1	Q_{eta} - measurements with a total absorption detector
2	composed of through-hole HPGe detector and
3	anti-Compton BGO detector
4	
5	Hiroaki Hayashi ^{a,1} , Michihiro Shibata ^b , Osamu Suematsu ^a , Yasuaki Kojima ^c ,
6	Kiyoshi Kawade ^a , Akihiro Taniguchi ^d
7	^a Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan
8	^b Radioisotope Research Center, Nagoya University, Nagoya 464-8602, Japan
9	^c Graduate School of Engineering, Hiroshima University, Higashi-Hiroshima 739-8527,
10	Japan
11	^d Research Reactor Institute, Kyoto University, Kumatori 590-0494, Japan
12	

¹Corresponding author. E-mail: hayashi.hiroaki@b.mbox.nagoya-u.ac.jp; FAX: +81 52 789 2573

13 Abstract

14	A total absorption detector, which is composed of a through-hole type HPGe
15	detector coupled with a surrounding annular anti-Compton BGO detector, has been
16	developed for β^- -decay energy (Q_β) measurements of nuclei far off the β -stability line.
17	This detector can measure radiations with an almost 4π solid angle by putting
18	radioactive sources in the middle of the HPGe detector and also can suppress the
19	scattered photons with the anti-Compton BGO scintillation detector. The systematic
20	uncertainty was determined to be 30 keV by measuring 19 nuclei having $Q_{\beta}s$ between 3
21	and 8 MeV by means of conventional square root plot analysis. The $Q_{\beta}s$ of
22	mass-separated fission products ¹⁴⁷ La and ¹⁴⁸ La were successfully determined to be
23	5366(40) and 7732(70) keV, respectively.
24	<i>Keywords</i> : Q_{β} ; total absorption detector, HPGe detector, anti-Compton BGO scintillator,

25 235 U(*n*,*f*), 147 La, 148 La, on-line mass separator

26 PACS : 29.40.Mc; 23.40.-s; 25.85.Ec

27

28 1. Introduction

Atomic masses of unstable nuclei are fundamental and important physical quantities
related to nuclear structure and nucleosynthesis in astrophysics. Moreover, the masses

31	of fission products are important for the evaluation of decay heat in nuclear power
32	plants. Nevertheless, most β -decay energies (Q_{β}) of nuclei far off the β stability line
33	have not been determined experimentally due to the low intensities of available beams.
34	Available theoretical mass values and the systematics have uncertainties of at least 500
35	keV. It would be meaningful to determine the values experimentally with smaller
36	uncertainty, even as high as 100 keV. Presently, ion-trap methods coupled with on-line
37	mass separators (ISOL) are used at various facilities. Alternately, Q_{β} measurement is
38	also considered to be a precise method. However, in Q_{β} measurement by applying β - γ
39	coincidence method, information on a decay scheme is necessary. In the nuclei of
40	interest, and especially in newly observed isotopes, most of their decay schemes are not
41	known, making β - γ coincidence measurement quite difficult. On the contrary, a total
42	absorption detector can be extremely effective because the Q_{β} can be determined
43	independently of the decay scheme information. This is because the energies of the β
44	and γ rays can be summed up to the Q_{β} , and the end-point of the measured spectrum
45	corresponds to a Q_{β} . It means the Q_{β} can be determined independently of the decay
46	scheme information. Hence, a total absorption detector composed of twin large volume
47	BGO scintillators has been developed and installed in two ISOL facilities (KUR-ISOL
48	and Tokai-ISOL). The $Q_{\beta}s$ of fission products of $^{235}U(n,f)$ or $^{238}U(p,f)$ including new

49 isotopes have been successfully determined independently of the decay scheme 50 information [1-4]. This total absorption detector has higher efficiency but less energy 51 resolution in comparison with the Ge detector, so the deduced $Q_{\beta}s$ include uncertainties 52 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_{β} of ¹⁴⁷La and ¹⁴⁸La that were separated from fission products using an on-line separator were proposed to be 5366(40) and 7732(70) keV, respectively.

