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NEW DETERMINATION METHOD OF RADON-220 IN MINERAL SPRINGS BY EXTRACTION-LIQUID SCINTILLATION COUNTER

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^{220}Rn in mineral springs was extracted into toluene. The α and β or γ rays spectra of ^{220}Rn and its descendants in toluene were measured by LSC or γ spectrometer and the establishment of equilibrium between ^{212}Pb and its descendants was confirmed. Based on this fact a new determination method of ^{220}Rn is developed, consisting of the extraction of radon and the application of integral counting with LSC to ^{212}Pb in radioactive equilibrium with the descendants.

There have been a few studies to determine the content of ^{220}Rn (hereafter called as Tn, thoron) in mineral springs[1)2)3)] by using IM type fontactoscope or KY type Lauritzen electroscope. However, the determination was extremely difficult due to the short half-life of Tn (54.5 seconds). The fact that Tn is often found in mineral springs in spite of its extremely short life makes the authors suspect that it is in equilibrium with parent nuclides having longer half-lives, probably with ^{224}Ra(3.64d). The authors estimated that the equilibrium between ^{228}Th(1.91y) and ^{224}Ra and between ^{224}Ra and Tn establishes in 27.8 days and 11.4 min. respectively. Thus thoron seems to occur more frequently and abundantly in mineral springs than has ever been reported.

The authors have proposed a new determination method of radon-222 in mineral springs which consists of extraction of radon with toluene(PPO 4g/l and POPOP 0.1g/l, hereafter called LS) and the integral counting with a liquid scintillation counter (LSC)[4)]. Thoron, being an isotope of radon, should be accompanied with radon-222 when extracted into LS for determination. The authors attempted to develop a new determination method of thoron following the similar procedures mentioned above.

The formation of decay products (^{212}Pb γ ray 0.239 MeV, 40%) of Tn extracted into LS was examined by γ ray spectrometer in the same way[5)]. Fig. 1 shows the growth decay curve of ^{212}Pb(ThB, 10.64 hrs) obtained by continual counting with the LSC. The rapid

decrease of counting rate in the initial period and the gradual attainment of maximum suggests that the extracted Tn and daughter ^{216}Po(ThA) disappear quickly, and ThB and its descendants establish a radioactive equilibrium in 3.67 hours after the extraction.

The energy spectrum of Tn and its descendants formed in the LS was investigated with LSC connected with 2048 channels pulse height analyzer. The α ray spectra of Tn (6.280 MeV) and ^{216}Po (ThA, 6.775 MeV) measured immediately after the extraction shows a strong single peak (Fig.2). The spectrum obtained 26 hours after the extraction shows that both Tn and ThA have disappeared, and that two α ray peaks of ^{212}Bi(ThC, 6.047 MeV, 33.7%) and ^{212}Po(ThC' 8.780 MeV,66.3%) appeared. β ray spectra observed was accounted by the formation of ThB, ThC (66.3%) and ThC" (^{208}Tl,33.7%) which forms the basic part of these two α ray spectra. ThB was found to reached the radioactive equilibrium with its descendants in about 4 hours. This establishment of the equilibrium suggests that Tn can be determined from ThB which has a remarkably longer half-life (10.64 hrs) compared to that of Tn (54.5 seconds). The activity of equilibrium mixture of ThB was measured by the integral counting method, which gives their absolute counting rate (1α + 2β). The authors[6)7)] has successfully proved the potentiality of this method in the determination of radon. The sensitivity of counting was three times better than counting only α particle of Tn.

Now the following procedures are developed taking into account the possibility of simultaneous extraction of thoron and radon. One litre sample water was shaken with LS of 25 ml. LS layer was carefully transferred into a counting vial[8)]. The temperature of water on extraction and the volume of LS transferred were recorded. The integral counting was started 4 hours after the extraction. The radioactive equilibrium was established for both Tn and Rn when the counting was started. The initial time of the decay curve was chosen at 4 hours after the extraction.

