

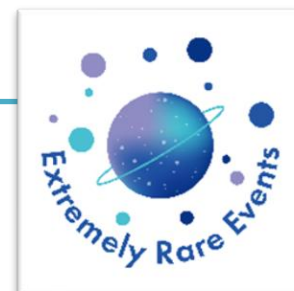
# Radon removal using silver-ion exchanged zeolite

Yasuo TAKEUCHI (Kobe University)



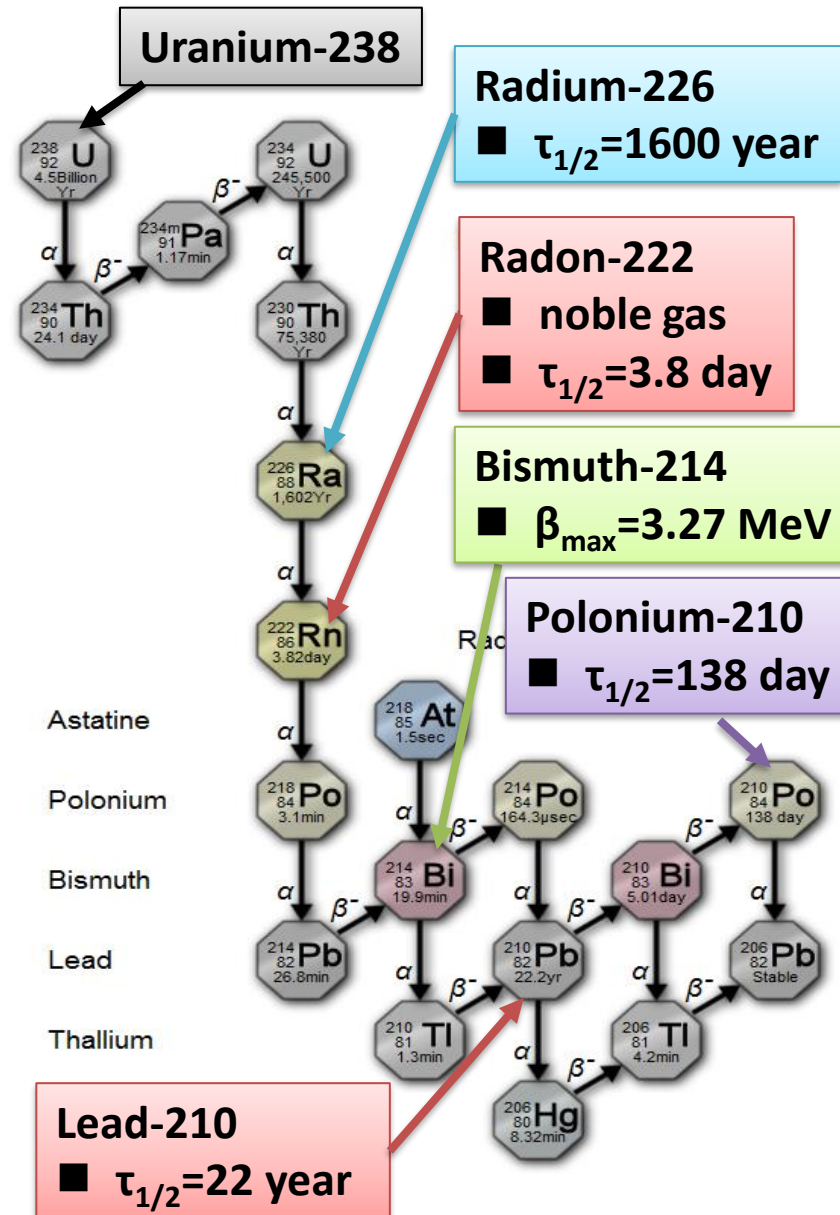
- Radon removal using adsorbents
- Silver-ion exchanged zeolite (Ag-zeolite)
- Summary of recent activities
- Measurement system & results
- Summary

Research Project D01: Deployment of Ultra-Low Radioactivity Technology  
Supported by JSPS KAKENHI Grant Numbers 24H02243 and 24K21549.



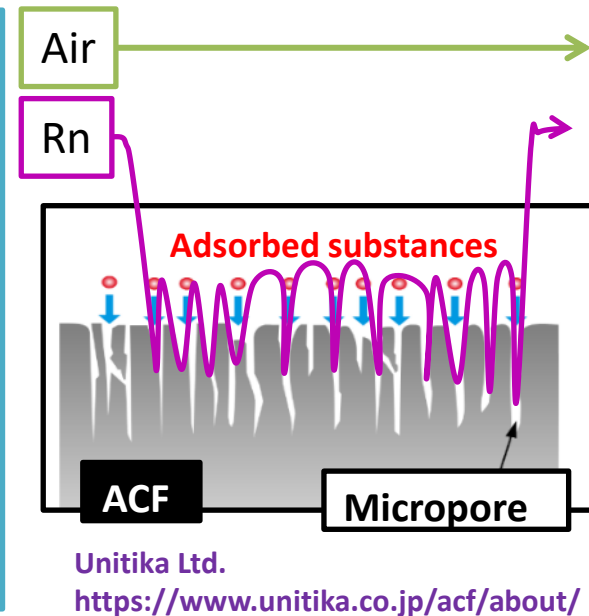
# Radon-222 (Radon, Rn)

- Radon is a radioactive noble gas and therefore **chemically inert**.
- The parent nucleus of  $^{222}\text{Rn}$  is  $^{226}\text{Ra}$  in the uranium chain, and since it exists at the ppm level in ordinary soil, **Rn concentrations can become particularly high in underground environments**.
- Because  $^{222}\text{Rn}$  has a half-life of **3.8 days**, Rn produced outside the detector can enter the detector volume and decay there.
- Rn can be a **major common background** in underground experiments.
- The conventional method for removing Rn has been **physical adsorption** using materials such as activated carbon, which contains a very large number of **micropores**, to trap and eliminate Rn.
  - Cooled activated carbon is used to enhance adsorption efficiency.

