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Feasibility study on signal separation for spontaneous alpha decay in $LaBr_3$:Ce scintillator by signal peak-to-charge discrimination

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A novel analysis method named peak-to-charge ratio (V_p/Q_{total}) discrimination, aiming at background rejection especially for alpha decay self-activity in $LaBr_3$:Ce scintillators has been developed. This method is based on a waveform analysis using the peak-to-charge ratio in the output waveform of a photomultiplier tube. The discrimination of alpha-induced events was achieved by using a threshold function based on the error propagation of the V_p/Q_{total} value. The accidental rejection ratio of gamma-induced events was evaluated to be 0.17%. Furthermore, a total absorption peak spectrum processed with the V_p/Q_{total} discrimination method for ^{68}Ga 1.883 MeV gamma rays, where the energy was overlapped with background alpha events, reproduced exactly the same result as that of the background subtraction method. The difference in measured peak counts of both methods was 0.716%, and the statistical error in the V_p/Q_{total} discrimination method and background subtraction was 4.81% and 8.70%, respectively. Thus a higher-accuracy measurement could be achieved using the V_p/Q_{total} discrimination method. The present study demonstrates that the V_p/Q_{total} discrimination method is a promising method for background rejection of the spontaneous alpha decay in $LaBr_3$:Ce scintillators. [http://dx.doi.org/10.1063/1.4928115]

I. INTRODUCTION

Recent developments of inorganic scintillators have been remarkable in the field of radiation measurement. In particular, the $LaBr_3$:Ce scintillator yields excellent energy resolution (<3% at 662 keV) with intrinsic time resolution of about 107 ps.[1,2] These characteristics are useful for gamma ray spectrometry,[3] development of gamma cameras,[4] and time of flight measurements.[5] According to previous studies, $LaBr_3$:Ce scintillators have also been used for lifetime measurement of unstable nuclei[6,7] and observation of the response to fusion neutrons.[8] Furthermore, $LaBr_3$:Ce scintillators have large stopping power in spite of their relatively lower effective atomic number, since it is possible to make a large crystal with high density (5.3 g/cm^3).[9–11] They are also suitable for high energy gamma ray measurements (e.g., cosmic ray, characteristic gamma ray due to nuclear reactions[12] and environmental radiation[13]). Additionally, fluorescence characteristics of $LaBr_3$:Ce scintillators have only a small dependency on thermal changes[14] and have strong radiation-resistance.[15,16] In recent years, $CeBr_3$ scintillators with high energy resolution (4.0% at 662 keV) and fast timing resolution of 119 ps with a self-activity much lower than the $LaBr_3$:Ce scintillator have also been developed.[17–19] However, the characteristics of $CeBr_3$ scintillators are slightly inferior to $LaBr_3$:Ce scintillators at present. Yet, self-activity of $LaBr_3$:Ce crystals is considered to be a serious defect in $LaBr_3$:Ce scintillators.[20] The major components of this self-activity are 1436 keV gamma rays from electron capture disintegration (66.4%) of ^{138}La nuclei with 32 keV K-X ray of Ba atom, and 789 keV gamma rays with 255 keV electron from beta decay (33.6%) of ^{138}La nuclei. Moreover, the self-activity background due to alpha decay from Ac-series nuclei is distributed at a range of 1.5–3 MeV in the energy spectrum and a characteristics gamma ray induced from the alpha decay is also emitted almost at the same time. Much research has been carried out on particle identification of alpha-decay nuclei using coincidence measurement of alpha-decay events and its induced characteristic gamma ray.[1,20] This inevitable background can be negligible if the count rate of true events is sufficiently larger than self-activity background; however, it is difficult to observe the true event when a low count rate is taken into account. In general, large volume $LaBr_3$:Ce crystals are used for high energy gamma ray measurements, but the event rate of self-activity increases with crystal volume. Therefore, research on the self-activity suppression carries a significant importance for the development of large volume $LaBr_3$:Ce crystals.

