## Mercury Vessel



\*Mitigation of cavitation damage by microbubbles

|                     |                      |
|---------------------|----------------------|
| Length :            | 12 m                 |
| Weight :            | 315 ton              |
| Mercury Volume :    | 1.5 m <sup>3</sup>   |
| Mercury Flow rate : | 41m <sup>3</sup> /hr |

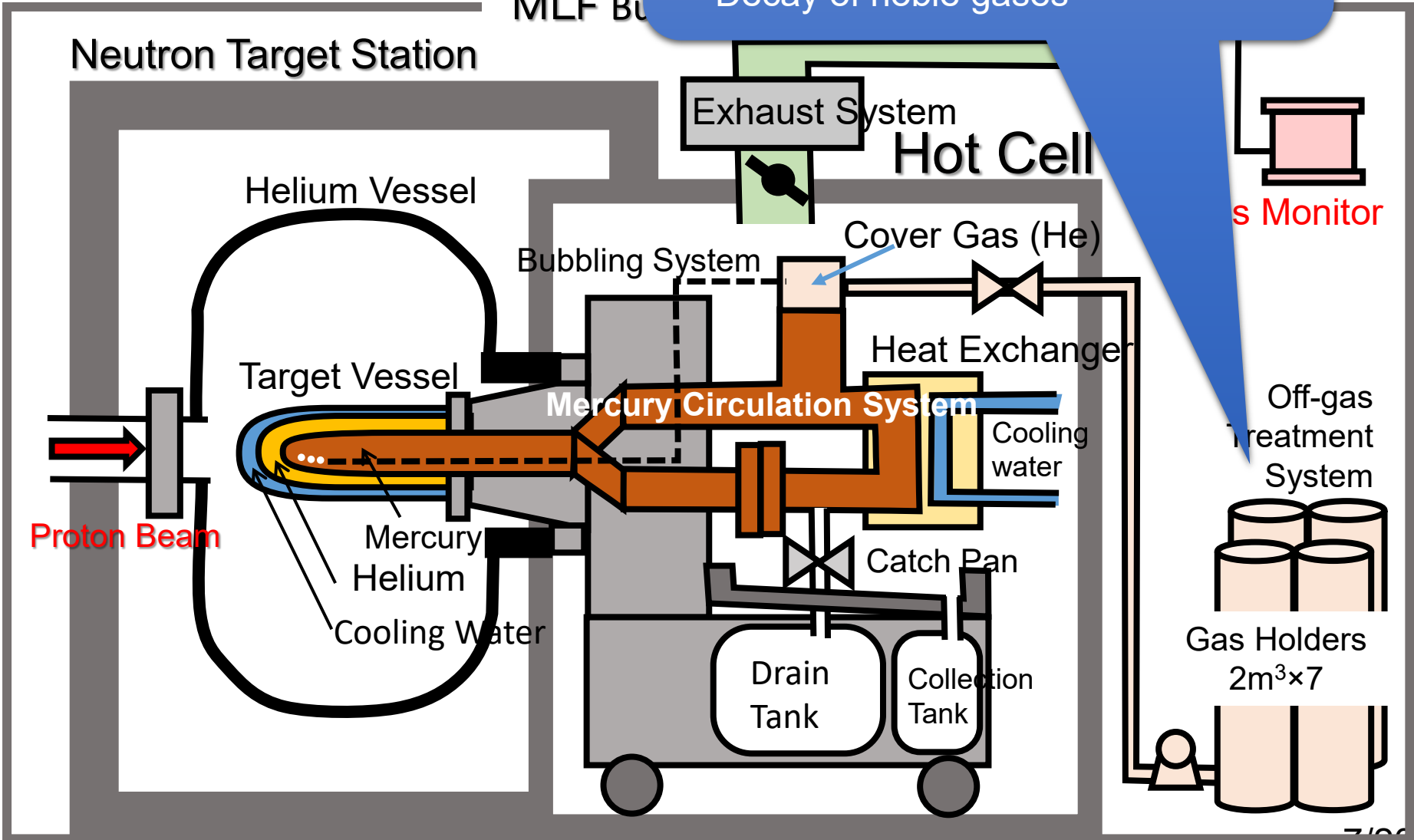
## Mercury Circulation System



# Concept of the R Confinement

## 【Off-gas treatment System】

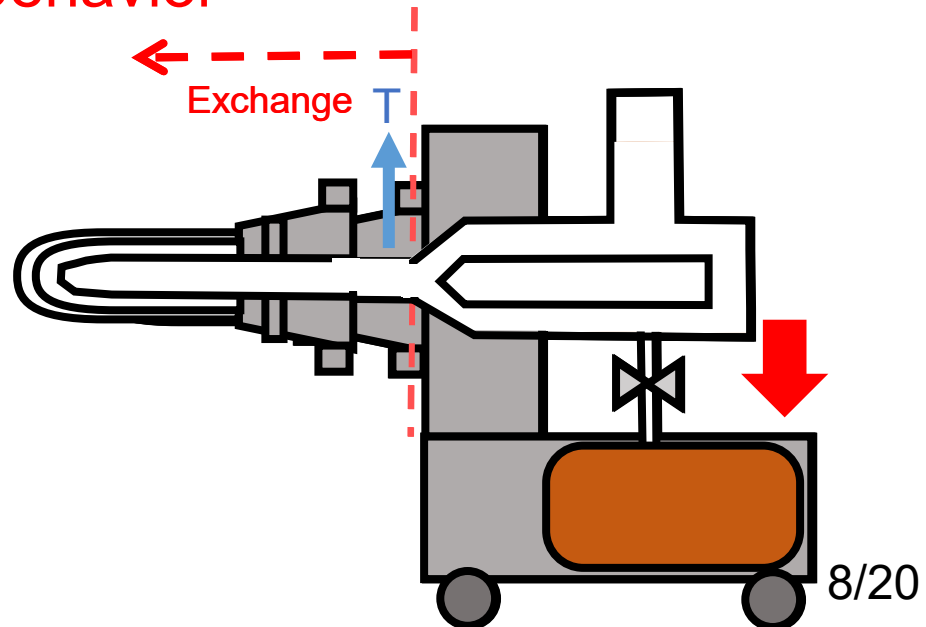
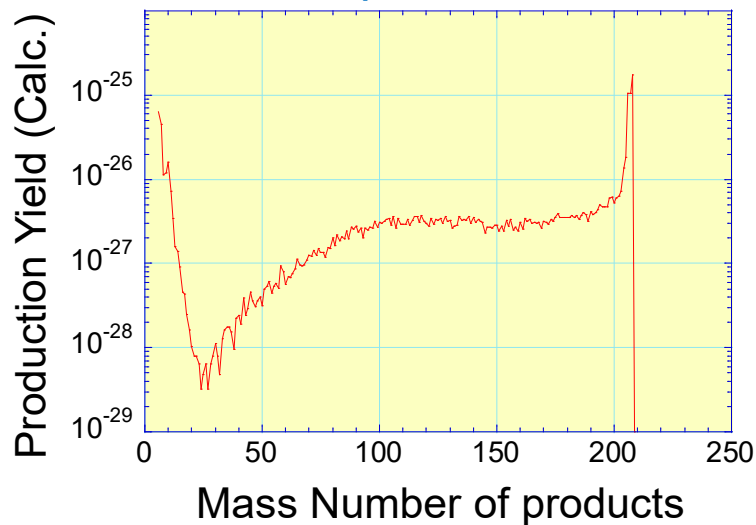
- Removal of mercury
- Removal of tritium
- Decay of noble gases



# Why We need to Understand the Tritium Behavior?

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