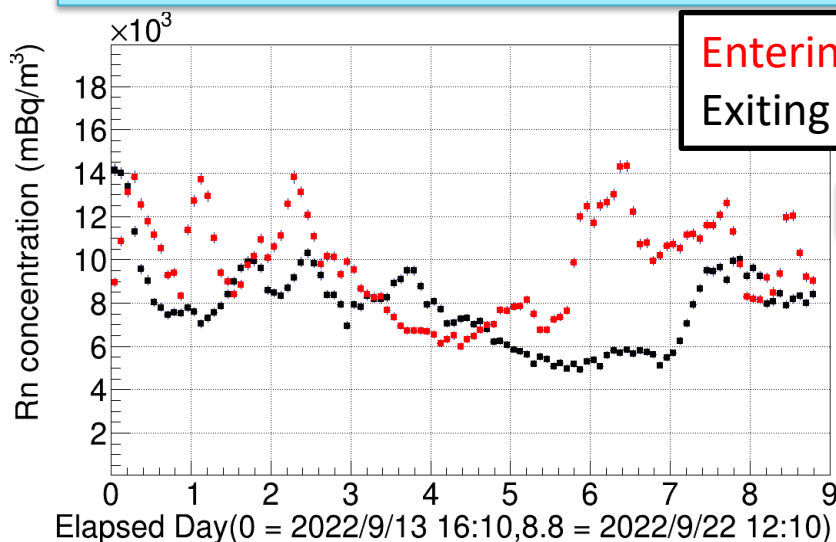


# Radon removal using adsorbents

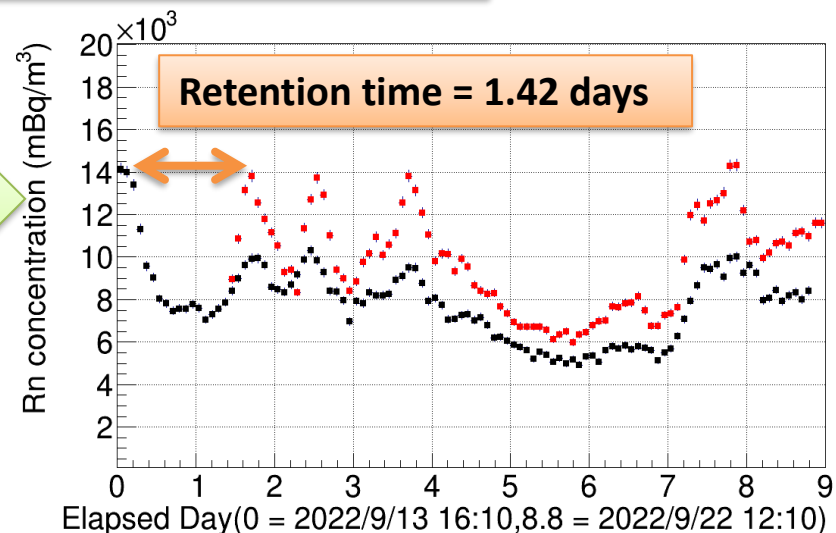
- Radon (Rn) atoms are adsorbed in the pores.
- Rn flow velocity slows down in the adsorbent column.
- Rn **decays in the column** during this slowing down.
- The time between Rn entering and exiting the column is called **retention time (RT)**.



## Rn in room-air measurement with Activated Carbon Fiber (ACF)



Shifted  
red  
points  
by 1.42  
days



# Rn adsorption coefficient (K)

$$\text{Rn adsorption coefficient (K)} = \text{Retention time (RT) [day]} * \text{Flow rate (F) [m}^3\text{/day]} / \text{Mass of adsorbent (m) [kg]}$$

## 2.2.1. K constant

Radon amount cannot be measured via standard gravimetric or volumetric techniques used in the field of adsorption. Therefore, an alternative observable — radioactivity (hereinafter referred to as *activity*) — is used to characterize radon adsorption in a way similar to Henry's law as follows [66,67]:

$$A = KC \Rightarrow K = \frac{A}{C}, \quad (3)$$

where A is the activity of radon adsorbed per unit of mass of adsorbent [Bq/kg], C is the activity concentration of radon in gas [Bq/m<sup>3</sup>]. The constant K, commonly referred to as *adsorption constant* or *K constant*, *K factor* [m<sup>3</sup>/kg], quantifies the affinity of radon for a specific adsorbent under given conditions (temperature, gas composition, etc.). K can be experimentally determined by measuring radon activity in the adsorbent and the gas once thermodynamic equilibrium between the two is established.

The K-constant can be measured using either static or dynamic experimental setups, depending on whether the adsorbent interacts with a static gas phase or a flowing gas stream. For dynamic experiments, the K constant can be calculated using Eq. (3), or based on chromatographic approach (see 2.2.5). Below, we provide a comparison of these methods, discuss technical setups and implemented technical solutions, and briefly mention alternative approaches to measuring radon adsorption.

## Separation and Purification Technology

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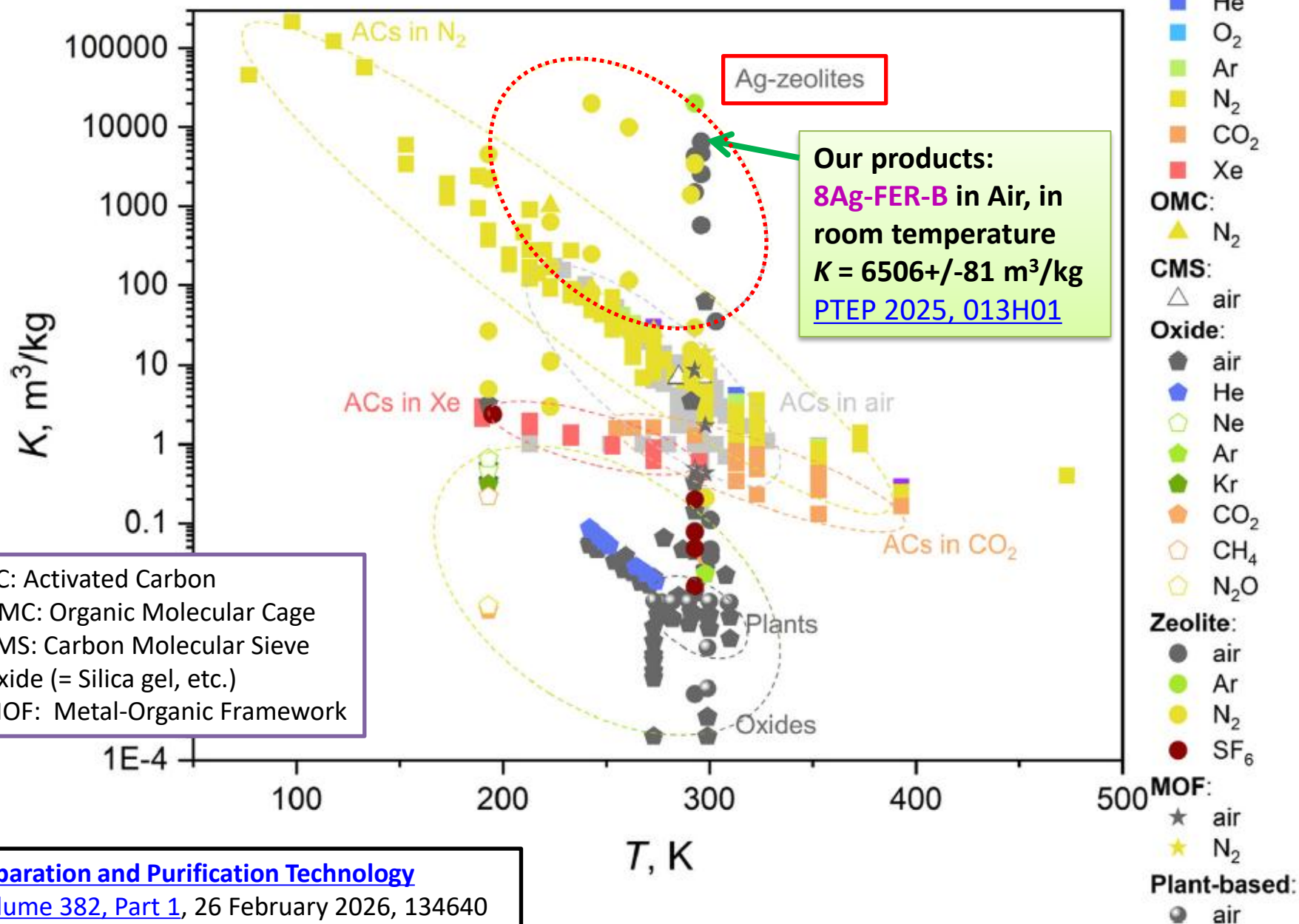
K: Rn adsorption coefficient [m<sup>3</sup>/kg]  
A: Adsorbed Rn [Bq/kg]  
C: Rn concentration [Bq/m<sup>3</sup>]

