

COMPARISON OF A MODIFIED INTEGRAL COUNTING METHOD AND EFFICIENCY TRACING METHOD FOR THE DETERMINATION OF ^{222}Rn BY LIQUID SCINTILLATION COUNTING

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ABSTRACT. We have modified the integral counting method of liquid scintillation counting (LSC) by extrapolating the integral pulse-height spectrum to the detection threshold of the LS system. We have used this method to determine ^{222}Rn in samples. We have compared the results with those obtained by the conventional integral counting method (CICM) and the efficiency tracing method (ETM). The modified integral counting method (MICM) gives more accurate results than the CICM and the ETM. We discuss here the possibilities and limitations of the efficiency tracing method.

INTRODUCTION

Several investigators have discussed the advantages of liquid scintillation counting (LSC) for measuring the absolute disintegration rate of ^{222}Rn . The use of the integral counting method (ICM), and the fact that ^{222}Rn is appreciably soluble in a liquid scintillator, make the method particularly useful (Horrocks 1964; Goldstein 1965; Homma & Murakami 1977). Homma, Murase and Takiue (1987) and Murase *et al.* (1989) have measured the effect of air luminescence on determining ^{222}Rn by LSC and evaluated the error inherent in the conventional LSC determination of ^{222}Rn . However, commercially available LS spectrometers are not always suitable for integral counting. Thus, we thought it would be interesting to determine ^{222}Rn by the efficiency tracing method (ETM), which has been used for the absolute determination of beta emitters.

In this study, we compared two methods of determining ^{222}Rn by an LS spectrometer, by ICM and by ETM. We modified the ICM by extrapolating the integral counting curves, not to zero pulse-height, but to the detection threshold of an LS spectrometer, which refers to an average energy required to produce a measurable pulse. Although comparing the modified ICM (MICM) with the ETM was the primary aim of this study, data obtained by the MICM and the conventional ICM (CICM) were also compared and evaluated.

METHODS

Seven ^{222}Rn samples were prepared without quencher as follows: ^{222}Rn was pipetted from 0.1 M HCl solution of $^{226}\text{RaCl}_2$ and added to 100 ml of ice-cooled liquid scintillator. About 14 ml of the solution were put into each of 7 counting vials; then about 6 ml of scintillator solution were added to each vial to make a total volume of 20 ml. These samples, sealed air-tight with silicon rubber stoppers, were allowed to remain for 3.5 h before measurement. During this time, the daughters, ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po , reached transient equilibrium with ^{222}Rn . As the counting vials were full of liquid scintillator, practically all the ^{222}Rn in the vial was considered to be dissolved in the liquid scintillator.

Upon completing the measurement of the non-quenched ^{222}Rn sample, 6 quenched ^{222}Rn samples were prepared by adding successively increasing amounts of CHCl_3 to the 6 non-quenched samples using a syringe through silicon rubber stoppers. Thus, 1 non-quenched and 6 quenched ^{222}Rn samples were prepared. The liquid scintillator consisted of 4 g PPO (2,5-diphenyloxazole) in 1 liter of toluene.

The non-quenched and quenched ^{222}Rn samples were measured with an Aloka LS spectrometer, Model LSC-3500 (Aloka Co., Ltd., Tokyo, Japan) using the MICM, details of which have been described elsewhere (Homma, Murase & Handa, ms.). In brief, the MICM determines the absolute disintegration rate by extrapolating the integral pulse-height spectrum, not to zero pulse height, but to detection threshold. After determination, the sample was also measured using an Aloka program for ETM. To check the level of quench, the value of the external standard channel ratio of each sample was measured using an external ^{137}Cs gamma source.