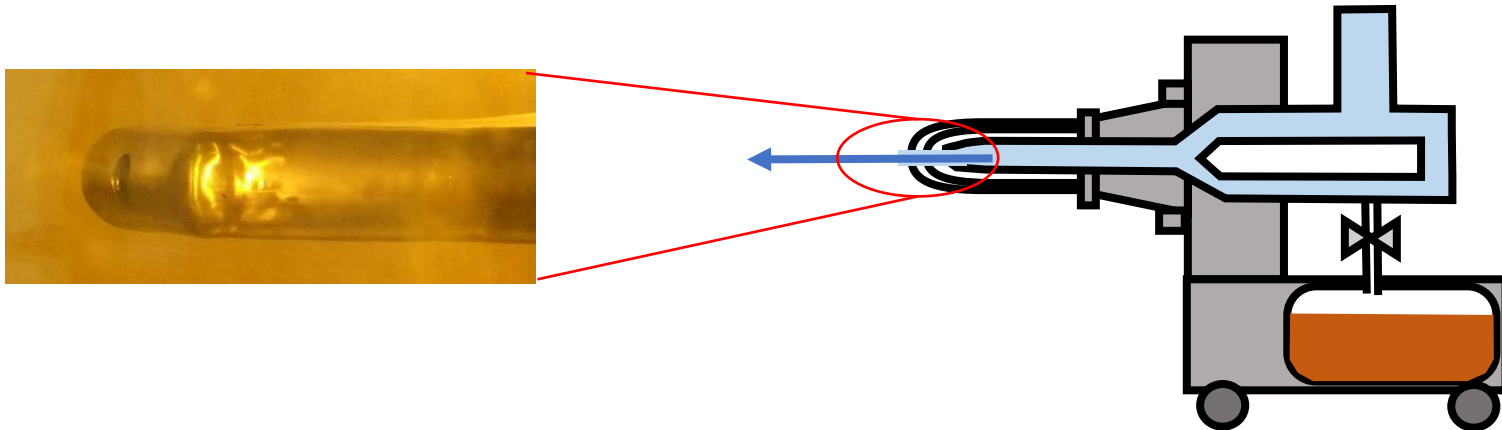
Mass Yield for spallation reactions





# First observation of tritium release

- Tritium release from the target vessel was observed on the specimen cut on 2011 for the first time.
  - Presented on the previous J-PARC Symposium in 2019.
- It was shown that the release behavior could be explained as diffusion from the stainless steel.
  - Comparing the observed tritium concentration to the simplified physical model in which an exact solution of a one-dimensional diffusion equation can be applied.
- The chemical form of the released tritium was HTO
  - Isotope Exchange:  $T + H_2O \rightarrow H + HTO$

