Determination of radon leakage from sample containers for gamma spectrometry measurement of ^{226}Ra

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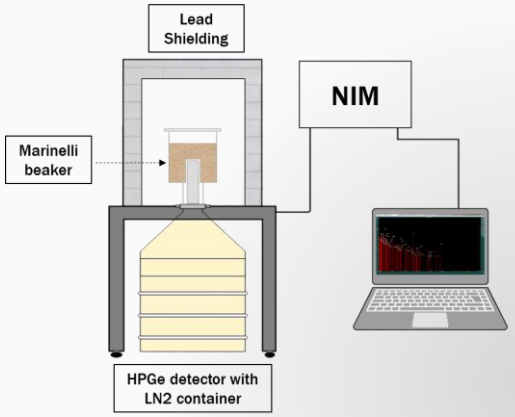
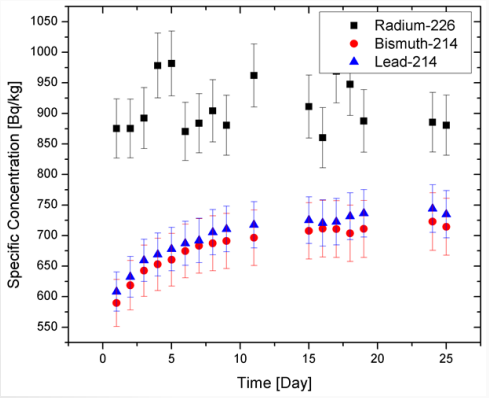
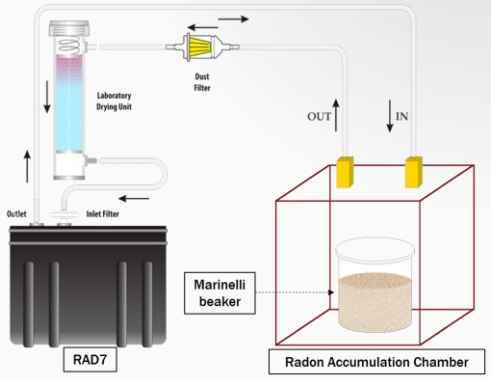
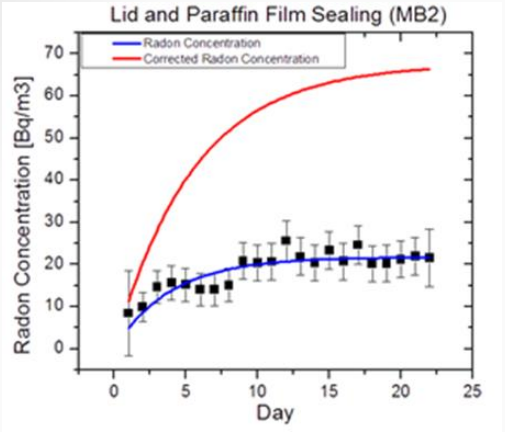
Introduction

Abstract









Marinelli Beaker Release Fraction (MB_{LR})

01 Introduction

Background Theory

Measuring Radium in Soil Sample by HPGe

Table 1. Advantages and disadvantages of gamma spectrometry methods

Gamma spectrometry	Advantage	Disadvantage
▷ Direct measurement : analyzing Ra-226 by its 186 keV peak.	✓ Versatile ✓ easy to use ✓ non-destructive ✓ relatively cheap method ✓ Repeatability ✓ easy sample preparation ✓ easier spectrum analysis	✓ weak yields γ-line 186.2 keV (3.59%), ✓ an interference with 235U direct line 185.7 keV
▷ Indirect measurement : analyzing Ra-226 by its progenies' gamma rays using radioactive equilibrium.		✓ longer time needed to achieve secular equilibrium (at least 21 days) ✓ radon leakage from the measurement container causes the equilibrium cannot be reached.

