富山大学研究推進機構水素同位体科学研究センター研究報告 36：21－29，2016.

論　文

光電子増倍管の印加電圧を制御する改良積分法の液体シンチレーション計測への応用

原　正憲[a]，坂口　春菜[a]，中山　将人[a]，阿蘇　司[b]，庄司　美樹[c]，
古澤　孝良[d]，加藤　結花[d]，吉村　共之[d]

[a]　富山大学　研究推進機構　水素同位体科学研究センター
〒930-8555　富山県富山市五福 3190
[b]　富山高等専門学校　電子情報子工学科
〒939-0293　射水市海老江練合　1-2
[c]　富山大学　研究推進機構　研究推進総合支援センター　生命科学先端研究支援ユニット
〒930-0194　富山市杉谷 2630
[d]　日立製作所
〒181-8622　東京都三鷹市牟礼　6-22-1

Modified integral counting method by controlling high voltage of photomultipliers for liquid scintillation counting

Masanori Hara[a], Haruna Sakaguchi[a], Masato Nakayama[a], Tsukasa Aso[b], Miki Shoji[c],
Takayoshi Furusawa[d], Yuka Kato[d], Tomoyuki Yoshimura[d]

[a] Hydrogen Isotope Research Renter, Organization for Promotion of Research,
University of Toyama, Gofuku 3190,
Toyama 930-8555, Japan
[b] Electronics and Computer Engineering, Toyama National College of Technology,
Ebie-neriya 1-2, Imizu city, Toyama 933-0293, Japan
[c] Life Science Research Center, Organization for Promotion of Research, University of Toyama, Sugitani 2630, Toyama 930-0194, Japan
[d] Hitachi, Ltd.,
6-22-1 Mure, Mitaka city, Tokyo 181-8622, Japan

(Received January 27, 2017; accepted July 24, 2017)

Abstract

The modified integral counting method by control of the high voltage of photomultipliers (MICM-HV) was investigated to show its applicability for radioactivity analysis. In the MICM-HV, pulse height spectra of the sample are measured at various high voltages of photomultipliers in the liquid scintillation counter. The spectra are converted to integral spectra, which are extrapolated to give the convergence point. The counting rate at the convergence point corresponds to the disintegration rate of the sample. The MICM-HV determines the disintegration rate with one cocktail sample and the method requires no unquenched standard sample.

Keywords: Modified integral counting method, high voltage, photomultiplier, scintillator

1. **Introduction**

Liquid scintillation counting (LSC) is widely used for the radioactivity analysis of beta emitters, whereas LSC requires a quenching correction to determine a counting efficiency. Several quenching correction methods have been proposed. The quenching correction is usually carried out using a quench correction curve which is made by a quenched standard set [1]. Quenched standard sets for both ^{3}H and ^{14}C can be purchased from some suppliers, whereas standards sets for other nuclides need to be prepared by oneself. On the other hand, no quenched standard is required in direct methods [1] such as the integral counting method [2] and the efficiency tracing method [3]. Homma et al. have proposed the modified integral counting method (MICM) [4, 5]. The MICM has been used routine works of moderate precision, because no complicated procedure is necessary. In this method, the disintegration rate is obtained from the integral count rate of the sample at the zero detection threshold (ZDT). However, ^{3}H unquenched standard sample which the activity of ^{3}H is known is required to determine the ZDT. The pulse height spectrum of ^{3}H unquenched sample is measured, and the spectrum obtained is transformed to the integral spectrum,

$$IC(Chi) = \sum_{Ch=Chi}^{Ch_{\max}} C(Ch) \qquad (1)$$

where $IC(Chi)$ is the integral counts from a given integral channel (Chi) to the maximum channel (Ch_{max}) and $C(Ch)$ is the count rate at a given channel number, respectively. The integral spectrum is extrapolated to the disintegration rate of the standard sample and the ZDT is found. Subsequently, the pulse height spectrum of an unknown sample is measured and it is converted to the integral spectrum by Eq. (1). The integral spectrum of the sample is extrapolated to the ZDT, the disintegration rate of unknown sample is determined the count rate at the ZDT. To know the disintegration rate of unknown sample the MICM requires an unquenched standard of ^{3}H. The MICM with various quenched samples (MICM-VQ) requires several quenched samples having same amount of radioactive material[6]. In the MICM-VQ these spectra are converted to integral spectra by Eq. (1) and these integral spectra are extrapolated to find the convergence point which corresponds to the ZDT, because the integral counts at the ZDT does not depends on the quenching of the sample. The integral count rate at the ZDT corresponds to the disintegration rate of sample. However, this technique can not be applied for one sample.

