DETERMINATION OF THE RADON DAUGHTERS CONCENTRATION IN AN ATMOSPHERE AND DISTRIBUTION OF RADON AND DAUGHTERS IN VARIOUS ENVIRONMENTS

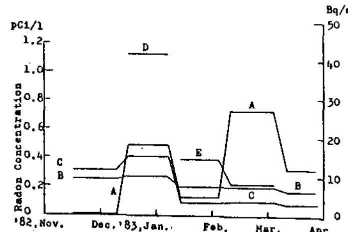
Ryuhei Kurosawa
Science and Engineering Research Laboratory, Waseda University
Tokyo
Tadashi Mutoo
Yoshihisa Kitahara
Power Reactor and Nuclear Fuel Development Corporation
Tokyo

1. Radon and Radon Daughters Concentrations in Various Environment

Internal radiation exposure for members of the public may be mainly caused by the inhalation of radon daughters. The radon and its daughters concentration in an atmosphere is maintained by emanating radon from the soil, building materials and underground water,etc, The observed emanation rate from soil surface, surrounding our laboratory in the Tokyo, is 0.0030—0.0093 $Bq/m^2.s$ and specific activity of radium-226 in that soil is estimated as 30.3 ± 5.2 Bq/kg.

In Japan, a large amount of plaster-board which is including some amounts of artificial phosphogypsum, is used for the important components of dwelling houses instead of wooden board owing its fireproof property. For instance, about 840 Bq/kg of radium-226 is observed in these plaster boards. Therefore, in the case of Japnese dwelling houses, main source of radon is estimated that the remained radium in the plaster board which is supplied from a waste of phosphoric acid manufactures and the probrem of emanated radon from igneous rock, such as granite, or other uranium containing hard materials is not serious source because of the Japanese dwelling house construction.

In the case of an uranium mine,the sources of radon are exhaust air of the mine itself and deposited mill tailings. In the Ningyo-toge mine, the mill tailings and residues of heap leaching are buried to the worked out open pit and covered with the satisfied thickness (:2.8m) of soil grained shale mixture. Therefore,the radon emanation rates from the covered soil surface are indicated comparative low value, averaged of these values is estimated as 0.057 ± 0.012 $Bq/m^2.s$.



A : Wooden house, located at 21 Km NW direction from centre of the Tokyo.
B : Wooden house, located at nearby centre of the Tokyo.
C : Made by prefabricated concrete, located at 19 Km WNW direction from centre of the Tokyo.
D : Closed room of the second basement in concrete building.
E : Tenth floor of the above building.

Fig.1 Variation of the radon concentrations in various dwelling houses of the Tokyo.

The continuous measurement of the averaged radon concentration in an atmosphere of the open cut working area of the Ningyo-toge mine and that of the dwelling houses in vicinity of the Tokyo is carried out by the passive type radon monitors. This monitor consists of 1.4 l stainless steel vessel with the radon removal filter, collecting centre electrode (covered with the thin aluminum foil) and high voltage supply (-800 v). The alpha radiations of the collected radon daughters are recorded on the cellulose nitrate film (LR-115, Type II) which is located inside of the aluminum foil of centre electrode. The collecting efficiency of this monitor is estimated as 70 % by experimentally. The results of the measurement are shown in Fig. 1 and Fig. 2.

The typical results of investigation of the individual radon daughters concentration in the various locations are indicated in Table 1.

Table 1. Radon daughters concetrations in various environments.

Locations	Conditions of atmosphere	Radon daughters concentrations		
		Ra-A Bq/m³	Ra-B Bq/m³	Ra-C Bq/m³
Indoor of dwelling house in western suburbs of Tokyo	Natural ventilation, in winter	6.14± 0.96	3.63±1.33	3.89±0.74
	Under the somewhat obstructed ventilation, in winter	7.07±0.96	6.62±1.37	2.63±0.74
		6.96±1.00	6.88±1.48	3.55±0.74
Outdoor of suburbs of Tokyo	Wind speed is 1-2 m, in winter	3.03±0.67	3.18±1.30	3.29±0.67
Open cut mining area, in Ningyo-toge	Down stream of the wind, without working	29.0± 6.4	13.9± 1.9	12.2± 1.9
Heap leaching site	Wind speed is 1-2 m	16.8± 4.4	0.3 ± 1.0	6.7 ± 1.3
Environmental area	Ningyo-toge area	5.8 ± 3.7	3.3 + 1.0	4.2 ± 1.1