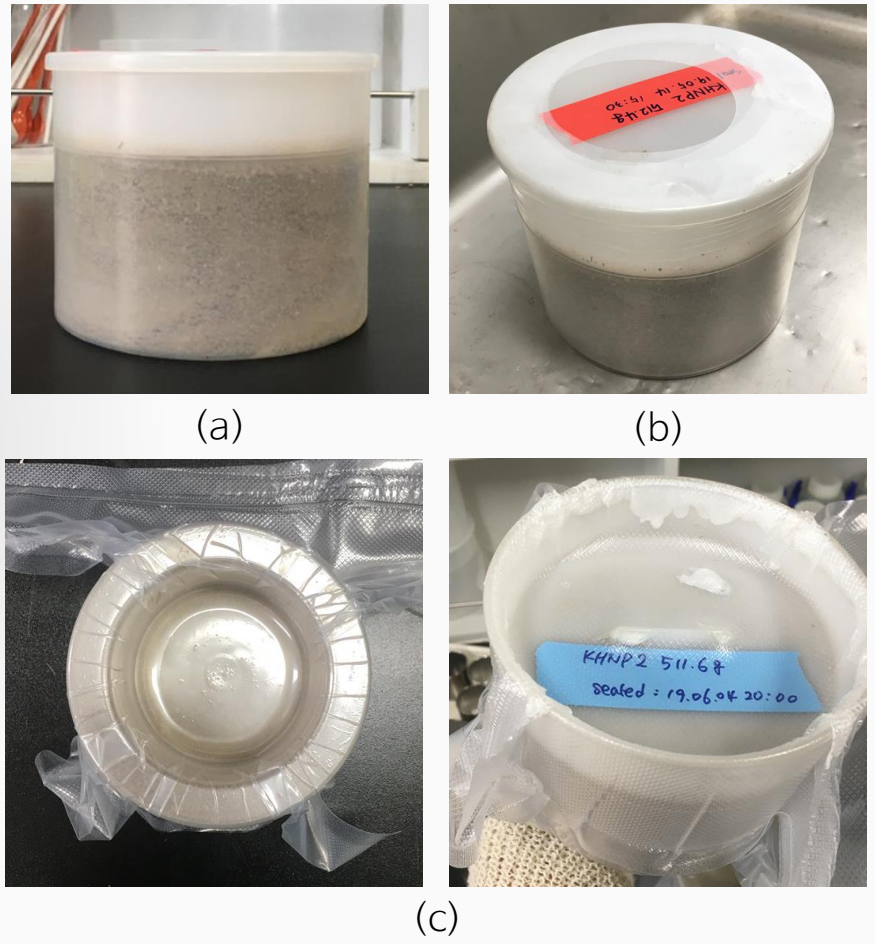
Materials and Methods

Materials and Methods

Samples and sealing methods

Table 2. Samples and sealing methods for HPGe and radon chamber measurement

Soil sample	Uljin soil sample Uljin 1: 522.6 gram (Radon Chamber) Uljin 2: 512.4 gram (HPGe)
Reference material	IAEA 434 Phosphogypsum, 250 gram
Marinelli beaker	Polypropylene Snap-on lid with inner lid Vol: 450 mL
Sealing method	MB0: open MB1: only lid without sealing MB2: lid and sealed with paraffin film MB3: sealed with vacuumed plastic bag



(a) (b) (c)

Figure 1. Marinelli beaker sealing method (a) lid without sealing, (b) lid and paraffin film sealing, (c) vacuumed plastic bag sealing

