NEW THREE-COUNT TECHNIQUE FOR SHORT-LIVED RADON DECAY PRODUCTS IN AIR

Deyuan Tian[1] and Zhizhao Lu[2]

[1]Lab. of Industrial Hygiene, Ministry of Public Health, Beijing 100088
[2]Suzhou Medical College, Suzhou 215007, Jiangsu, People's Republic of China

ABSTRACT

Up to the present, radon and its short-lived decay products in air are usually monitored by means of α detection. But radon progenies, including RaB (^{214}Pb) and RaC (^{214}Bi) which are β and γ emitters, contribute about 90 % to the equilibrium equivalent radon concentration (EECRn). Therefore, this paper introduces a new three-count technique by a β detector in the light of radioactive decay law and its boundary conditions during sampling and counting times to solve the Bateman equation. β (even low level β) instruments have been fairly popularized domestically and internationally. It can be used not only as an instrument for radon and its daughters in air, but also as a monitor for β airborne activity in the environment. This new method taps further the latent power of the present instrument and realizes various uses for a unit.

Keywords : radon, three-count technique, equilibrium equivalent radon concentration (EECRn), thoron

INTRODUCTION

It is well known that natural radon and its short-lived decay products in airborne state may cause serious radiation harm to human health such as inducing lung cancer to those who were exposed to them for a long period. Therefore, detailed studies for how to monitor them were conducted. Today, along with the improvements in science and technology, the monitoring procedures for radon progenies have made an encouraging progress. Tsivoglou et al.[1] proposed the Tsivoglou method (a three-point method) for monitoring radon daughters in air on the basis of the count-ratemeter in 1950s. Thomas[2] then improved the three-count technique (or modified Tsivoglou method) on the foundation of the scaler in 1970s. In addition, there are some rapid methods for monitoring the radon progenies in air[3], which are all by means of α-particle detection. However, β and γ rays are emitted in the decay process of radon daughters and reports concerning the detection method of them were not seen so far.

PRINCIPLE

Radon and its short-lived decay products emit α, β and γ rays during the decay process. Their main decay diagram is shown in Fig.1. Radon (^{222}Rn) decays to form ^{218}Po (RaA), ^{214}Pb (RaB), ^{214}Bi (RaC), ^{214}Po (RaC′), ^{210}Pb(RaD), etc., consecutively. The former four decay products are called radon short-lived daughters because their half-lives are less than 30 min. RaD and the subsequent nuclides have much longer half-lives, for instance 22.3 y for RaD. They are quite stable as compared with the former ones. For this reason, it can be regarded as if RaD and the latter are not to decay, and the multi-progeny decay for radon and its daughters may be simplified to a four-progeny decay. On the other hand, the half-life of nuclide RaC′ is very short, only 164 μs. The radioactivity equilibrium between RaC and RaC′ appears right away. It can be said that RaC is simultaneously emits one β- and one α-particles and decays immediately to RaD, not through RaC′. Hence, the four-progeny decay is further simplified to a three-progeny decay.

$$^{226}Ra \xrightarrow[1602a]{\alpha,\ 4.6019MeV(5\%);\ \alpha,\ 4.7846MeV(95\%)} {}^{222}Rn(Rn) \xrightarrow[3.825d]{\alpha,\ 5.489MeV} {}^{218}Po(RaA) \longrightarrow$$

$$\xrightarrow[3.05min]{\alpha,\ 6.0026MeV} {}^{214}Pb(RaB) \xrightarrow[26.8min]{\beta,\ \gamma} {}^{214}Bi(RaC) \xrightarrow[19.7min]{\beta,\ \gamma} {}^{214}Po(RaC') \rightarrow$$

$$\xrightarrow[164\ \mu s]{\alpha,\ 7.687MeV} {}^{210}Pb(RaD) \xrightarrow[22.3a]{\beta,\ \gamma} \ldots\ldots$$

Fig. 1 Main decay diagram of radon and its short-lived decay products

Radon short-lived progenies emit two α-particles in all. For this reason, radon daughter monitoring in air is almost by means of α-particle detection because of its high α-energy and α-pulse signal amplitude, very low background, and the simple structure and easy manufacture of α-detector. On the other hand, the equilibrium equivalent radon concentration (EECRn) is given by:[4]

$$EECRn = 0.105\ C_{RaA} + 0.516\ C_{RaB} + 0.379\ C_{RaC}\ Bq/m^3 \quad (1)$$

where C_{RaA}, C_{RaB} and C_{RaC} are the concentrations of radon progenies, RaA, RaB and RaC, respectively. If the three radon daughter concentrations in air are known, EECRn can be obtained immediately from Eq. (1), and their dose to the public can be evaluated. The RaC′ concentration, $C_{RaC'}$, does not contribute to EECRn as seen from Eq. (1).

Two radon progenies, RaB and RaC, which contribute about 90 % to EECRn are not α-emitter, but are both β and γ emitters. Hence, it is unsuitable to say that α-particle counting for detecting RaA and RaC′ is the best choice because radiation doses are caused mainly by α-radiation. On the contrary, β or γ counting is an optimal selection.