2. New Detecting System for Individual Radon Daughters Concentration

The presented system has been developed to determine the extreme low concentration radon daughters in an atmosphere with the comparble short measuring time. The features of this system are application of the continuous counting during the collecting operation and adoption of the additional beta counting informations of Ra-B and Ra-C to the alpha-spectroscopy technique. The system consists of "Ruggedized" silicon surface barrier (SSB) detector (ORTEC CR-28-450-100), low noise charge sensitive preamplifier, linear amplifier, multichannel or three channels pulse height analyser, special type filter holder and pumping system. The block-diagram is shown in Fig.3. The features of this detecting device are the cleanable front electrode and the much less lightsesitive characteristic than other SSB detectors. The actual depletion layer thickness of this detector is estimated as 198 μm from the resistivity of detector and applied bias voltage. It is sufficient thickness for the detection of beta rays of radon daughters. The detector is mounted in the filter holder (shown in Fig. 4) and be able to detect the emitted alpha and beta rays from the collected daughter nuclides on the membrane filter (millipore DA) during collecting operation. The geometrical arrangement of the detector and the filter is indicated in Fig.5. A typical out put pulses height distribution of the linear amplifier is shown in Fig. 6. These pulses are divided to the

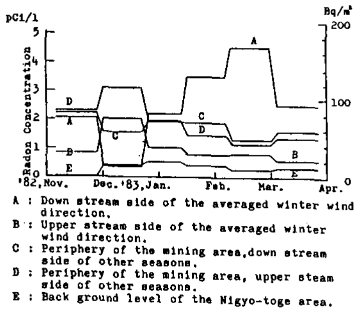


Fig. 2 Variation of radon concentrations at various locations in Ningyo-toge mining area.

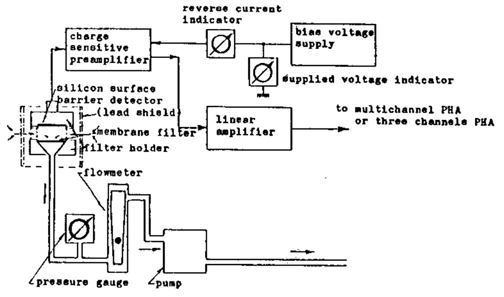


Fig. 3 The blockdiagram of the detecting system.

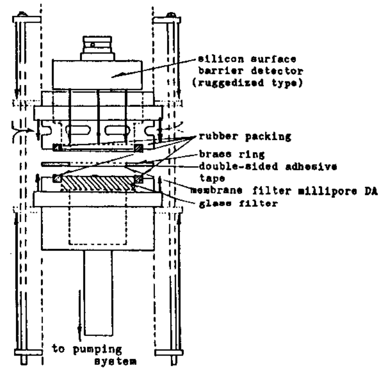


Fig. 4 Sectional view of the special filter holder.

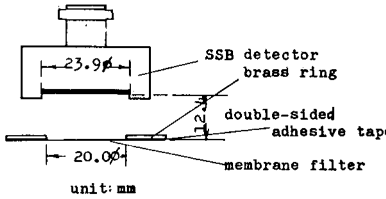


Fig. 5 Geometrical arrangement of the detector and filter.

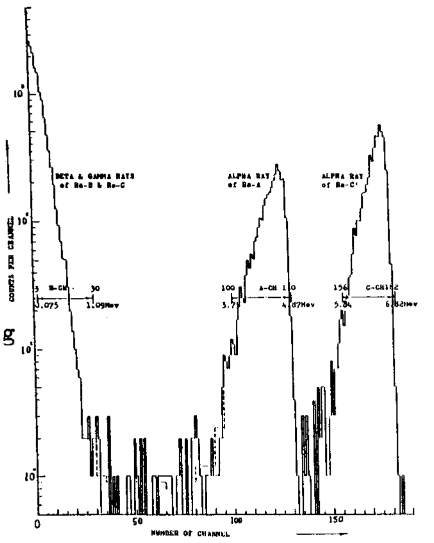


Fig. 6 Pulse height distribution of the detecting system.