Research by Hoel *et al.*[21] and Crespi *et al.*[22] suggested that there exists a difference between alpha and gamma induced events in the vicinity of the peak voltage in the output waveform of a photomultiplier tube (PMT) equipped with a $LaBr_3$:Ce scintillator. However, to the best of our knowledge, almost no research has been done in the field of pulse shape discrimination (PSD) for rejected alpha-decay events. This is because the waveform difference obtained is almost imperceptible. In research by Crespi *et al.*,[22] self-activity rejection of alpha-decay was achieved by comparing the total integrated charge and the partial integrated charge in a suitable time domain in the vicinity of the peak voltage. In the present work, the domain of integration and threshold constant in the whole energy range have been determined discretionally, and a *post hoc* analysis is necessary for the optimization

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of these parameters. Furthermore, there is a possibility that the rejection accuracy of alpha-induced events depends on the domain size of the charge integration, as the threshold for the discrimination of alpha and gamma induced events changes with this size. Therefore, the signal peak-to-charge ratio V_p/Q_{total} determined uniquely in all waveforms was noted in this work. Generally, Q_{total} is proportional to V_p and for gamma ray events at adequate gamma ray energy region,[23,24] while the V_p/Q_{total} value is expected to be a constant. However, we observed different values between alpha and gamma induced events. By using this difference, we developed an alpha-event discriminating method dedicated for $LaBr_3$:Ce scintillators using the V_p/Q_{total} value. In this paper, we will report the V_p/Q_{total} discrimination method and demonstrate the effectiveness of an $LaBr_3$:Ce scintillation detector in gamma ray spectrometry.

II. MATERIAL AND METHOD

A. Data acquisition system and noise reduction

As shown in Figure 1, the electric signal waveforms from a PMT (R6231-100, Hamamatsu Photonics) equipped with a $LaBr_3$:Ce scintillator (38S382/B380, BrilLanCe™ Saint-Gobain) were measured using an oscilloscope (Wave Runner 64xi, LeCroy, 5 GS/s) without electronic devices such as a shaping amplifier. The raw waveforms were stored to an external storage device. The values of V_p and Q_{total} were obtained from the analysis of the measured waveforms using in-house software which has several functions such as offset correction, a moving average for waveform smoothing, a frequency analysis using Fast Fourier Transform (FFT), a digital low pass filter using FFT with inverse FFT (IFFT), and spectrum generation for all measured data. In particular, the function related with noise suppression is important for the V_p/Q_{total} value, since the V_p is sensitive to high-frequency noise. Therefore, the high-frequency noise was suppressed using a moving average with a time width of 2 ns, and a digital low pass filter using FFT and IFFT suppressed the noise component of >50 MHz in the entire waveform.

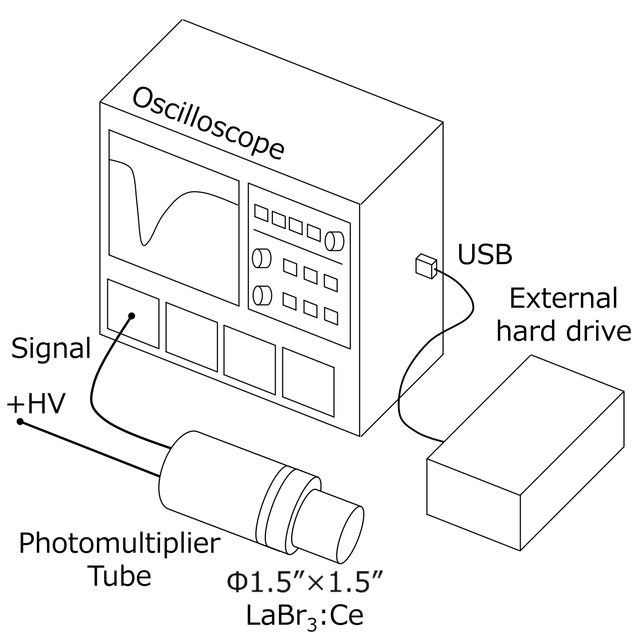

FIG. 1. Schematic diagram of the data acquisition system in the experiment.