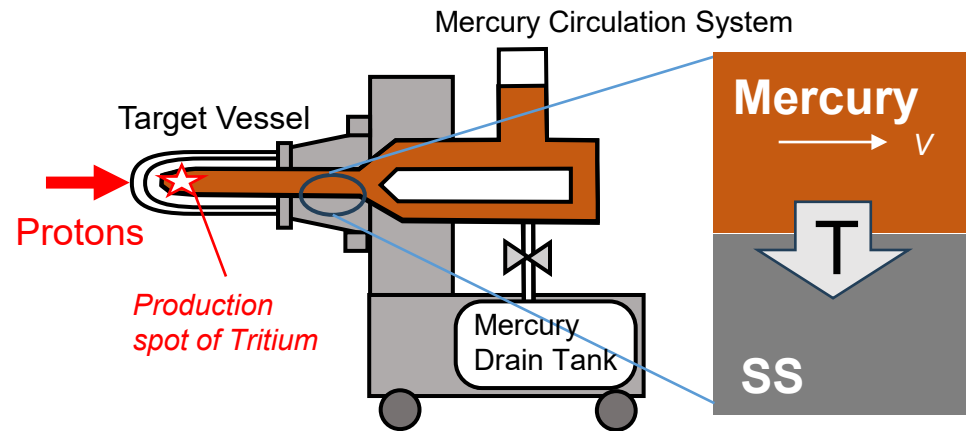


# Outline of this work

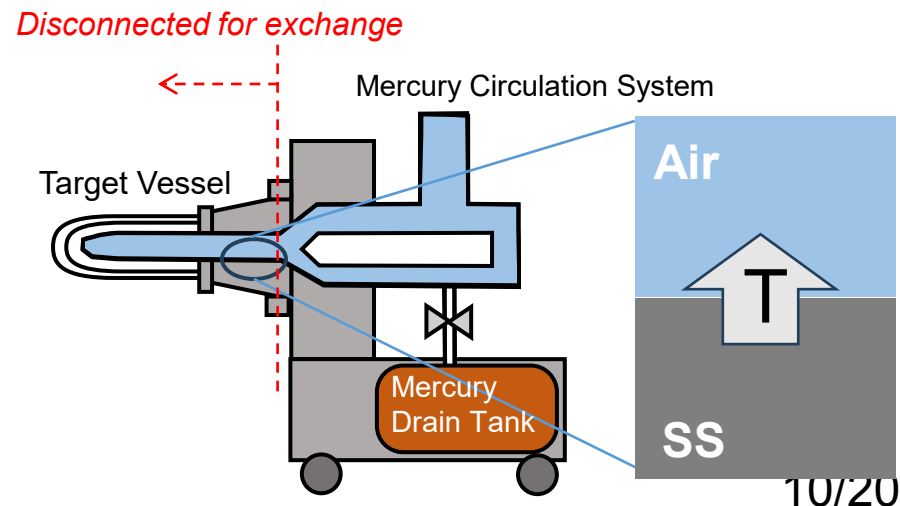
- In this work, **a new physical model** has been **developed** to evaluate the **averaged behavior of tritium** by assuming the whole surface between mercury and the stainless steel as **one physical system**.
- The evaluations are performed through **whole process from (1)Absorption to (2)Release** by solving the one-dimensional diffusion equation **numerically**.

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

## (1) Absorption Process (Beam-Operation Period)



## (2) Release Process (Maintenance Period)



# (1) Absorption Process

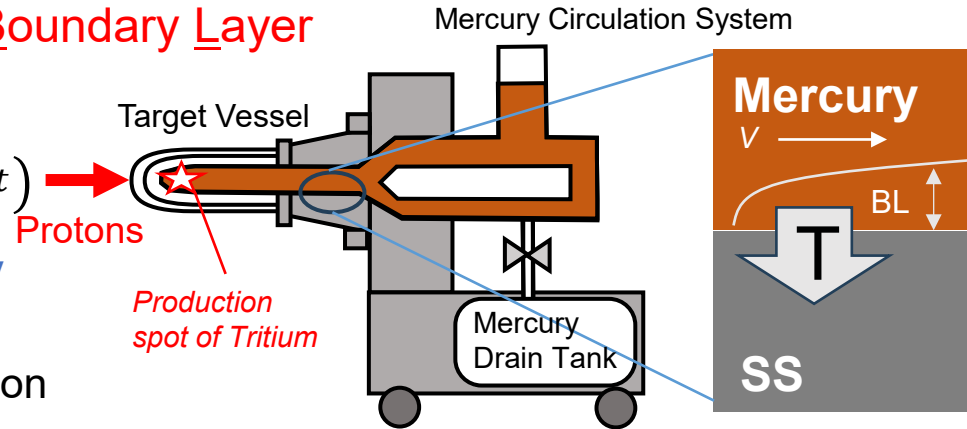
• Tritium concentration in mercury is calculated supposing as

- Homogeneous distribution in mercury
- Gradient of the concentration in the Boundary Layer

$$V \frac{dC_T(t)}{dt} = a_T - \alpha S C_T(t)$$

$$\rightarrow C_T(t) = \frac{a_T}{\alpha S} \left( 1 - e^{-\frac{\alpha S}{V} t} \right)$$

- $C_T(t)$ : Tritium concentration in mercury
- $a_T$ : Production rate of tritium
- $t$ : Time from start of the beam operation
- $V$ : Volume of mercury (=1.5 m<sup>3</sup>),
- $S$ : Effective area of SS (=10 m<sup>2</sup>)
- $\alpha$ : Mass transfer coefficients of tritium from mercury to SS



$$\frac{\ln 2}{\left(\frac{\alpha S}{V}\right)} \sim 2,000 \text{ sec}$$

• Tritium diffuses into SS in accordance with the diffusion equation

- Diffusion coefficients:  $D = 1.2 \times 10^{-10} \text{ cm}^2/\text{s} (80 \text{ }^\circ\text{C})$
- Duration of the beam operation: 5,000 hours (~208 days)

