A BI4π COUNTING METHOD FOR THE MEASUREMENT OF ACTIVITY*

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ABSTRACT

In this article, a Bi4π counting method comprised of Bi4π counter and a new fundamental measurement method was described, which can measures rather more radionuclides and can achieve accuracy of 0.03% to 0.06% (1σ), in principle. The correctness of this method is demonstrated by measurements of ^{60}Co sources which results of activities compared with 4π β − γ coincident method are in agreement within the measurement errors, and it is readily achieved to accuracy of 0.2%. Finally, the range of the applications for this Bi4π counting method among one hundred or more important radionuclides is also discussed.

Key words: Activity Bi4π method Measurement 4π Counter

I . INTRODUCTION

Th uncertainties of 0.03% level are required for the establishment of primary standards, scientific reasearch, military and nuclear engineering[1]. The radioactive laboratory was established by BIPM in 1960 and the great efforts in promoting and leading the radionuclide metrology of the world for the improvement of measurement accuracy. In 1940's, for the defined-geometry activity measurement methods, its uncertainty was 2%, 1% for 4π β counting methods in 1950's, and since 1960's, the uncertainty of the 4π β − γ coincident method can be 0.3%. However, no results in the measurements of the most radionuclides can be achieved to the accuracy of 0.03%, and in general, remained at the level of 0.3%[2] except ^{60}Co and ^{139}Ce.

With the 4π β − γ coincident methods are using the γ − rays in measuring activity, in principle, it could overcome the β − ray absorption of souces, and achieve to the high accuracy; however, due to the restriction of this methods itself and complexity of nuclide decay schemes, the 4π β − γ methods have to take the extrapolation techniques to determine the activities. But the extrapolation function is sometimes disturbingly nonlinear, therefore, it is expected that the research of the extrapolation methods[3] would pare the way for achieving higher accuracy.

The Bi4π counting method is a new fundamental measurement method[4] that the source measured is put into a small 4π − particle detector which, is put into the central well hole of a large 4π γ detector,then this Bi4π countor measures simultaneously all the particles and gamma− and/or X− photons emitted from each decays of nuclides in the measured source. This work has measured the ^{60}Co source using the new method with the small 4π β proportional counter as the 4π − particle detector. The efficiency− loss of the 4π β detector mainly from self− absorption

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of sources is about of 7% which has been compensated greatly by the 4π γ detector with a high efficiency of about 90%, thus, it makes the efficiency– loss of Bi4π detector reduced to about of 0.7% that has been corrected with extrapolation method. Thus the uncertainty of measurement reduced further.

II. INSTRUMENTATION

The Bi4π instrumentation used in this work is shown in Fig.1, 4π geometry proportional counter used as 4π particle detector being worked with flowing mixed gas of 10% methane and 90% argon. An aluminium tube with a 40mm internal diameter is used as a cathode of the counter. The anodes of counter are two parallel wires of tungsten, coated with gold, whose diameter are 25μ m. The source measured is put on the support plate between the two anode wires. The 4π β detector is inserted in the central well hole of 4π γ detector. The 4π γ detector is with outside diameter of 550mm, internal diameter of 110mm, and the hight of plastic scintillator is of 550mm. The whole Bi4π detector is housed in a suitable lead shielding.

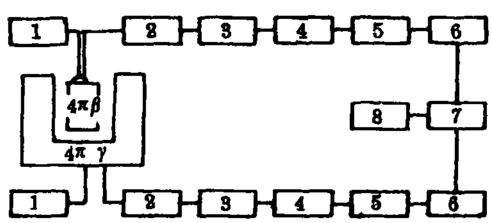

Fig. 1 Block diagram for Bi4π instrumentation

1. High voltage 2. Preamplifiers/mixers 3. Amplifiers 4. Discriminators
5. Dead– time circuits 6. Delay units 7. Adders 8. Scalers

The output pulses of β – and γ – channels are added by the adder in the sum– coincident mode, and then the pulses from adder are counted by a scaler. The adder give one output signal for every one of not coincident signal from β – or γ – channel and for each a pair signals of the β – and γ – channels which arived to the adder simultaneously, but the time difference should be less than the width of the pulses being fed to the adder.