Total amount of processed gas  
volume for the adsorbed Rn [m<sup>3</sup>]:  
 $RT * F$

Total amount of adsorbed  
Rn [Bq]:

$$\begin{aligned} A * m &= K * m * C \\ &= RT * F * C \\ \Rightarrow K &= (RT * F) / m \end{aligned}$$

# Comparison of the $K$ coefficient



# Silver-ion exchanged zeolite (Ag-zeolite)

- **Ag-zeolite: Silver ions are introduced into zeolite (by ion exchange, etc.).**
  - Zeolite: a porous crystalline aluminosilicate (多孔質の結晶性アルミノケイ酸塩)
- **Commercialized** as an antibacterial material, etc.
- **2020~: Certain Ag-zeolites show excellent adsorption properties for xenon in air.**
- **2023~: Some Ag-zeolites have very high adsorption performance for radon in air.**
  - Mechanism is not solved yet (chemical adsorption by metallic silver or silver ion ???)

| Adsorbent type               | $m$ [kg]              | $RT$ [day] | $K$ [m <sup>3</sup> /kg] |
|------------------------------|-----------------------|------------|--------------------------|
| Ag-ZSM-5 [1] (Ag-MFI)        | $8.85 \times 10^{-3}$ | 10.69      | 3500                     |
| Ag-ETS-10 [1]                | $16.3 \times 10^{-3}$ | 19.16      | 3400                     |
| Ag-ETS-10 (18°C) [2]         | $2 \times 10^{-4}$    |            | 1400                     |
| 8Ag-FER-B [3] (our products) | 0.020                 | 30.12      | 6506                     |
| Activated Carbon Fiber [4]   | 4.65                  | 1.9        | 6.47                     |
| Activated Carbon (AC) [4]    | 26.9                  | 10.16      | 4.96                     |
| Cooled AC@SK (-60°C) [5,6]   | 18.8                  | 16.5       | 379                      |

- [1] [Sci Rep 13, 6811 \(2023\)](#)
- [2] [PTEP 2024, 023C01](#)
- [3] [PTEP 2025, 013H01](#)
- [4] [Takagi thesis, Kobe \(2024\)](#)
- [5] [PLB 452 \(1999\) 418](#)
- [6] [NIM A 867 \(2017\) 108](#)

Measurements  
under **room**  
**temperature**

Ag-ZSM-5: Zeolite Socony Mobil-Five, or Ag-MFI  
Ag-ETS-10: Engelhard Titano-Silicate, a zeolite analog  
AC: Activated Charcoal  
ACF: Activated Carbon Fiber

In this Transformative Research Areas (A) , Research Project D01: Deployment of Ultra-Low Radioactivity Technology, we are studying on **the application of Ag-zeolites in underground experiments with strong support from experts.**

# Our studies on Ag-zeolites

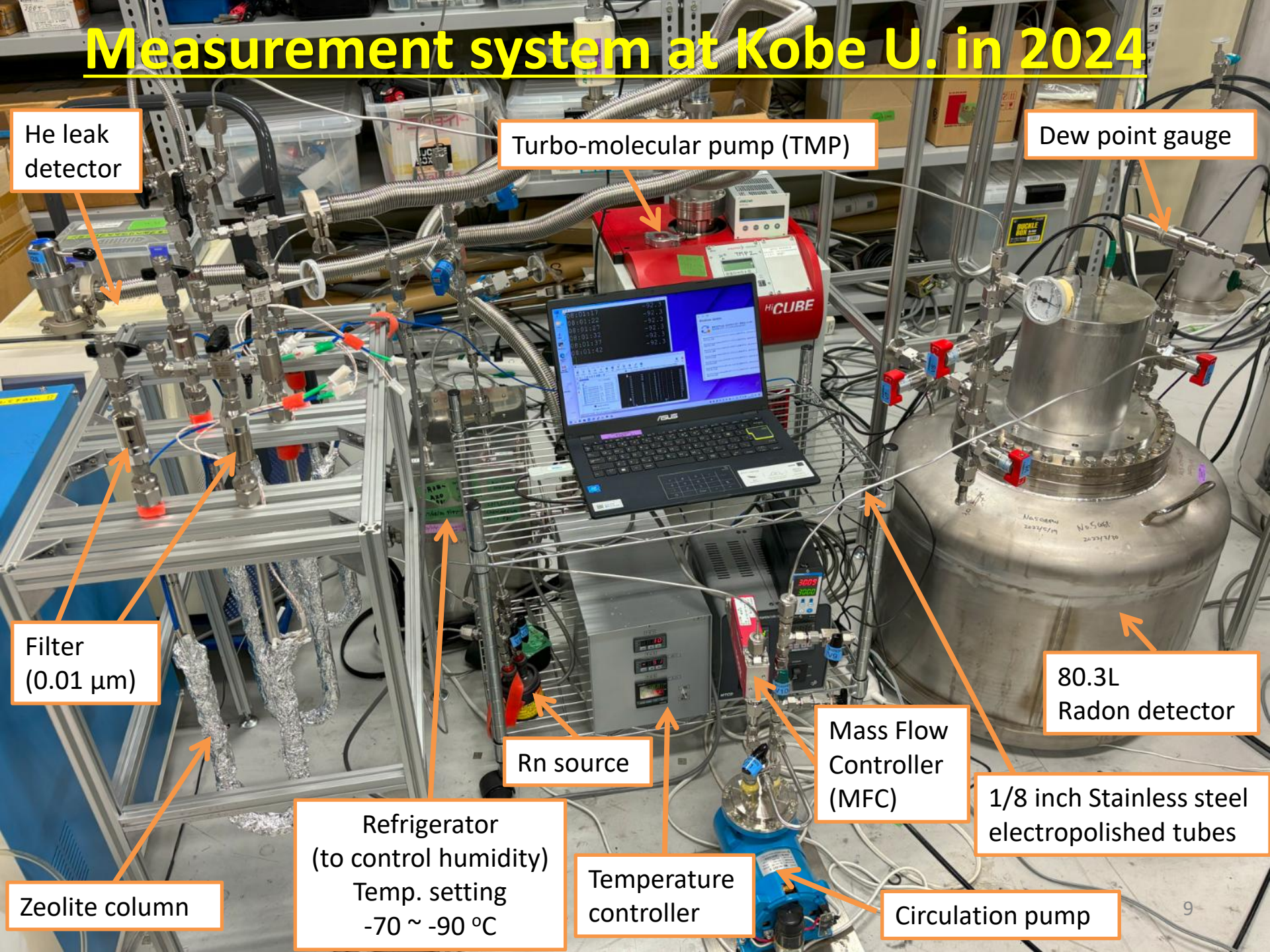
- Cooperating people/companies :
  - T. Wakiyara, M. Matsukura (Dept. of Chemical System Engineering, U. Tokyo), : K. Iyoki (Grad. School of Frontier Sciences, U. Tokyo): Leading experts in zeolite research field
  - S. Hirano (TOSOH CORPORATION): Manufacturer of various types of zeolite
  - A. Taniguchi (Sinanen Zeomic Co., Ltd.): Pioneering company of silver addition
  - (H. Ogawa (Nihon U., Kobe U.): Expert on low-radioactivity zeolite synthesis) (until Dec. 2025)
  - K. Nagatsu, J. Ichinose (QST): Information sharing on Rn removal from air
- From Xe in Air adsorption, the experts suggested: FER > MFI > ... > MOR > ...
- We produced Ag-zeolites with raw zeolites made by Tosoh Corporation, and silver ions were introduced by Sinanen Zeomic Co., Ltd.
- In 2025, we found 8Ag-FER-B has a good radon adsorption performance.
  - T. Sone , et al., PTEP 2025, 013H01, <https://doi.org/10.1093/ptep/ptae181>
- We also produced prototype samples of other Ag-zeolites: Ag-MFI, Ag-MOR, 22Ag-FER-D, 20Ag-Y-D, ...
  - We purchased Ag-ETS-10 and conducted radon adsorption tests with it.
- So far, 8Ag-FER-B still shows the best Rn adsorption performance in air among our tests.
- 8Ag-FER-B was also tested (or under tests) in Air, Ar, Xe, CO<sub>2</sub>, and CF<sub>4</sub>.