The conventional procedure was used to analyse the composite decay curve. There was a large difference of half-lives between Rn and ThB (Fig. 3). The linear part of composite decay curve shows the activity of Rn only. This part extrapolated backward to the initial time and subtracted from the composite decay curve gives the decay curve for ThB. This decay curve of ThB was then extrapolated backward to get an apparent counting rate of ThB, N cpm. The corrected counting rate of ThB, N_3(dps) was obtained by the following equation.

$$N_3(\mathrm{dps}) = \frac{N}{3 \times 60} e^{\lambda t} \frac{\lambda_3 d \cdot B}{(1-e^{-\lambda_3 d}) E_{ff} \cdot A \cdot C} \quad \cdots\cdots\cdots\cdots\cdots\cdots (1)$$

where t: time at the moment of counting from the initial time, $\frac{\lambda_3 d}{1 - e^{-\lambda_3 d}}$: correction term for decayed activity during counting period d, A : correction term for LS recovery, B : correction term specified for the experimental system used[4], accounting for solubility of radon in water and toluene and the fraction of activity distributed into air layer in counting vial, C : correction term to normalize the volume of sample water to one liter, E_{ff} : counting efficiency (=1) λ_3 : decay constant of ThB.

The relation between the number of Tn atom N_1^0 on the time of extraction and that of ThB 4 hours after the extraction can be approximatly given by Bateman's equation[9]. The number of Tn and ThA atoms which appeared in the first two terms in the equation, can be neglected against that of ThB. The equation is expressed as,

$$N_3(\text{dps}) = \lambda_3 N_1^0 \times 0.7717 = 1.396 \times 10^{-5} N_1^0 \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

The activity of Tn can be expressed in Ci unit with the value N_3.

$$\text{Tn(Ci)} = 2.46 \times 10^{-8} N_3 \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

When some difficulties were met in analyzing the composite decay curve or when it was required to obtain the results in a short time, the Bunney plot method[10] can be utilized (Fig. 4). The activity of Tn can also be calculated with these equations from the value obtained by the plot.

In Table 1, the amount of Tn in mineral springs determined by the present method is shown with the amount of Rn determined simultaneously. The present method enables us to determine the short-lived nuclide Tn with high accuracy and good feasibility. This was accomplished since the method is based on the radioactive equilibrium of ThB which decays with long half-life of 10.64 hrs. In case 6.8×10^{-8}Ci/l of Tn is present in the sample, the counting can be done at longest about 71 hours after the extraction. The present method is simple and is very powerful. With this method, the absolute counting can be made three times more sensitive. The details of the new method will be reported elsewhere[11].

Finally the occurrence of actinon (half-life 3.92 sec) must be mentioned. Actinon is a Rn isotope of actinum series and may also be extracted into LS. However, the effect of this isotope was negligible. The natural abundance of An is small and its short half-life results in the rapid disappearance of the descendants.

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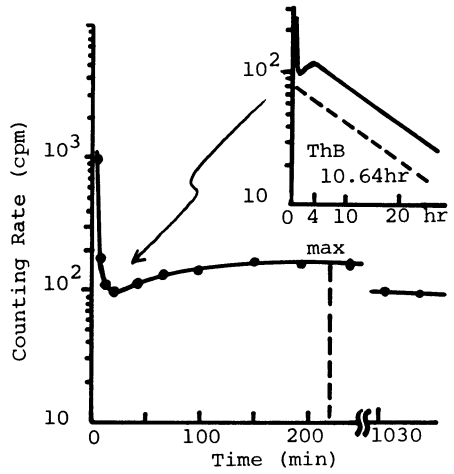

Fig 1 Growth decay curve of Rn-220 with its descendants (LSC).