RESULTS AND DISCUSSION

Figure 1A (1) shows a representative example of differential pulse-height spectrum for a non-quenched ^{222}Rn sample measured with an Aloka LSC-3500 LS spectrometer. The observed net integral counting rate for the same sample was plotted against pulse height; the resultant integral pulse-height spectrum was extrapolated to the detection threshold to obtain the true disintegration rate of the sample (Fig. 1B (1)). Differential and integral pulse-height spectra for the other non-quenched ^{222}Rn samples, which show similar spectra, have been omitted here. Table 1 lists the disintegration rates of seven non-quenched ^{222}Rn samples determined by the MICM. We assumed the detection threshold of the LS system to be 2.30 ± 0.10 keV, based on our analysis of the integral pulse-height spectrum of standardized ^3H samples. Details of the determination of the detection threshold have been published elsewhere (Homma, Murase & Handa, ms.). Alternatively, the ^{222}Rn samples were measured either by the CICM, which extrapolates these integral pulse-height spectra to zero pulse height, or by the ETM. Table 1 lists results.

The differential pulse-height spectra and the integral pulse-height spectra for the six quenched ^{222}Rn sample (Samples 2–7) are shown in Figure 1 (2)–(7). The disintegration rates of the six quenched ^{222}Rn samples were determined by the MICM, CICM and ETM. The percent difference of the disintegration rates determined by these methods from those for non-quenched samples determined by the MICM are listed in Table 1.

As shown in Table 1, the percent difference of results obtained by the CICM from those obtained by the MICM was -1.9% for the non-quenched samples and -2.7% for the quenched samples. The ICM has been one of the most useful methods for determining ^{222}Rn by LSC (Homma & Murakami

TABLE 1. Comparison of the MICM, the CICM and the ETM in determining ^{222}Rn in samples

Sample	MICM (dpm)*	% Difference from the dpm of non-quenched ^{222}Rn samples determined by MICM					ESCR [†] value
		CICM* (%)	ETM* (%)	MICM** (%)	CICM** (%)	ETM** (%)	
1	33152.6 ± 81.4	-1.9	0.0	-0.8	-2.8	-2.0	27.66
2	34396.7 ± 82.9	-2.0	-0.5	-1.0	-2.9	-1.7	27.22
3	31259.2 ± 79.1	-2.0	-0.2	-1.3	-2.1	-1.3	26.22
4	33194.3 ± 81.5	-1.8	-1.1	-0.7	-2.2	-1.9	25.30
5	33797.2 ± 82.2	-1.9	-0.6	-0.7	-3.2	-1.8	25.16
6	32489.9 ± 80.6	-1.8	-0.9	-1.6	-3.8	-1.8	24.72
7	34415.4 ± 83.0	-1.9	-0.8	-1.2	-2.2	-1.2	24.48
Average		-1.9	-0.6	-1.0	-2.7	-1.7	