# Recent activities on Ag-zeolites

- Rn removal system with 8Ag-FER-B for Hyper-Kamiokande
  - P08: H. Bambara
- Tests with cooled 8Ag-FER-B (and Ag-ETS-10)
  - P05: Y. Inaba
- Radon adsorption tests in Air and in Ar with various test samples.
  - Ag-MFI, Ag-MOR, Ag-FER (3Ag-FER, 8Ag-FER-B, 8Ag-FER-B\_2, 8Ag-FER-D, 22Ag-FER-D), 20Ag-Y-D, ...
    - Contains 0.5-1.0% potassium (K) (~2025)
    - Almost no potassium (2026~)
  - P10: H. Nishimura
- Topics in this Transformative Research Areas (A) with Research Project D01
  - Rn removal in air (C01, A01, B03), QST (Medical field)
  - Rn removal in CF<sub>4</sub> gas (B03)
  - Xe adsorption in air (for Xe recovery) (A01, B02)
  - Removal of Ar, Kr, Rn, moisture, and oxygen from Xe (B02)
  - Removal of moisture from liquid scintillator (A01)
  - Removal of Kr from N<sub>2</sub> (A01)
  - Xe recovery from air with copper-ion-exchanged zeolites → P39: Y. Gando
- Overseas: Consultations, collaborative research, and/or purchase (8Ag-FER-B)
  - Korea, New Zealand, Australia, Austria, Italy, Hong Kong, and Spain.

Publicly Offered Research

# Measurement system at Kobe U. in 2024



He leak detector

Turbo-molecular pump (TMP)

Dew point gauge

Filter (0.01  $\mu\text{m}$ )

Zeolite column

Refrigerator (to control humidity)  
Temp. setting  
-70 ~ -90  $^{\circ}\text{C}$

Rn source

Temperature controller

Mass Flow Controller (MFC)

Circulation pump

80.3L Radon detector

1/8 inch Stainless steel electropolished tubes

# Zeolite column

2024/08/08



Silver zeolite pellets  
(just after delivery)  
8Ag-FER-D, 20g

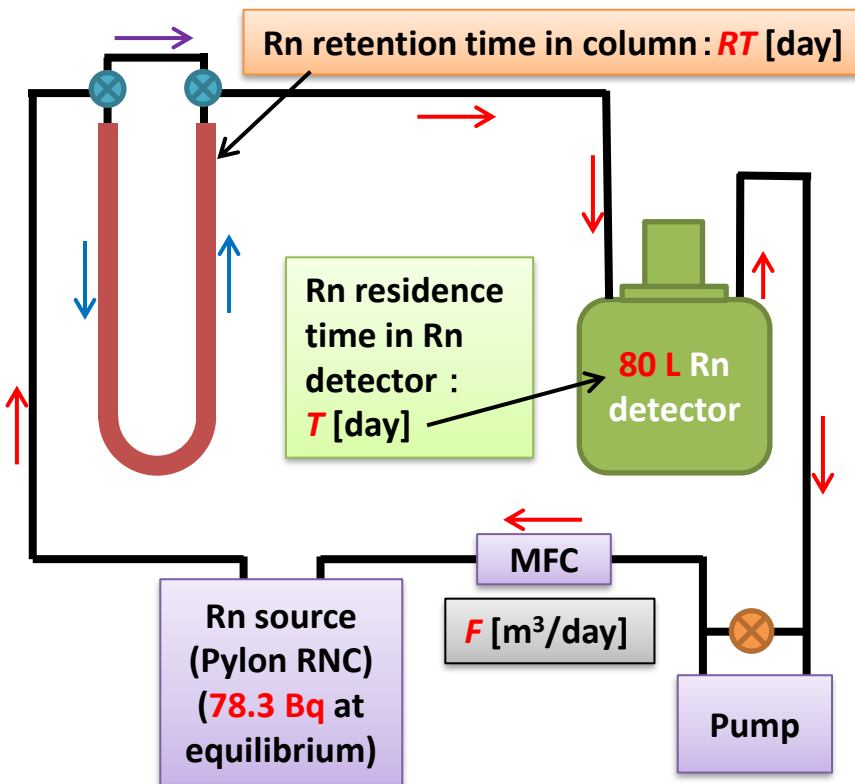
Before heat insulation:  
1/2 inch SUS BA tube  
1 m + Ribbon heater  
(150 W, 1.5 m)