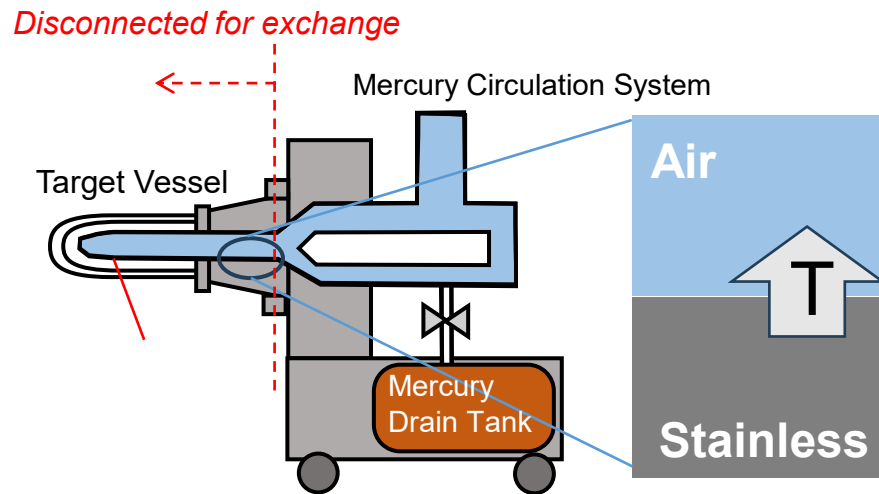
$$\frac{\partial C(x, t)}{\partial t} = D \frac{\partial^2 C(x, t)}{\partial x^2}$$



$$\frac{C(x, t + \Delta t) - C(x, t)}{\Delta t} = D \frac{C(x + \Delta x, t) - 2C(x, t) + C(x - \Delta x, t)}{\Delta x^2}$$

## (2) Release Process

- Using **the depth distribution at the end of the beam operation in SS** as initial condition.
- The tritium release rate is calculated by assuming that tritium which reaches at the SS surface is released to air.
- $D=6.7 \times 10^{-12} \text{ cm}^2/\text{s}$  (30 °C)



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

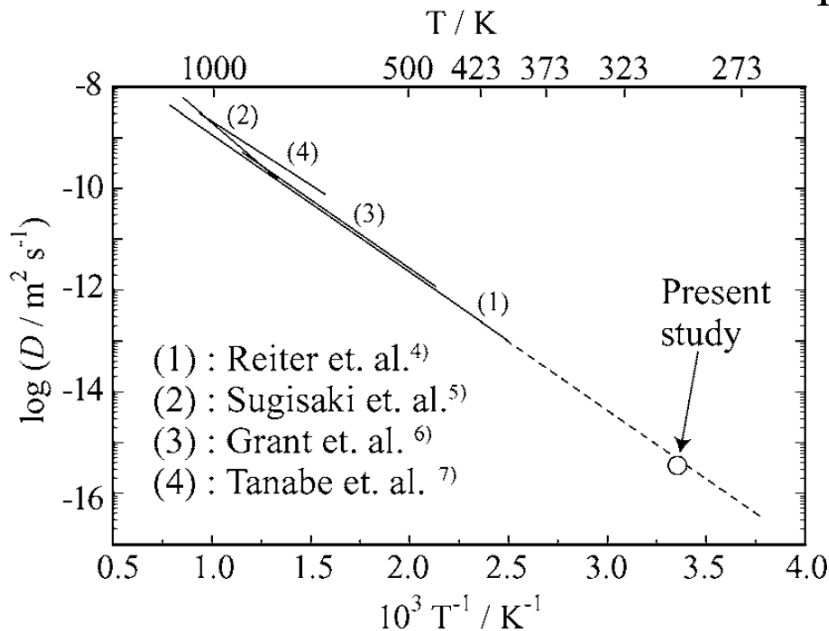
$$\Rightarrow \frac{C(x, t + \Delta t) - C(x, t)}{\Delta t} = D \frac{C(x + \Delta x, t) - 2C(x, t) + C(x - \Delta x, t)}{\Delta x^2}$$

# Evaluation of Diffusion Coefficient (Tritium in Stainless Steel)

F. Reiter, K. S. Forcey and G. Gervasini: EUR15217 EN (1993)

$$D [\text{m}^2/\text{s}] = 5.9 \times 10^{-7} e^{-\frac{51.9 \times 10^3}{RT}}$$

$T$ : Temperature[K],  $R$ : Gas Constant(=8.314 J/K)



Y. Torikai et al., "Chronic Release of Tritium from Stainless Steel 316", Proceeding of 47<sup>th</sup> Annual Symposium of the Vacuum Society of Japan (2007)



