The Air Luminescence Count for the Rapid Determination of ^{222}Rn in a Liquid Scintillation Spectrometer

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A liquid scintillation spectrometer has been applied to the determination of the counting efficiency for the air luminescence produced by standardized ^{222}Rn and its daughters. Based on the counting efficiency, known amounts of ^{222}Rn can be prepared in 3.5 h. Moreover, the ^{226}Ra source from which ^{222}Rn is collected does not necessary have to be a standard one. In contrast, the conventional method for preparing known amounts of ^{222}Rn usually requires a long waiting period (ca. 15–30 days).

The passage of α-particles in air results in the ionization and excitation of nitrogen.[1] Several α-emitters in air was determined by measuring the luminescence from excited nitrogen with a liquid scintillation spectrometer.[2–4] However, the counting efficiency for ^{222}Rn and its daughters (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po) was determined with a specially modified liquid scintillation spectrometer which could not be universally utilized. In the present study, therefore, ^{222}Rn and its daughters were standardized by measuring γ-rays from ^{214}Pb with a well Ge detector, and transferred into air in the counting vial. Then, counting efficiency for the air luminescence was determined with a commercially available liquid scintillation spectrometer giving much attention to Cherenkov counting efficiency due to β-particles from ^{214}Pb and ^{214}Bi and the counting efficiency for the short-lived daughters, some of which are supposed to be attached to wall of the counting vial.

Until now, ^{222}Rn collected from a standard ^{226}Ra source was determined based on the secular equilibrium between ^{226}Ra and ^{222}Rn. However, the determination using the standard ^{226}Ra source requires a long waiting period (ca.15–30 days) to grow ^{222}Rn from the ^{226}Ra source. In addition, it is necessary to confirm that all of ^{222}Rn from the ^{226}Ra source is removed. A significant advantage of the proposed method is that the time required for the preparation of known amounts of ^{222}Rn is only 3.5 h. In addition, it must be emphasized that the ^{226}Ra source from which ^{222}Rn is collected, does not necessary have to be standardized one. This is because we do not determine ^{222}Rn based on the secular equilibrium between ^{226}Ra and ^{222}Rn, but need only standardize the ^{222}Rn that is actually transferred and stored in the counting vial by measuring the air luminescence count. Therefore, we need not confirm that all of the ^{222}Rn from the ^{226}Ra source was removed. Exactly the luminescence in this study should be termed "nitrogen luminescence". However, "air luminescence" was employed in conformity with the expression used in the preceding papers.[2,3]

Standardization of ^{222}Rn sample was carried out as follows: ^{222}Rn was collected from the air space above $^{226}RaCl_2$ solution with a syringe and added to a quartz pipe of 7-mm internal diameter and 40-mm long, both ends of which were closed silicon rubber stoppers. ^{222}Rn in the pipe was allowed to remain for 3.5 h before measurements. During the time, the ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po came to transient equilibrium with ^{222}Rn. Then the activity of ^{222}Rn was determined through its daughter ^{214}Pb: γ-rays from ^{214}Pb were measured with a Ge well detector, Model GCW1521 (Canberra Industries Inc., CT, USA). This system had a 1.9-keV resolution (FWHM) at 1.33 MeV and relative efficiency of 17.1%. The detector was absolutely calibrated using a set of DAMRI/LMRI γ-ray standard sources (Saclay, France).

The energy and abundance of γ-ray from ^{214}Pb used for the determination were 0.3519 MeV[5] and 36.74%,[5] respectively. The full energy peak efficiency for 0.3519 MeV γ-ray was 25.4 ± 0.4%.

The full energy peaks were counted to a minimum of 1800 counts to limit the statistical error to less than ca. 2.4%. The background was observed to be constant within statistical error. The dead time losses were always less than 1%. No peaks which seem to be due to sum effects were observed. Thus ^{222}Rn in the quartz pipe was standardized based on the ^{214}Pb-activity.

