DETERMINATION OF ABSOLUTE RATE OF β-DISINTEGRATION FROM A P^{32}-SOURCE USING THE 4π-COUNTER TECHNIQUE

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INTRODUCTION

The determination of the absolute rate of β-ray disintegration from radio-active substances is an important problem in nuclear physics. With the discovery of radio-isotopes and their wide application in tracer physics these determinations have acquired added importance. The methods usually employed for this purpose consist of measurement with calibrated end-window counters, the use of coincidence technique with β, γ tube counters, counting with β-ray proportional counters of known efficiency, etc. One serious difficulty in these methods, however, is that the result of each method depends largely on its geometry. Results of different measurements of the same or different methods are sometimes quite difficult to compare. If fairly strong γ-rays are emitted by a radio-isotope, the problem becomes still more complicated. The counts produced by these γ-rays or their internal conversion products cannot be easily distinguished from the real β-counts. In particular, the absolute measurement of β-activity from radionuclides using end-window counters is beset with certain inherent difficulties, e.g. large correction factors due to the emission of secondary electrons, the back-scattering, self-absorptions and absorption of β-rays by the counter window and the air gap. To avoid these difficulties a new technique was developed by Haxel and Houtermans (1948) in which the β-ray source was placed inside a G. M.-counter on a very thin support. As β-particles going practically in all the 4π-directions could be counted in this arrangement, the counter used was called the 4π-counter. The technique was later employed by Seliger and Cavallo (1951) for the absolute standardization of radio-isotopes. Houtermans, Meyer-Schützmeister and Vincent (1952) used it for the absolute calibration of both the high and low energy β-emitters. The use of 4π-counter eliminates entirely some of the difficulties mentioned above and reduces the rest to a minimum. It thus improves the accuracy of the result. The usual accuracy attained in the orthodox methods of β-ray counting probably lies between 5 and 10%. The method of 4π-counter using coincidence technique as described below seems to be capable of pushing this accuracy to about 2% or even less. We shall describe here the results obtained in a preliminary measurement undertaken in connection with the determination of certain reaction cross-sections using activation method and discuss the present limitations.

THE 4π-COUNTER AND ITS WORKING PRINCIPLE

Fig. 1 (*a*) shows the 4π-counter designed by us. A brass cylinder has been divided into two halves, Z_1 and Z_2, by means of a brass partition sheet S (1 mm. thick) lying in the middle of the cylinder along its axis. Two tungsten wires W are stretched parallel to the axis in the two halves which thus constitute two separate counters. The source of β-rays under investigation lies in the middle of the partition sheet S (Fig. 1(*b*)) on a very thin support. All the β-rays emitted from the source reach either the counter Z_1 or Z_2. If we neglect the absorption of β-rays in

the thin layer of the source and its mount, then any disintegration process in which at least one β-particle is emitted will always be recorded by the counter as a single