66

67 2. Experimental

As shown in Fig. 1, the newly developed detector is composed of two detectors. 68 One is a custom-made HPGe detector in which the large volume single crystal 69 $(84 \text{mm}^{\phi} \times 90 \text{mm}^{t})$ has a 20 mm diameter through hole in the center. This allows 70 71 radiation to be measured with an almost 4π solid angle and to be measured energy sum of β -ray and γ -ray following the β -decay. The Ge crystal is covered with aluminum 72 housing and the thickness of the well is 0.4 mm in order to reduce the energy loss and 73 absorption as low as possible. The other detector is the 25mm thick anti-Compton BGO 74 detector which surrounds the HPGe detector. The energy resolution of the HPGe 75 76 detector was approximately 2.5 keV at the 1332 keV γ -ray. Measurements were taken as follows: a singles spectrum using the HPGe detector and a coincidence spectrum with 77 the BGO detector were measured simultaneously. Here, coincidence events correspond 78 to incomplete energy absorption events, namely Compton components of y-rays or 79 bremsstrahlung photons associated with β -particles. By subtracting the coincidence 80 spectrum from the singles spectrum, total absorption events can be extracted. Therefore, 81 the deduced spectrum principally corresponds to the superimposed spectrum of a fully 82 absorbed β -ray, and the end point of the spectrum corresponds to the Q_{β} . 83

84

In practice, β -ray spectra are distorted by the energy distribution resulting from

5

85	energy straggling by the Al housing, a dead layer including some materials on the
86	surface of the crystals. Hence, in order to determine the effective energy loss
87	experimentally, many nuclei having well-determined $Q_{\beta}s$ were measured, and the
88	measured end point energies were compared to their evaluated values. Nine
89	radioisotopes ²⁷ Mg, ³⁸ Cl, ⁴² K, ⁵² V, ⁵⁶ Mn, ⁷² Ga, ⁹⁰ Y, ¹³⁹ Ba and ¹⁴² Pr were prepared by the
90	(n,γ) reactions at the pneumatic tube facility in the Kyoto University Reactor (KUR) and
91	ten fission products of ⁹¹⁻⁹⁴ Rb, ^{93,95} Sr and ¹³⁹⁻¹⁴² Cs were provided by the on-line mass
92	separator at the KUR (KUR-ISOL) [e.g. 5, 6]. On the (n,γ) experiment, radioactive
93	sources in liquid form were dropped on thin filter paper. On the ISOL experiment, the
94	mass-separated radioactive beams were implanted into a thin Mylar tape, and the
95	sources on the tape were moved periodically with predetermined time interval from the
96	collecting port in a vacuum chamber to the measuring position in the air using
97	differential pumping. The tape was computer-controlled and the reproducibility of the
98	source position was within 1 mm. On the ISOL experiment, the detector was shielded
99	from the background neutrons with 5 cm lead blocks, 5 mm thick 40% boron-doped
100	rubber sheets and 20 cm thick paraffin blocks. These shields reduced the background
101	from 830 to 120 cps, approximately. This background rate was nearly constant during
102	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 ²⁴Na and capture γ -rays of the surrounding materials such as Fe, H, and N.

106

107 3. Results

In this paper, the results obtained through conventional analyzing method, 108 namely square root plots method, is described. The total absorption spectrum was 109 deduced from the singles spectrum by subtracting the coincidence spectrum multiplied 110 by a factor of 1.4 for every nucleus. This factor corresponds to the inverse of the 70% 111 solid angle of the BGO scintillation detector and it was consistent with not only the 112 measurements, but also with the simulations by the Monte Carlo code GEANT4 [8] for 113 mono-energetic γ -rays. The background spectra in singles and coincidences were also 114 subtracted from the each measured spectrum. Two typical total absorption spectra of 115 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), 116 respectively, together with singles and coincidence spectra. Here, the spectrum was 117 analyzed with the energy bin of 20 keV in convenience. Generally, in energy spectra 118 measured with Ge detector, the energy can be determined within the uncertainty of 1/5119 120 channel or much better, in this case 4 keV, under the good statistics condition. The

121	energy broadening represented as the FWHM of response for 3-8 MeV mono-energetic							
122	electrons, which occurs mainly by the housing of Aluminum well, is evaluated to be 70							
123	keV by the Monte Carlo simulation GEANT4, approximately. Moreover, as							
124	mentioned later, the uncertainty of the analysis is evaluated to be 30 keV practically.							
125	Therefore, the uncertainty originated from the bin does not have influence on the							
126	analysis, and the energy bin of this 20 keV is enough to analyze the spectra with much							
127	better precision than that of the BGO detector.							
128	The square root plot method is often the preferred method of Q_{β} analysis for							
129	measurements with a plastic scintillation detector and can also be applied to the spectra							
130	measured with the total absorption BGO detector [1, 2]. As shown in the inset of Fig. 2,							
131	the square root plot of the spectrum shows an almost straight line near the end-point.							
132	It suggests the distortion of the response for the electrons does not have influence so							
133	much near the end point energy at least 500 keV, and the root plot analysis is enough to							
134	analyze the measured total absorption spectrum in this detector. The negative counts							
135	in square root plots show putting the minus sign when the counts become negative after							
136	subtracting the coincidence spectra. The fact that the counts scatter around zero in high							
137	energy background region after subtracting means the spectra were subtracted properly.							
138	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.