Materials and Methods

HPGe measurement system

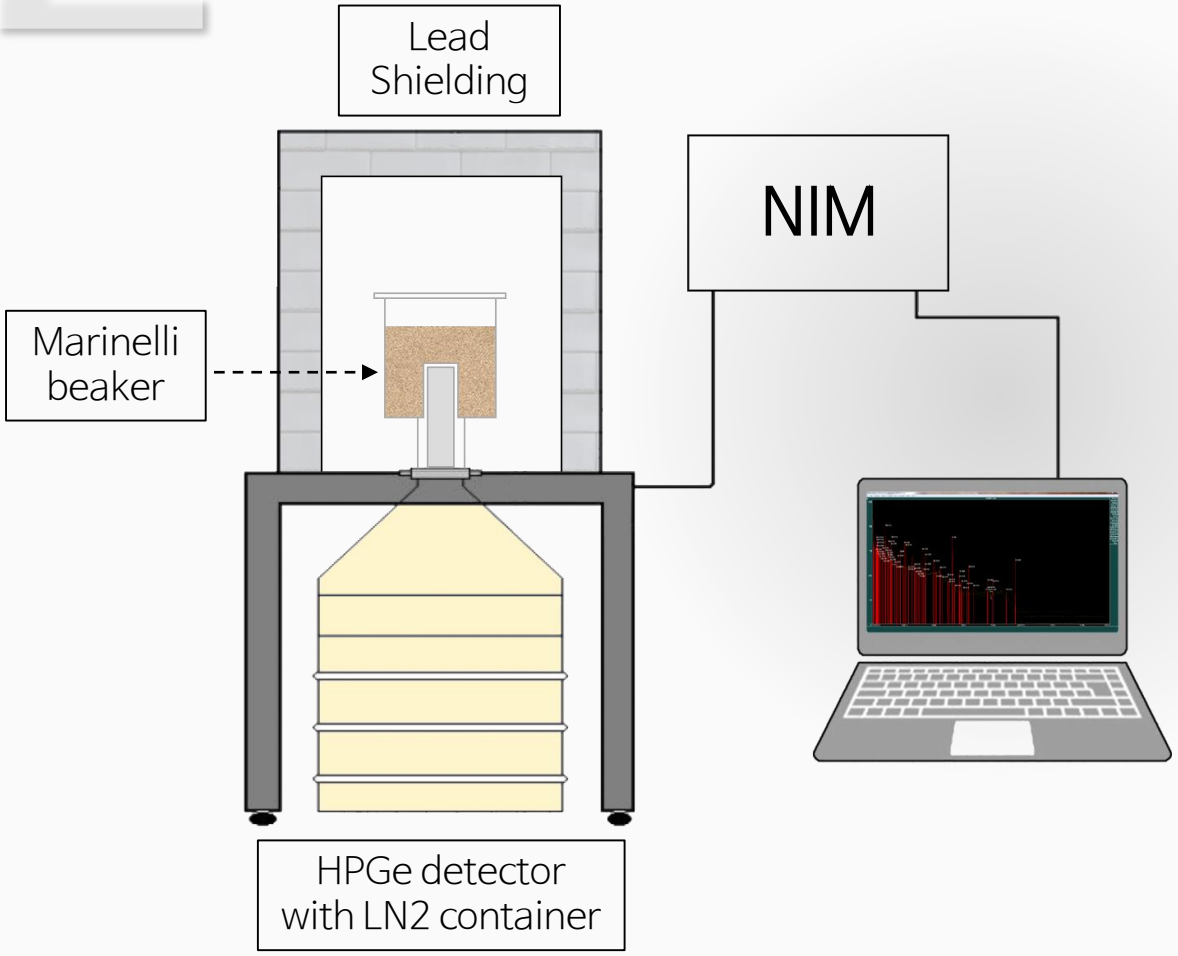


Figure 2. Experimental set up for radon leakage measurement.

Detector specification

- ORTEC GEM 15P4 coaxial HPGe
- 70mm diameter endcap; equipped with 16384 channels MCA
- 0.82 keV for the 122 keV-peak resolution and is 15% for the 1.33 MeV Co-60 peak relative efficiency .

Energy and efficiency calibration

Using CRM volume source in 450mL Marinelli Beaker of agar medium, certified by KRISS, consists of ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y with nominal density of 1.001 g/cm3.

Measurement set up

- Samples' gamma ray spectra were taken by HPGe detector for each of 86400 seconds during 21 days.

Materials and Methods

Radon chamber measurement system

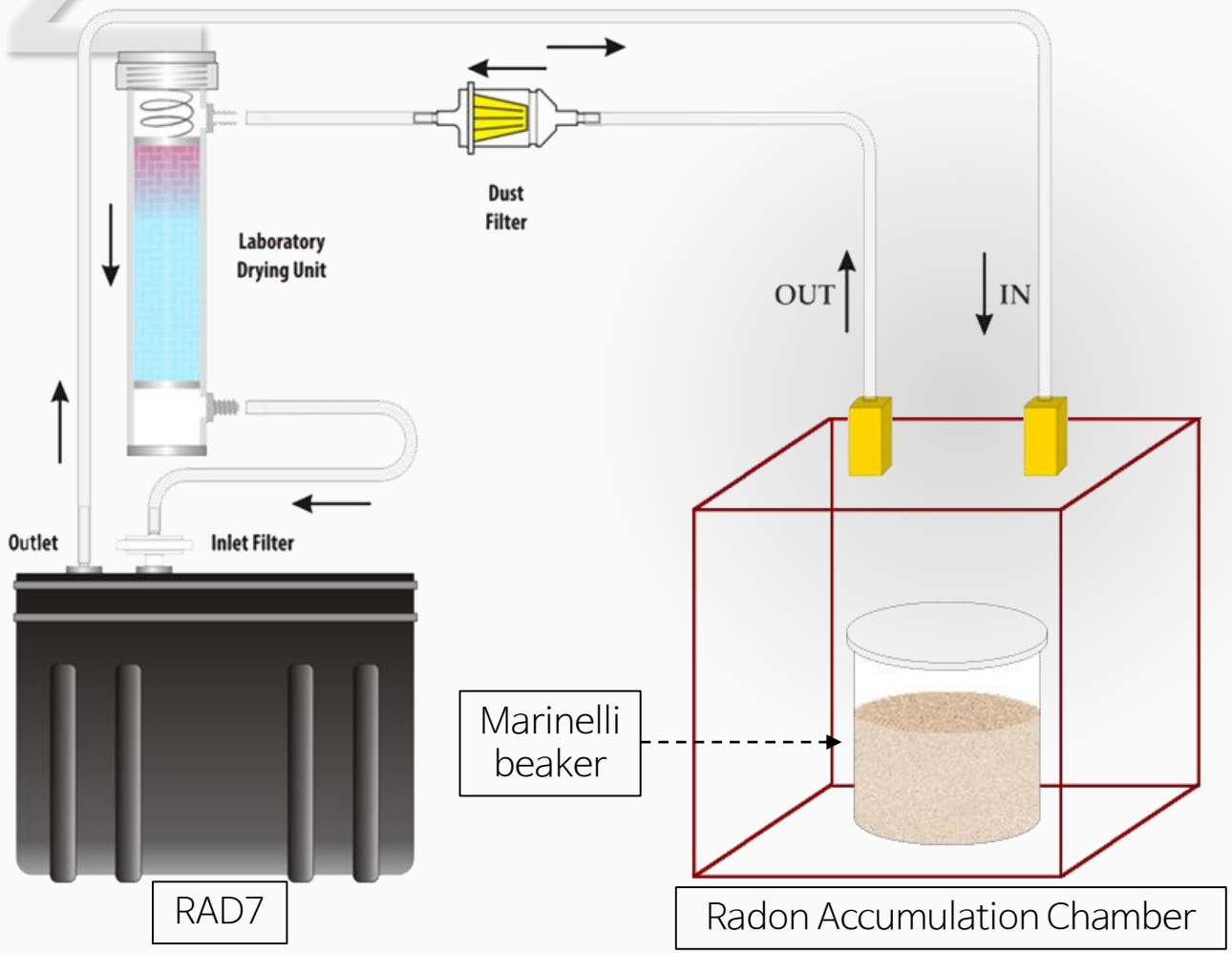


Figure 3. Experimental set up for gamma ray spectrum measurement.