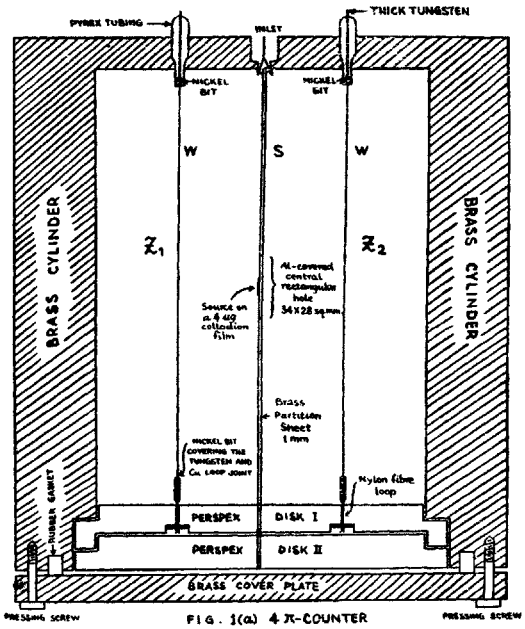

FIG. 1(a) 4 π-COUNTER

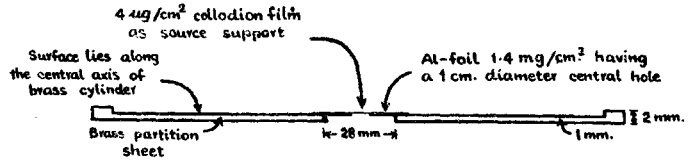

Fig. 1 (b). THE CENTRAL PARTITION

event. For such an event either Z_1 or Z_2 will respond. On the other hand, both Z_1 and Z_2 would discharge simultaneously on account of a scattered electron from the counter wall or the counter gas. Complexity of the decay scheme of the active substance also would give rise to such simultaneous discharge on account of an accompanying γ-quantum or the internal conversion electron. However, by noting the individual counting rates N_1 and N_2 of the two counters Z_1 and Z_2 and the coincidence rate N_{12}, the true β-disintegration rate N can be found:

$$N = N_1 + N_2 - N_{12} \quad .. \quad .. \quad .. \quad .. \quad (1)$$

This value of N should also be obtained directly if the counting rate $N_{||}$ is recorded with Z_1 and Z_2 connected in parallel, provided the interval between the β and γ-emissions involved is less than the resolving time of the counting systems. It is clear from this discussion that the absolute β-counting with 4π-counter is almost independent of the complexity of the disintegration scheme.

Measurements and Procedural Details

(a) *The counting arrangement:* The 4π-counter was filled with the usual argon-alcohol mixture and the characteristics studied. A 200-volt plateau with a

slope of ~ 15%/100 volt was obtained for each of the counters (Fig. 2). The counter was placed in a Pb-housing and the backgrounds, singles as well as coinci-

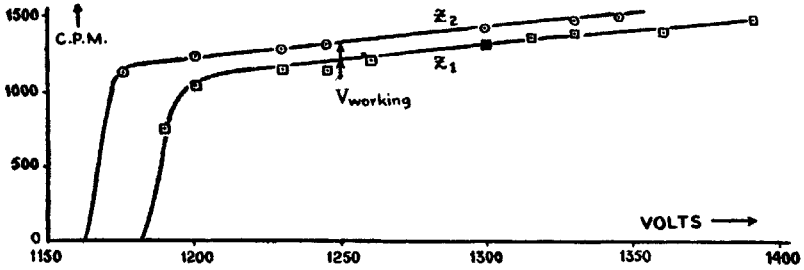

FIG. 2 — 4π - COUNTER CHARACTERISTICS

dence rates, were determined. For each of the two channels there was a cathode-follower type preamplifier and a two-stage main amplifier which were designed and

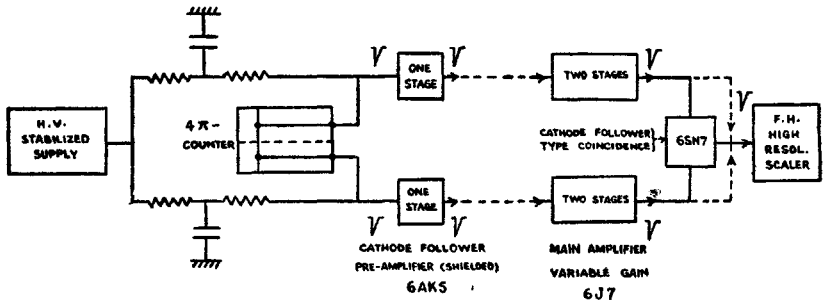

FIG. 3. Block diagram of the 4π-counter arrangement including the coincidence circuit used.

constructed to suit the present measurements. The amplified pulses in the two channels could either be directly counted or the coincidence pulses recorded.

A high resolution Frieske-Hoepfner decade scalar was employed for the final recording of the counts. Fig. 3 shows the block diagram of the electronic equipment used.