B. Alpha-event discrimination using V_p/Q_{total}

Although the background suppression method for self-activity has been discussed in previous research,[22] it is still difficult to assess the rejection accuracy quantitatively. This is because a threshold for discriminating alpha and gamma events could not be determined theoretically as described in Section I. In the present work, the peak-to-charge ratio V_p/Q_{total} is expected to be constant for gamma events, and a different constant value is expected for alpha events. However, the V_p/Q_{total} value fluctuated according to the uncertainties of V_p and Q_{total} values, and a function of $\sigma_{V_p/Q_{total}}$ (Equation (1)) deduced from an error propagation of V_p/Q_{total} value was therefore used for discriminating alpha and gamma events. Since the $\sigma_{V_p/Q_{total}}$ function varies according to the supplied voltage to the PMT, the parameters for the function should be obtained as a calibration. This process requires a gamma ray source. However, $LaBr_3$:Ce self-activity generates a gamma ray emission statistically sufficient for the calibration,

$$\sigma_{V_p/Q_{total}} = \sqrt{\left(\frac{1}{Q_{total}}\sigma_{V_p}\right)^2 + \left(\frac{V_p}{Q_{total}^2}\sigma_{Q_{total}}\right)^2}. \qquad (1)$$

C. Evaluation of alpha event rejection using V_p/Q_{total} discrimination method

In order to confirm the efficacy of alpha-event discrimination, a quantitative assessment was performed using an external gamma ray source. Since the energy range of background alpha-decay is distributed from 1.5 to 3 MeV (gamma equivalent), a Ge-68/Ga-68 source (CS-6-14, 12.4 kBq, Siemens Medical Solutions) which emits 1.883 MeV gamma rays from ^{68}Ga was used as the external gamma-ray source. The rejection accuracy of alpha-induced events was assessed by comparing the energy spectrum between the V_p/Q_{total} discrimination method and conventional method (background subtraction). The ^{68}Ga source was placed 0.6 m away from the $LaBr_3$:Ce scintillation detector. Waveforms of 60 000 events were used for the comparison.

III. RESULT

A. Noise reduction to improve measurement accuracy

The raw waveform data at a detection energy of 511 keV and the waveform of high-frequency noise suppressed by a moving average with a time width of 2 ns and 50 MHz digital low pass filter using FFT with IFFT are shown in Figures 2(a.1)–2(a.3). Since components of instantaneous noise affect the raw waveform, the values of V_p obtained from raw data was about 4.7% larger than noise-suppressed waveform of 511 keV gamma ray, and in the case of V_p obtained from 1.883 MeV gamma ray which is emitted form ^{68}Ga was 0.71% larger. The signal to noise ratio in the V_p value

