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Radioactivity Concentration Index Evaluation of Construction Materials by Gamma-ray Spectroscopy

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INTRODUCTION

➢ Natural Occurring Radioactive Material (NORM)

- Construction materials contain NORM such as potassium (^{40}K), long-lived uranium (^{238}U), and thorium (^{232}Th)
- Long-lived ^{238}U and ^{232}Th follow decay chain respectively producing radionuclides (^{222}Rn, ^{228}Ac, ^{220}Rn, etc.) until reaching a stable isotope (^{206}Pb and ^{208}Pb)
- Exhalation of ^{222}Rn from construction materials causes inhalation of ^{222}Rn accounting for more than 50% of annual effective radiation dose [1]
- To manage radon concentration, "Guidelines for reduction and management of radon in building materials" apply radioactivity concentration index to building materials [2]

➢ Assessment of Radioactivity Concentration Index

- Standard gamma-ray spectroscopy method (KS A ISO 18589-2, 18589-3)
  - ✓ Pretreating and sealing samples for 1 month to reach radiative equilibrium

METHODS AND RESULTS

➢ Radioactivity Concentration Index (I) and Gamma-ray Spectroscopy

- Radioactivity concentration of nuclides is calculated from the gamma-ray spectroscopy of three nuclides

$$I = \frac{A_{226Ra}}{300} + \frac{A_{232Th}}{200} + \frac{A_{40K}}{3000} \quad [2]$$

$$A_x = \frac{N_{net,x}}{\eta_x \cdot \varepsilon_{int,x} \cdot T \cdot m} \; (Bq \cdot kg^{-1})$$

(A_x : radioactivity concentration, m : mass)

- ✓ For ^{40}K, it is easier to measure due to relatively shorter half-life, and higher branching-ratio than ^{238}U and ^{232}Th
- ✓ For ^{232}Th and ^{226}Ra, overlapped photopeak exists respectively
  - ➢ In addition to direct determination of radioactivity concentration, indirect determination by analyzing radionuclides in decay chains, using the fact that the radionuclides of the decay chain follow radiative equilibrium (Fig. 1)
  - ➢ Overlapped photopeak at ~186 keV is corrected by determining ^{235}U concentration from both 143.8 keV gamma-ray peak of ^{235}U and natural abundance of ^{238}U concentration [4]
  - ➢ Likewise, photopeak at ~63.5 keV is corrected by determining ^{232}Th concentration from the activity concentration of ^{228}Ac
- Non-destructive gamma-ray spectroscopy has advantages over standard method
  - ✓ Taking less time and more efficient to measure due to no pretreatment and sealing
  - ✓ No structural changes in construction materials
  - ✓ Enabling to compare radioactivity concentration index with ^{222}Rn exhalation

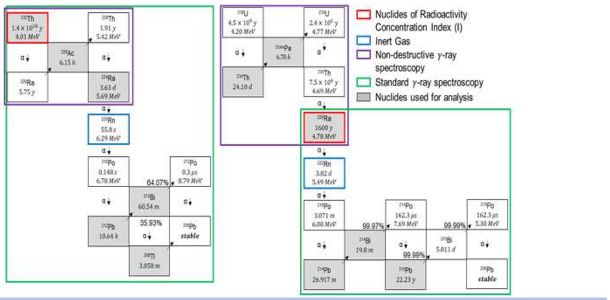

Fig. 1. Decay chain of ^{232}Th (left) and ^{238}Ra (right) : Radionuclides to determine radioactivity concentration index in each method

➢ Measurement System

- Background radiation shielding : lead bricks and 10 mm thickness copper box (Fig. 2)
- Background radiation decreased, and small peaks can be measured (Fig. 3)

Fig. 2. Schematic view (left) and configuration (right) of the system

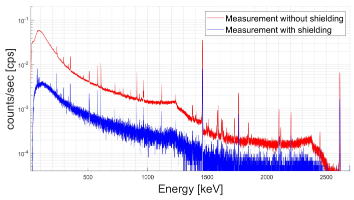

Fig. 3. Effects of shielding systems on the background radiation spectra

- Minimum Detectable Activity (MDA)

$$N_D = 2.706 + 4.653\sigma_B$$

$$MDA = \frac{N_D}{\eta \varepsilon_{abs} T} \quad [3]$$

(σ_B : standard deviation, η : branching ratio, ε_{abs} : absolute peak efficiency, T : measured time)

- ✓ MDA of ^{40}K is decreased by more than half from 59.389 to 24.851 Bq after shielding

	^{226}Ra 186 keV	^{228}Ac 911 keV	^{40}K 1,461 keV
MDA	7.98 Bq	2.68 Bq	24.85 Bq

➢ Detector Calibration

- 9 gamma standard sources : ^{241}Am, ^{133}Ba, ^{109}Cd, ^{57}Co, ^{60}Co, ^{137}Cs, ^{152}Eu, ^{54}Mn, ^{22}Na
- Energy calibration : $Energy = 0.181 \times Channel + 1.433$
- Efficiency calibration
  - ✓ Dead layer of HPGe crystal is corrected to verify the validity of Monte Carlo N-Particle Transport Code (MCNP6.2), according to experiment results (Fig. 4)
  - ✓ Absolute peak efficiency considered for solid angle and self-attenuation by construction materials through MCNP6.2 simulation (Fig. 5)