3Ag-FER, 20g

8Ag-FER-D, 20g

8Ag-FER-B, 20g

# Evaluation method for ***RT*** and ***K***



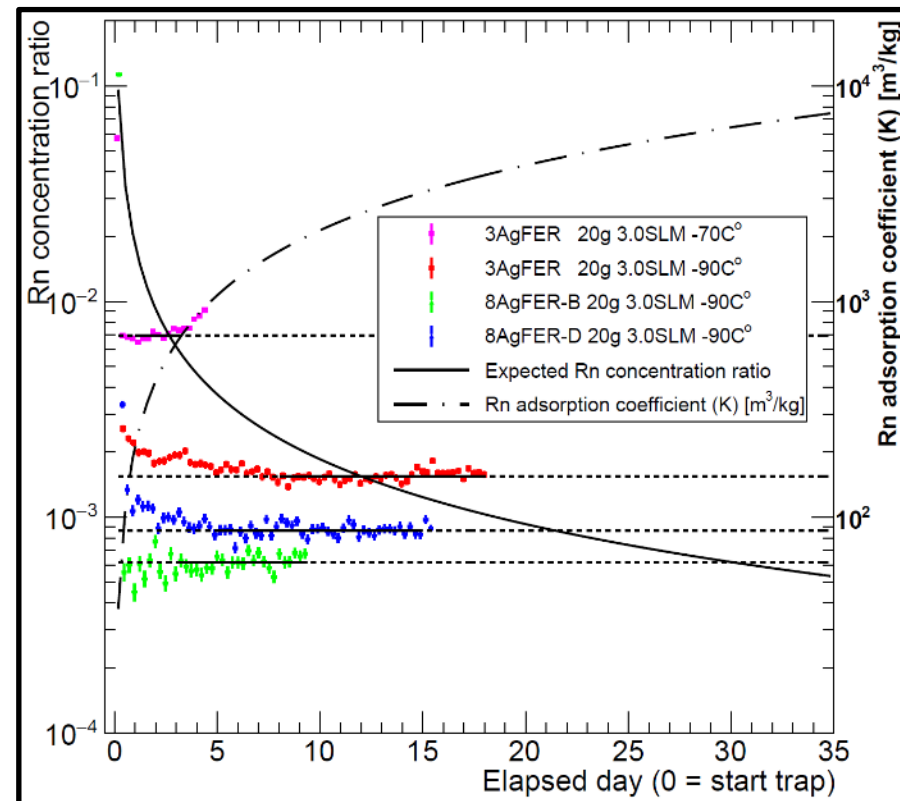
## Operating Procedure:

1. Bypass the zeolite column and circulate the gas (check for radiation equilibrium)
2. (Replace and) bake the zeolite column under vacuum (**200~300 °C**, several hours)
3. Circulate the gas **through the zeolite column** (Rn adsorption measurement)
4. Check the **Rn concentration ratio**, then bake (remove Rn in the column). → Return to 1.

- Rn source always connected → 78.3 Bq of Rn is distributed in the system.
- Assume that the ratio of the amount of Rn in the zeolite column to that in the Rn detector is equal to the ratio of Rn residence time in each device. (Ignore the volume of piping)
  - Fraction of Rn in the Rn detector = **Rn concentration ratio** ( $Rn_{ratio}$ )
  - $$Rn_{ratio} = \frac{T}{RT+T} = \frac{0.080}{F \cdot RT + 0.080}$$
- On the other hand,  $Rn_{ratio}$  can be measured by  $Rn_{ratio} = \frac{\text{Rn concentration via column}}{\text{Rn concentration at column bypass}}$
- Evaluate  $RT$  from the measured  $Rn_{ratio}$ , then  $K (= \frac{RT \times F}{m})$  from  $RT$ .

# Results from [PTEP 2025, 013H01](#)

- **Humidity**:  $-90^{\circ}\text{C}$  ( $2 \times 10^{-4} \text{ g/m}^3$ ) is better than  $-70^{\circ}\text{C}$  ( $5 \times 10^{-3} \text{ g/m}^3$ ).  
(Absolute humidity)
- **Amount of Ag**: 8% is better than 3%.
- **Ag introduction method**: type B (special ion-exchange method) is superior.



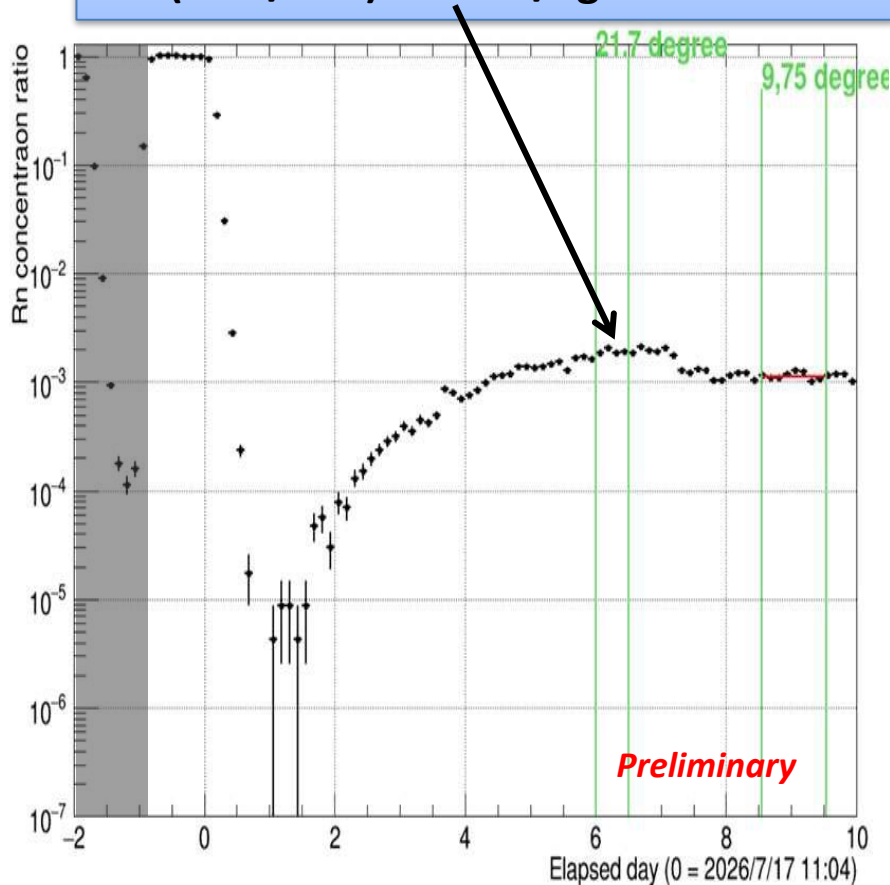
| Sample      | Refrigerator setting  | Adsorbent mass [kg] | Retention time [day] | Radon adsorption coefficient [ $\text{m}^3/\text{kg}$ ] |
|-------------|-----------------------|---------------------|----------------------|---------------------------------------------------------|
| (This work) |                       |                     |                      |                                                         |
| 3Ag-FER     | $-70^{\circ}\text{C}$ | 0.020               | $2.65 \pm 0.04$      | $573 \pm 4$                                             |
| 3Ag-FER     | $-90^{\circ}\text{C}$ | 0.020               | $11.97 \pm 0.14$     | $2563 \pm 14$                                           |
| 8Ag-FER-B   | $-90^{\circ}\text{C}$ | 0.020               | $30.12 \pm 0.48$     | $6506 \pm 81$                                           |
| 8Ag-FER-D   | $-90^{\circ}\text{C}$ | 0.020               | $21.38 \pm 0.27$     | $4618 \pm 35$                                           |