Radon accumulation chamber

- Acrylic material
- Dimension= 30 cm X 30 cm X 30 cm
- Sealed with rubber
- Effective Volume= 23.41L
- Averaged BKGRND air radon = 12.38 Bq/m^3

Chamber tightness test

- Measuring decaying radon concentration in the chamber.
- Radon leakage from chamber was determined by comparing the theoretical decay graph of radon and the measured decay.

Measurement set up

- Radon leakage from Marinelli Beaker was measured by RAD7 detector each of every hour continuously for 21 days

Results and Discussion

HPGe measurement

Results and Discussion

HPGe measurement result

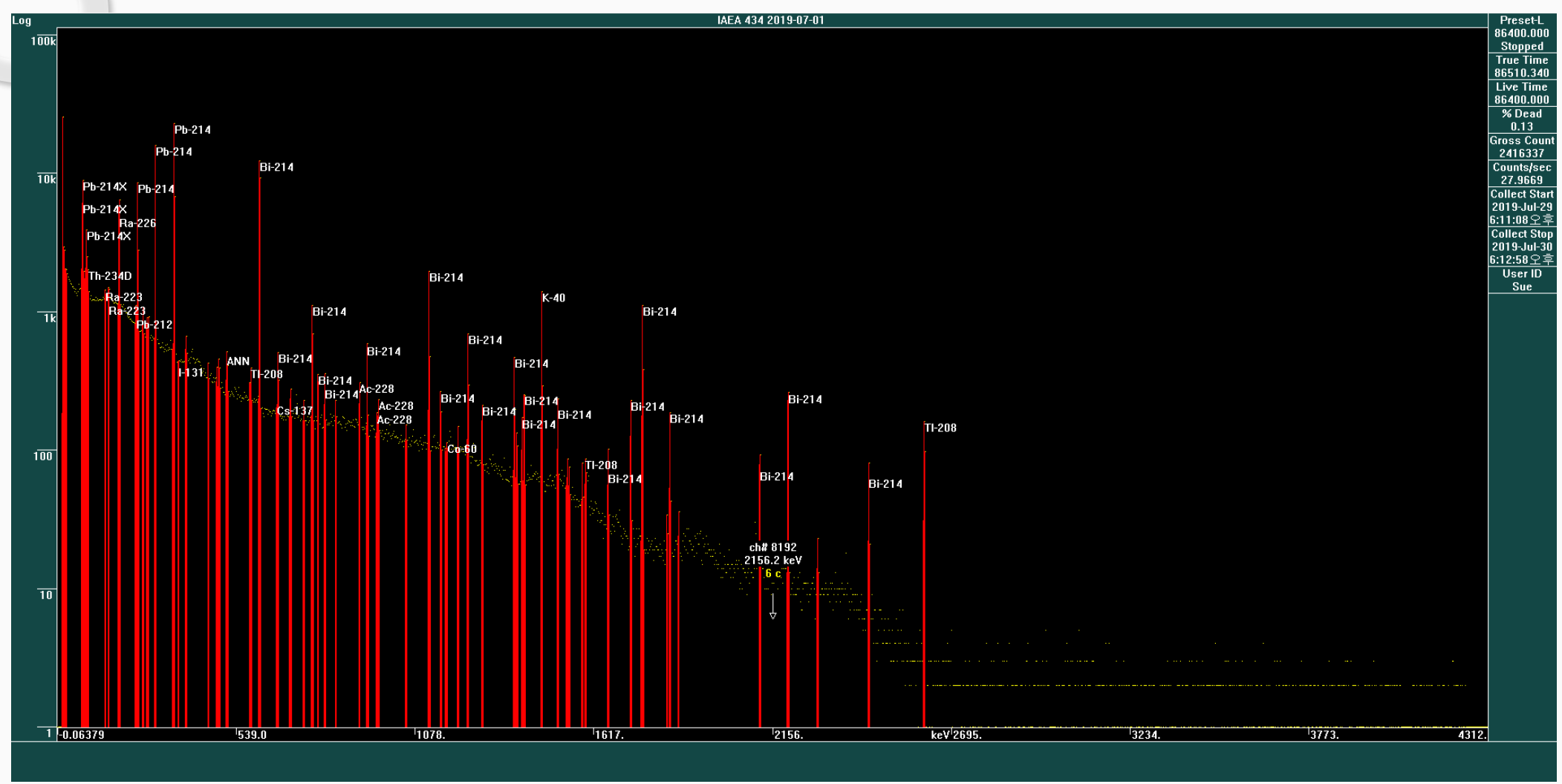


Figure 4. IAEA-434 reference material gamma spectrum measured by HPGe and analyzed by Aptec program

