$Q_\beta$- measurements with a total absorption detector
composed of through-hole HPGe detector and
anti-Compton BGO detector

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Abstract

A total absorption detector, which is composed of a through-hole type HPGe detector coupled with a surrounding annular anti-Compton BGO detector, has been developed for $\beta^-$-decay energy ($Q_\beta$) measurements of nuclei far off the $\beta$-stability line. This detector can measure radiations with an almost $4\pi$ solid angle by putting radioactive sources in the middle of the HPGe detector and also can suppress the scattered photons with the anti-Compton BGO scintillation detector. The systematic uncertainty was determined to be 30 keV by measuring 19 nuclei having $Q_\beta$s between 3 and 8 MeV by means of conventional square root plot analysis. The $Q_\beta$s of mass-separated fission products $^{147}$La and $^{148}$La were successfully determined to be 5366(40) and 7732(70) keV, respectively.

Keywords: $Q_\beta$; total absorption detector, HPGe detector, anti-Compton BGO scintillator, $^{235}$U($n,f$), $^{147}$La, $^{148}$La, on-line mass separator

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1. Introduction

Atomic masses of unstable nuclei are fundamental and important physical quantities related to nuclear structure and nucleosynthesis in astrophysics. Moreover, the masses
of fission products are important for the evaluation of decay heat in nuclear power
plants. Nevertheless, most β-decay energies ($Q_{\beta}$) of nuclei far off the β stability line
have not been determined experimentally due to the low intensities of available beams.
Available theoretical mass values and the systematics have uncertainties of at least 500
keV. It would be meaningful to determine the values experimentally with smaller
uncertainty, even as high as 100 keV. Presently, ion-trap methods coupled with on-line
mass separators (ISOL) are used at various facilities. Alternately, $Q_{\beta}$ measurement is
also considered to be a precise method. However, in $Q_{\beta}$ measurement by applying β-γ
coincidence method, information on a decay scheme is necessary. In the nuclei of
interest, and especially in newly observed isotopes, most of their decay schemes are not
known, making β-γ coincidence measurement quite difficult. On the contrary, a total
absorption detector can be extremely effective because the $Q_{\beta}$ can be determined
independently of the decay scheme information. This is because the energies of the β
and γ rays can be summed up to the $Q_{\beta}$, and the end-point of the measured spectrum
corresponds to a $Q_{\beta}$. It means the $Q_{\beta}$ can be determined independently of the decay
scheme information. Hence, a total absorption detector composed of twin large volume
BGO scintillators has been developed and installed in two ISOL facilities (KUR-ISOL
and Tokai-ISOL). The $Q_{\beta}$s of fission products of $^{235}$U(n,f) or $^{238}$U(p,f) including new
isotopes have been successfully determined independently of the decay scheme information [1-4]. This total absorption detector has higher efficiency but less energy resolution in comparison with the Ge detector, so the deduced $Q_{\beta s}$ include uncertainties of 60–100 keV.

In order to determine $Q_{\beta s}$ with higher accuracy and reliability, another total absorption detector composed of HPGe and BGO scintillation detectors has been recently developed. The efficiency of this total absorption detector is less than that of the BGO total absorption detector [1], but the energy resolution is much higher. Moreover, energy calibration can be carried out up to the high energy region by the observed γ-rays (e.g. prompt γ-rays). Consequently, the deduced $Q_{\beta s}$ are much more reliable.

The performance of the detector, namely that it could measure $Q_{\beta s}$ with systematic uncertainties of within 30 keV under good statistics, was confirmed by measuring the well-evaluated $Q_{\beta s}$ between 3 and 8 MeV using conventional square root plot analysis. Finally, the $Q_\beta$ of $^{147}$La and $^{148}$La that were separated from fission products using an on-line separator were proposed to be 5366(40) and 7732(70) keV, respectively.
2. Experimental