143 The comparison between the deduced end-point energies ($E_{\beta-max}$) obtained through energy calibration carried out with γ -ray peaks and the literature Q_{β} values [7] is 144 shown in Fig. 3. The effective energy loss by this analyzing method was experimentally 145 determined to be 180 keV between 2 and 8 MeV with an uncertainty of 30 keV. The Q_{β} 146 of newly measured nuclei can be deduced by adding this effective energy loss to the 147 $E_{\beta-max}$ of the spectra. The origin of the energy loss was evaluated as follows. According 148 to the Monte Carlo simulation for mono-energetic electrons, the energy loss by Mylar 149 150 tape was evaluated to be 20 keV, approximately. So, that by 0.4 mm^t Al housing and dead layer including some materials on the crystal well was deduced to be 160 keV, 151 approximately. On the other hand, that by only 0.4 mm^t Al housing was evaluated to 152 be 130 keV, approximately, then, that by dead layer including surface materials was 153 154 evaluated to be 30 keV. This effective energy loss for the (n,γ) experiment was almost the same as that for the ISOL experiment. The uncertainty obtained through the 155 156 conventional analysis is comparable with, or better than, that of the BGO total

absorption detector [3].

The Q_{β} of some nuclei around A=150 were measured with the detector at the 158 KUR-ISOL. The periods for collection-measurement of the tape transport system for 159 ¹⁴⁷La (T_{1/2}=4.0 s [9]) and ¹⁴⁸La (T_{1/2}=1.4 s [9]) were set at 8 s - 8 s and 3.4 s - 3.4 s, 160 respectively, to reduce each daughter activity. The total measurement period for each 161 nucleus was longer than 15 hours. As shown in Fig. 4(a) and (b), the $E_{\beta-max}$ of ¹⁴⁷La and 162 ¹⁴⁸La were analyzed to be 5186 keV and 7552 keV, respectively, by means of the square 163 root plot as described above. Finally, after adding the effective energy loss of 180 keV, 164 the $Q_{\beta}s$ were deduced to be 5366(40) and 7732(70) keV for ¹⁴⁷La and ¹⁴⁸La, respectively. 165 The uncertainties were derived from the effective energy loss and each statistic. The 166 deduced value for ¹⁴⁸La has a relatively larger uncertainty which consists in the 60 keV 167 168 statistical one of 60 keV and the systematic one of 30 keV, approximately.

169

170 4. Discussion

The results and the previously proposed values are summarized in Table 1. The value determined for ¹⁴⁷La 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 ¹⁴⁸La is also in agreement with the result that was 180 measured with the BGO total absorption detector [12]. Previously, three experimental 181 values of >5862(100) [13], 7255(55) [14] and 7650(100) keV [12] were proposed. As 182 described in ref. 14, the value of 5862 keV [13] were re-evaluated to be 7310(150) keV 183 from the information of decay study. In the previous systematics in 1995 [15], the value 184 of 7262(50) keV was proposed. It seems to stand on the ref. 14 only. The last 185 systematics of 7260(50) keV for ¹⁴⁸La by Audi et al. [7], seems to exclude the value of 186 7650(100) keV [12]. In the case of the neutron-rich La isotopes, it had proposed that 187 there is disagreement in the two neutron separation energies (S_{2n}) between the 188 experimental values [12] and the systematics determined by Audi et al. [15]. [16] 189 Recently, Clark et al. [17] also proposed that the experimentally determined atomic 190 masses of ^{147,148}La by Canadian ion trap were more than 400 keV larger than those in 191 192 Audi et al. [15]. These results are consistent with the present results. Such large

