B I P M
F-92310 SEVRES

Rapport BIPM-80/5

Report on some experiments carried out with the 4π β-γ coincidence counting equipment of BIPM

by X.Z. Wu, C. Veyradier, C. Colas, P. Bréonce and A. Rytz

A. Measurement of the γ efficiency of two 4π proportional counters

1. Introductory remarks and theoretical background

The sensitivity to γ rays of so-called pillbox proportional counters has been measured in the past by several authors [1 to 5]. The partial disagreement of their results reflects the differences which may exist between individual counters and the difficulties of such measurements.

In what follows we present results obtained with two 4π proportional counters of similar shape but different dimensions. Both counters were built so as to allow three different anode lengths and three different diameters of the inserts coaxial with the anode. However, the limited time did not permit to vary these parameters.

The method applied was the one described in [2] in which several suitable β-γ emitting nuclides, i.e. with simple decay schemes and sufficiently low β end-point energy with respect to the energy of the associated photons, were used.

The activity N_o of a source is calculated according to the equation

$$\frac{N_\beta N_\gamma}{N_c} = N_o \left[1 + \frac{1-\varepsilon_\beta}{\varepsilon_\beta} \left(\varepsilon_{\beta\gamma} - \frac{\varepsilon_c}{\varepsilon_\gamma} \right) \right] ,$$

where N_β, N_γ and N_c are the observed count rates in the β, γ and coincidence channels, respectively, corrected for dead time, background and accidental coincidences.

ε_β is the efficiency of the proportional counter to β rays,

ε_γ " " γ counter to γ rays,

$\varepsilon_{\beta\gamma}$ " " proportional counter to γ rays,

ε_c is the probability of obtaining a coincidence without detection of a β particle.

The apparent disintegration rate $N_\beta N_\gamma / N_c$ was calculated by means of equation 4 in [6].

If the γ channel is set so as to accept only the (highest) photoelectric peak, we have, since $\varepsilon_c/\varepsilon_\gamma \ll \varepsilon_{\beta\gamma}$,

$$\frac{N_\beta N_\gamma}{N_c} = N_o \left(1 + \frac{1-\varepsilon_\beta}{\varepsilon_\beta}\,\varepsilon_{\beta\gamma}\right).$$

The parameter $\varepsilon_\beta \approx N_c/N_\gamma$ can be varied, for instance by covering the source with absorber foils. If a sufficient range for ε_β can be obtained without altering $\varepsilon_{\beta\gamma}$, the quantity $N_\beta N_\gamma/N_c$ varies linearly versus $(1-\varepsilon_\beta)/\varepsilon_\beta$ with slope $N_o\,\varepsilon_{\beta\gamma}$ and intercept N_o.

The number of radionuclides which can be used for the measurement of $\varepsilon_{\beta\gamma}$ is limited by considerations of decay scheme and availability. We have chosen the nuclides indicated in Table 1 which were purchased from LMRI (Saclay).

Table 1 – Radionuclides used for measuring $\varepsilon_{\beta\gamma}$

Nuclide	Half-life (d)	Principal γ energy (keV)	Results shown in Figs. no.	Number of sources measured
^{7}Be	53	478	-	4
^{54}Mn	312	835	3,4	20
^{60}Co	1 925	1 332	1,2	28
^{88}Y	107	1 836	5,6	20
^{95}Nb	35	766	7,8	25

2. Experimental procedures

The two proportional flow counters were made of aluminium and had anode lengths of 35 and 120 mm, respectively. Methane or argon/methane (9 : 1) was used as counting gas.

The sources were prepared following the technique in current use at BIPM which includes weighing of the drops of solution dispensed. Thus accurate activity ratios were known for the sources of each radionuclide.

The β efficiency of each counter was reduced in steps by covering the sources with gold-coated VYNS films or thin aluminium foils. Efficiency functions could thus be determined either with a single source used with absorbers of gradually increasing thickness or with a set of sources which were covered with absorbers of different thicknesses. The latter method allowed the two counters to be compared more closely, by means of the same set of sources.

The results of these measurements are presented in Figs. 1 to 8. In the case of ^{7}Be $\varepsilon_{\beta\gamma}$ could be obtained directly by observing the count rate divided by the γ-emission rate which was derived from the activity concentration as indicated in the certificate of LMRI. The sources were sandwiched between gold-coated VYNS films which prevented the very soft Auger electrons ($\approx$ 45 eV) from being counted.

3. Discussion of the results

The values for $\varepsilon_{\beta\gamma}$ obtained with the two counters are summarized in Table 2 and Fig. 9. The very high values for ^{95}Nb may be ascribed to a deterioration of the source mounts due to the high activity of this solution. They are not shown in Fig. 9 and should probably be discarded.

