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Author	本間, 義夫(Honma, Yoshio) 村上, 悠紀雄(Murakami, Yukio)
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Study on the Applicability of the Integral Counting Method for the Determination of ^{226}Ra in Various Sample Forms Using a Liquid Scintillation Counter*

YOSHIO HOMMA, YUKIO MURAKAMI

本間義夫，村上悠紀雄

The liquid scintillation counting(abbreviated LSC hereafter)exihibits some advantages that it has no possibility of internal contamination and that no self-absorption correction is nesessary, which make the sample preparation easier for counting compared with the other methods. However, an intrinsic problem always associated with LSC is the correction against quenching effect which causes the reduction in the counting efficiency because of the energy loss contribute to the overall decrease in photon yield of scintillation system. Therefore it is important to obtain as high counting efficiency as possible by adequate sample preparation. The correction methods commonly applied are the channel-ratio methods, external or internal standard methods, all of which necessitate the use of standardized source of the radionuclide concerned. In the present study, the authors extended the integral counting technique to the case when several radionuclides are present in equilibrium which were formed after extration of Rn into toluene. Namely, in order to make the absolute counting of ^{226}Ra possible, it was tried to determine simultaneously the three different α-particles from ^{222}Rn, RaA and the RaC′ and the two β-particles from RaB and RaC in equilibrium state. The applicability of this technique was verified by examining the pulse height distribution of ^{226}Ra and ^{222}Rn, the integral counting rate-bias curve and the time required to establish the radioactive equilibrium between ^{222}Rn and its daughters.

^{226}Ra sample solutions were prepared according to a widely employed procedure as briefly described in the following. Ra occuring in natural water was coprecipitated with barium sulfate formed in it. The ashes of bone or eggs were decomposed with nitric acid. The resulting solutions were treated with barium sulfate to coprecipitate radium by adjusting the pH of the solutions with acetate buffers. Alkaline elements, most of calcium and their phosphates were removed during this process. The barium sulfate thus fomed was dissolved in 30% EDTA solution in alkaline medium to transter radium into the Curie flask.

Integral counting-bias curve

The integral counting tehnique was applied to radon sample separated from the ^{226}Ra standard solution. The counting rate was taken at the window width of 50 to ∞, 100 to ∞, and 1000 to ∞ with changing the gain at 10, 40, 60 and 90%. Then the integral counting-bias curve was constructed as shown in Fig. 1. The lower limit of the window was set at 50, not smaller than this, to avoid nois counting, while the upper limit was set at ∞ because some definite counting was still obtained even at the window width 1000 to ∞.

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The gain was determinative factor the slope of the integral counting–bias curve as shown in Fig 1. Namely the curve shifts upward with increasing the gain, hence making the slope flat the higher gains which can reduce the error caused by extrapolation of the curve to zero bias. Conclusively, the setting of gain at 90% is a reliable choice for this purpose. It was clearly shown that the curve shifts downward as the amount of the quencher increases. Consequently, when the quenching effect is exepected to be significant, the gain should be maximized to reduce the extrapolation error by flattening the curve.

Since the integral counting rates obtained were in a good agreement with the calculated values within the experimental error, 2 to 7%, the counting efficiency was regarded as 100%. Here the decay rate of ^{226}Ra in the equilibrium was calculated as 6 times of that in isolated state, since the decay rate of ^{226}Ra, ^{222}Rn, RaA, RaB, RaC and RaC′ should all be equalized. The ^{226}Ra standard solution accompanied water, nitric acid and barium chloride carrier into the scintillation solution, which will give the quenching effect to a significant extent. However a 100% counting efficiency was obtained at the gain 90% by the extrapolation method. The proposed method to transfer only radon gas separated from ^{226}Ra solution, free from CO_2, air and other accompanied materials if any, should be influencbd to a lesser extent by the quenching effect. The integral counting rate by the extrapolation method should consequently be equal to the absolute decay rate because 100% counting efficiency should be possible for the mixed system of ^{222}Rn and its daughter nuclides.

Concerning to the possible effect of γ rays from the mixed system of the counting efficiency, it was concluded insignificant because of the following reasons. Transitions between excited nuclear states generally take within 10^{-10} sec, which is shorter both than the resolving time of the coincidence counting circuit of LSC used(10^{-8} sec)and than the fluorescence decaying time of the LSC(2.8 to 3.0 · 10^{-9} sec). And the γ rays are always in coincidence with either α or β particles, resulting in no effect on the counting efficiency.

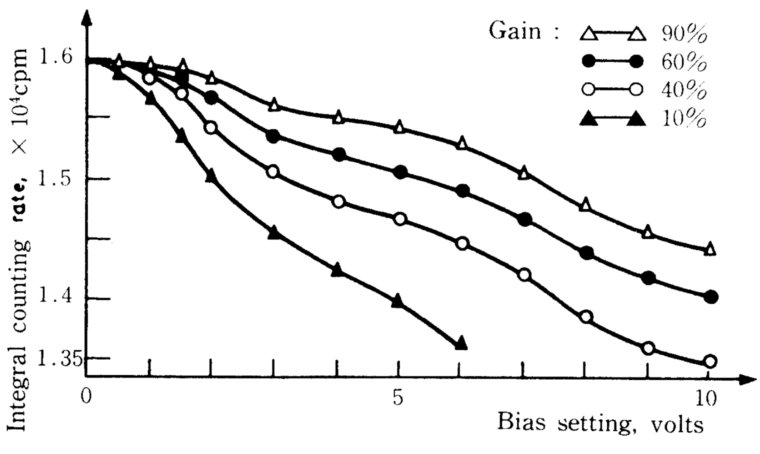

Fig. 1 Integral counting rate curves for ^{222}Rn