As shown in Fig. 1, the newly developed detector is composed of two detectors. One is a custom-made HPGe detector in which the large volume single crystal (84mm$^\phi \times 90$mm$^t$) has a 20mm$^\phi$ diameter through hole in the center. This allows radiation to be measured with an almost $4\pi$ solid angle and to be measured energy sum of $\beta$-ray and $\gamma$-ray following the $\beta$-decay. The Ge crystal is covered with aluminum housing and the thickness of the well is 0.4 mm in order to reduce the energy loss and absorption as low as possible. The other detector is the 25mm thick anti-Compton BGO detector which surrounds the HPGe detector. The energy resolution of the HPGe detector was approximately 2.5 keV at the 1332 keV $\gamma$-ray. Measurements were taken as follows: a singles spectrum using the HPGe detector and a coincidence spectrum with the BGO detector were measured simultaneously. Here, coincidence events correspond to incomplete energy absorption events, namely Compton components of $\gamma$-rays or bremsstrahlung photons associated with $\beta$-particles. By subtracting the coincidence spectrum from the singles spectrum, total absorption events can be extracted. Therefore, the deduced spectrum principally corresponds to the superimposed spectrum of a fully absorbed $\beta$-ray, and the end point of the spectrum corresponds to the $Q_{\beta}$.

In practice, $\beta$-ray spectra are distorted by the energy distribution resulting from
energy straggling by the Al housing, a dead layer including some materials on the surface of the crystals. Hence, in order to determine the effective energy loss experimentally, many nuclei having well-determined $Q$ were measured, and the measured end point energies were compared to their evaluated values. Nine radioisotopes $^{27}$Mg, $^{38}$Cl, $^{42}$K, $^{52}$V, $^{56}$Mn, $^{72}$Ga, $^{90}$Y, $^{139}$Ba and $^{142}$Pr were prepared by the $(n,\gamma)$ reactions at the pneumatic tube facility in the Kyoto University Reactor (KUR) and ten fission products of $^{91-94}$Rb, $^{93,95}$Sr and $^{139-142}$Cs were provided by the on-line mass separator at the KUR (KUR-ISOL) [e.g. 5, 6]. On the $(n,\gamma)$ experiment, radioactive sources in liquid form were dropped on thin filter paper. On the ISOL experiment, the mass-separated radioactive beams were implanted into a thin Mylar tape, and the sources on the tape were moved periodically with predetermined time interval from the collecting port in a vacuum chamber to the measuring position in the air using differential pumping. The tape was computer-controlled and the reproducibility of the source position was within 1 mm. On the ISOL experiment, the detector was shielded from the background neutrons with 5 cm lead blocks, 5 mm thick 40% boron-doped rubber sheets and 20 cm thick paraffin blocks. These shields reduced the background from 830 to 120 cps, approximately. This background rate was nearly constant during the measurements. In both experiments, the source preparations and measurements were
iterated many times in order to obtain sufficient counting statistics. The total counting rate was kept below 1.5 kcps to reduce pulse pile-up. Energy calibration was carried out with $^{24}$Na and capture $\gamma$-rays of the surrounding materials such as Fe, H, and N.

3. Results

In this paper, the results obtained through conventional analyzing method, namely square root plots method, is described. The total absorption spectrum was deduced from the singles spectrum by subtracting the coincidence spectrum multiplied by a factor of 1.4 for every nucleus. This factor corresponds to the inverse of the 70% solid angle of the BGO scintillation detector and it was consistent with not only the measurements, but also with the simulations by the Monte Carlo code GEANT4 [8] for mono-energetic $\gamma$-rays. The background spectra in singles and coincidences were also subtracted from the each measured spectrum. Two typical total absorption spectra of $^{93}$Sr($Q_{\beta} = 4.2$ MeV) [7] and $^{92}$Rb($Q_{\beta} = 8.2$ MeV) [7] are shown in Fig. 2(a) and (b), respectively, together with singles and coincidence spectra. Here, the spectrum was analyzed with the energy bin of 20 keV in convenience. Generally, in energy spectra measured with Ge detector, the energy can be determined within the uncertainty of 1/5 channel or much better, in this case 4 keV, under the good statistics condition. The
energy broadening represented as the FWHM of response for 3-8 MeV mono-energetic electrons, which occurs mainly by the housing of Aluminum well, is evaluated to be 70 keV by the Monte Carlo simulation GEANT4, approximately. Moreover, as mentioned later, the uncertainty of the analysis is evaluated to be 30 keV practically. Therefore, the uncertainty originated from the bin does not have influence on the analysis, and the energy bin of this 20 keV is enough to analyze the spectra with much better precision than that of the BGO detector.