Table 2 - Results of the measurements of $\varepsilon_{\beta\gamma}$, the γ efficiency of the β detector (values are given in %)

Nuclide	This work small counter	This work large counter	Williams + Campion [2]	Merritt and Taylor [3]
^{7}Be	0.12 ± 0.01	0.15 ± 0.01	0.25 ± 0.02	0.15 ± 0.01
^{54}Mn	0.37 ± 0.03	0.47 ± 0.03	*0.41 ± 0.04	0.21 ± 0.03
^{60}Co	0.44 ± 0.01	0.65 ± 0.04	0.44 ± 0.02	0.40 ± 0.09
^{88}Y	0.51 ± 0.05	0.55 ± 0.05	0.51 ± 0.03	0.38 ± 0.02
^{95}Nb	0.60 ± 0.04	0.68 ± 0.03	0.34 ± 0.03	0.16 ± 0.10

* impurity suspected

The results of a further experiment, shown in Fig. 10, were obtained by measuring four sources in the large counter and by using either one or the other half-counter, or both as usual (two sources only). The close agreement with the corresponding value in Table 2 suggests that this method may be employed for a rapid check of $\varepsilon_{\beta\gamma}$ when the sources available are thin enough.

From Table 2 it is seen that the large counter has always a higher $\varepsilon_{\beta\gamma}$ value than the small one. This difference which is significant except for ^{88}Y may be due to the increased sensitive volume of the large counter. It will be noted that most of the values of $\varepsilon_{\beta\gamma}$ for the small counter are in fair agreement with those determined by Williams and Campion [2].

B. Activity measurements of ^{60}Co sources at high count rates

1. Measurements carried out with zero mean delay between correlated β and γ pulses

In 1975/76 a high-count rate experiment using ^{60}Co sources was organised by NPL as an international comparison on behalf of BIPM [7]. The results obtained showed clearly that accurate measurements call for improved coincidence formulae and removal of (or appropriate correction for) a mean delay between correlated β and γ pulses, as soon as higher count rates are involved.

We describe here an experiment in which a set of thirteen ^{60}Co sources was measured under conditions granting the best attainable delay matching. Source activities ranged from 6 to 180 kBq. Their ratios were accurately known from the drop masses dispensed.

The time distribution of the γ pulses with respect to the β pulses was recorded by means of the circuit shown in Fig. 11. Typical distributions obtained with sources of ^{60}Co and of ^{54}Mn are presented in Figs. 12 and 13, respectively.

Let Y_r be the number of counts registered in the r^{th} channel of the multichannel analyzer. The channel number $\bar{x}$ of the centroid of the distribution between channel numbers $r = n_1$ and n_2 is given by

$$\bar{x} = \left(\sum_{n_1}^{n_2} r\, Y_r\right) \Big/ \left(\sum_{n_1}^{n_2} Y_r\right) .$$

The conversion of channel numbers to time delay was found by replacing the β channel by a pulse generator set at 1 kHz. The width of the rectangular distribution thus obtained was adjusted to cover 100 channels. Therefore, one channel corresponded to a delay of 10 ns. After determining the position $\bar{x}$ of the centroid, the switch shown in Fig. 11 was set to position 2 permitting to start the time-to-amplitude converter (TAC) by a β pulse and to stop it by the same pulse, after a delay δ . This delay was changed until all the pulses registered fell into the channel $\bar{x}$. This adjustment could be done with a precision of much less than one channel width. It differed according to the nuclide and its activity. Moreover, it had to be checked and corrected from time to time.

The procedure described above does not take into account distortions of the distribution by accidental coincidences. The adjustable delay δ was therefore corrected by adding $\Delta = 2(1-\mathcal{E}_\gamma) N_o x_2^2$, where x_2 may be approximated, according to [7], by one quarter of the width of the distribution.

The results of the measurements carried out under conditions of optimal delay matching are presented in Fig. 14. It can be seen that, up to 110 kBq, there was excellent agreement between the measured and the expected values. For higher count rates the measured activities were high by about 0.4 % which is still very satisfactory, considering the fact that the dead-time corrections amounted to more than 50 %.

2. Effects of delay mismatch

Measurements were taken with a weak (7.6 kBq) and a strong (78 kBq) source of ^{60}Co. For each source the delay δ was varied in steps until the calculated activity started to deviate strongly from the value at $\delta = 0$ (see Fig. 15). In each case the activity N_o was calculated using three different formulae.

a) Campion's formula [1]

$$N_o = \frac{N_\beta N_\gamma \left[1 - \tau_r (N'_\beta + N'_\gamma)\right]}{\left[N_c - 2\tau_r N'_\beta N'_\gamma\right](1-\tau N'_c)} ,$$

where

N'_β, N'_γ, N'_c are the observed count rates in the respective channels,

N_β, N_γ, N_o are the corresponding rates corrected for background,

τ_β, τ_γ are the dead times of the respective channels,

τ is the shorter of τ_β or τ_γ,

τ_r is the coincidence resolving time.