following three groups. These groups are (1) beta channel (B-CH, 0.075 to 1.1 Mev-eq), (2) Ra-A alpha channel (A-CH, 3.7 to 5.0 Mev-eq) and Ra-C channel (C-CH, 5.8 to 6.9 Mev-eq). The counts of B-CH provides for the usefull information concerning to the Ra-B and Ra-C activities. The counting efficiency of each of the groups for the various radiations of daughter nuclides are estimated experimentlly as the followings, Each of the efficiency of the A-CH for alpha rays of Ra-A and Ra-C' is 10.61 % ($=\eta_{AA}$) and 0.0799 % ($=\eta_{CA}$), that of B-CH for alpha ray of Ra-A and beta rays of Ra-B and Ra-C is 0.184 % ($=\eta_{AB}$), 8.47 % ($=\eta_{BB}$) and 10.04 % ($=\eta_{BC}$) and that of C-CH for alpha ray of Ra-C' is 10.04 % ($=\eta_{CC}$), respectively. The analytical approachs to the determination of net counts of each of the channels in the appropriate collecting time are obtained from the integrated forms of the Bateman's equations as follows;

$$I_{AA} = \frac{C_a V}{\lambda_a}\left(T_c - \frac{1}{\lambda_a}(1 - e^{-\lambda_a T_c})\right)$$

$$I_{AB} = \frac{C_a V}{\lambda_a}\left(\frac{\lambda_b}{(\lambda_b - \lambda_a)}\left(T_c - \frac{1}{\lambda_a}(1 - e^{-\lambda_a T_c})\right) + \frac{\lambda_a}{(\lambda_a - \lambda_b)}\left(T_c - \frac{1}{\lambda_b}(1 - e^{-\lambda_b T_c})\right)\right)$$

$$I_{AC} = \frac{C_a V}{\lambda_a}\left(\frac{\lambda_b \lambda_c}{(\lambda_b - \lambda_a)(\lambda_c - \lambda_a)}\left(T_c - \frac{1}{\lambda_a}(1 - e^{-\lambda_a T_c})\right) + \frac{\lambda_a \lambda_c}{(\lambda_a - \lambda_b)(\lambda_c - \lambda_b)}\left(T_c - \frac{1}{\lambda_b}(1 - e^{-\lambda_b T_c})\right) + \right.$$

$$+ \frac{\lambda_a \lambda_b}{(\lambda_a-\lambda_c)(\lambda_b-\lambda_c)}(T_c - \frac{1}{\lambda_c}(1 - e^{-\lambda_c T_c})))$$

$$I_{BB}= \frac{c_b v}{\lambda_b}(T_c - \frac{1}{\lambda_b}(1 - e^{-\lambda_b T_c}))$$

$$I_{BC}= \frac{c_b v}{\lambda_b}(\frac{\lambda_c}{(\lambda_c-\lambda_b)}(T_c - \frac{1}{\lambda_b}(1 - e^{-\lambda_b T_c})) + \frac{\lambda_b}{(\lambda_b-\lambda_c)}(T_c - \frac{1}{\lambda_c}(1 - e^{-\lambda_c T_c})))$$

$$I_{CC}= \frac{c_c v}{\lambda_c}(T_c - \frac{1}{\lambda_c}(1 - e^{-\lambda_c T_c}))$$

$$C_A = \eta_{AA} I_{AA} + \eta_{CA} I_{CC}, \quad C_B = \eta_{BB} I_{BB} + \eta_{BC}(I_{BC} + I_{CC}) + \eta_{AB} I_{AA}, \quad C_C = \eta_{CC}(I_{AC} + I_{BC} + I_{CC})$$

Where c_a, c_b and c_c are the averaged concentration (activities) of the Ra-A, Ra-B and Ra-C (=Ra-C'), λ_a, λ_b and λ_c are the decay constant of the Ra-A, Ra-B and Ra-C, v is the collecting rate (= flow rate) and T_c is the collecting period of time (=counting time).
I_{AA} is the activity of Ra-A nuclide, collected as Ra-A.
I_{AB} is the activity of Ra-B nuclide, collected as Ra-A.
I_{AC} is the activity of Ra-C or Ra-C' nuclide, collected as Ra-A.
I_{BB} is the activity of Ra-B nuclide, collected as Ra-B.
I_{BC} is the activity of Ra-C or Ra-C' nuclide, collected as Ra-B.
I_{CC} is the activity of Ra-C or Ra-C' nuclide, collected as Ra-C.
C_A, C_B and C_C are the net counts of A-CH, B-CH and C-CH.

The following equations are provided for the determination of the individual daughter nuclides concentrations and these statistical error estimations.