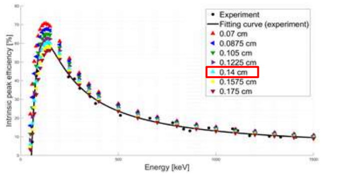

Fig. 4. Intrinsic peak efficiency of the detector by correcting dead layer thickness

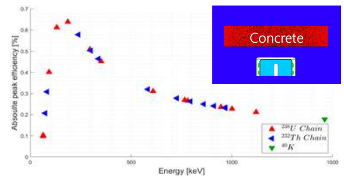

Fig. 5. Absolute peak efficiency of the detector and concrete configuration

➢ Radioactivity Concentration Index (I)

- Non-destructive gamma-ray spectroscopy of concrete sample is conducted.
- Overlapped peaks are corrected such as around 186.2 keV and 63.5 keV
- Activity concentration of ^{226}Ra and ^{232}Th ranges respectively from 56.3 to 74.4 Bq·kg^{-1} and from 42.6 to 61.0 Bq·kg^{-1}
- Activity concentration of ^{40}K is 834.8 Bq·kg^{-1}
- Radioactivity concentration index (I) for concrete sample (size : 20 × 20 × 5 cm^3) ranges from 0.682 ± 0.020 to 0.819 ± 0.028
  - ✓ The index of ^{226}Ra, ^{232}Th, and ^{40}K accounts for 28.7, 34.0, and 37.1% respectively
  - ✓ Although activity concentration index of ^{40}K is more than 10 times higher than that of the others, radioactivity concentration index of ^{40}K has same order with others

	Radionuclide	Decay Chain	Energy (keV)	Branching Ratio (%)	Activity Concentration (Bq/kg)	Radioactivity Concentration Index (I)		
^{226}Ra	^{226}Ra	^{238}U	186.2	3.56	56.3 ± 4.0	0.188 ± 0.013	0.188 – 0.248	0.682 – 0.831
	^{234}Th		63.3	3.75	70.8 ± 16.9	0.236 ± 0.028		
			92.6	5.58	74.4 ± 3.6	0.248 ± 0.012		
^{232}Th	^{232}Th	^{232}Th	63.8	0.26	51.1 ± 2.6	0.256 ± 0.013	0.213 – 0.305	
	^{228}Ac		270.2	3.55	50.9 ± 2.7	0.254 ± 0.014		
			328.0	3.04	42.6 ± 2.9	0.213 ± 0.015		
			338.3	11.40	48.6 ± 0.9	0.243 ± 0.005		
			409.5	2.02	55.7 ± 4.2	0.278 ± 0.021		
			463.0	4.45	51.8 ± 2.0	0.259 ± 0.010		
			794.5	4.25	45.3 ± 2.4	0.226 ± 0.012		
			835.7	1.70	61.0 ± 5.0	0.305 ± 0.025		
			911.2	26.20	49.8 ± 0.7	0.249 ± 0.003		
			964.8	4.99	55.1 ± 2.1	0.275 ± 0.011		
			969.0	15.90	50.5 ± 0.9	0.253 ± 0.005		
^{40}K	^{40}K	-	1460.8	10.55	834.83 ± 4.2	0.278 ± 0.0014	0.278	
-	^{235}U	^{235}U	143.8	10.94	1.83 ± 0.8	-	-	-
			185.7	57.00	-	-	-	-

- Radioactivity concentration index (I) is obtained by several gamma-ray peaks and has discrepancy
- Here are several conjectures of discrepancy in results
  - ✓ 92.6 keV gamma-ray peak of ^{226}Ra is overlapped with X-ray of ^{232}Th
  - ✓ 835.7 keV gamma-ray peak of ^{228}Ac has less precision due to 840.4 keV gamma-ray peak of ^{228}Ac
  - ✓ Peaks with low branching-ratio have more discrepancies
- Accurate results can be obtained by sealing the construction sample to obtain radioactive equilibrium for the Rn decay products

CONCLUSIONS

- Non-destructive gamma-ray spectroscopy is conducted considering self-attenuation effect
- Obtained radioactivity concentration index of concrete sample is less than 1, which is the recommended limit in the guideline
- Validation of non-destructive gamma-ray spectroscopy must be verified in the future.
- As inhalation of ^{222}Rn closely related to concentration of ^{226}Ra, the level of ^{222}Rn exhalation should be compared with the radioactivity concentration index in the future

REFERENCES

[1] L. Fior, et al., Activity measurements of radon from construction materials, Applied Radiation and Isotopes, Vol. 70, Issue 7, pp.1407-1410, 2012.
[2] Ministry of Environment, Ministry of Land, Infrastructure and Transport, Nuclear Safety and Security Commission, "Guidelines for Reduction and Management of Radon in Building Materials", p.2, Nov. 2019.
[3] G. F. Knoll, Radiation Detection and Measurement, John Wiley & Sons, New York, pp.95-98, 2010.
[4] N.P. Petropoulos, et al., Photon attenuation, natural radioactivity content and radon exhalation rate of building materials, Journal of Environmental Radioactivity, Vol. 61, Issue 3, pp.257-269, 2002.

ACKNOWLEDGMENTS

This research was supported by the Korea Land and Housing Corporation (LH).