b) Smith's formula [8] for $\tau_\beta = \tau_\gamma = \tau$

$$N_o = \frac{N'_\beta N'_\gamma \left[N_\beta e^{N_\beta \tau} e^{(N_\gamma - N_\beta) h'_\gamma} - N_\gamma e^{N_\gamma \tau} e^{(N_\beta - N_\gamma) h'_\beta} + \frac{N''_c}{p_\beta\, p_\gamma} (e^{N_\beta \tau} - e^{N_\gamma \tau})\right]}{N''_c (N_\beta e^{N_\beta \tau} - N_\gamma e^{N_\gamma \tau})}$$

where

$h'_\beta = h_\beta + \delta$, $\quad h'_\gamma = h_\gamma - \delta$,

h_β and h_γ being the resolving times in the respective channels,

δ is the mean delay,

$N''_c = N'_c - (h'_\beta + h'_\gamma) N'_\beta N'_\gamma - B_c$,

$p_\beta = 1 - \tau N'_\beta$; $p_\gamma = 1 - \tau N'_\gamma$,

N_β , N_γ , N_c are count rates corrected for background and dead time,

B_β , B_γ , B_c are the background rates.

c) For $\delta = 0$ the Smith formula becomes identical with the formula of Cox and Isham [9].

For a count rate of about 8 000 s^{-1} the resulting activity value was found to be practically independent of delay when using Smith's formula and nearly so with Campion's formula. It should be added that Smith's formula is valid only for $|\delta| < h$.

However, with sources ten times stronger, the behavior is quite different (see Fig. 15). As the formulae by Campion and by Cox and Isham both assume a zero delay, there is a poor agreement with their predictions. On the other hand, our results indicate that Smith's formula correctly takes into account the effect of delay mismatch. The small residual variation may be due to time jitter.

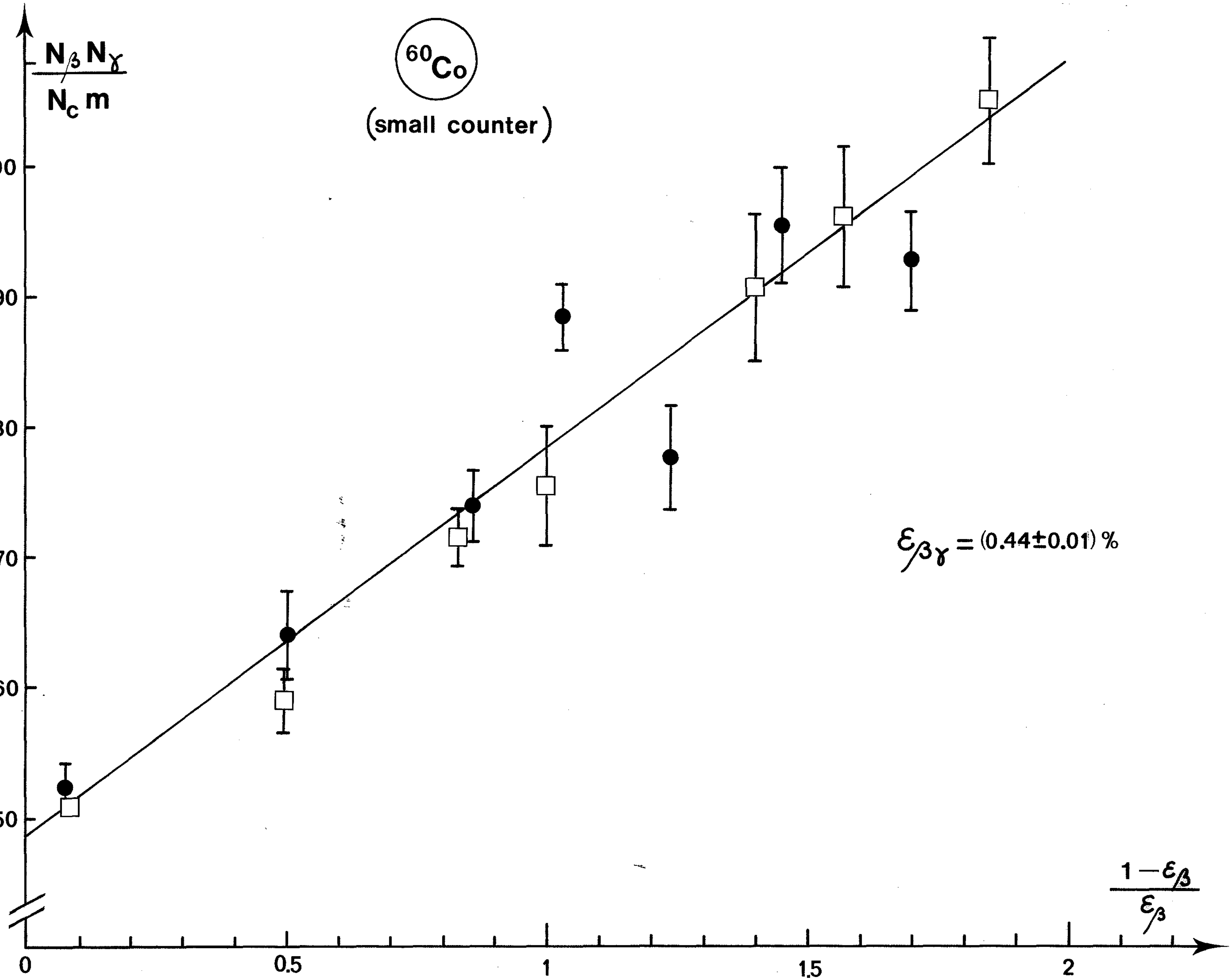

Figure 1 – γ sensitivity of the small counter for ^{60}Co; $\varepsilon_{\beta\gamma}$ = slope/intercept of the least-squares adjusted straight line, CH_4.