(*b*) *The preparation of the active-source:* To get the active P^{32}, 4-litres of CS_2 were exposed to Cd-filtered fast neutrons from a 100 mgm. (Ra α + Be) source under a spherical geometry. Fig. 4 shows the arrangement of bombarding CS_2. *F* is a 4-litre flask containing carbon di-sulfide. It has got a ground-joint through which a glass tube T_1 runs to the middle of the flask. The source *S* is surrounded by 1 mm. of cadmium and is put at the bottom of another glass tube T_2. *A* and *B* are the copper foils (2″ × 1″), separated by ~2·5 cm., between which an electric field (~100 V/cm.) is applied (Hevesey, 'Radio-active Indicators'). The exposure was continued for about a month. P^{32}-ions (both positive and negative) produced migrate to the copper electrodes and get deposited there; the total yield of active P^{32} in this method of separation is expected to be about 95%.

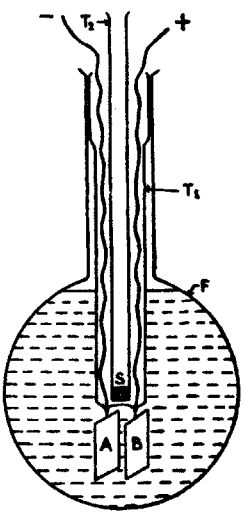

FIG. 4. Fast neutron irradiation of CS_2 under electric field.

To separate the active deposit of P^{32} from the copper electrodes, the latter were completely dissolved in conc. nitric acid, a suitable quantity of sodium ammonium

hydrogen phosphate added as a carrier and the copper precipitated out as CuS. Magnesia mixture was then added to the clear filtrate and the active P^{32} precipitated as magnesium ammonium phosphate.

(*c*) *Mounting the source in the* 4π*-counter:* To mount the source at the centre of the partition of the 4π-counter the following procedure was employed: The brass partition had a central rectangular hole 3·4 cm. × 2·8 cm. A very thin Al-foil (1·4 mgm./cm.^{2}) was pasted to cover this hole on that surface of the partition which lies in the middle (axial plane) of the 4π-counter. The Al-foil carried a central circular hole 1 cm. in diameter. This hole was covered with a very thin film of collodion. Collodion, 10% by weight, was dissolved in amyl acetate and a solution formed. A drop of this solution was put over a clean water surface by a micropipette. A nearly circular film was formed after a little time as the amyl acetate evaporated. The film was then mounted on to the platform surface having the Al-foil and was dried under radiation. The unwanted portion of the film was then removed by cleaning with amyl acetate. The thickness of the film thus formed was estimated to be $\sim 4\,\mu$ gm./cm.^{2}. To mount the active source on to the film at the central hole, we proceeded as follows. A weighed quantity of the active phosphate salt was dissolved in a known volume of dilute HCl and then by means of a micropipette a drop of the solution was put on the film with utmost care. At the same time the volume occupied by that drop was known from the micropipette graduations, and hence the mass of the active deposit estimated. The drop was then dried by placing the partition in a thermostat working at $\sim$ 50°C. After cooling, the mass of the active salt was also found with an accurate chemical balance. The two observations agreed quite well and gave the mass of the active deposit as $\sim$ 1·1 mgm.

(*d*) *The counting procedure:* Next the 4π-counter was cleaned with absolute alcohol and the partition with the source at its centre was introduced into it. The counter was then filled with the usual argon-alcohol mixture and the characteristics taken. The 4π-counter was laid flat along the axis, the half containing the source deposit being kept at the top. In the following we shall refer to the source half as the top half (Z_1) and the other as the bottom half (Z_2). The counter was shielded by means of a suitable lead-housing. Measurements of the single counting rates N_1 and N_2 from the two halves, the coincidence rate N_{12} and the parallel rate $N_{\parallel}$ (with the two halves connected in parallel) were then started using the electronic circuits already mentioned (Fig. 3). Each set of measurements involved 5 minutes' observations of the N_1, N_2 and $N_{\parallel}$ and 30 minutes' observation of the coincidence rate N_{12}. Four sets of such measurements were taken, out of which one set had to be discarded as unreliable due to some readjustment of electronic parameters done. For each of the sets a corresponding observation of the counting characteristics of the two halves was taken in order to be able to apply corrections due to the slope of the characteristics as well as for any possible change in the Geiger threshold, the working voltage being kept fixed for all sets of measurements at $\sim$ 1240 volts. Corrections were then applied for the finite slope of the plateau and all the rates were converted to those corresponding to the Geiger threshold of each of the two halves. The resolving time of the coincidence circuit was determined to be 50 ± 1 μsec. and the corrections due to chance coincidences applied to the coincidence rate N_{12}.