III. PRINCIPLE

Hoppes has reviewed various fundamental measurement methods (basic calibrations) for measurements of activity[5]. In essence, all the methods including those with calculated efficiencies are of the "single radiation measurement method" except the high efficiency 4π γ counter[6,7] which should be "the multiradiation measurement method", apart from that uses cascade radiations in measurement, e.g. various coincident methods, as which are still to measure the α – or β – spectrum efficiency ε_s, and the activity $A = R_s/\varepsilon_s$, R_s is of net rate for the associated measured particles. Practically, different single radiation measurement methods are equivalent to the different methods used to determine ε_s. The high– efficiency or the Bi4π γ counter as the multiradiations measurement method measures the net rate R_t, the activity $A + R_t/\varepsilon_t$, ε_t is total efficiency for the integral rate of all γ – rays. In the cases, the decays are of multi– cascades of coincident γ – rays for which the total efficiency ε_t will increase greatly that make the accuracy of measurement results able improved largely. This suggests that if there is a detector which can detect all radiations including particles of decay from source measured should be called all– radiations detector that is limit case of them. If the particle detecting efficiency ε_p of that all– radiations detectors is high then, the accuracies of that detectors would be greatly improved; because the total efficiency will be great, and very nearly to unity, thus the efficiency– loss, $\triangle\varepsilon = 1-\varepsilon_t$, should be very small, and obviously the uncertainty (approximately proportional to $\triangle\varepsilon$) will also be very small. The Bi4π detector is one of all– radiations detectors.

1. Equations

We can take the Bi4π counter as a 'one' detector, e.g. all– radiations detector, that required to follow these conditions: β – and γ – channels with the unique and equal nonextension dead times; the resolution time is smaller than the dead times. In essence, the high efficiency 4π γ detectors are also the all– radiations detectors, apart from that their β – efficiencies (through the bremsstruhlung radiations) is relatively small and of about 4%[7].

Therefore the equations of Bi4π counter should be the similar to that for high– efficiency 4π γ detectors. For the radionuclide with q different branches, the k– th branch intensity is P_k, $k = 1, 2, ...q$, that the efficiency is obviously given by:

$$\varepsilon_t = \sum_{k=1}^{q} P_k \varepsilon_t^{(k)} \tag{1}$$

where

$$\sum_{k=1}^{q} P_k = 1 \tag{2}$$

and $\varepsilon_t^{(k)}$ is the total efficiency of k– th branch. If the k– th branch decay is the mode of β – γ cascade radiations (like the case of γ – γ cascade transitions), it is well known that the $\varepsilon_t^{(k)}$ is expressed by the equation:

$$\varepsilon_t^{(k)} = \varepsilon_\beta^{(k)} + \varepsilon_\gamma^{(k)} - \varepsilon_\beta^{(k)} \varepsilon_\gamma^{(k)} \tag{3}$$

where $\varepsilon_\beta^{(k)}$ and $\varepsilon_\gamma^{(k)}$ are the total efficiencies of the β – ray and γ – transitions respectively for the k– th branch decay. Here it does not restrict k– th gamma transition being of the single or cascade transition only that there is no the long– lived excited state.

Eq(3) may be alternatively insteaded of by one of the following two comvenient equivalent equations:

$$\varepsilon_t^{(k)} = \varepsilon_\beta^{(k)} + (1 - \varepsilon_\beta^{(k)}) \varepsilon_\gamma^{(k)} \tag{4}$$

$$\varepsilon_t^{(k)} = 1 - (1 - \varepsilon_\beta^{(k)})(1 - \varepsilon_\gamma^{(k)}) \tag{5}$$

eq. (1) using eqs. (5) and (2) is:

$$\varepsilon_t = 1 - \sum_{k=1}^{q} P_k (1 - \varepsilon_\beta^{(k)})(1 - \varepsilon_\gamma^{(k)}) \tag{6}$$

The total efficiency ε_t can be obtained by the eq.(6) and conventional method using the 4π β detector efficiency $\varepsilon_\beta^{(k)}$ and 4π γ efficiency $\varepsilon_\gamma^{(k)}$ equivalent to the level efficiency e_i referred by Winkler[8], then the activities can be got readily.

2. Extrapolation method

The extrapolation method as a experimental method to determine the activities is very suitable to the nuclides decaying by single branch.