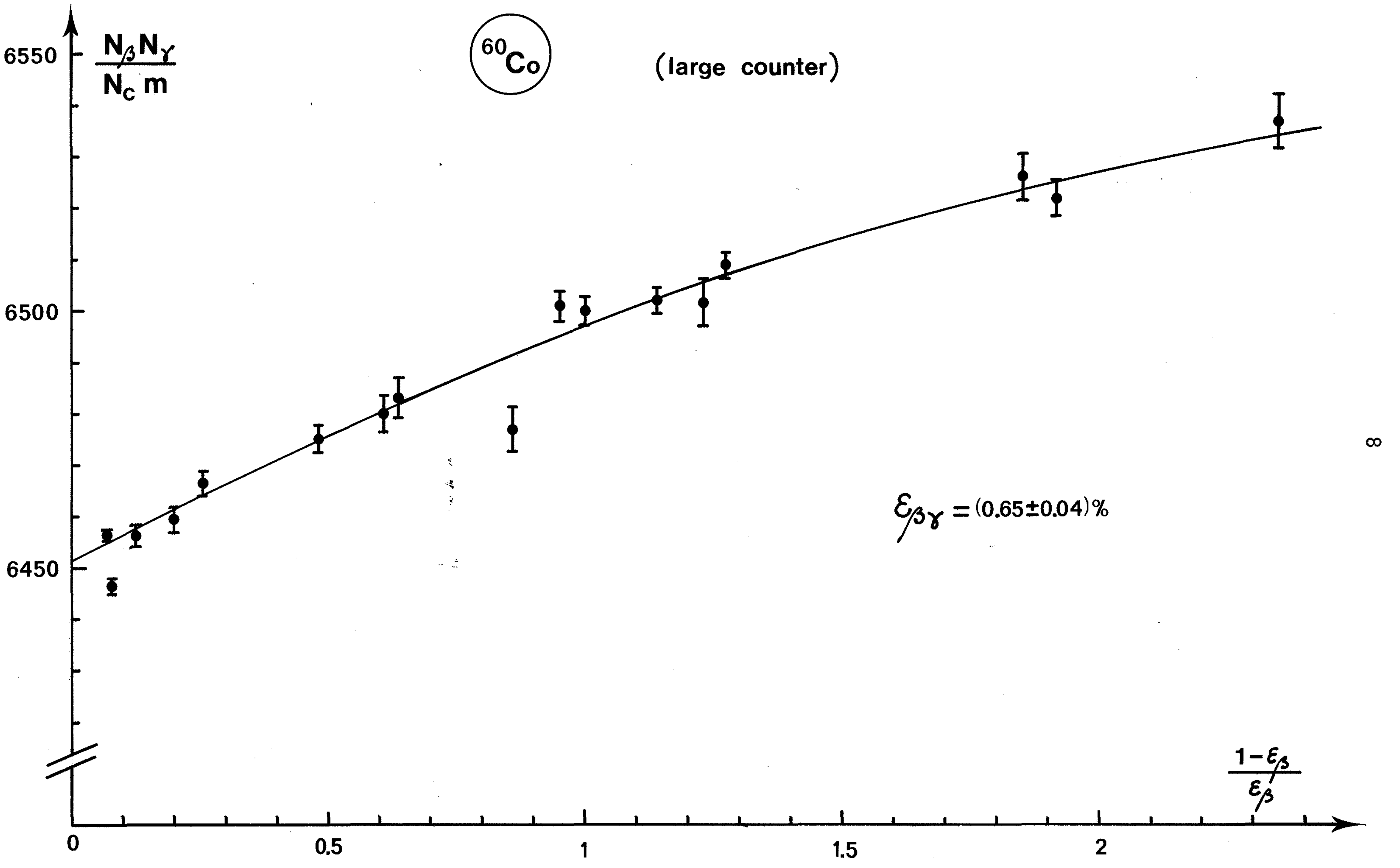

Figure 2 - $\varepsilon_{\beta\gamma}$ of the large counter for ^{60}Co, quadratic adjustment, CH_4.