193	differences might indicate some nuclear effects. Spectroscopic study is necessary to
194	further understanding of the nuclear structure. As mention above, the systematics
195	strongly depends upon the experimental values. It means the experimental values are
196	important for the proposition of reliable systematics. In further analysis taking
197	accounts of the energy broadening owing to energy straggling, the tailing region in high
198	energy side could be included in the analyzing region, and then the uncertainties will be
199	expected to be reduced. The results will be described in detail in a forthcoming paper.
200	In conclusion, a novel total absorption detector for Q_{β} determination was
201	developed. It is composed of a large volume HPGe detector having a through-hole and a
202	BGO anti-Compton detector. The ability of the detector to determine Q_{β} with high
203	accuracy and independently of decay scheme information was demonstrated. The $Q_{\beta}s$ of
204	the ¹⁴⁷ La and ¹⁴⁸ La were determined to be 5366(40) and 7732(70) keV, respectively.
205	

206 Acknowledgements

This work was carried out under the Research Collaboration Program of the Research
Reactor Institute, Kyoto University. This work was partially supported by the Ministry
of Education, Science, Sports and Culture, Grant-in-Aid for Scientific Research (B),
No.15360504.

211 References

- 212 [1] M. Shibata et al., Nucl. Instr. and Meth. A459 (2001) 581.
- 213 [2] M. Shibata et al., J. Phys. Soc. Jpn. 71 (2002) 1401.
- 214 [3] H. Hayashi et al., E. Phys. J. A 34, (2007) 363.
- 215 [4] H. Hayashi et al., submitted in J. Phys. Soc. Jpn.
- 216 [5] A. Taniguchi et al., Nucl. Instr. and Meth. A351 (1994) 378.
- 217 [6] Y. Kawase and K. Okano, Nucl. Instr. and Meth. B37/38 (1989) 116.
- 218 [7] http://www.geant4.org/geant4/, e.g. S. Agostinelli et al., Nucl. Instr. and Meth.
- 219 A506 (2003) 250.
- 220 [8] G. Audi and A.H. Wapstra, Nucl. Phys. A729 (2003) 337.
- 221 [9] Chart of the Nuclides 2004, Japanese Nuclear Data Committee and Nuclear Data
- 222 Center, Japan Atomic Energy Research Institute.
- 223 [10] M. Graefenstedt et al., Proc. of 5th Int. Conf. Nuclei far from stability, NUFAST-5,
- 1987, p.30, Edited by Ian S. Towner et al. AIP Conf. Proc. 164.
- 225 [11] T. Ikuta et al., J. Phys. Soc. Jpn. 64 (1995) 3244.
- 226 [12] M. Shibata et al., Proc. of the Third Int. Conf. on Atomic masses and Exotic nuclei,
- Hämeenlinna, Finland, 2-7, July, 2001, p.479, Edited by J. Äystö et al. Springer.
- 228 [13] D. S. Brenner et al., Phys. Rev. C 26 (1982) 2166.

- 229 [14] M. Graefenstedt et al., Z. Phys. A336 (1990) 247.
- 230 [15] G. Audi and A.H. Wapstra, Nucl. Phys. A595 (1995) 409.
- 231 [16] O. Suematsu et al., KURRI Prog. Rep. (2001) 37. Published by the Research
- 232 Reactor Institute, Kyoto University. ISSN 0919.
- 233 [17] J. A. Clark et al., Nucl. Phys. A 746 (2004) 342c.

	_			Q	p_{β}		
	Nuclide	Experimental				Systematics	
	-	Present		Previous		1995 ^a	2003 ^b
	¹⁴⁷ La	5366(40)	4945(55) ^c	5150(40) ^d	5370(100) ^g	4945(55)	5180(40)
	¹⁴⁸ La	7732(70)	7310(150) ^e	7255(55) ^f	7650(100) ^g	7262(50)	7260(50)
	a) taken f	rom ref.15	b) taken from re	f.7 c) taken	from ref.10	d) taken from r	ef.11

Table 1 The comparison of the experimental and systematics Q_{β} for ¹⁴⁷La and ¹⁴⁸La.

a) taken from ref.15b) taken from ref.7c) taken from ref.10e) taken from ref.13f) taken from ref.14g) taken from ref.12

237 List of Figures

Fig. 1 Schematic view of the total absorption detector. The size is indicated in mm scale.

240

Measured spectra of mass-separated ⁹³Sr(a) and ⁹²Rb(b). The solid and broken Fig. 2 241 lines indicate the singles and coincidence spectra, respectively. Dots indicate the 242 243 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 244 square root plot of the total absorption spectrum. The region between two dotted 245 lines indicates the analyzed region. It shows almost straight line near the 246 247 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 248 counts scatter after subtracting around zero in high energy region. It means the 249 250 spectra were subtracted properly.

251

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.

255

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