Then, the quartz pipe was put in the liquid scintillation counting vial and after sealing with a silicon rubber stopper, the counting vial was shaken until the quartz pipe in the vial was broken. The sample that contains the equilibrium mixture of ^{222}Rn and its daughters is hereinafter referred to as "standard ^{222}Rn sample". Table 1 lists nine standard ^{222}Rn samples of a wide variety of ^{222}Rn concentrations prepared in the similar manner described above. Unfortunately, γ-ray spectrometry is not suitable for routine measurements of low-level ^{222}Rn in the environment, because they generally require long counting time.

Table 1. The E_α for the standard ^{222}Rn samples

Activity of the standard ^{222}Rn sample[a] / Bq	The air luminescence count rate / cps	The E_α / %
656.25 ± 2.15	1396.75 ± 1.53	42.57 ± 0.15
527.40 ± 1.67	1152.78 ± 1.39	43.72 ± 0.15
189.22 ± 0.64	391.75 ± 0.81	41.41 ± 0.16
133.06 ± 0.54	282.22 ± 0.69	42.42 ± 0.20
69.22 ± 0.34	149.99 ± 0.50	43.40 ± 0.26
30.49 ± 0.16	65.49 ± 0.33	42.96 ± 0.32
10.25 ± 0.10	22.12 ± 0.20	43.15 ± 0.57
6.57 ± 0.09	13.75 ± 0.11	41.88 ± 0.67
2.96 ± 0.07	6.23 ± 0.08	41.92 ± 1.14
		av 42.6 ± 0.2

[a] Based on the activity of ^{214}Pb.

Upon completion of γ-ray measurements, the air luminescence counts of the standard ^{222}Rn samples were measured with an Aloka liquid scintillation spectrometer, Model LSC-3500 (Aloka Co. Ltd. Tokyo, Japan) for 30 min. Background counts were measured under identical conditions using a counting vial which was full of air. The counting efficiency of the standard ^{222}Rn sample by the air luminescence method, E_α, is given by the relation

E_α = [A_{air} (cps) / S (Bq)] × 100%,

where A_{air} is the air luminescence count rate of the standard ^{222}Rn sample and S is the total disintegration rates of the standard ^{222}Rn sample. It is noted that the total disintegration rates of the ^{222}Rn sample are 5 times the disintegration rates of ^{222}Rn. The average counting efficiency for the air luminescence method, the E_α, was found to be 42.6 ± 0.2% (Table 1). It is interesting to note that, although the liquid scintillation spectrometer and the method were different from those in the preceding paper,[3] the E_α obtained in the present study is in good agreement the previously reported value (42.0 ± 0.2%).[3]

The counting efficiency for the E_α measured in this way is the average of the following three counting efficiencies, i.e., the counting efficiency for α-emitters which are uniformly distributed in air in the counting vial and do not attached to the wall of the counting vial (unattached daughters), the counting efficiency for α-emitters which are attached to the wall of the counting vial, and the Cherenkov counting efficiency due to β-particles from ^{214}Pb and ^{214}Bi in the counting vial.

The counting efficiency for α-emitters uniformly distributed in the counting vial was obtained as follows: First we measured the α-emission rate of a ^{210}Pb–^{210}Bi–^{210}Po source electroplated on silver disc (13-mm diameter) with 2π proportional counter for 10 min. The α-emission rate was 40.795 ± 0.260 α-particles / s in a solid angle of 2π steradians. Then the source was mounted at different positions in the counting vial (Figure 1), and the air luminescence counts due to α-particles from ^{210}Po were counted with the liquid scintillation spectrometer to a minimum of 20000 counts to obtain a standard statistical error of <ca. 0.7%. The background was observed to be constant within statistical error. The counting efficiency for α-emitters which were uniformly distributed in the counting vial, is given by

(the air luminescence count rate / the α-emission rate) × 100%.