In this study, the determination method of the disintegration rate of one sample cocktail without an unquenched standard was developed. Several integral spectra of one sample were obtained by the control of the high voltage of photomultipliers (PMTs). These spectra were extrapolated to find the ZDT like as the MICM-VQ. The disintegration rate of the sample was obtained from the integral counts at the ZDT. The method developed was named as the modified integral counting method by the control of high voltage of PMTs (MICM-HV). The disintegration rates obtained by the MICM-HV were compared with those by the MICM-VQ.

2. Experimental

The quenched standard set of ^{14}C of toluene based scintillator (denoted as TL set) and that of Ultima Gold (denoted as UG set) were purchased as sample cocktails. The quenched standard sets were composed of 10 samples which contained same amount of radioactive material. The disintegration rate of TL set and of UG set were determined to be 128700 dpm and 122200 dpm by suppliers, respectively. The uncertainty of both sets was ±1.3% in 99% confidence level. The five samples of TL set are named from TL A to TL E with increasing quenching, and the samples of UG set are from UG 1 to UG 5.

The pulse height spectra from scintillation cocktails were measured by the liquid scintillation counter, Aloka LSC-LB5. This scintillation counter has three of PMTs for the scintillation counting with the coincidence unit to reduce the background counts. This system is equipped the 4000 channel multichannel analyzer to register the pulse height spectrum of scintillation cocktail. The channel range for measuring the ^{14}C spectrum was adjusted from 0 to 200 keV. The pulse height spectra of samples were registered for 10 min when the high voltage of PMTs was controlled in the range from 1500 V to 1540 V.

3. Results and discussion

3.1. MICM with various quenched samples

Nakayama et al. reports MICM-VQ can be applied for various liquid scintillators such as toluene and Ultima Gold [7]. The disintegration rates of TL and UG sets were determined by the MICM-VQ. Fig. 1 shows the pulse height spectra of UG set at 1533 V for PMTs. The abscissa and the ordinate are the channel number and the counting rate, respectively. The pulse height of UG 1 with the lowest quenching level is extended to 2500 channel. However, the pulse height of quenched samples decreased with increasing the quenching. On the other hand, the peak positions of pulse height spectra were kept. These spectra were converted to the integral spectra with using

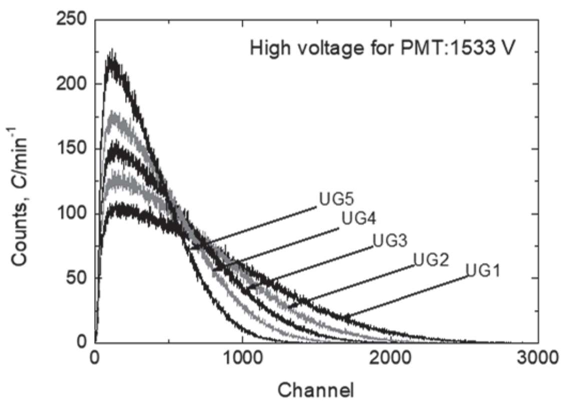

Figure 1. Pulse height spectra of UG set. These spectra were obtained under 1533V of the high voltage of photomultipliers in LSC-LB 5.

Eq. (1). They are shown in Fig. 2. The value of *IC* decreased with increasing *Chi*. The fitting function was obtained from the integral spectrum where a second order function was applied. The fitting was done for the higher channel above the peak around 150 channel when the high channel was set to be satisfied $R^2 \geq 0.9997$. The convergence point was found at -13 channel and the disintegration rate of samples was evaluated to be 123.8 ±0.2 kdpm. Since low energy beta particles can generate insufficient number of photons to detect, a channel number of a convergence point has a negative value. The uncertainty of obtained value is the standard deviation of fitting functions at the convergence point. The counting rate at the convergence point agreed with the assayed value of UG set. The evaluations of the disintegration rate of both TL and UG sets at other values of high voltage of PMTs were listed in Table 1 and 2, respectively. The disintegration rate evaluated agreed with the assayed value by the supplier. Within the measurement range the high voltage of PMTs does not influence on the evaluation of the disintegration rate by the MICM-VQ.