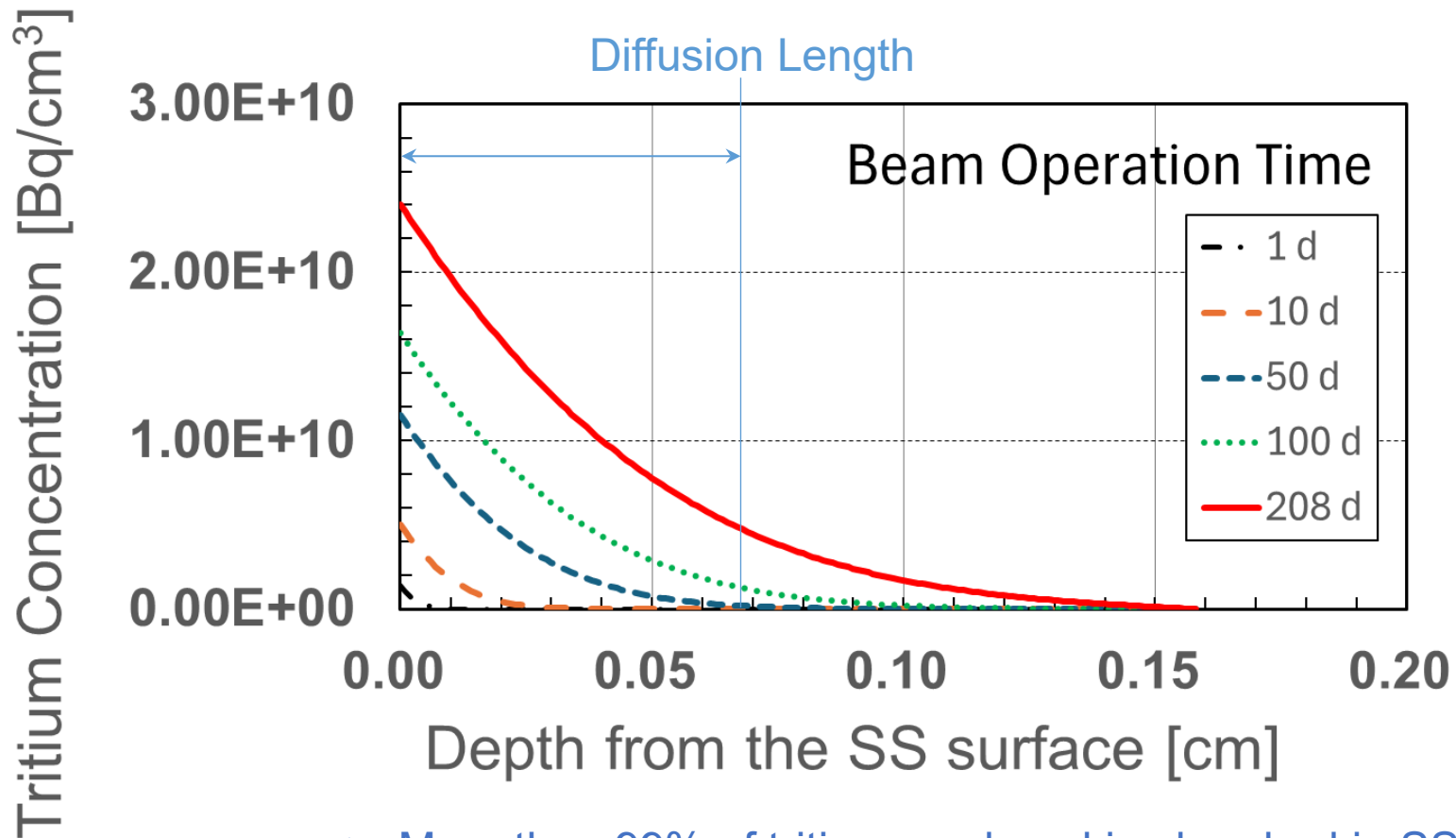
| Period         | Temperature      | D [cm <sup>2</sup> /s] |
|----------------|------------------|------------------------|
| Beam Operation | 80 °C<br>(353 K) | 1.2×10 <sup>-10</sup>  |
| Maintenance    | 30 °C<br>(303 K) | 6.7×10 <sup>-12</sup>  |

**Fig. 5** Comparison of the diffusion coefficients appeared in the literatures with that determined from the chronic tritium release rates at 298 K for series C (○).

# Result (1) Absorption Process

Depth distribution of concentration

$$\text{Diffusion Length} = \sqrt{2Dt} = \sqrt{2 \times 1.23 \times 10^{-10} \text{ cm}^2/\text{s} \times 5000 \text{ h}} = 0.067 \text{ cm}$$