# Comparison of 8Ag-FER-B & Ag-ETS-10

P05: Y. Inaba

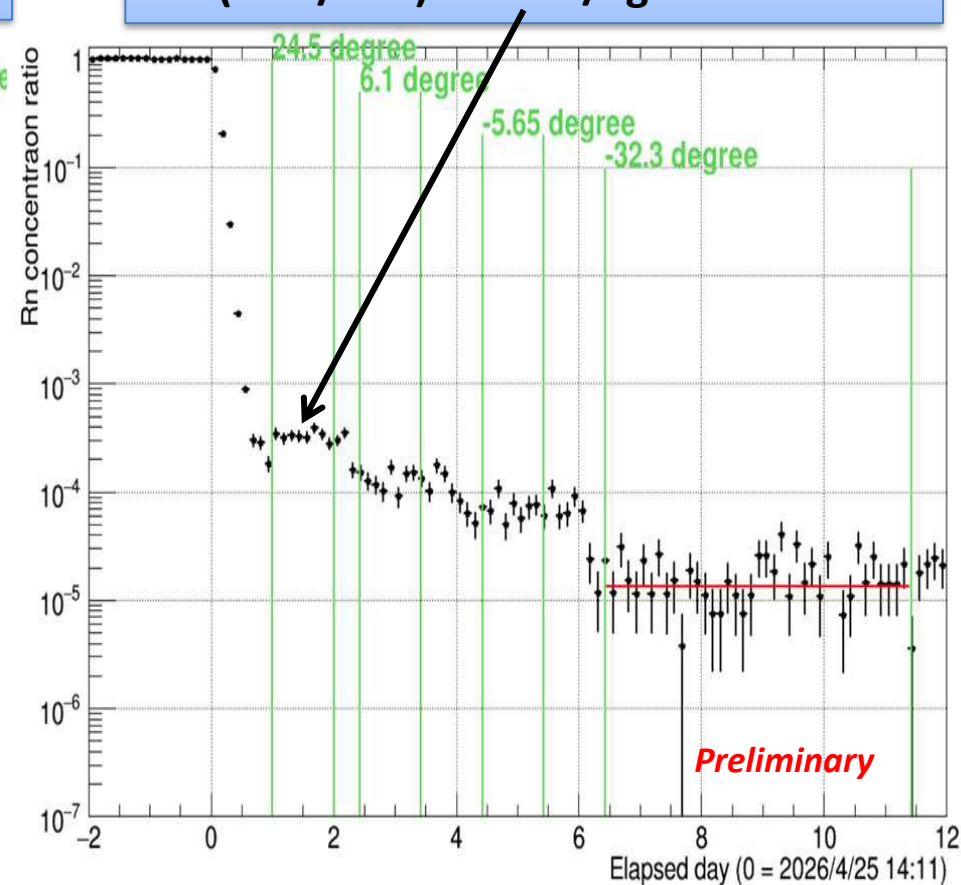
## Ag-ETS-10

G1 Air, 21.7 °C, 6.0g, Atmospheric pressure  
1.5 ~ 2.1(??) L/min (under checking)  
 $K = (9.0 \pm 0.2) \times 10^3 \text{ m}^3/\text{kg}$



## 8Ag-FER-B

G1 Air, 24.5 °C, 6.0g, Atmospheric pressure  
1.5 L/min  
 $K = (4.1 \pm 0.2) \times 10^4 \text{ m}^3/\text{kg}$



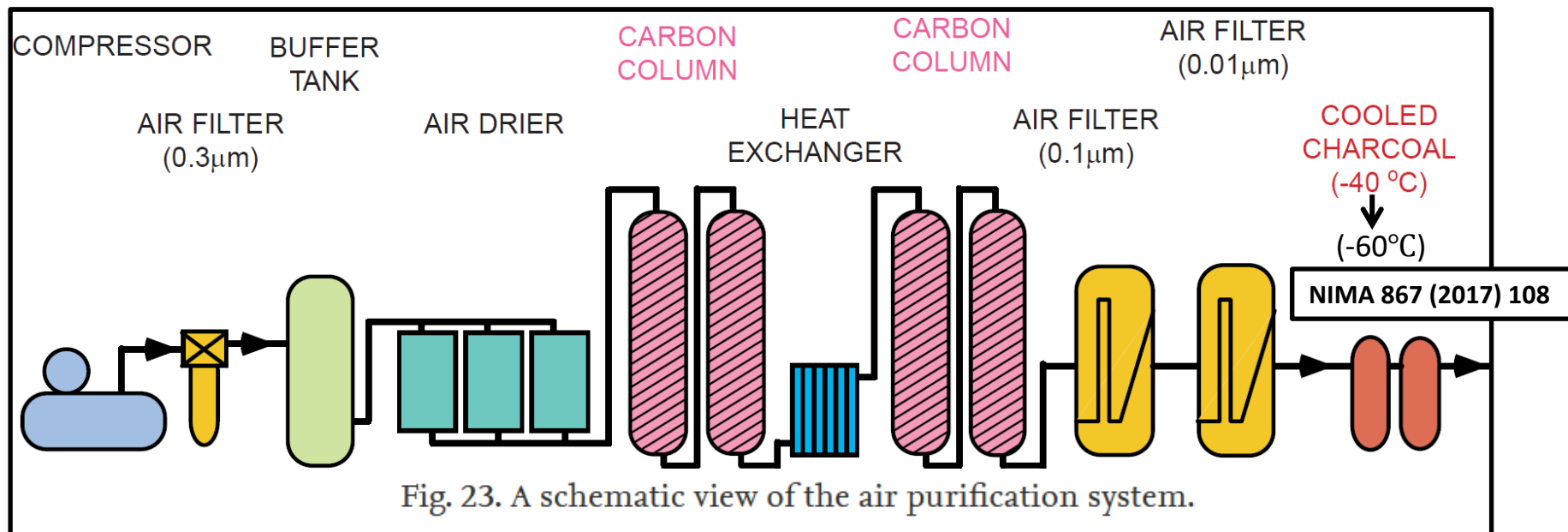
Ag-ETS-10 shows stronger initial Rn reduction, but 8Ag-FER-B seems better in the steady state. (under checking)