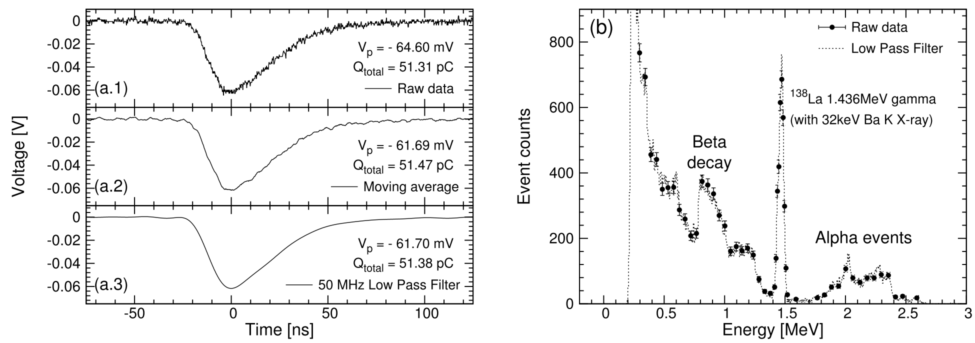

FIG. 2. (a.1) The raw waveform data measured by the data acquisition system for a detection energy of 511 keV, and the noise-suppressed waveform using (a.2) the moving average with a time width of 2 ns and (a.3) 50 MHz digital low pass filter using FFT with IFFT analysis. (b) Comparison of self-activity spectrum of LaB_3:Ce before (raw data) and after noise-suppression by digital low pass filter.

improves with increasing the injected gamma ray energy. The value of V_p obtained from the moving average and digital low pass filters were almost identical since high-frequency noise was adequately suppressed. On the other hand, no significant difference in the total integrated charge Q_{total} was observed among the raw data and the two smoothing methods. Figure 2(b) shows the energy spectra consisting of about 10^5 Q_{total} data with and without noise suppression. An almost identical energy spectrum was obtained. This supports the fact that the Q_{total} value is not affected by the noise reduction technique using a low pass filter.

B. Demonstration of the V_p/Q_{total} discrimination method using $LaBr_3$:Ce self-activity

Figure 3(a.1) shows the energy (Q_{total} equivalent) dependence of the V_p/Q_{total} value for self-activity of the $LaBr_3$:Ce scintillator. Two components of the V_p/Q_{total} value at the energy range of 1.5– MeV were observed. According to Crespi *et al.*, the waveform of alpha-induced events contains a higher value of peak intensity than the gamma-induced events in the same detection energy.[22] Hence, the alpha-induced events should be distributed at a higher value than the gamma-induced events. For the lower component of V_p/Q_{total}, it is considered to be a gamma-induced event from environmental radiation such as ^{208}Tl (2.615 MeV). On the other hand, the V_p/Q_{total} value in Fig. 3(a.1) decreases with increase in energy, although V_p/Q_{total} was expected to be a constant. The reason for the decline seems to be due to saturation of V_p as shown in Figure 3(b.1). The linearity between V_p and Q_{total} was only kept in the low energy region. In order to correct the saturated V_p, the saturation curve of V_p was assumed to be in the form of Equation (2) which consists of a linear term αQ_{total} (α = const.) and a saturation term $1 + \beta Q_{total}$ (β = const.). Equation (3) was deduced by assuming that the corrected peak voltage V_{pCorr} is proportional to Q_{total} such as $V_{pCorr} = \alpha Q_{total}$,

$$V_p = \frac{\alpha Q_{total}}{1+\beta Q_{total}} (\alpha = const., \beta = const.), \tag{2}$$

$$V_{pCorr} = \frac{V_p}{1-(\beta/\alpha)V_p}. \tag{3}$$

Note that these parameters α and β which are necessary for the saturation correction were obtained by a fitting analysis with Equation (2) in the experimental value without alpha-induced events. The coefficient of correlation R^2 between the measurement value and Equation (2) was found to be 0.9976, which shows a strong correlation between them. Figs. 3(a.2) and 3(b.2) show that the saturation was corrected properly using Equation (3). The optimized threshold function on the V_p/Q_{total} value was configured theoretically using its standard deviation of $\sigma_{V_p/Q_{total}}$. As mentioned for Equation (1), $\sigma_{V_p/Q_{total}}$ is a function of Q_{total} in approximate inverse proportion. Threshold functions of 1 to 3 $\sigma_{V_p/Q_{total}}$ were obtained by the calibration using a fitting analysis with $\sigma_{V_p/Q_{total}} = kQ_{total}^{-1} + l$ (k = const., l = const.). The fitting curves adequately expressed their measured values as shown in Figure 4(a). In this analysis, since the component of alpha-induced events ranged from 1.5 to 3 MeV, only the data lower than 1.4 MeV were used. Figure 4(b) shows that for each threshold function with