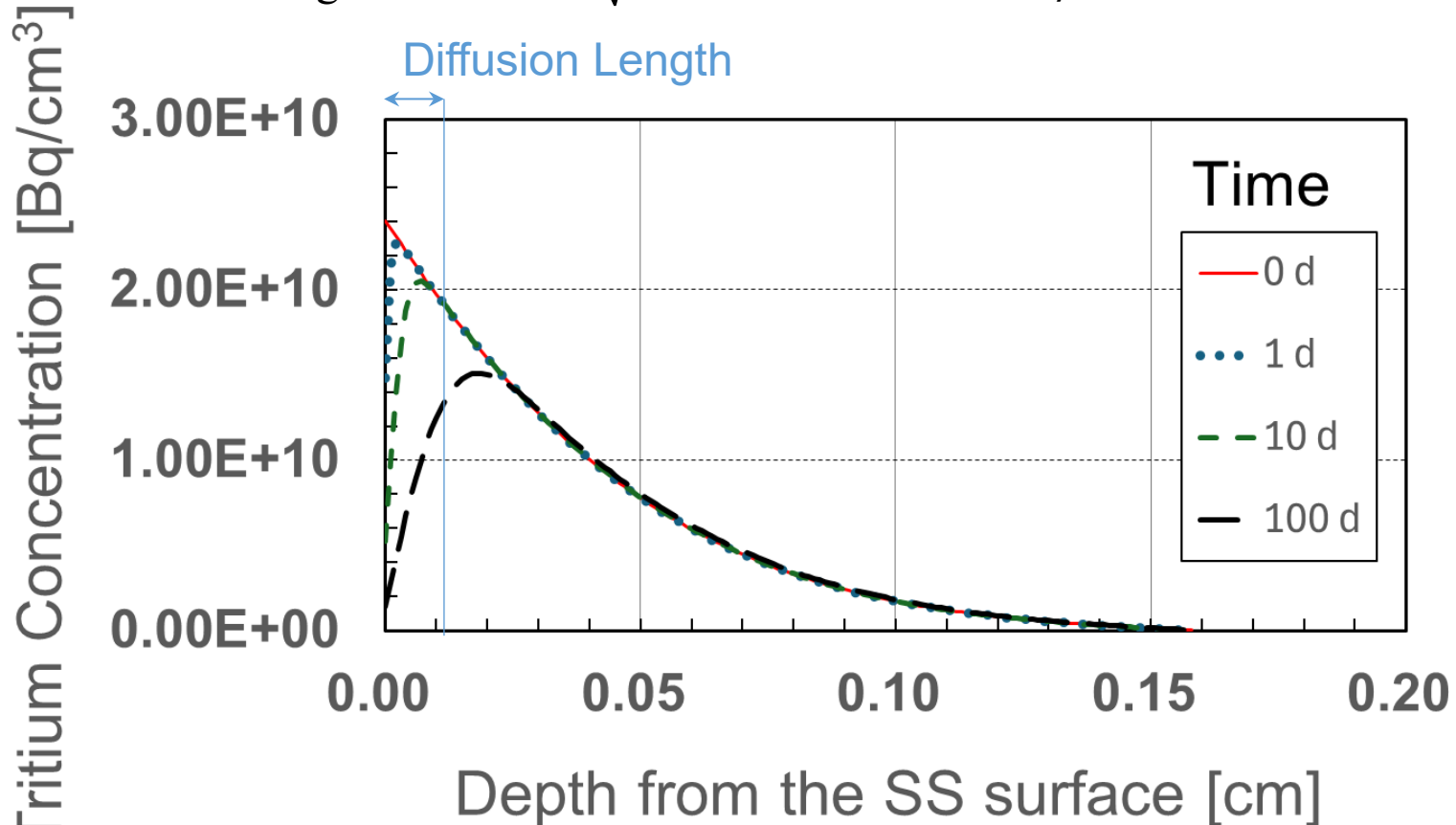


=> More than 99% of tritium produced is absorbed in SS.

# Result (2)-1 Release Process

Depth distribution of concentration

$$\text{Diffusion Length} = \sqrt{2Dt} = \sqrt{2 \times 6.66 \times 10^{-12} \text{ cm}^2/\text{s} \times 100 \text{ d}} = 0.011 \text{ cm}$$