Results and Discussion

HPGe measurement result (IAEA-434)

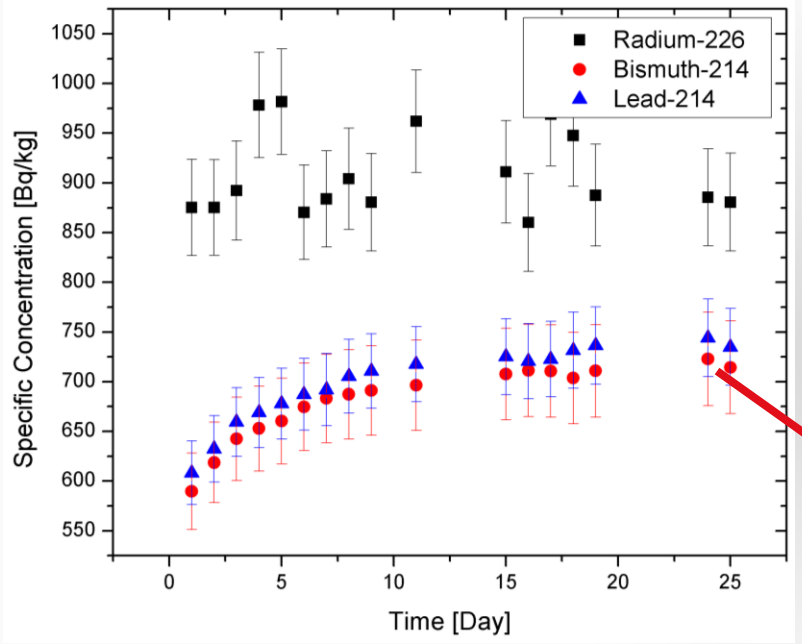


Figure 5. IAEA-434 reference material measurement result (MB3).

Table 2. IAEA-434 reference material description.

Radionuclide	Certified Value [Bq/kg]	Uncertainty
^{210}Pb	680	58
^{226}Ra	780	62
^{230}Th	211	9
^{234}U	120	9
^{238}U	120	11

24th day measured result

^{226}Ra : 885.5 ± 48.8

^{214}Bi : 722.8 ± 47.8

^{214}Pb : 744.2 ± 39.1

Calculated concentration

^{226}Ra : 776.1 ± 62Bq/kg

^{214}Pb : 766.0 ± 61 Bq/kg

2% difference

- Resulted direct measurement value of ^{226}Ra was not agreed well with certified value of IAEA-434 possibly caused by low efficiency of P-type HPGe for lower gamma energy.
- Radon progeny result (indirect measurement) is used for further analysis.

Results and Discussion

HPGe measurement result (Soil)