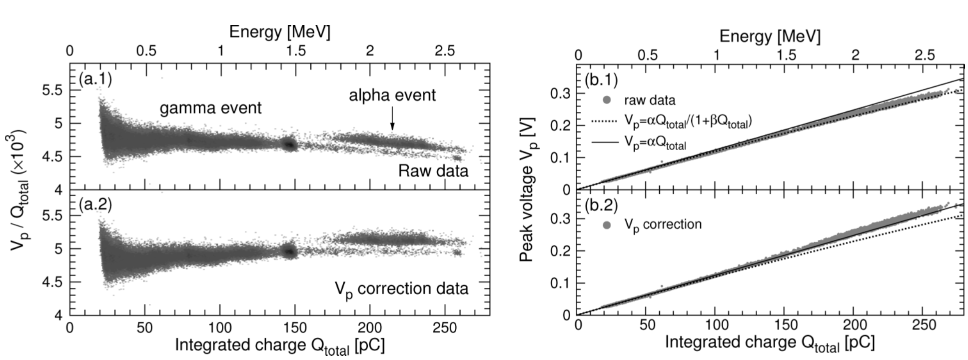

FIG. 3. (a.1) Energy (Q_{total} equivalent) dependence of the peak-to-charge ratio V_p/Q_{total} in raw data waveform. (b.1) The energy (Q_{total}) dependence of the V_p with the ideal linear function and the saturation curve used for the fitting analysis. (a.2), (b.2) Saturation correction of V_p using the Equation (3).

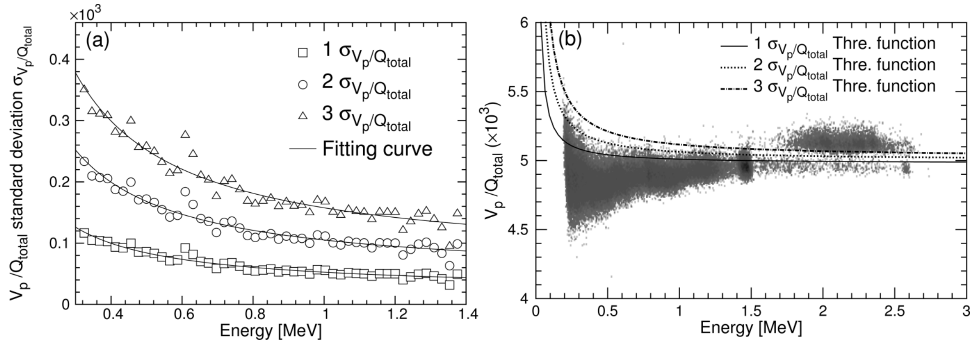

FIG. 4. (a) Energy dependence of the V_p/Q_{total} standard deviations 1 to 3 $\sigma_{V_p/Q_{total}}$ with fitting curves. (b) The corrected V_p/Q_{total} value with each threshold function.

V_p/Q_{total} scatter plot in Figure 3(a.2), alpha-induced events seem to be adequately discriminated using the threshold function. Figures 5(a.1)–5(a.3) shows the energy spectrum with and without alpha event discrimination in $LaBr_3$:Ce self-activity using the present method. It is shown that the alpha-decay event can be discriminated effectively. On the other hand, the accidental rejection of gamma-induced events occurred by the threshold function as shown in Figures 5(b.1)–5(b.3). Accidental gamma-rejection depends on the threshold function; the probability of gamma-rejection for 1, 2, 3 $\sigma_{V_p/Q_{total}}$ was evaluated as 4.661%, 0.809%, and 0.172%, respectively, in the energy range of <1.5 MeV where alpha-induced events are not contained.