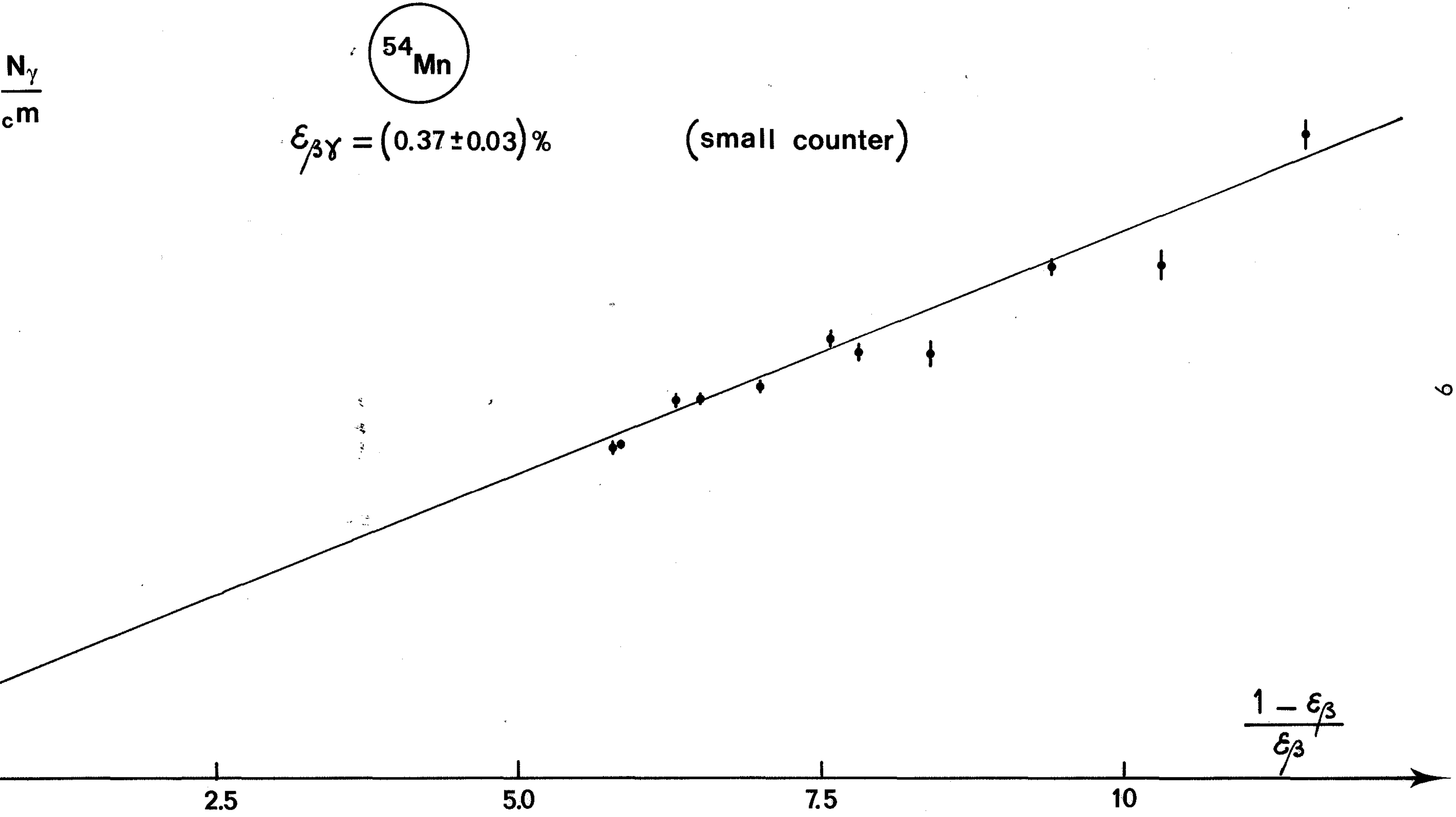

Figure 3 - $\varepsilon_{\beta\gamma}$ of the small counter for ^{54}Mn, linear adjustment, Ar/CH_4 (9:1).