Table I shows the different counting rates observed after these have been corrected for the following: (i) Background rates N_1^b, N_2^b, N_{12}^b and $N_{\parallel}^b$ observed without the presence of any source inside the 4π-counter. (ii) Finite slope of the counting characteristics of the individual halves (as checked before and after each set of measurement) and conversion of rates to Geiger threshold, V_g, since the two halves were found to have slightly different Geiger thresholds, but were worked at the same voltage. (iii) Correction to N_{12} due to chance coincidences.

TABLE I

(Ra α+ Be) source removed on 21.v.55 at 1445 hrs.
Weight of the active salt [$Mg\ NH_3\ (PO_4)_2$]+ carrier = $0{\cdot}0011 \pm 0{\cdot}0001$ gm.

Set	Date and time	N_1 cpm.	N_2 cpm.	N_{12} cpm.	$N_\parallel$ cpm.	$(N_1+N_2-N_{12})$ cpm.	Mean of last two columns 'N' cpm.	Absolute activity after correcting for decay, N_0 cpm.
I	1.vi.55 1740 hrs.	1170 ± 40	1290 ± 40	102 ± 5	2300 ± 70	2358 ± 85	2330 ± 55	3990 ± 95
II	2.vi.55 1315 hrs.	1060 ± 45	1260 ± 40	99 ± 5	2180 ± 65	2221 ± 90	2200 ± 55	3920 ± 100
III	3.vi.55 1145 hrs.	1030 ± 45	1160 ± 35	100 ± 5	2070 ± 70	2090 ± 85	2080 ± 55	3880 ± 105

Corrections due to the absorption and back-scattering of β-rays at the collodion film-mount were neglected since the film was very thin ($\sim 4\,\mu$ gm./cm.^{2}); the self-absorption of β-rays in the thin source deposit was also negligible. It will be observed from the table that the counting rates in the parallel position, $N_\parallel$, and that corresponding to $N_1+N_2-N_{12}$ agree within the limits of experimental error, as expected. The mean counting rates, N, in column 8 are then corrected for the P^{32}-source decay ($T = 14{\cdot}3$ days) and the initial rates of disintegration thus obtained are shown in the last column.

The mean value of 'N_0' comes out to be 3930 ± 60 cpm. for $1{\cdot}1 \pm 0{\cdot}1$ mgm. of the active salt of P^{32} (with carrier). The absolute β-activity of the P^{32}-source (with carrier) comes out to be $(3{\cdot}57 \pm 0{\cdot}39) \times 10^6$ *disintegrations/minute/gm.* This corresponds to the specific activity of the source of $\approx 1{\cdot}6 \pm 0{\cdot}2\,\mu$ *curie.*

The probable error of β-ray counting rate here is only about 1·5% which is mainly due to the slope and change of operating characteristics of the two halves of the 4π-counter. This is a substantial improvement over the accuracy of the end-window counter measurements. However, a large probable error ($\pm 12\%$) in the value of the specific activity of the source occurs. This is mainly due to the inaccuracy in the present mass measurement. The accuracy of the mass measurement can be very much improved in our future measurements. It is hoped that in the final result an accuracy better than 2% may then be attained. In the present set of measurements the superiority of the 4π-counter in the absolute β-disintegration rate measurements over other conventional methods is at least clearly demonstrated.

DISCUSSION OF RESULTS

The accuracy of the 4π-counter measurements deserves careful examination, as the absolute rate of β-disintegration determined here depends entirely on that. As enumerated in the Introduction, the 4π-counter method has got several advantages over the end-window counter method, as only very few simple corrections are needed in the former. However, it should be pointed out that in the present measurements some discrepancy still exists regarding the counting characteristics of the two halves of the 4π-counter. As Fig. 2 shows, these characteristics are relatively displaced from each other slightly. This may be due to some geometrical non-alignment in the construction of the two halves. It, however, makes the individual rates of the halves somewhat uncertain.