C. Assessment of alpha-discrimination accuracy

In the present experiment, the rejection accuracy of alpha-induced events was assessed using an external radiation source (^{68}Ga 1.883 MeV). Figures 6(a) and 6(b) show the energy spectrum for the background subtraction and V_p/Q_{total} discrimination methods, respectively. The energy spectrum without radiation source was normalized by the integrated event counts in the energy range of >2 MeV for background subtraction. As shown in Figure 6(b), only the ^{68}Ga 1.883 MeV gamma ray peak remained clear by using the 3 $\sigma_{V_p/Q_{total}}$ threshold function. Similarly, Figure 6(c) shows the enlarged energy spectra dedicated for ^{68} Ga 1.883 MeV gamma rays for detailed comparison. The V_p/Q_{total} discrimination method accurately reproduced the result as that using the background subtraction method. In both methods, the difference in measured peak counts was 0.716%. And the difference in parameters obtained by the fitting analysis with a gauss function in the spectrum such as peak counts, mean energy, full width at half maximum (FWHM) was 1.694%, 0.021%, and 0.165%, respectively, as shown in Table I. Additionally, statistical errors in measured total peak counts for the V_p/Q_{total} discrimination method and subtraction method were 4.81% and 8.70%, respectively, at a range of $\pm 3\sigma$. A reduction in statistical error was thus observed using the former method.

IV. DISCUSSION

A. Calibration of threshold function and correction for V_p saturation

The V_p/Q_{total} value increases along with the supplied voltage of the PMT because the pulse width in the PMT declines with the rise in supplied voltage. Accordingly, the threshold function must be calibrated for each measurement condition especially for the supplied voltage. The actual supplied voltage must be taken into account for a successful calibration, but the V_p/Q_{total} difference between alpha and gamma events could be observed in the range of typical voltage (500–1000 V) for gamma-ray spectrometry using a $LaBr_3$:Ce scintillator. Once the calibration is done for a specific supplied voltage, the stability and reproducibility of the threshold function is confirmed with the fixed supplied voltage. Moreover, the advantage of this calibration process

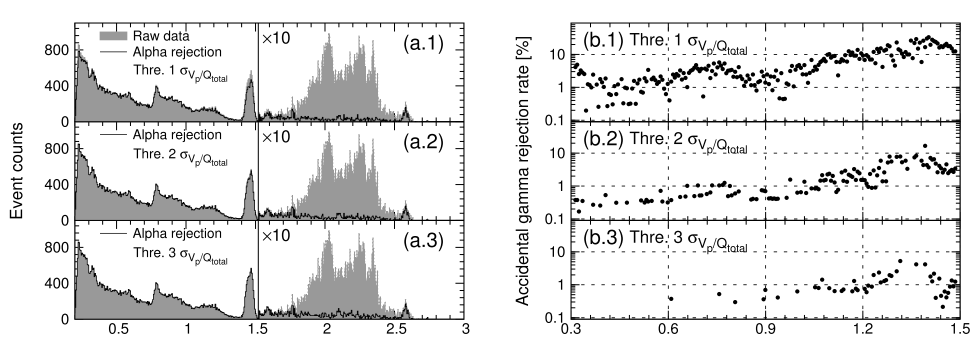

FIG. 5. (a) Energy spectrums with and without alpha event rejection using each threshold functions in $LaBr_3$:Ce self-activity. For the clarity of the present method effect, event counts increase by a factor of ten in the energy range of alpha self-activity. (b) The accidental gamma rejection rate in the V_p/Q_{total} discrimination method in the energy range of 1.5 MeV, where alpha events are not existent. The energy range without a data point means an accidental gamma rejection of 0%.