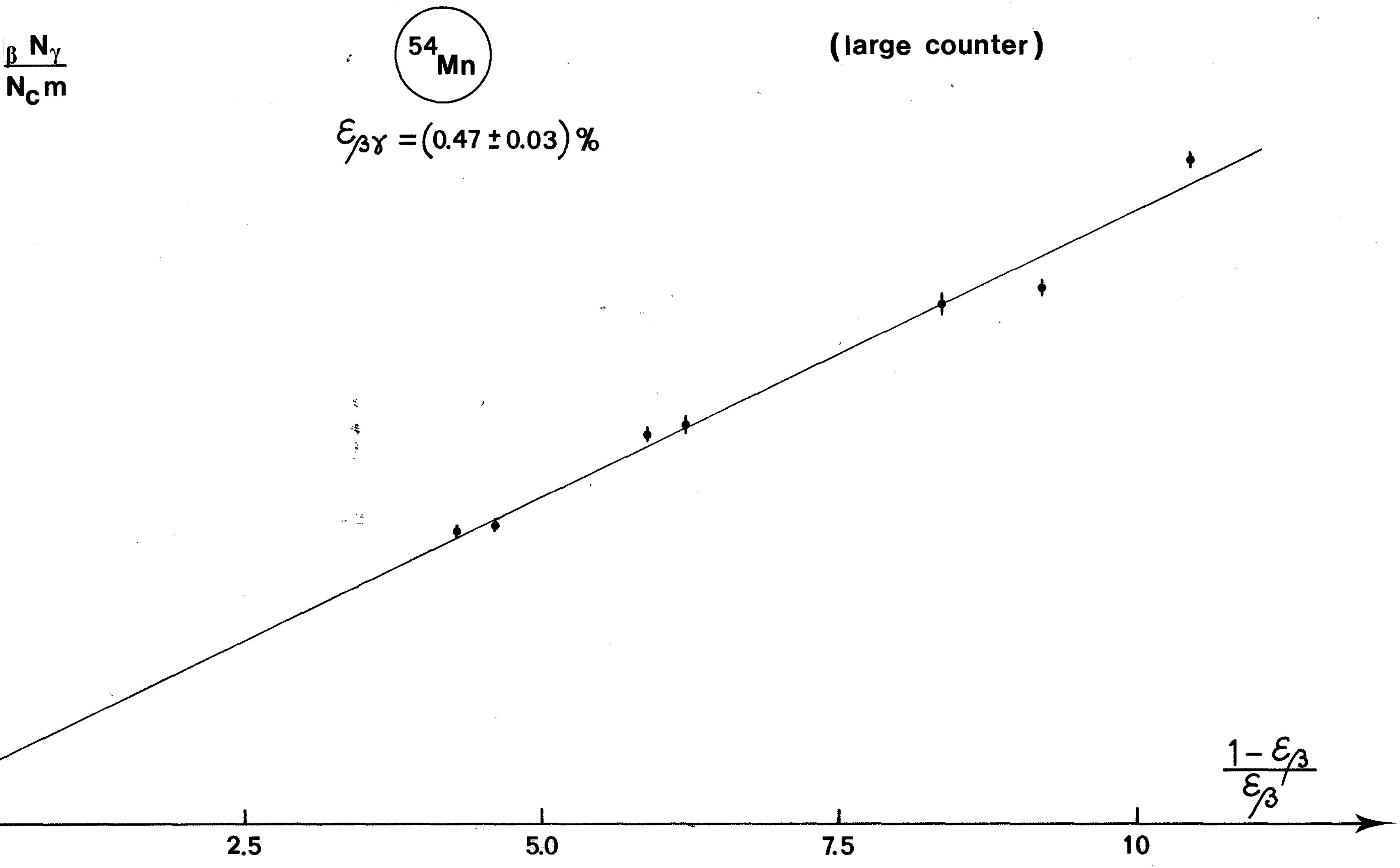

Figure 4 - $\mathcal{E}_{\beta\gamma}$ of the large counter for ^{54}Mn, linear adjustment, Ar/CH_4.