Better sealing

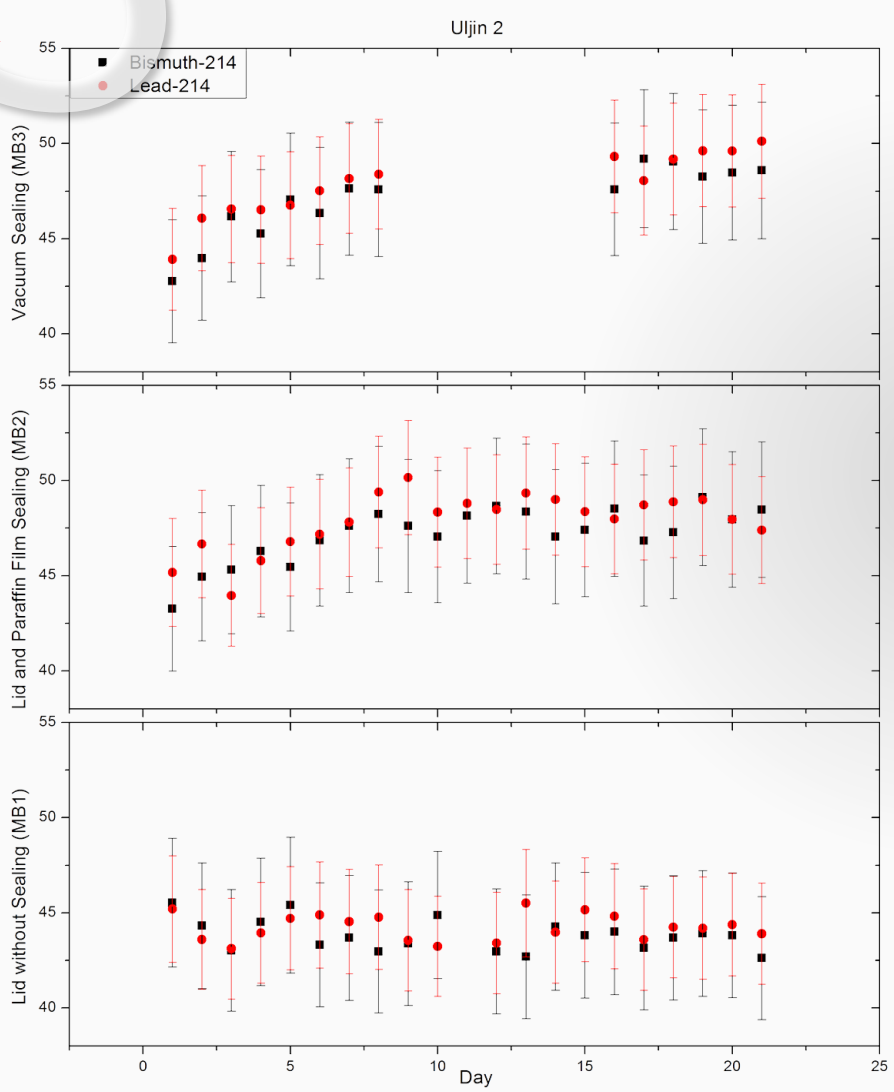


Figure 6. ^{214}Bi and ^{214}Pb build-up for different sealing methods measured by HPGe.

Direct measurement (186 keV of ^{226}Ra)

- Results from 3 different sealing methods show similar results with averaged values which were 108.91, 110.04 and 106.31 Bq/kg respectively for MB1, MB2, and MB3.

← Interference from 185.7 keV peak of ^{235}U .

Indirect measurement (^{214}Bi and ^{214}Pb peaks)

- Results show large discrepancy between ^{226}Ra and ^{222}Rn progenies.

← Radon leakage from beaker so that equilibrium cannot be reached.

- Radon progenies slightly built-up for MB2 and MB3, while for MB1 did not.

Free radon inside leak tight Marinelli beaker (MB3)

$$C_{MB} = EF * C_{Ra} * \frac{m}{V}$$

$$C_{MB} = 0.124 * 50.12\ {}^{Bq}/_{Kg} * 1.156\ {}^{g}/_{mL} = 7184 Bq/m^3$$

Radon Chamber measurement

Results and Discussion

Chamber tightness test result

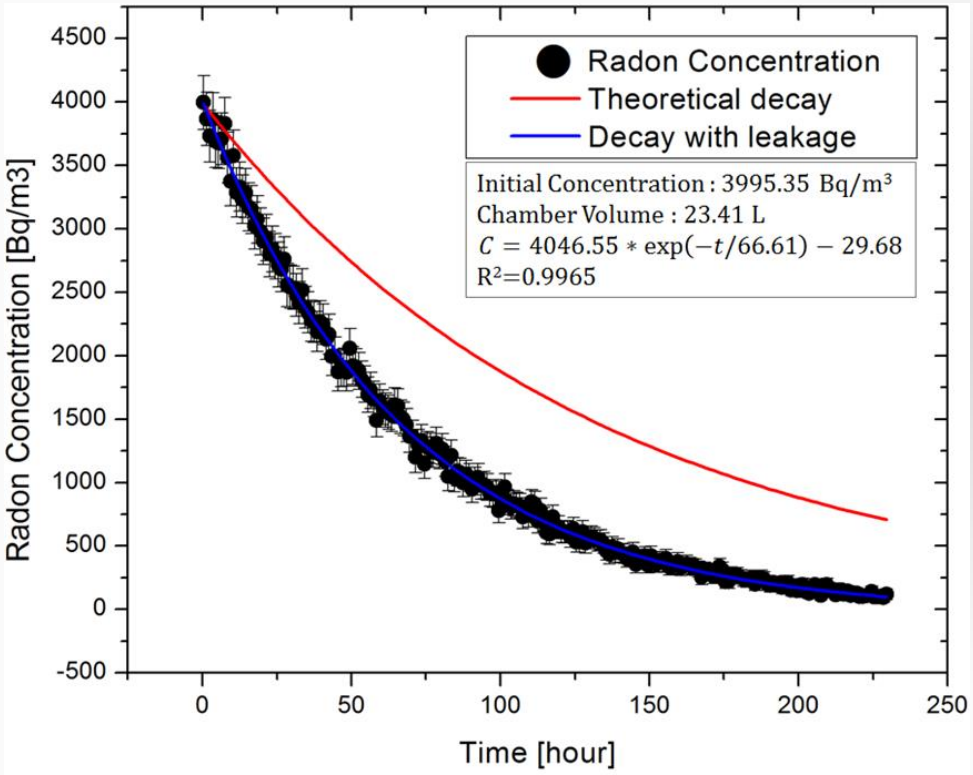


Figure 7. Chamber tightness test result.