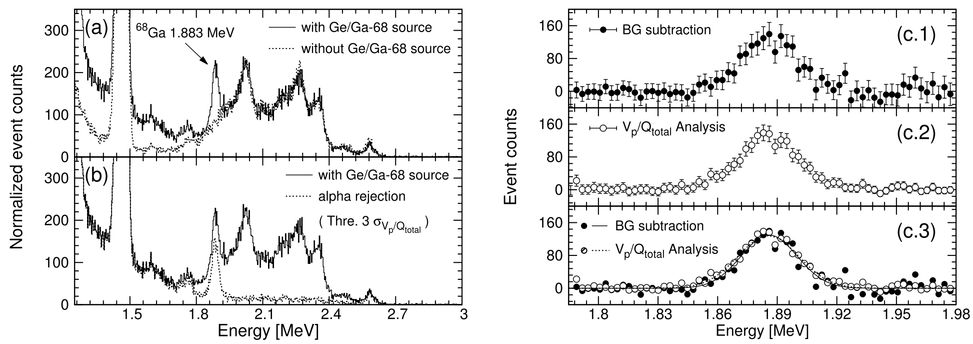

FIG. 6. (a) Normalized energy spectrums with and without Ge-68/Ga-68 source. (b) The energy spectrum of rejected alpha-induced events using the V_p/Q_{total} discrimination method with 3 $\sigma_{V_p/Q_{total}}$ threshold function. The total absorption distribution of (^{68}Ga 1.883 MeV) rejected alpha-decay background, (c.1) by the BG subtraction method, and (c.2) by V_p/Q_{total} discrimination method. (c.3) Comparison of the total absorption distribution between two methods.

does not require an external radiation source because the $LaBr_3$:Ce scintillator itself has self-activity. This makes the calibration process much easier and can be performed just before a measurement.

In Figures 3(a.1) and 3(b.1), the saturated V_p was seen as a function of Q_{total}. As the V_p decreases with increase in the pulse width of PMT output due to the space charge effect (increase in the electron transit time spread in the PMT), the saturation becomes more obvious in higher supplied voltage. The analytical solution of this saturation curve is extremely complex; however, the saturated V_p fitted using Equation (2) could reproduce exactly the experimental values.

B. System dead time of the data acquisition

In an energy range of about 2.6 MeV, the contribution of alpha events from ^{215}Po in the $LaBr_3$:Ce crystal should be observed. However, the contribution was not seen in the energy spectrum in Figure 2(b). The alpha decay of the ^{215}Po with shorter lifetime of 1.785 ms occurs immediately after the disintegration of the parent nuclide such as ^{219}Rn. The reason for the unrecorded alpha event from ^{215}Po is due to long dead time of the present data acquisition system. The acquisition system requires at least 50 ms for storing measured waveform, and this latency rises to about 200 ms with increasing a number of stored waveform to the external hard drive. The dead time for this data acquisition system is difficult to measure precisely due to low reproducibility. However, event losses for alpha and gamma are not essential in discriminating alpha event because the event loss occurred not only for alpha event from ^{215}Po nuclei but also following events. For that reason, it was difficult to estimate the real measured time. We therefore used event counts for normalization to compare the subtraction method and V_p/Q_{total} discrimination methods in Section III C.

C. Detection of low-rate environmental gamma ray

As shown in Figure 7, the contribution of ^{208}Tl (2.614 MeV) gamma rays was clearly detected. This means that Thorium series nuclei exist in the measurement environment. In the raw data, ^{214}Bi (1.764 MeV) of Uranium series could not be identified; however, they were clearly identified using the V_p/Q_{total} discrimination method with the 3 $\sigma_{V_p/Q_{total}}$ threshold function. On the other hand, ^{212}Bi (1.620 MeV) of Thorium series nuclei could be observed in both cases of raw data and alpha rejection as shown in Fig. 7. For the subtraction method, the actual background spectrum also depends on the surrounding materials and it would therefore be quite difficult to eliminate from it the ubiquitous presence of ^{212}Bi. With a background spectrum containing ^{212}Bi peak, it will thus be difficult to identify ^{212}Bi in the actual measurement. Therefore, the V_p/Q_{total} discrimination method can be considered a more practical approach for surveying environmental radiation.