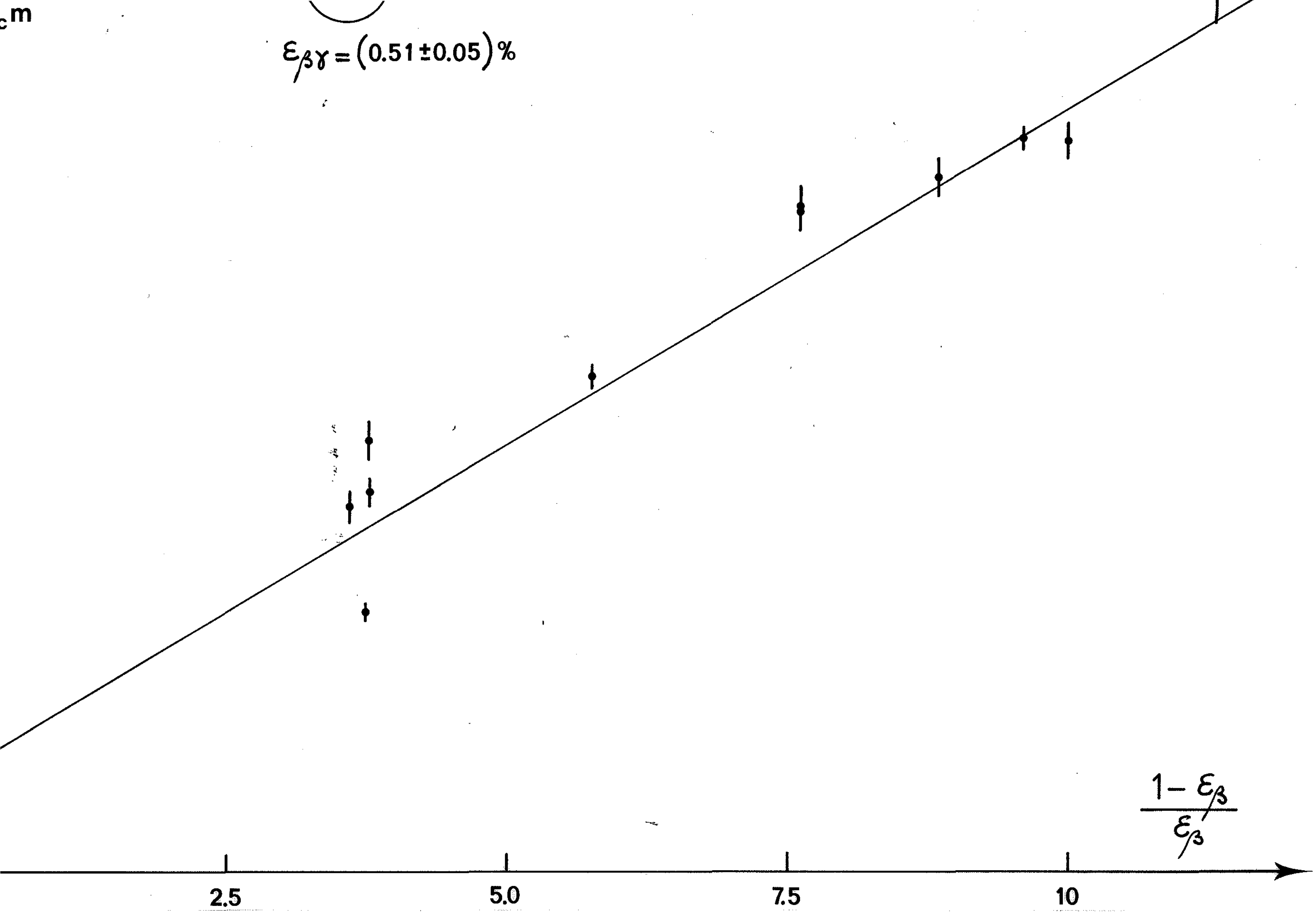

Figure 5 – $\varepsilon_{\beta\gamma}$ of the small counter for ^{88}Y, linear adjustment, Ar/CH_4.