Radon Build-up Formula

- The radon accumulation chamber has a non negligible radon leakage, therefore the radon buildup must be corrected using the following equation (Scholten et al, 2013):

$$C_{(t)} = C_\infty \times (1 - e^{-\lambda \times (1+a_v) \times t})$$

$$\bar{C} = C_\infty \times (1 + a_v)$$

- Normalized leak rate:

$$a_v = \frac{\lambda_{eff}}{\lambda_{Rn}} = 1.97$$

Results and Discussion

Radon chamber measurement result (Soil)

$C_\infty = equilibrium\ radon\ concentration\ in\ chamber\ (measured)$
$\bar{C} = non\ leakage\ radon\ concentration\ in\ chamber$
$(calculated\ with\ correction\ factor\ of\ \lambda_V\ )$

Less sealing
More leakage

Leak-rate corrected radon concentration in radon chamber : $C = \bar{C} \times (1 - e^{-\lambda \times t})$

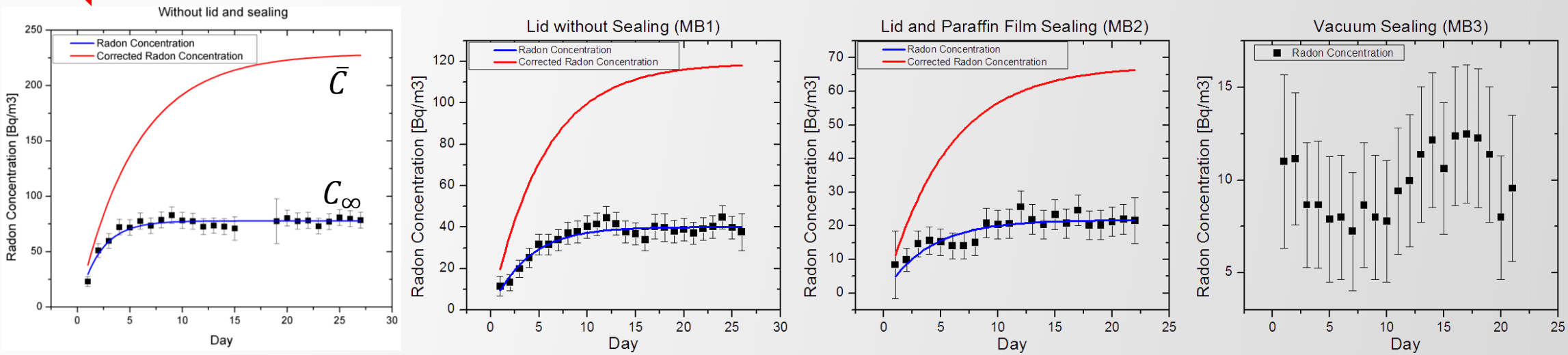
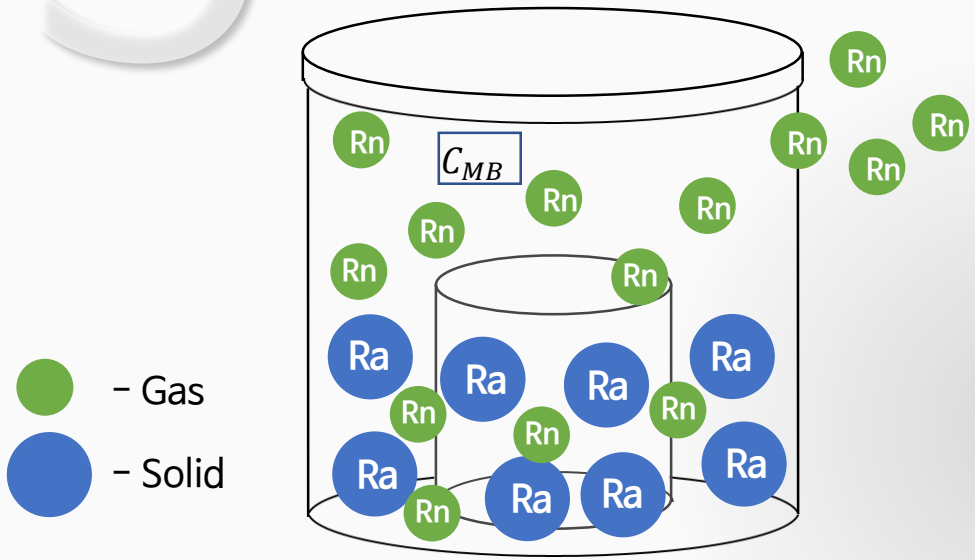


Figure 8. Radon chamber measurement result.