Another uncertainty that exists in our case is the occurrence of a higher counting rate N_2 for the lower half as compared to N_1, the counting rate of the source half

Z_1. Normally N_1 is expected to be somewhat higher than N_2 on account of a possible small absorption in the supporting film. The observed behaviour of the two halves is, however, in the reversed order. This may arise due to two causes: (i) greater sensitive volume of the half Z_2 compared to that of Z_1, (ii) improper crystallization of the active salt (with the carrier) causing the presence of a non-active sedimentation-layer over the insignificant amount of the actual active salt layer. According to Houtermans, Meyer-Schützmeister and Vincent's (1952) absorption measurements, the situation existing in our case is equivalent to having a 1·2 mg./cm.^{2} thick Al-absorber in front of the source in the upper half Z_1. This hypothetical layer would scatter back β-rays into the lower half Z_2 and would also absorb a part of the radiation going into the half Z_1. The result would be an increase in the rate N_2 and a simultaneous decrease in N_1. The presence of this effect, however, appears to be rather unlikely, because the addition of carrier generally ensures a uniform distribution of the active matter throughout the solution layers before and during their subsequent evaporation. There is no reason to think that the method of depositing the active salt on the supporting film in any way disturbs the uniformity of the distribution. The effect of unequal sensitive volumes of Z_1 and Z_2 is likely to be the predominant cause. The error due to this can be largely eliminated by taking the mean of the counting rates N_1 and N_2, as has been done here.

Another factor is the loss of counting efficiency caused by the use of a non-conducting film as a source mount. Since the two positively charge collecting wires are not completely shielded from one another, the region covered by the source mount will be of weak electric field. Low energy β-particles, which are completely absorbed within this weak field region, may not be detected. The correction factor needed due to this cause can be estimated (Mann and Seliger, 1953), but comes out to be quite small and well within the present experimental errors.

Self-absorption of β-rays in the source layers is the limiting factor in the use of 4π-counting technique. It can be minimized but never completely eliminated. All other factors being equal, the amount of self-absorption depends on the particular nuclide and the energy of the β-rays emitted by it, being high for the low energy β-emitters. In our case the amount of carrier has been kept to the minimum consistent with the proper chemical behaviour of the solution. This naturally reduces the self-absorption also to the minimum.

Another possible source of error in measuring P^{32}-activity is the presence of a 25-day P^{33}-activity as reported by Jenson and Nichols (1951) and confirmed by Sheline, Holtzman and Fan (1951). They, however, estimated the P^{33}-activity initially present as $\sim$1% of the total activity with pile-produced P^{32}. In our case the amount of P^{33} developed, if any, will be negligible.

The present preliminary measurements, when perfected are likely to be of importance, since the method of 4π-counting has got versatile utility, e.g. absolute standardization of radio-active (β-ray) sources, determination of efficiency of the G. M.-counters, absolute cross-section of some nuclear reactions involving β-disintegration rate measurements, etc.

In conclusion, the authors wish to express their sincere gratitude to Prof. D. S. Kothari, Head of the Physics Department, and Prof. R. C. Majumdar, University of Delhi, for encouragement and facilities given for carrying out this work. Their thanks are also due to Mr. L. K. Rangan for helping them in the electronic problems and to Dr. K. R. Kar, Chemistry Department, University of Delhi, in guiding them through the chemical separation of the P^{32}-source. This work forms part of a research scheme under the Department of Atomic Energy, Government of India, and has been supported by their financial help for which the authors are thankful,

Abstract

A preliminary report is presented for absolute measurement of β-ray disintegration rate by using a 4π-type of counter and coincidence method. The principle involved in the counter working and the simplification introduced in the elimination of the usual sources of error of β-ray counting and the present limitations of the method are discussed. A radiophosphorous P^{32}-source prepared in the laboratory shows a source strength of $1{\cdot}6 \pm 0{\cdot}2$ μ curie per gramme and a mean disintegration rate of 3930 ± 60 cpm. per 1·1 mgm. of the active salt as measured by this method.

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