The Bi4π counting rate $R_{4\pi}$ for the single decay is given by:

$$R_{4\pi} = A \varepsilon_i^{(k)} = A \varepsilon_i \quad (7)$$

where A is activity. The eq. (7) using eq.(4) will be:

$$R_{4\pi} = A \varepsilon_\beta + A(1-\varepsilon_\beta)\varepsilon_\gamma \quad (8)$$

the ε_β and ε_γ are activity efficiencies of samples to be measured for 4π β – and 4π γ – detectors respectively. Eq. (8) is practically a linear equation:

$$R_{4\pi} = a + b R_{4\pi\gamma} \quad (9)$$

with $a = A\varepsilon_\beta,\quad R_{4\pi\gamma} = A\varepsilon_\gamma,\ B = (1-\varepsilon_\beta)$

and $$A = a/(1-b) \quad (10)$$

By changing ε_γ with regulating the discriminating threshold level one gets a set of experimental points ($R_{4\pi\gamma i}$, $R_{4\pi i}$), i = 1, 2, ...n. Using linear fitting to these points to get a and b then the activity A is obtained with equation (10).

3. The equation for dead time counting losses

The counting losses introduced by the dead times of both β – and γ – channels are no longer independent each other due to the existing of adder and simultaneously counting then, the dead– time losses of both channels can not been compensated by the function of Bi4π counter that the efficiency– losses could be compensated by the function of Bi4π counter that the efficiency– losses could be componsated each other, therefore, the dead– time losses must be corrected explicitly.

One makes the dead times of β – and γ – channels are equal by regulating the dead– time circuits and are denoted by τ, and resolution times of adder are remained smaller than the τ. With the $N'_{4\pi}$ denoting the counts of Bi4π counter collected during the time T, and the real count rate is:

$$R^0_{4\pi} = \frac{N'_{4\pi}/T}{(1-\tau\ N'_{4\pi}/T)} \quad (11)$$

IV. TEST

1. Extrapolation curves

Table 1

Data of extrapolation curves (1/s)

i	1	2	3	4	5	6
$R_{4\pi\ i}$	3342.9	3337.1	3309.1	3262.0	3198.0	3151.3
$R_{4\pi\gamma\ i}$	2975.5	2729.8	2362.4	1669.6	925.3	403.4

The Bi4π net rates, $R_{4\pi\ i}$ and the 4π γ detector net counting rates, $R_{4\pi\gamma\ i}$ have been obtained and are shown in Table 1. The $\tau = \tau_\beta$ = 9.47 μs are used in the corrections of dead– time losses with Eq. (11), the $\tau_{\gamma} = 9.0\ \mu$ s is used for calculations of $R_{4\pi\gamma i}$.

The extrapolation curve drawn using data from Table 1 is shown in Fig.2, and for the clearness, $R_{4\pi\gamma i}$ are in stead of ε_{γ}. It is saw that there is a fine linearity. It is noted that the values of ε_{γ} points are approximately distributed on the whole range of γ – efficiency (0—1), whereas the associated counting rates $R_{4\pi}$ are only changed from 3151 to 3342 which are only equivalent about the 93% to 99% of the activity value (3383.5Bq), but the increments of extrapolation is only about of the 1%. The small increments of extrapolation make the influence of the non– regularity of the curve form and the errors of ε_{γ} on the extrapolative values would be greatly decreased.