The square root plot method is often the preferred method of $Q_0$ analysis for measurements with a plastic scintillation detector and can also be applied to the spectra measured with the total absorption BGO detector [1, 2]. As shown in the inset of Fig. 2, the square root plot of the spectrum shows an almost straight line near the end-point. It suggests the distortion of the response for the electrons does not have influence so much near the end point energy at least 500 keV, and the root plot analysis is enough to analyze the measured total absorption spectrum in this detector. The negative counts in square root plots show putting the minus sign when the counts become negative after subtracting the coincidence spectra. The fact that the counts scatter around zero in high energy background region after subtracting means the spectra were subtracted properly. The spectrum also shows tailing to the high energy side at the end-point owing to
energy broadening. In the root plot analysis, the region of interest for analysis was chosen to be about 500 keV below 100–200 keV from the end-point of each spectrum in order to exclude this tailing effects. This method was employed for all spectra independently of each decay scheme.

The comparison between the deduced end-point energies ($E_{\beta\text{-max}}$) obtained through energy calibration carried out with $\gamma$-ray peaks and the literature $Q_\beta$ values [7] is shown in Fig. 3. The effective energy loss by this analyzing method was experimentally determined to be 180 keV between 2 and 8 MeV with an uncertainty of 30 keV. The $Q_\beta$ of newly measured nuclei can be deduced by adding this effective energy loss to the $E_{\beta\text{-max}}$ of the spectra. The origin of the energy loss was evaluated as follows. According to the Monte Carlo simulation for mono-energetic electrons, the energy loss by Mylar tape was evaluated to be 20 keV, approximately. So, that by 0.4 mm$^\dagger$ Al housing and dead layer including some materials on the crystal well was deduced to be 160 keV, approximately. On the other hand, that by only 0.4 mm$^\dagger$ Al housing was evaluated to be 130 keV, approximately, then, that by dead layer including surface materials was evaluated to be 30 keV. This effective energy loss for the (n,$\gamma$) experiment was almost the same as that for the ISOL experiment. The uncertainty obtained through the conventional analysis is comparable with, or better than, that of the BGO total
The $Q_{\beta}$ of some nuclei around $A=150$ were measured with the detector at the KUR-ISOL. The periods for collection-measurement of the tape transport system for $^{147}$La ($T_{1/2}=4.0$ s [9]) and $^{148}$La ($T_{1/2}=1.4$ s [9]) were set at 8 s – 8 s and 3.4 s – 3.4 s, respectively, to reduce each daughter activity. The total measurement period for each nucleus was longer than 15 hours. As shown in Fig. 4(a) and (b), the $E_{\beta\text{-max}}$ of $^{147}$La and $^{148}$La were analyzed to be 5186 keV and 7552 keV, respectively, by means of the square root plot as described above. Finally, after adding the effective energy loss of 180 keV, the $Q_{\beta}$s were deduced to be 5366(40) and 7732(70) keV for $^{147}$La and $^{148}$La, respectively. The uncertainties were derived from the effective energy loss and each statistic. The deduced value for $^{148}$La has a relatively larger uncertainty which consists in the 60 keV statistical one of 60 keV and the systematic one of 30 keV, approximately.

4. Discussion

The results and the previously proposed values are summarized in Table 1. The value determined for $^{147}$La is not in agreement with the previously reported values of 4945(55) keV [10] and 5150(40) keV [11] over their uncertainties. However, it is close to the value of 5150(40) keV. The value of 4945(55) keV [10] was adopted by Audi et al.
in 1995. The preliminary proposed value of 5370(100) keV [12], which was measured with the BGO total absorption detector, is in good agreement with the present result. The last evaluation in 2003 [7]; 5180(40) keV; seems to take ref. [11] into account intensively, but not ref. [12]. Anyhow, the value of 4945(55) keV is quite small compared to the present result.