Results and Discussion

Radon chamber measurement result



$$Marinelli\ beaker\ release\ fraction$$
$$MB_{LR} = \bar{C}/C_{MB}$$
C_{MB} = 7184Bq/m³

Table 4. Comparing Marinelli beaker release fraction with 4 measurements

Sample and sealing methods	$\bar{C}$ ($^{Bq}/_{m^3}$)	Marinelli beaker Release Fraction
Uljin1 - MB0	225	0.031
Uljin1 - MB1	115	0.016
Uljin1 - MB2	65	0.009
Uljin1 - MB3	10 (~Background)	≪

Conclusion

04 Conclusion

1. **Direct measurement** and **indirect measurement** by P-type HPGe shows different concentration between soil and reference material which may caused by:
   - Lower efficiency for low gamma energy measurement (〈 200 keV)
   - Interference from other ^{235}U gamma peak to ^{226}Ra gamma peak
   - Radon leakage from Marinelli beaker that inhibit secular equilibrium between Radium and Radon progenies
2. IAEA-434 indirect measurement results using MB3 sealing show very low difference with certified value (±2 %) therefore **MB3 can be considered as radon leak tight**.
3. The ^{214}Pb concentrations in soil obtained by HPGe indirect measurement were **MB3 〉 MB2 〉 MB1**. This result was verified by radon chamber measurement which showed accumulated radon concentration(leaked radon) **MB3 〈 MB2 〈 MB1**.
4. **Radon release fraction (MB_{LR})** were determined as MB3 〈 MB2 〈 MB1. For low ^{226}Ra activity samples the three sealing methods have low radon leakage rate (〈 5 %). Further study will be needed for relatively high ^{226}Ra activity samples.
5. The results show that simple and cheap sealing method using vacuumed plastic bag can effectively minimize the leakage.

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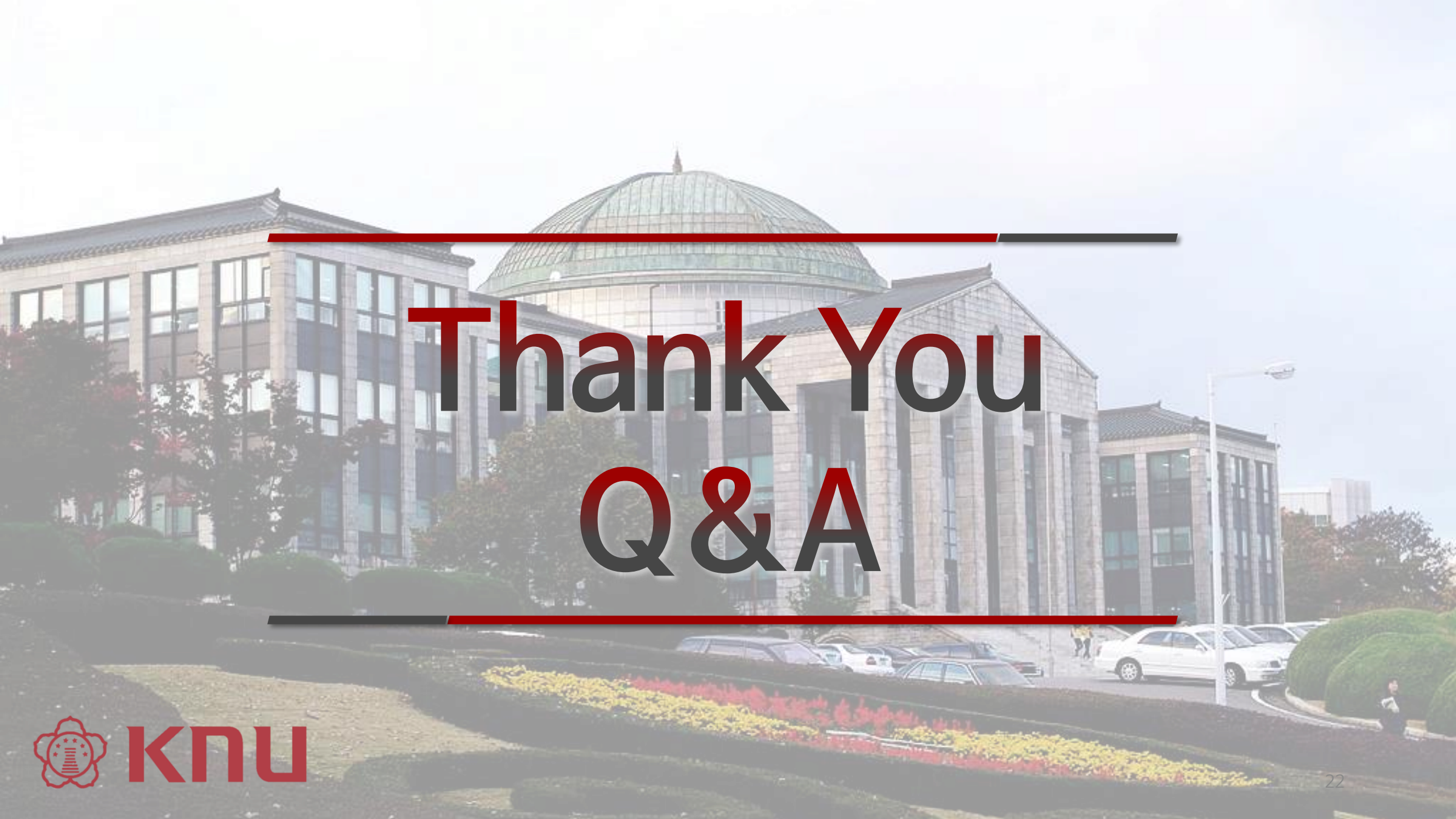
Thank You
Q&A