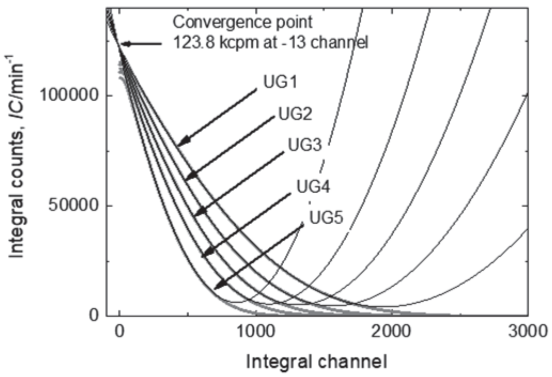

Figure 2. The integral spectra of UG set. The integral spectra were obtained from the pulse height spectra with using Eq. (1). The high voltage of PMTs was adjusted at 1533 V. The bold and thin lines indicate the obtained spectra and the fitting curves, respectively.

Table 1. Obtained value of Toluene quenched set (TL) with MICM-VQ.

High voltage (V)	Convergence point (Chi)	Disintegration rate (kdpm)	[obtained]/[assayed]
1500	-1	128.4±0.3	1.00
1510	-2	128.4±0.4	1.00
1520	-1	128.2±0.4	1.00
1533	-3	128.1±0.3	1.00
1540	1	127.3±0.2	0.99

Table 2. Obtained value of Ultima Gold quenched set (UG) with MICM-VQ.

High voltage (V)	Convergence point (Chi)	Disintegration rate (kdpm)	[obtained]/[assayed]
1500	-15	124.2±0.3	1.02
1510	-13	123.9±0.2	1.01
1520	-13	123.9±0.1	1.01
1533	-13	123.8±0.2	1.01
1540	-12	123.4±0.2	1.01

3.2. MICM by control of high voltage of PMTs

The MICM-VQ requires several samples, which contain same amount of radioactive material, to determine the ZDT. If the several spectra are obtained from one sample, the disintegration rate of its sample will be determined by using the MICM. By controlling the high voltage of PMTs, the pulse height from PMTs can be regulated. The control of high voltages works like an addition of quencher. Therefore, several pulse height spectra can be obtained from one sample by the control of the high voltages. To confirm the applicability of the MICM by the control of the high voltage of PMTs (MICM-HV), the pulse height spectra of both TL and UG sets were obtained under several different values of high voltage.

Fig. 3 shows the dependence of pulse height spectra on the high voltages of PMTs. These spectra were obtained from UG1 which was a less quenched sample in UG set. The pulse height spectra were stretched to the higher pulse height side with increasing high voltage of the PMTs. However, the peak position of the pulse height spectra was maintained around 150

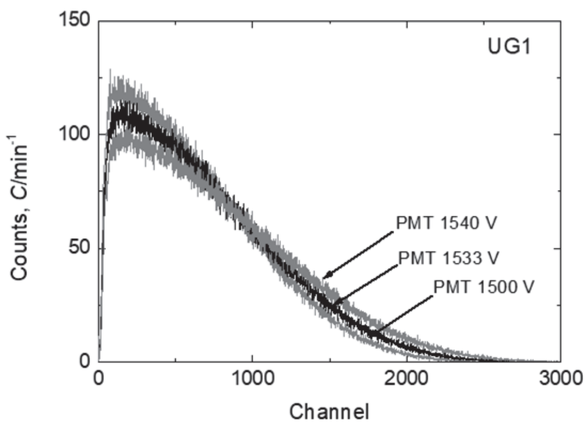

Figure 3. Pulse height spectra of UG 1 at various high voltages of PMTs in LSC-LB5.

channel. This change in the spectra with the variation of high voltages was similar to the effect of the quenching as shown in Fig. 1. In other words, the control of the high voltage of PMTs can provide a set of spectra like a set of quenched samples. These spectra obtained were converted to the integral spectra with using Eq. (1) as shown in Fig. 4. All integral spectra monotonically decreases. These spectra were fitted by a second order function. The fitting curves from the integral spectra obviously show the convergence point at 7 channel. The counting rate at the convergence point was found to be 121.2