Based on the radioactive decay law and relevant boundary conditions during sampling and counting periods, the Bateman equations were solved and got integral gross β counting which includes three unknowns hiddenly, namely the RaA, RaB and RaC concentrations in air. That is a ternary first-order equation. It is well known that only a set of three simultaneous equations can be solved to get a sole answer when there are three unknowns in it. For this reason, three equations with three observed values are required. Three activity measurements on the filter (i.e., three countings) are to be conducted in the experiment for three different time intervals after the end of sampling. Hence, this method is called the three-count technique. A set of simultaneous linear equations obtained from three countings was set up and solved by the inverse matrix method. The formulas for radon progeny concentration in the air to be measured by the gross β coun-ting method are derived via a computer. According to the UNSCEAR definition[5], the formula of Potential Alpha Energy Concentration (PAEC) is led in. The formula coefficients are a series of complicated exponential operations which are functions of sampling and counting times. A computer program was written to calculate all coefficients rapidly so long as each time parameter inputs are given. These coefficients are simplified constants and do not need to be computed every time for a certain procedure with fixed time parameters.

However, it is possible not to start and/or stop the countings on time in actual situations for a variety of reasons. The data will become invalid at the moment. It is well known that the radon and its daughter concentrations in air change with time and place. Reappeared them is impossible yet even though they are determined again. Therefore, the above-mentioned program is onlined to realize the automatic measurements. Output data from various procedures can be typed out immediately in the light of both relative parameters and measured gross β countings.

RESULTS AND DISCUSSION

To compare conveniently with the Thomas three-count technique[2], the same procedures as his were used, i.e., 5 min sampling time and 2~5, 6~20 and 21~30 min countings after the end of sampling to obtain the three gross β countings, N_1, N_2 and N_3, respectively. If the total β counting efficiency G (including the instrument and filter efficiencies, self-absorption and backscattering corrections on the filter, etc.), sample flow rate V L/min and background counting rate N_o cpm (counts per minute) are known, the formulas for the radon progeny concentrations, PAEC and EECRn are:

$$C_{RaA} = (-60.9933N_1 + 29.6053N_2 - 27.9909N_3 + 20.4233N_o) / GV \ Bq/m^3 \quad (2)$$

$$C_{RaB} = (+8.50301N_1 - 4.71163N_2 + 5.32176N_3 - 7.44207N_o) / GV \ Bq/m^3 \quad (3)$$

$$C_{RaC} = (-3.08556N_1 + 2.90511N_2 - 3.84836N_3 + 3.22043N_o) / GV \ Bq/m^3 \quad (4)$$

$$PAEC = (-175.253N_1 + 97.9325N_2 - 90.9556N_3 - 26.6961N_o) / GV \ 10^{-10} \ J/m^3 \quad (5)$$

$$EECRn = (-3.18617N_1 + 1.77840N_2 - 1.65155N_3 - .475117N_o) / GV \ Bq/m^3 \quad (6)$$

Generally, the flow rate V, efficiency G and background rate N_o are determined previously, and their errors can be managed to be reduced as much as possible or neglected. Statistical counting errors can not be decreased and are dominant in the environmental level. The resultant concentration uncertainties are shown below according to the error resultant rule when only statistics are considered:

$$S_{RaA} = \sqrt{3720.19N_1 + 876.476N_2 + 783.490N_3 + 268734N_o} / GV \ Bq/m^3 \quad (7)$$

$$S_{RaB} = \sqrt{72.3011N_1 + 22.1994N_2 + 28.3211N_3 + 7295.81N_o} / GV \ Bq/m^3 \quad (8)$$

$$S_{RaC} = \sqrt{9.52067N_1 + 8.43965N_2 + 14.8099N_3 + 2939.46N_o} / GV \ Bq/m^3 \quad (9)$$

$$S_{PAEC} = \sqrt{30713.5N_1 + 9590.78N_2 + 8272.93N_3 + 2826321N_o} / GV \ 10^{-10} \ J/m^3 \quad (10)$$

$$S_{EECRn} = \sqrt{10.1517N_1 + 3.16270N_2 + 2.72761N_3 + 932.190N_o} / GV \ Bq/m^3 \quad (11)$$

where S_{RaA}, S_{RaB}, S_{RaC}, S_{PAEC} and S_{EECRn} are the resultant uncertainties for the RaA, RaB and RaC concentrations, PAEC and EECRn in the air, respectively.

The experiments showed that the results of this new three-count technique agree well with that of the Thomas' method.

SUMMARY

Almost all monitors and dosimeters commercially available now for radon and its progenies in the air employ generally the method of detecting α-particle. Their efficiencies are low and detector areas are small normally although the method is good. It means these instruments are well suited for integrated or continuous long time measurements, but not for the rapid and sensitive grab ones at low levels in the environment. This paper develops for the first time a new three-count technique by gross β countings to determine the radon daughters in the air. The formula for calculating EECRn is led firstly in so as to realize the combination monitoring of radon and its progenies. This sdudy fully utilizes the high efficiency and large area of the β detector, and is a new pioneering work in radon and its daughter monitoring in the air. The new method taps further the latent power of these β instruments and realizes the various uses for a unit.

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