In spite of the different positions of the source in the counting vial, the results, listed in Table 2, show good agreement with each other. Therefore, the counting efficiency for the unattached α-emitters, which were uniformly distributed in the counting vial, could reasonably be taken as the average value of Table 1, i.e., 67.5 ± 0.4%.

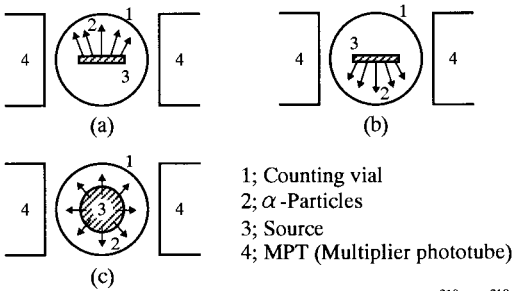

Figure 1. A plain figure for measurements of the ^{210}Pb-^{210}Bi-^{210}Po source in the counting vial. The source was placed; (a) and (b), at right angles to the surfaces of MPTs at the center of the vial; (c) on the bottom of the vial.

On the other hand, a part of the short-lived daughters of ^{222}Rn seem to be attached to the wall of the counting vial and are supposed to be counted with 2π geometry. Therefore, the counting efficiency for the α-emitters which are attached to the wall of the counting vial is ca. (1 / 2) × (67.5 ± 0.4) = 33.8 ± 0.2%. It is noted that the β-emitters do not produce the air luminescence count, and that ^{222}Rn, which has a relatively long half-life (3.825 d), is assumed to be unattached to the wall of the counting vial.

Table 2. Counting efficiency of the ^{210}Pb-^{210}Bi-^{210}Po source by the air luminescence method

Position of the source	The air luminescence count rate[a] / cps	Counting efficiency / %
(a)	27.653 ± 0.283	67.78 ± 0.73
(b)	27.245 ± 0.283	66.79 ± 0.72
(c)	27.767 ± 0.283	68.06 ± 0.73
		av 67.5 ± 0.4

[a]Corrected for the Cherenkov count rate due to β-particles from ^{210}Pb and ^{210}Bi.

Let δ be the average ratio of the number of the daughters which are attached to the wall to the total number of the daughters, and let A be the individual activity of ^{222}Rn and the daughters after transient equilibrium between ^{222}Rn and the daughters was established; then the count rate due to ^{222}Rn, ^{218}Po and ^{214}Po can be expressed

$$A(67.5 \pm 0.4) + 2A(67.5 \pm 0.4)(1 - \delta) + 2A(33.8 \pm 0.2)\delta. \quad (1)$$

As have been reported in a preceding paper,[6] β-particles from ^{214}Pb (av β^- 0.220 MeV)[5] and ^{214}Bi (av β^- 0.641 MeV)[5] gave Cherenkov radiation by interacting with the wall of the counting vial: The Cherenkov counting efficiencies for ^{214}Pb and ^{214}Bi were found to be 3.7 ± 0.2% and 32.9 ± 1.3%, respectively.[6] Thus the Cherenkov count rate due to β-particles from ^{214}Pb and ^{214}Bi were estimated to be

$$A[(3.7 \pm 0.2) + (32.9 \pm 1.3)]. \quad (2)$$

Hence the E_α (42.6 ± 0.2%) is given in terms of equations (1) and (2) as

[(1) + (2)] / 5A = 42.6 ± 0.2%,

where 5A is the total disintegration rate of ^{222}Rn and its daughters, so that, δ = 38.7 ± 2.8%. Errors were calculated as the sum of uncertainties of the standard solutions and standard statistical error.

A detailed investigation concerning three counting efficiencies which constitute the E_α would be desirable. However, it should be emphasized that the E_α obtained in this study is based on the experimental method, therefore, it can be applied to the preparation of known amounts of ^{222}Rn, which are needed for calibration of detectors measuring ^{222}Rn or a number of experiments without elaborate instrumentation or technique.

References

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