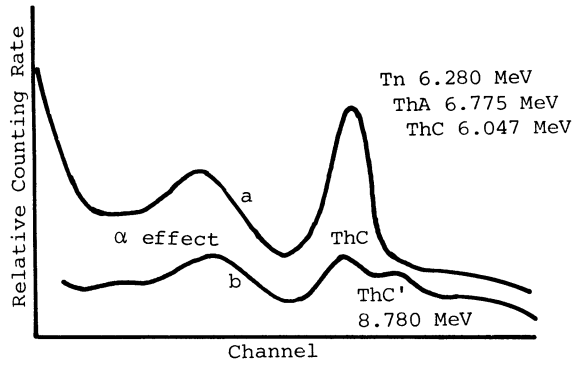

Fig 2 α,β-spectrum of Tn and its descendants obtained by liquid scintillation counter connected with 2048 channels pulse height analyzer.
a: few sec after the extraction
b: 26 hrs. after the extraction

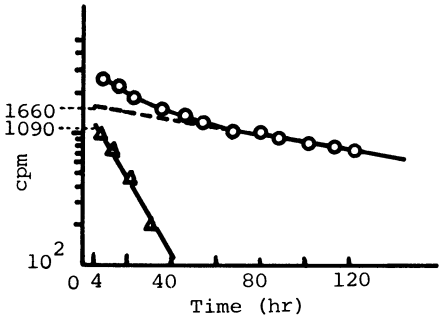

Fig 3 Analysis of Composite Decay Curve of Rn + Tn Mixture

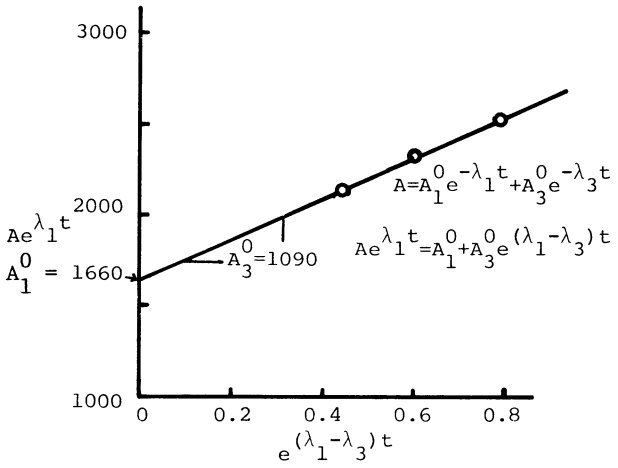

Fig 4 Example of Bunney Plot

Table 1. Tn and Rn in mineral springs determined by Extraction-LSC Method

Name of Springs	Orifice temp.°C	at the initial time ThB cpm	at the initial time Rn cpm	at the extraction time Tn $\times 10^{-8}$Ci/l	at the extraction time Rn $\times 10^{-10}$Ci/l
Hisuinoyu	29.0	5012.2±178.7	21647.3± 52.2	68.5 ±2.4	20.1 ±0.1
Ohashi nakanoyu	30.0	6863.4±188.9	23155.1± 54.0	93.8 ±2.6	21.5 ±0.1
Ichiyoso A	30.3	3622.0±148.2	91758.6±107.4	49.5 ±2.0	85.2 ±0.1
Kawaranoyu	25.2	2568.3± 40.2	5632.6± 26.7	35.1 ±0.6	5.23±0.02
Furokaku iwaburo	22.5	266.3± 50.5	11092.9± 37.4	3.64±0.7	10.3 ±0.03
Koyabara	39.0	334.4± 39.5	6429.6± 28.5	4.57±0.5	5.97±0.03
Ikeda No. 3	19.0	3139.0±322.7	441561.7±235.4	42.9 ±4.4	410.0 ±0.2
Shigaku No. 4	37.5	103.9± 19.1	1443.2± 13.7	1.42±0.3	1.34±0.01
Fujinoyu	47.0	166.8± 43.1	7754.3± 31.3	2.28±0.5	7.20±0.03

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