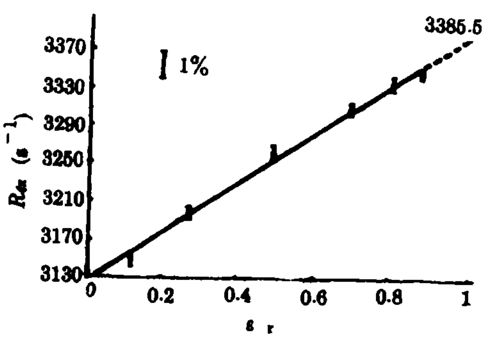

Fig.2 Extrapolation curve for determination of activity

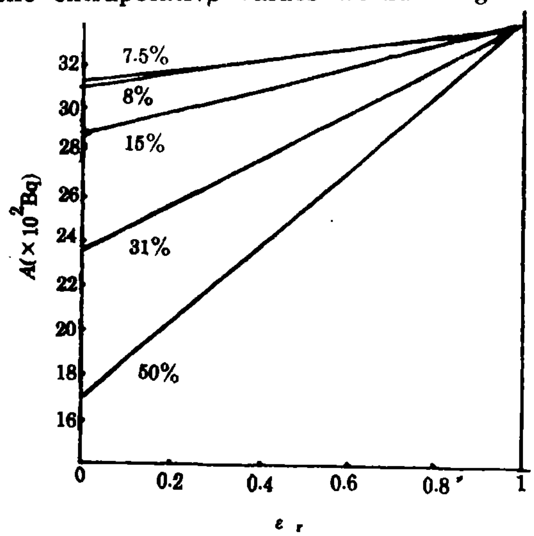

Fig.3 Tolerance of Bi4π method to self– absorption

$R_{4\pi}$: Bi4π counting rate ε_{γ}: 4π γ detection efficiency

2. Tolerance to the self– absorption

Whether 4π β method of 4π $\beta - \gamma$ coincident method requires that self– absorption of source to be measured must be as possible as small, and otherwise there will be a great measurement error. The self– absorptions of ^{60}Co sources can be reduced to 3—7% with the recent source preparation techniques, however it is still not satisfied.

For examining the tolerance of the Bi4π method to self– absorption of sources, the efficiency of 4π β detector has been decreased to 50% by reduction of working voltages that equivalently the self– absorption of source is 50%. The activity measurements are carried out, of which a set extrapolation curves are shown in Fig.3. It has been seen that the results of measurements for activity with Bi4π method are in good agreement in the range of self– absorption from 7.6% to 50%, as the extrapolation lines intersect into one point and the spread is less than $\pm 0.2\%$. This remained spread will be analysed and eliminated in the next subparagraph.

3. Corrections

The 4π γ detector is of a scintillation counter which can produces spurious counts; if the dead time of γ – channel is smaller than that of the β – channel, the component of spurious of γ – detector in the Bi4π count rates will increase with the increasing of the fraction of γ – counts, $1-\varepsilon_\beta$. Another possible reason inducing this phenomenon is that the non– equilibrium of dead times for two channels rises diference of count loss effects. In Fig.3 the different self– absorptions are, practically, the different values of $1-\varepsilon_\beta$, the activities obtained with extrapolation curves are listed in Table 2.

Table 2

The activities obtained with measurement under the different fractions of gamma counts ($1-\varepsilon_\beta$)

$1-\varepsilon_\beta$	0.076	0.082	0.15	0.31	0.50
A(Bq)	3383.5	3389.5	3389.3	3395.5	3398.0
A	1	1.0016	1.0016	1.0034	1.0042

Fig.4 shows the relation curve of the fractions of spurious counts to the contribution fractions of the gamma counts $1-\varepsilon_\beta$, uncer the given condition the probability of spurious counts is constant, and so it should be a straight line. One has get the slope of line in Fig.4 which value is 0.019, when $1-\varepsilon_\beta = 0.076$ then, the fraction of spurious counts is 0.0015. According to the possibleswing of slope of line in Fig.4 estimated uncertainty is of $0.3\times0.0015 = 0.0005$.

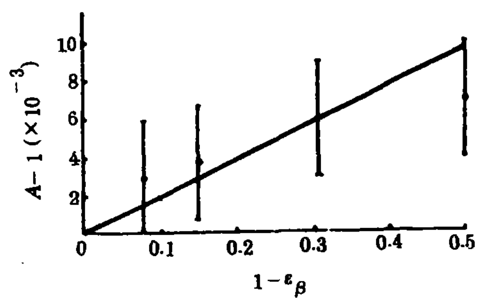

Fig.4 (A– 1) fraction of γ – spurious counts Vs ($1-\varepsilon_\beta$) contribution fraction of γ – counts

In the paper[9] it will talk about the 4π β counter spurious counts which contribute to the Bi4π measurement results with fraction of 0.003 ± 0.001.

The total fraction of spurious counts to the Bi4π counts is of 0.0045 with a combined uncertainty of 0.12%.

4. Results of activity

The first result of A = 3383.5 Bq is given in the Fig.2 for the activity measurements of No 8643 of ^{60}Co source, the measurement results of other 5 times are 3386.2, 3379.3, 3383.3, 3393.5 and 3377.4 Bq respectively. The arithmetic average value of six results is of 3383.9 Bq. The final activity value is of 3368.6 Bq corrected with 0.0045 of spurious count fraction given in the subparagraph 3. The uncertainty of category A for single measurement is of s = 0.17%. The uncertainty of catogory A for mean value is of

$(1/\sqrt{6})\,s = 0.07\%$.