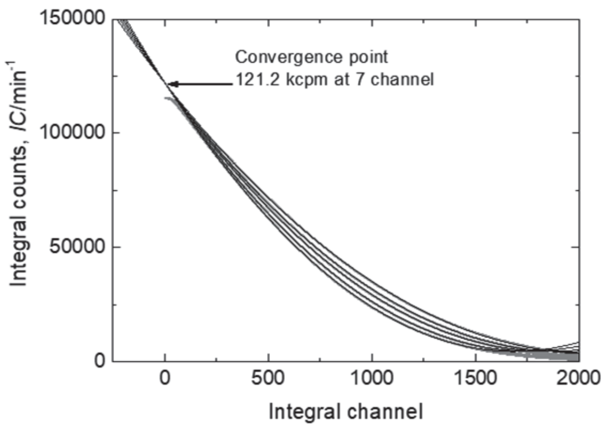

Figure 4. The integral spectra of UG 1 at various high voltages of PMTs. The bold and thin lines indicate the obtained spectra and the fitting curves, respectively.

Table 3. Obtained value of Toluene quenched set with MICM-HV.

Sample	Convergence point (ch.)	Disintegration rate (kdpm)	[obtained]/[assayed]
TL A	35	124.6±0.1	0.97
TL B	35	124.2±0.2	0.97
TL C	30	124.0±0.3	0.96
TL D	21	124.3±0.2	0.97
TL E	9	125.4±0.2	0.97

Table 4. Obtained value of Ultima Gold quenched set with MICM-HV.

Sample	Convergence point (ch.)	Disintegration rate (kdpm)	[obtained]/[assayed]
UG 1	7	121.2±0.1	0.99
UG 2	-23	125.3±0.2	1.03
UG 3	6	120.3±0.2	0.98
UG 4	-9	123.0±0.2	1.01
UG 5	-15	124.3±0.2	1.02

kcpm. The integral spectra of both TL and UG sets were obtained, and the disintegration rates were evaluated from the convergence point. These results of TL and UG were listed in Tables 3 and 4, respectively. In TL set, the disintegration rates evaluated were 3 % less than the assayed value. The values evaluated for UG set slightly deviated from the assayed value within ±3 %. These results indicate that the MICM-HV can be used for the radioactivity measurement of moderate precision. It is also worth to mention that the MICM-HV is able to determine the disintegration rate from only one sample and it was applicable for various scintillators.

4. Conclusions

A new method based on the MICM, MICM-HV, was developed for the measurement of disintegration rate of a single sample without unquenched standard. The pulse height spectra of a sample were measured under various high voltages of PMTs in the commercial liquid scintillation counter. The pulse height spectra obtained were converted to the integral spectra to find the convergence point, and the disintegration rate was determined from the integral counts at the convergence point. The disintegration rate evaluated by the MICM-HV agreed with the assayed value within 3 % of deviation. It indicates that the MICM-HV has a potential for the radioactivity analysis from one sample with moderate precision.

Acknowledgement

This work was supported by JSPS KAKENHI Grant Number 26610065.

References

[1] M.F. L'Annunziata, M. J. Kessler, Liquid scintillation analysis: principles and practice, Chpt. 5, M. F. L'Annunziata (Eds.), Handbook of Radioactivity Analysis. Academic press, California, (2003).

[2] G. Goldstein, Absolute liquid-scintillation counting of beta emitters., Nucleonics

23(1965) 67-69.
[3] M. Takiue, H. Ishikawa, Thermal neutron reaction cross section measurements for fourteen nuclides with a liquid scintillation spectrometer. Nucl. Instrum. Methods., 148(1978) 157-161.
[4] Y. Homma, Y. Murase, K. Hanada, The zero detection threshold of a liquid scintillation spectrometer and its application to liquid scintillation counting, Appl. Radiat. Isot., 45(1994) 341-344.
[5] Y. Homma, Y. Murase, K. Hanada, Absolute liquid scintillation counting of 35S and 45Ca using a modified integral counting method, J. Radioanal. Nucl. Chem., 187(1994) 367-374.
[6] M. Hara, M. Nakayama, K. Hirokami, T. Aso, Appropriate quenching level in modified integral counting method by liquid scintillation counting, J. Radioanal. Nucl. Chem., 310(2016)857-863.
[7] M. Nakayama, M. Hara, M. Matsuyama, K. Hirokami, Modified counting method with various quenched samples for different scintillators, Radio. Safety Manage., 16(2017)1-7.