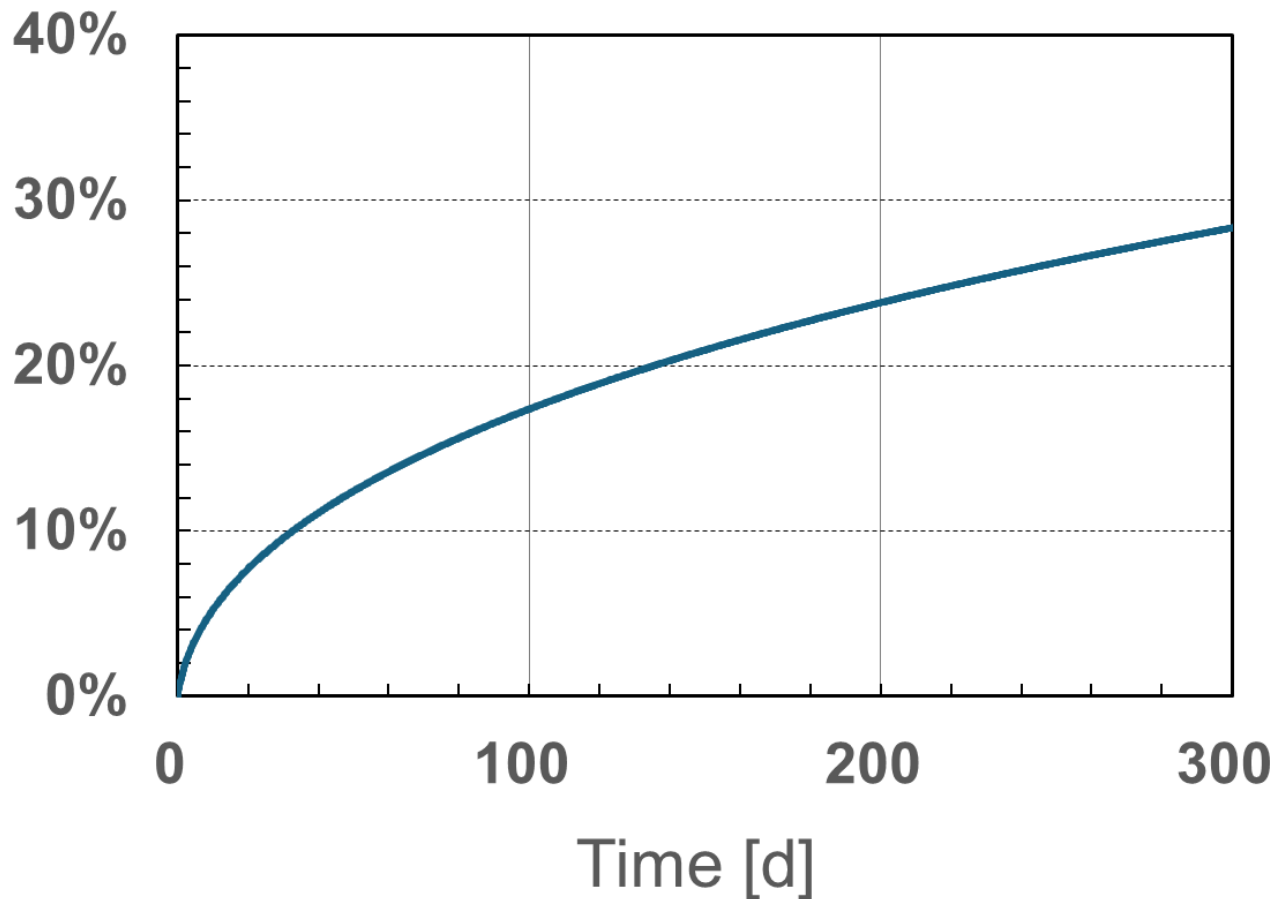


=> Due to difference of the diffusion coefficients between the both processes, coming from difference of the temperatures, only a part of the whole amounts of absorbed tritium can release.

# Result (2)-2 Release Process

Ratio of Tritium release to the total absorption

Ratio of accumulated tritium release to the total absorption

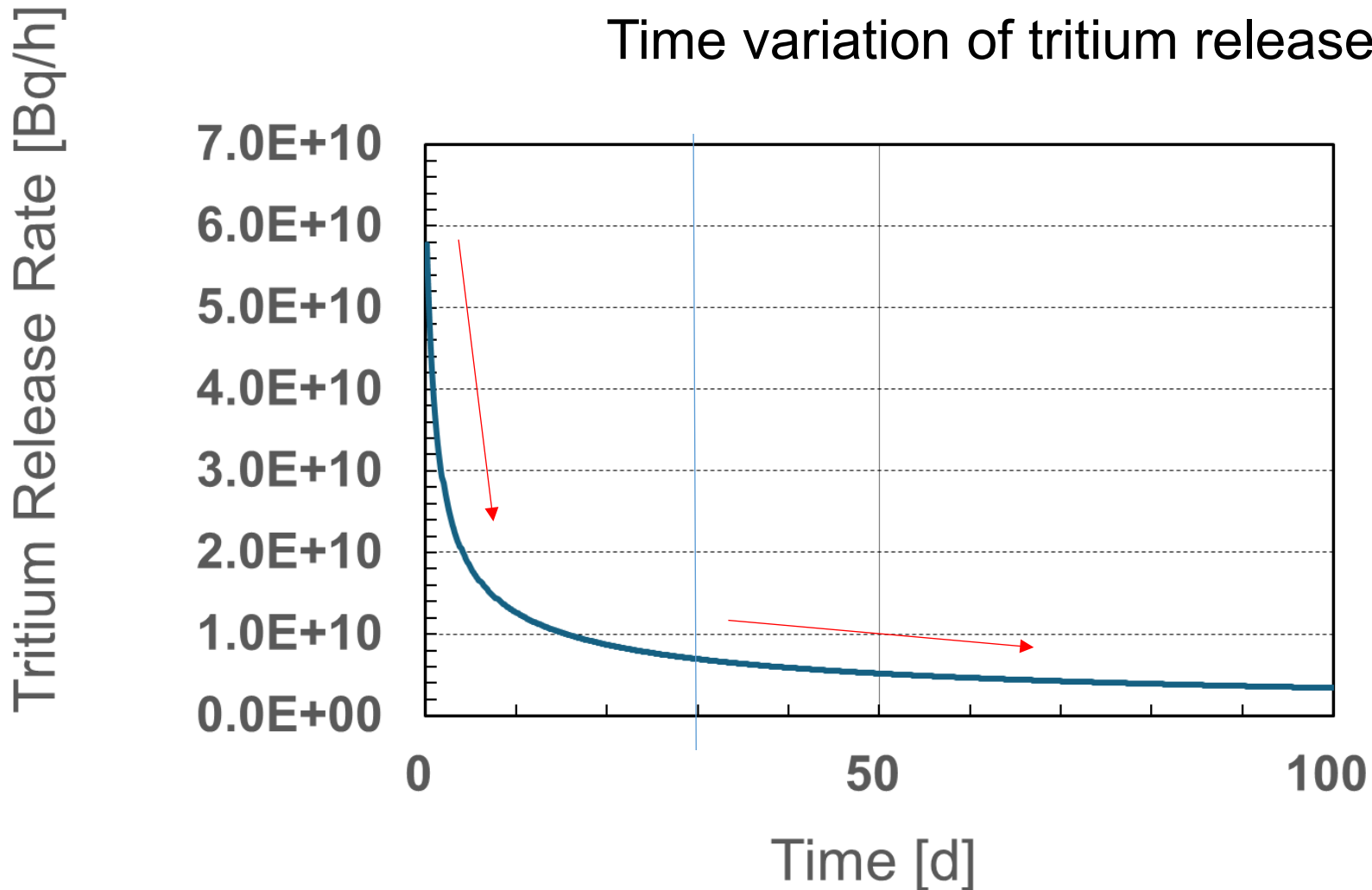


=> Only 30% of tritium can be released for about 1 year.