D. The V_p/Q_{total} discrimination method extensibility

Although the V_p/Q_{total} discrimination seems to be categorized in the PSD, the use of this method is not restricted only for alpha discrimination in LaBr3 self-activity. The PSD method is applied in discriminating neutron and gamma event using a plastic scintillator. Rise time to fall time ratio is analyzed by a partial integration window in the traditional method. Recently, oscilloscope and flash analog-to-digital

TABLE I. Total energy peak distribution of rejected alpha-decay background using the background subtraction method and V_p/Q_{total} discrimination method.

	Peak counts			
	Measurement[a]	Fitting	Mean energy (MeV)	FWHM (keV)
BG subtraction	139.6 ± 28.54	129.9 ± 6.34	1885.9 ± 0.7334	33.1 ± 3.62 (1.755%)
V_p/Q_{total} discrimination	138.6 ± 19.22	127.7 ± 4.46	1885.5 ± 0.5281	36.2 ± 3.31 (1.920%)
Difference (%)	0.716	1.694	0.021	0.165

[a]The error of measured value is a statistics error, and the error of other parameters mean a fitting error.

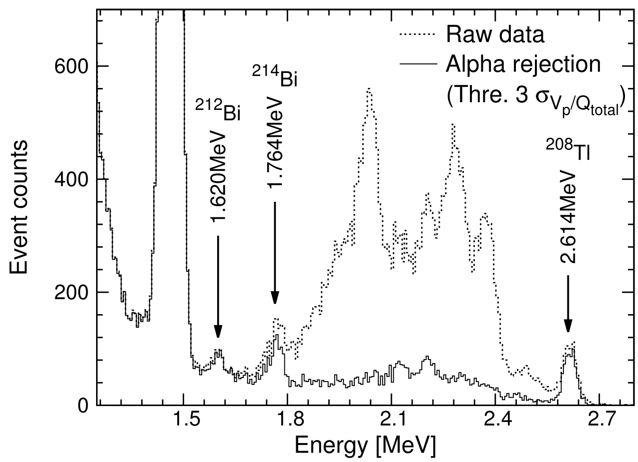

FIG. 7. The energy spectrum of environmental radiation obtained by using the $LaBr_3$:Ce scintillator with the V_p/Q_{total} analysis (3 $\sigma_{V_p/Q_{total}}$ threshold function).

converter (ADC) driven in GHz are available. Time resolution in few picosecond can thus be obtained. The V_p/Q_{total} discrimination method is expected to be simpler and more accurate than the conventional PSD methods.

For our future work, we are planning to make a hardware-based V_p/Q_{total} discrimination system using a flash ADC (ADC12J4000, Texas Instruments). The flash ADC can acquire 4 Giga sampling per second, implies a time resolution of 250 ps, and intrinsic latency of about 200 ns. However, the current V_p/Q_{total} discrimination method applies FFT/IFFT for noise reduction, which might take a few millisecond for calculation. Making the sampling rate lower is a possible solution for the realistic implementation of V_p/Q_{total} discrimination method, but discrimination accuracy may be worse depending on the time resolution because the lower sampling rate makes the peak voltage lower.

V. CONCLUSION

In the present research, alpha-decay events of the $LaBr_3$:Ce scintillator self-activity were discriminated by analyzing the peak-to-charge ratio V_p/Q_{total} of the PMT-output waveform. The V_p/Q_{total} discrimination method has demonstrated only small accidental gamma rejection, and the energy spectrum after alpha-rejection could be reproduced exactly as that of the background subtraction method with higher-accuracy. The presented method uses measurable parameters (V_p and Q_{total}) using flash ADC, and the entire process can be implemented with an electric circuit. The authors strongly believe that the V_p/Q_{total} discrimination method will be used as one of the standard techniques for gamma ray energy spectrometry using $LaBr_3$:Ce scintillators especially for environmental gamma ray surveys.

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