The correction of dead– time losses is approximately of $N_{4\pi}\,\tau_{\beta} = 3.3\times10^{3}\times9.47\mu$ s $= 3\%$,the error of dead time τ_{β} is $5\%(1\sigma)$, the uncertainty of category B induced by the dead– time error is of $5\%\times3\%=0.15\%$. With summing uncertainty of category A of 0.07%, uncertainty of category B of 0.15% and the uncertainty 0.12% in correction of spurious counts mentioned in subparagraph 3 the uncertainty of mean result of activity is obtained with a value of 0.2% using calculating mode of "root sum square".

5. Comparison

The activity values of three ^{60}Co sources have been determined with use of activity of No. 8643 ^{60}Co source measured by Bi4π method and relative measurements using 4π γ counter. The difference of the measurement result from Bi4π method and those from 4π β – γ coincident method do not exceed the 0.4%. The measurement uncertainty of these two used methods both are of 0.2%. Considering that the decay corrections and relative measuring method are used, thus it may say that the results of comparisons are in agreement within the measurement errors.

V. SUMMARY

The early defined geometry methods make the efficiency– loss exceeds over 90%. After that it has developed a 4π β counter which efficiency– loss has reduced to 1– 7%. The 4π β – γ methods determine experimentally th efficiency– losses (practically the efficiency). The accuracies of these methods for determining efficiency– losses are restricted then it is required to improve source preparation procedures to reduce the efficiency– losses. Whereas the big 4π γ detectors to decrease the efficiency– losses by the detecting all the γ – rays and avoid the difficults to reduce the β – self absorptions. The Bi4π method remains the high efficiency of 4π β detector for β – rays and, at the same time, use the Bi4π γ detector to compensate this efficiency– loss of 1— 7% that in turn would be further reduced to 0.1— 0.7% (as the practical example of ^{60}Co in this work). It makes the accuracy could be greatly raised. The Bi4π method is already finely approximated to ideal mode that counting rate equals to activity and the ideal detector.

In the case of measurements with very high efficiency (very small efficiency– loss), each and every radiations have become the assistors to count the decay for his own nucleus; and there is not free radiations which will affect the counting.

The extrapolation method proposed in this article has been demonstrated by the measurements of ^{60}Co. The possibility of this extrapolation method is due to that the all radiation counter proposed in this work is comprised of two different 4π detectors which can be in operating individually with maintaining their efficiencies being not changed. Generally, the extrapolations with straight line are desired because of their extrapolation values are considered more reliable. It is shown, in practice, that the measurements of ^{60}Co have very fine linearity. The straight line correlation coefficients γ for the extrapolation lines in Fi.g3 are 0.996, 0.996, 0.993, 0.995 and 0.99996 respectively from top to bottom. It can be predicted that, in the cases of complex decays, the linearity of extrapolation lines will not exist, as the component fractions of various γ spectra in Bi4π counts are different from the originals that of 4π γ counts due to the efficiency– losses of various β branches in Bi4π counts are not the same, in consequence the fractions of count changes of various γ spectra in extrapolating Bi4π counts are not the same as in γ channel too. However, the influences of the non– linearity of extrapolation function on the results of measurements would not be serious due to the smaller range of extrapolations, usually less than 0.5% of activity with the high efficiency of the Bi4π counter.

The Bi4π method provides a possibility for many radionuclides to get the accuracy of 0.03%, and it is especially effective to the measurements for radionuclides of β (α or X)– γ decay which are about 30 or more among the generally important radionuclides of about one hundred. In the case of measurements for radionuclides with part branch of pure β (α , or X)– ray and/or together with internal– conversion electrons, the Bi4π method has to introduce some corrections for the efficiency– loss in that branch and still has generally the higher accuracy than other methods. The Bi4π method requires only 0.5 h and 4 h to count the Bi4π counts achieving the statistical errors of 0.2% and 0.02% respectively in the case of this work, this could be not compared by any coincident methods.

When the Bi4π method, as a new method, measure a individual radionuclide, it must into account takes and controls the conditions according to the characteristics of that nuclide, if one intends the measurement to achieve high accuracy level. In general, it will propose high requirements of determinations of dead times, radioactive purity of sources to be measured and the less spurious counts from detectors, but, we are convinced that this can be achieved.

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