Similarly, the result for $^{148}$La is also in agreement with the result that was measured with the BGO total absorption detector [12]. Previously, three experimental values of >5862(100) [13], 7255(55) [14] and 7650(100) keV [12] were proposed. As described in ref. 14, the value of 5862 keV [13] were re-evaluated to be 7310(150) keV from the information of decay study. In the previous systematics in 1995 [15], the value of 7262(50) keV was proposed. It seems to stand on the ref. 14 only. The last systematics of 7260(50) keV for $^{148}$La by Audi et al. [7], seems to exclude the value of 7650(100) keV [12]. In the case of the neutron-rich La isotopes, it had proposed that there is disagreement in the two neutron separation energies ($S_{2n}$) between the experimental values [12] and the systematics determined by Audi et al. [15]. [16]

Recently, Clark et al. [17] also proposed that the experimentally determined atomic masses of $^{147,148}$La by Canadian ion trap were more than 400 keV larger than those in Audi et al. [15]. These results are consistent with the present results. Such large
differences might indicate some nuclear effects. Spectroscopic study is necessary to further understanding of the nuclear structure. As mention above, the systematics strongly depends upon the experimental values. It means the experimental values are important for the proposition of reliable systematics. In further analysis taking accounts of the energy broadening owing to energy straggling, the tailing region in high energy side could be included in the analyzing region, and then the uncertainties will be expected to be reduced. The results will be described in detail in a forthcoming paper.

In conclusion, a novel total absorption detector for $Q_\beta$ determination was developed. It is composed of a large volume HPGe detector having a through-hole and a BGO anti-Compton detector. The ability of the detector to determine $Q_\beta$ with high accuracy and independently of decay scheme information was demonstrated. The $Q_\beta$s of the $^{147}$La and $^{148}$La were determined to be 5366(40) and 7732(70) keV, respectively.

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References


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<td>5366(40)</td>
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<td>7732(70)</td>
<td>7310(150)$^e$</td>
<td>7255(55)$^f$</td>
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a) taken from ref.15  b) taken from ref.7  c) taken from ref.10  d) taken from ref.11  
e) taken from ref.13  f) taken from ref.14  g) taken from ref.12
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Fig. 1 Schematic view of the total absorption detector. The size is indicated in mm scale.

Fig. 2 Measured spectra of mass-separated $^{93}\text{Sr}(a)$ and $^{92}\text{Rb}(b)$. The solid and broken lines indicate the singles and coincidence spectra, respectively. Dots indicate the total absorption spectrum which was obtained by subtracting the coincidence spectrum from the singles one multiplied by a factor of 1.4. The inset shows the square root plot of the total absorption spectrum. The region between two dotted lines indicates the analyzed region. It shows almost straight line near the end-point. The negative counts in square root plots show putting the minus sign when the counts become negative after subtracting the coincidence spectra. The counts scatter after subtracting around zero in high energy region. It means the spectra were subtracted properly.

Fig. 3 Effective energy loss of the detector. The differences between the determined end-point energies and the evaluated $Q_{\beta}s$ show the effective energy loss of the HPGe detector.

Fig. 4 The square root plots and the deduced $E_{\beta\text{-max}}$ of $^{147}\text{La}(a)$ and $^{148}\text{La}(b)$. Each region between dotted lines is adopted for analysis.
movable tape

RI beam from ISOL

Differential Pumping

Photomultiplier Tube

Ge

BGO

0.4 mm Al

Cold finger

32

90

25

150
(a) $^{93}\text{Sr}$

Counts per energy

Energy (keV)

(a) $^{93}\text{Sr}$

Counts per energy

Energy (keV)

Counts per energy

Energy (keV)

Counts per energy

Energy (keV)

Counts per energy

Energy (keV)

Counts per energy

Energy (keV)

Counts per energy

Energy (keV)

Counts per energy

Energy (keV)

Counts per energy

Energy (keV)
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<td>10^4</td>
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<tr>
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<td>10^5</td>
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In the diagram:
- **Singles**
- **Coincidence**
- **Total absorption**

Inset inset:
- **Singles**
- **Coincidence**
- **Total absorption**

The graph shows the relationship between energy (keV) and the square root of the number of counts (N). The energy range is from 4000 to 5200 keV.

- **^{147}\text{La}**