# Result (2)-3 Release Process

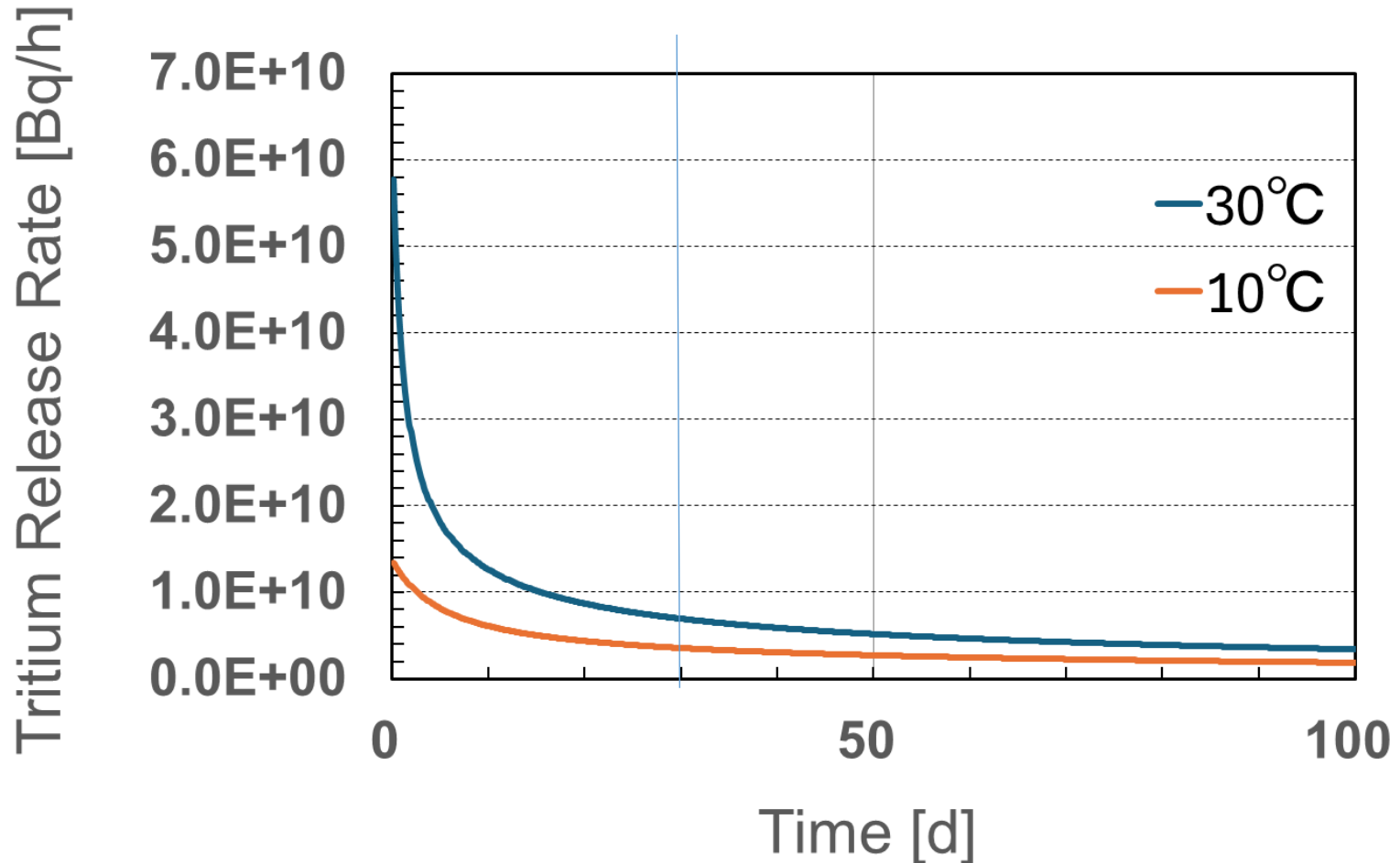
Time variation of tritium release rate



=> Even after 30 days, tritium with rate of  $7 \times 10^9$  Bq/h (more than  $10^{11}$  Bq/d) can be continuously released.

# Result (2)-4 Release Process

Time variation of tritium release rate



=> If the system temperature can be kept at 10°C, the release rate after 30 days decrease by 50% in comparison with that at 30°C.

# Future Development

This Work: Evaluated the **averaged** behavior by assuming the whole surface of the stainless steel as **one physical system**.

In the Future: Consider **tritium movement** in mercury using **fluid dynamics**  
=> Evaluate the absorption and release behaviors on stainless steel (SS) **in each part separately**.

This Work: All tritium that reaches the SS surface by diffusion during the release process is directly released into the air

In the Future: Consider **chemical effect of air humidity (water molecules)**

[Reference] In the presence of a small amount of oxide layer or in the presence of water vapor of several hundred ppm, almost all of the tritium releases in the form of water (HTO)" ←T. Yamanishi et al., Plasma Fusion Res. Vol.85, No.10 (2009)716-725

In the Future: Comparing the calculation to the actual system

=> Adjust the parameters required for the calculation

=> Precise Evaluation of tritium behavior for more complicated situation.

# Summary

- The tritium behavior in the spallation neutron target system with mercury was evaluated using a new physical model.
  - The evaluations are performed through **whole process from absorption to release** by solving the one-dimensional diffusion equation **numerically**.
  - **The temperature effect** for the tritium release rates were shown numerically.
- By developing the model further based on this one, it is expected that tritium behavior can be evaluated more precisely even in a system which is more complicated.