$c_a = (0.353\ 956\ C_A - 0.002\ 671\ 75\ C_C) \div v$, (collecting time : 10 min.)
$c_b = (-0.023\ 570\ 0\ C_A + 0.256\ 976\ C_B - 0.243\ 423\ C_C) \div v$
$c_c = (0.000\ 871\ 70\ C_A - 0.028\ 765\ 3\ C_B + 0.853\ 747\ C_C) \div v$
$WL = (1.115874\times10^{-4} C_A + 5.388\ 885\times10^{-4} C_B - 1.525\ 183\times10^{-4} C_C) \div v$
$\Delta c_a = (1.252\ 846\times10^{-1} \Delta C_A^2 + 7.137\ 750\times10^{-6} \Delta C_C^2)^{\frac{1}{2}} - v$
$\Delta c_b = (5.555\ 550\times10^{-4} \Delta C_A^2 + 6.603\ 684\times10^{-2} \Delta C_B^2 + 5.925\ 468\times10^{-2} \Delta C_C^2)^{\frac{1}{2}} \div v$
$\Delta c_c = (7.598\ 565\times10^{-7} \Delta C_A^2 + 8.274\ 452\times10^{-4} \Delta C_B^2 + 5.700\ 033\times10^{-2} \Delta C_C^2)^{\frac{1}{2}} \div v$
$\Delta WL = (1.245\ 175\times10^{-8} \Delta C_A^2 + 2.904\ 008\times10^{-7} \Delta C_B^2 + 2.326\ 183\times10^{-8} \Delta C_C^2)^{\frac{1}{2}} \div v$

$c_a = (0.083\ 678\ 7\ C_A - 0.000\ 631\ 61\ C_C) \div v$, (collecting time: 30 min.)
$c_b = (-0.007\ 897\ 8\ C_A + 0.033\ 444\ 9\ C_B - 0.031\ 644\ 4\ C_C) \div v$
$c_c = (0.000\ 389\ 89\ C_A - 0.010\ 390\ 96\ C_B + 0.038\ 740\ 5\ C_C) \div v$
$WL = (2.137\ 362\times10^{-5} C_A + 5.884\ 617\times10^{-5} C_B - 6.875\ 371\times10^{-6} C_C) \div v$
$\Delta c_a = (7.002\ 131\times10^{-3} \Delta C_A^2 + 3.989\ 274\times10^{-7} \Delta C_B^2)^{\frac{1}{2}} \div v$
$\Delta c_b = (6.237\ 507\times10^{-5} \Delta C_A^2 + 1.118\ 558\times10^{-3} \Delta C_B^2 + 1.001\ 371\times10^{-3} \Delta C_C^2)^{\frac{1}{2}} \div v$
$\Delta c_c = (1.520\ 155\times10^{-7} \Delta C_A^2 + 1.079\ 720\times10^{-4} \Delta C_B^2 + 1.500\ 830\times10^{-3} \Delta C_C^2)^{\frac{1}{2}} \div v$
$\Delta WL = (4.568\ 316\times10^{-10} \Delta C_A^2 + 3.462\ 872\times10^{-9} \Delta C_B^2 + 4.727\ 073\times10^{-11} \Delta C_C^2)^{\frac{1}{2}} \div v$

Where unit of c_a, c_b and c_c is the dpm/l, and v is the l/min. ΔC_A, ΔC_B and ΔC_C are the statistical counting error of the A-CH, B-CH and C-CH, and WL is the working level.

The relative counting error of B-CH is somewhat larger than that of other channeles owing high back ground counts of this channel. The back ground counts of B-CH is 6to7 cpm under the without shielding and is 2 to 3 cpm under the lead shielding. Some calculated precisions of the presented method is indicated in Table 2.

Table 2. The precisions for the various concentrations and collecting times (v = 16 l/min, back ground counts of the B-CH = 7 cpm.)

Concentrations	Collecting time	Ra-A	Ra-B	Ra-C	WL
1:1:1	10 min.	±18.2 %	±41 %	± 17.3 %	± 17.8 %
11.1 Bq/m³	30 min.	± 9.1 %	±12.9 %	± 8.7 %	± 4.4 %
0.002 94 WL	60 min.	± 6.3 %	± 5.9 %	± 6.7 %	± 1.8 %
1.11 Bq/m³	10 min.	±57.8 %	——	± 62.5 %	——
	30 min.	±28.9 %	±73.7 %	± 33.5 %	± 27.9 %
0.000 294 WL	60 min.	±20.0 %	±32.6 %	± 25.8 %	± 10.0 %