*Non-quenched

**Quenched

[†]ESCR = External standard channels ratios

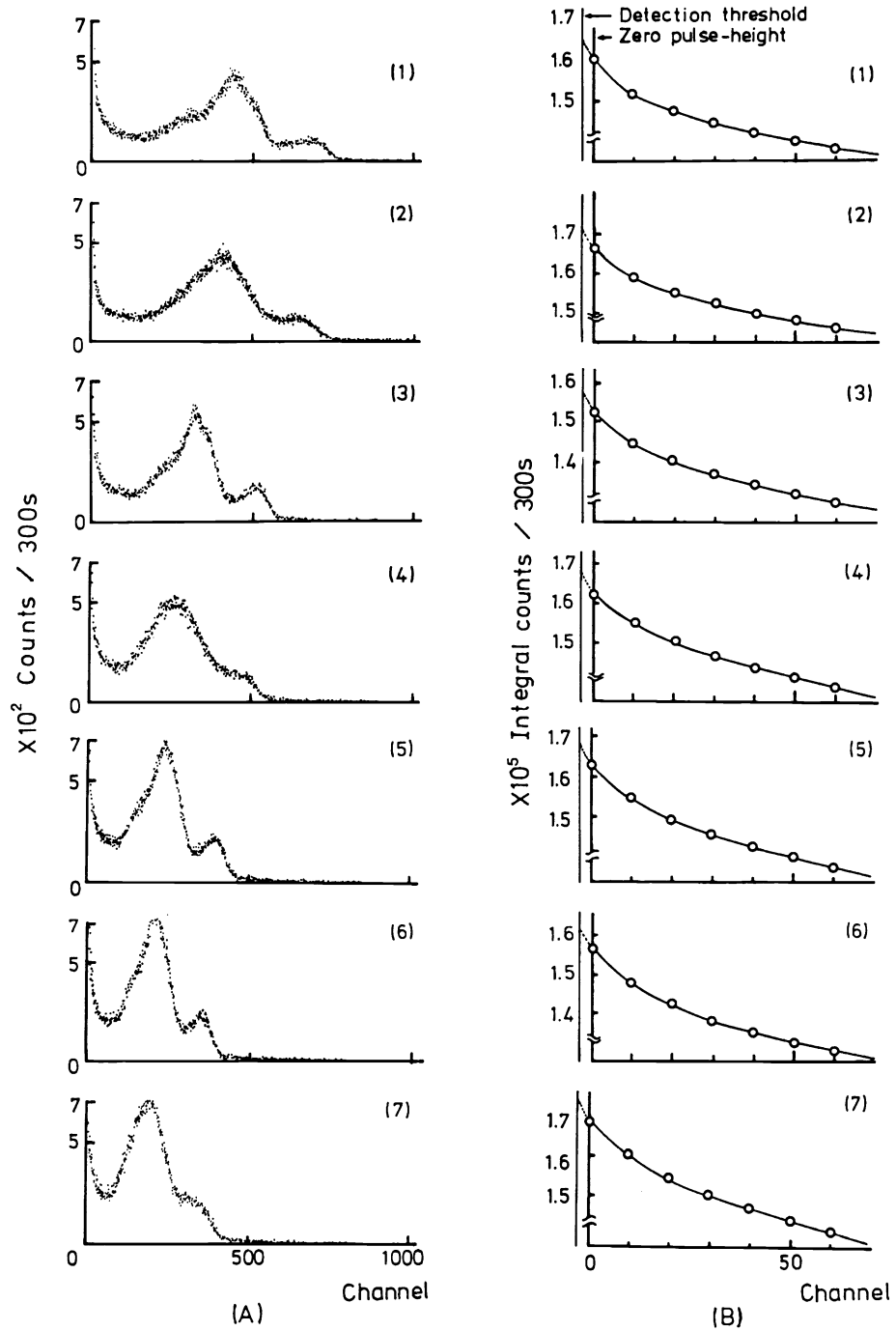


Fig. 1. A. Differential pulse-height spectra; B. Integral pulse-height spectra for ^{222}Rn dissolved in a liquid scintillator: (1) no CHCl_3 added; (2) 8 drops CHCl_3 added; (3) 16 drops CHCl_3 added; (4) 24 drops CHCl_3 added; (5) 32 drops CHCl_3 added; (6) 40 drops CHCl_3 added; (7) 50 drops CHCl_3 added.

1977). However, extrapolation of the integral pulse-height spectrum to zero pulse height gives the number of disintegrations that release energy equal to or greater than the detection threshold of the LS system, and not the absolute disintegration rate. It indicates only the fact that the difference between the true disintegration rate and the extrapolated intercept is negligible. Thus, it is clear that extrapolation of the integral pulse-height spectrum to the detection threshold gives more accurate results.

The averaged percent difference of the results obtained by the ETM from those by the MICM was -0.6% for the non-quenched samples and -1.7% for the quenched samples. The ETM is also useful for determining ^{222}Rn samples, provided that the quench of the sample is not so high (the ESCR value, which shows that the quench range of the sample is 27.94–24.48 (Table 1)). However, we emphasize that the ETM is mainly for determining pure β emitters. If the α peaks of the sample shift to lower pulse height due to quenching, and if the measurement by the ETM is made, at least in part, in the region of α peaks, unacceptable errors would result.

Advantages of the MICM are readily apparent. Not only can β emitters be standardized easily without preparing quench correction curves, but the method also has been used for the absolute determination of the equilibrium mixture of α and β emitters. Moreover, the average percent difference of the results for the quenched sample obtained by this method from those for the non-quenched samples by the same method was -1.0% . The MICM gives more accurate results than both the CICM and the ETM.

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