# Rn reduction in air in SK

- Granular AC (room temperature): 8 m<sup>3</sup>
- Cooled granular AC: 50 L @ -60 °C
- Air flow rate: 18 Nm<sup>3</sup>/hour
- Dew point: -65 °C@+1 kg/cm<sup>2</sup>
- Remaining Rn concentration
  - After “CARBON COLUMN”: ~20 mBq/m<sup>3</sup>
  - After “COOLED CHARCOAL”: ~1 mBq/m<sup>3</sup>

## NOTE:

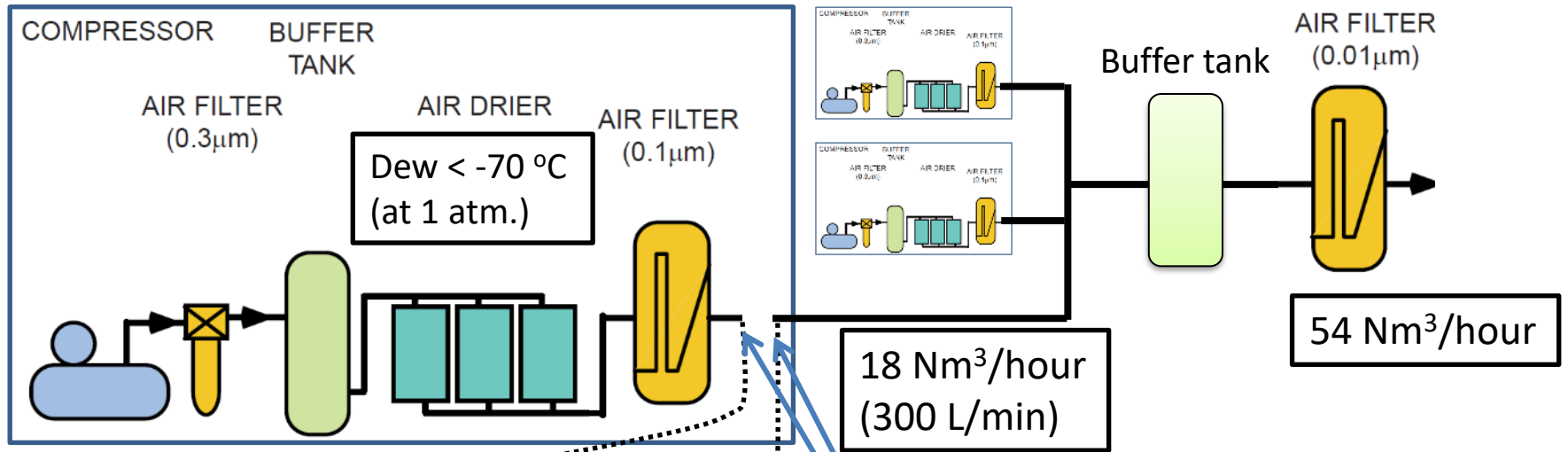
- Activated carbon has not been replaced.
- Baking is not done. (too large)
- Fresh air from outside mine is supplied into this area.



# Rn removal system for HK

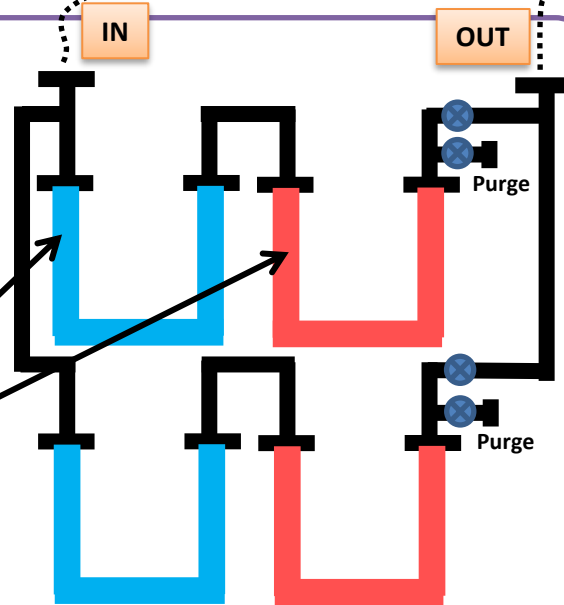
28-SEP-2026

Current HK air system design: (Essentially SK air system (w/o ACs)) x 3 set



Our plan: to add

- U-shape Vessel: 50A (6 cmφ) SUS BA pipe 2.8 m long
- Adsorbent: Zeolite to remove water (~4kg) 8Ag-FER-B (4kg)
- 1-inch piping: 20A10K flange Perfluoro O-ring

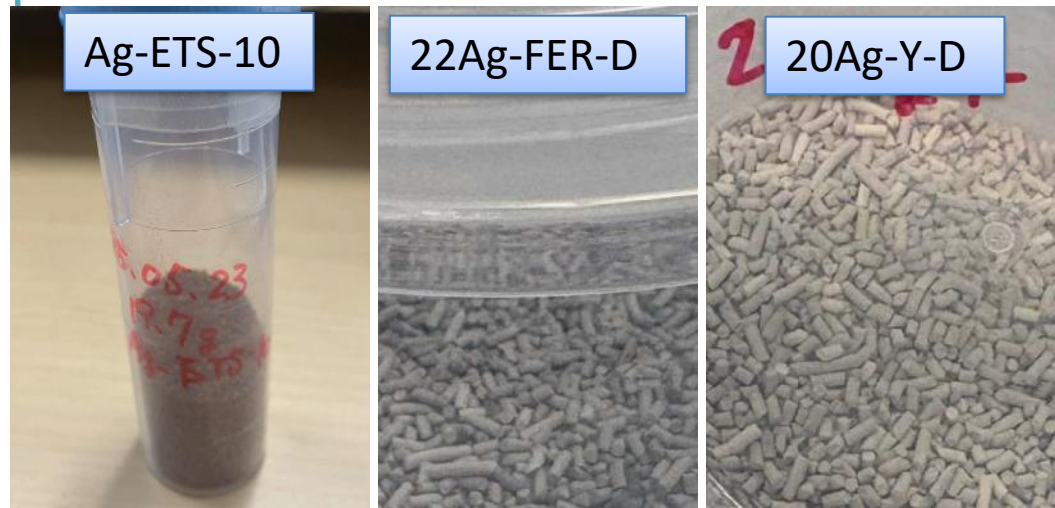


Pressure: 0.65~0.50 MPa (in), 0.30 MPa (out)

- Prepare two Ag-zeolite columns
- One column : Rn reduction
- Another column : Baking with a small dry-air purge
- Alternate these two operations
- Goal: ~50 Bq/m³ → < 1mBq/m³
- Test with a prototype Rn removal system is on going in Mozumi at ~100L/min → P08 H. Bambara

# Tests with other Ag-zeolites in Air

- **Ag-ETS-10**:  $K=3400$  [m<sup>3</sup>/kg] in a preceding study.
- **22Ag-FER-D**: Structurally, the FER framework allows ion exchange only up to about 8% at most. Although ion exchange may not occur, we prepared a test sample with the silver content increased to 22%.
- **20Ag-Y-D**: Y-type zeolite. 20% Ag. Y is easier to absorb moisture than FER. To check water effect.
- Tests under similar conditions (6 g sample, 1.0~2.1L/min flow rate, 300°C baking, -90°C refrigerator)



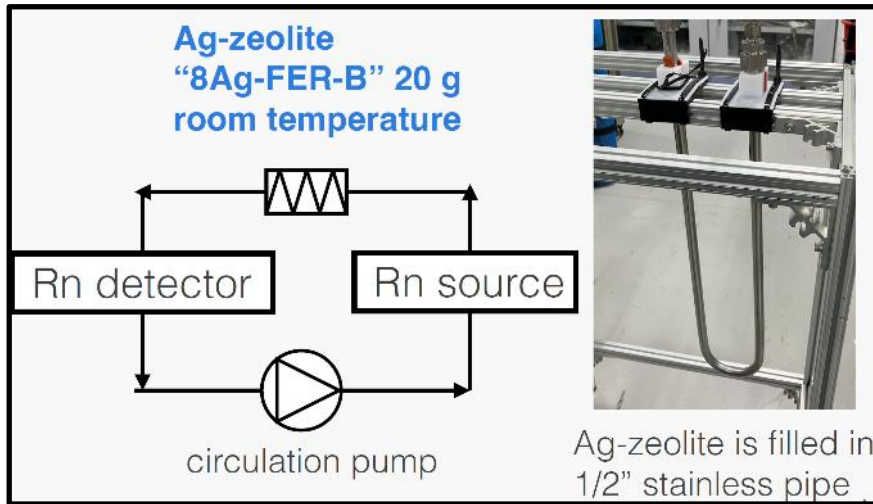
**Preliminary** results (Sep. 2026)  
(6 g, 300°C baking, room temperature)

| Ag zeolite | Flow [L/min] | K [m <sup>3</sup> /kg]      |
|------------|--------------|-----------------------------|
| 8Ag-FER-B  | 1.5          | (4.1+/-0.2)x10 <sup>4</sup> |
| Ag-ETS-10  | 1.5~2.1(?)   | (7.0+/-0.2)x10 <sup>3</sup> |
| 22Ag-FER-D | 1.0          | 35.6+/-0.2                  |
| 20Ag-Y-D   | 1.0          | 0                           |

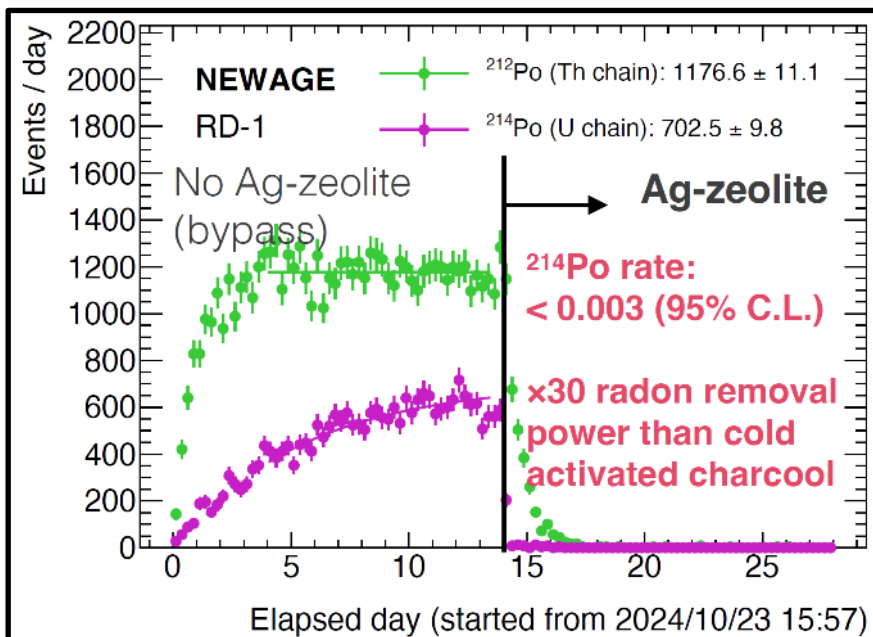
Cf. (20 g, 200°C baking, 3 L/min)  
8Ag-FER-B :  $K = 6506 \pm 81$  m<sup>3</sup>/kg

# Rn adsorption of 8Ag-FER-B in CF<sub>4</sub>

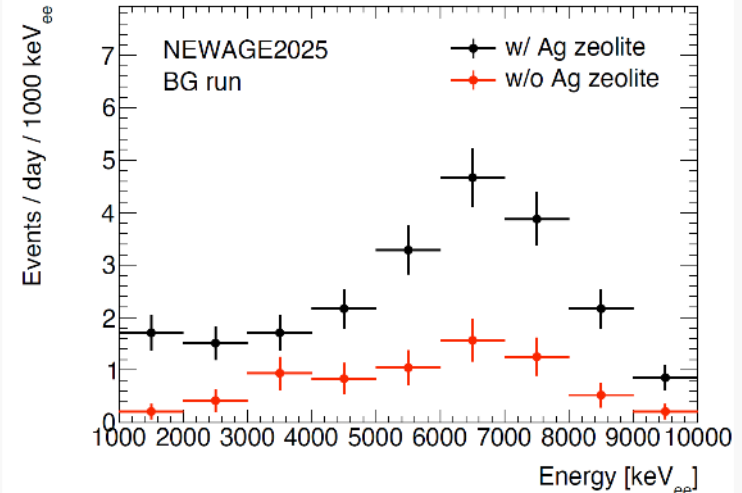
S. Higashino @CYGNUS2026



- NEWAGE group demonstrated 8Ag-FER-B reduced Rn in CF<sub>4</sub> gas
- This is one of the outcomes of the collaborative research within the Transformative Research Areas (A) “Extremely Rare Events” KAKENHI.



## Radon rate successfully reduced by 1/3



# Summary

- We developed silver-ion exchanged zeolite (**8Ag-FER-B**) with excellent Rn adsorption performance in collaboration **with experts**.
  - We also found that the Rn adsorption performance of 8Ag-FER-B is highly **sensitive to humidity**.
- In air, at room temperature, its Rn adsorption performance is approximately **1,300 times higher** than that of the granular activated carbon used in Super-Kamiokande. **8Ag-FER-B at room temperature** exhibits approximately **17 times higher** Rn adsorption performance than **cooled activated carbon at -60 °C**.
  - These results are for 200 °C baking; 300 °C baking yields better performance.
- Within the Transformative Research Areas (A) program “Extremely Rare Events”, **collaborative research** on Ag-zeolite is expanding.
- Interest from **overseas** is also increasing.