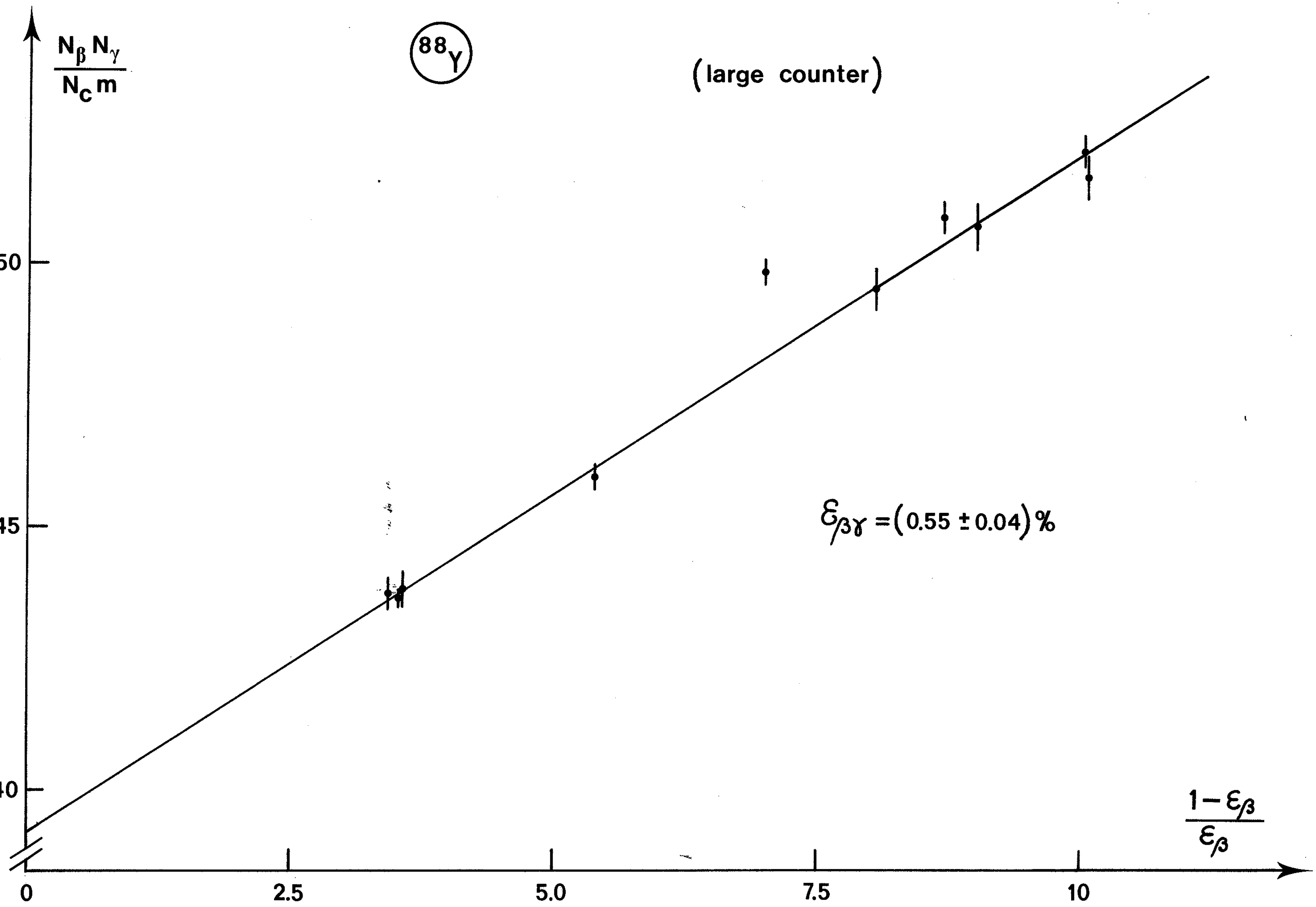

Figure 6 - $\varepsilon_{\beta\gamma}$ of the large counter for ^{88}Y, linear adjustment, Ar/CH_4.

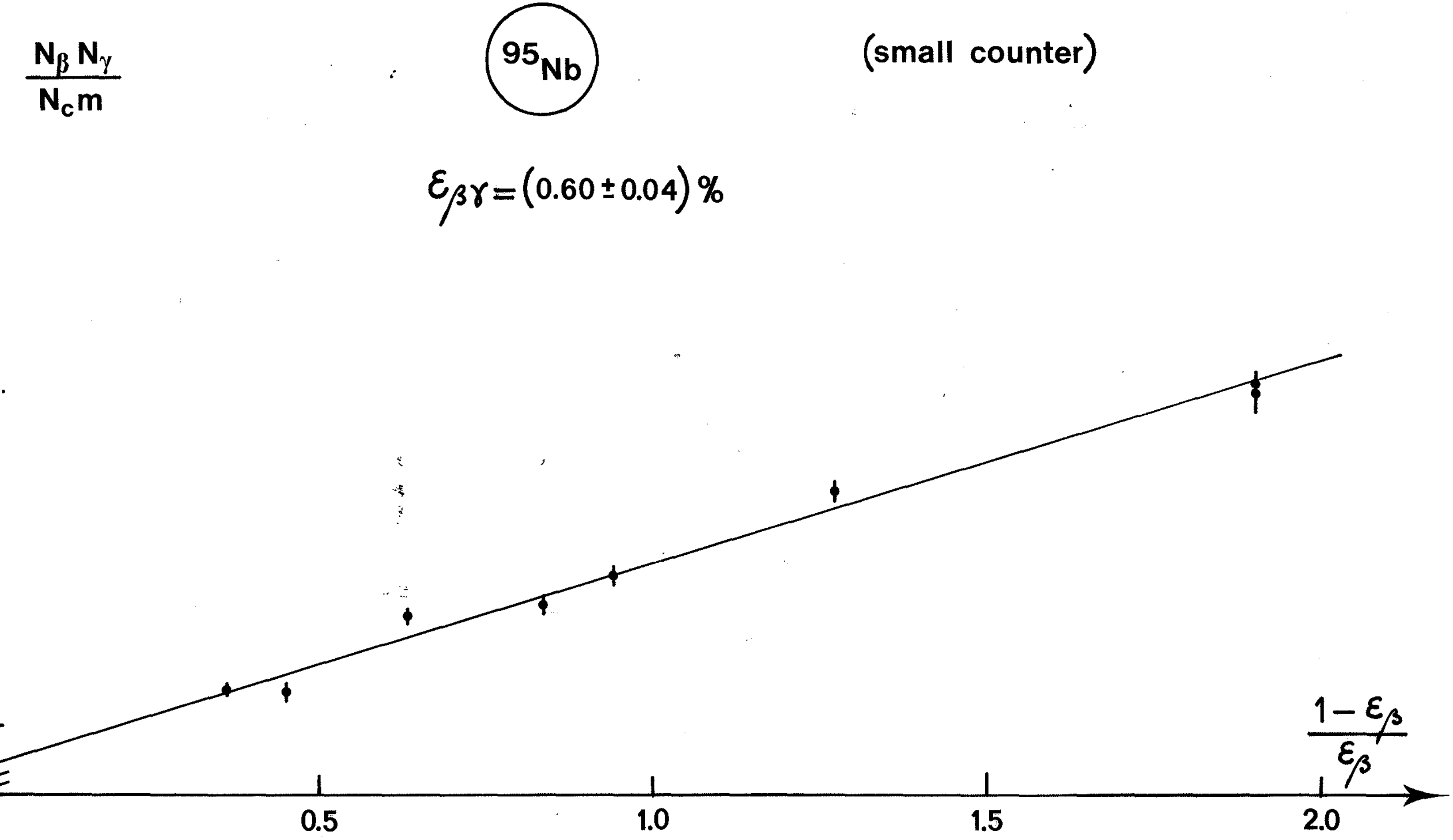

Figure 7 - $\varepsilon_{\beta\gamma}$ of the small counter for ^{95}Nb, linear adjustment, CH_4.

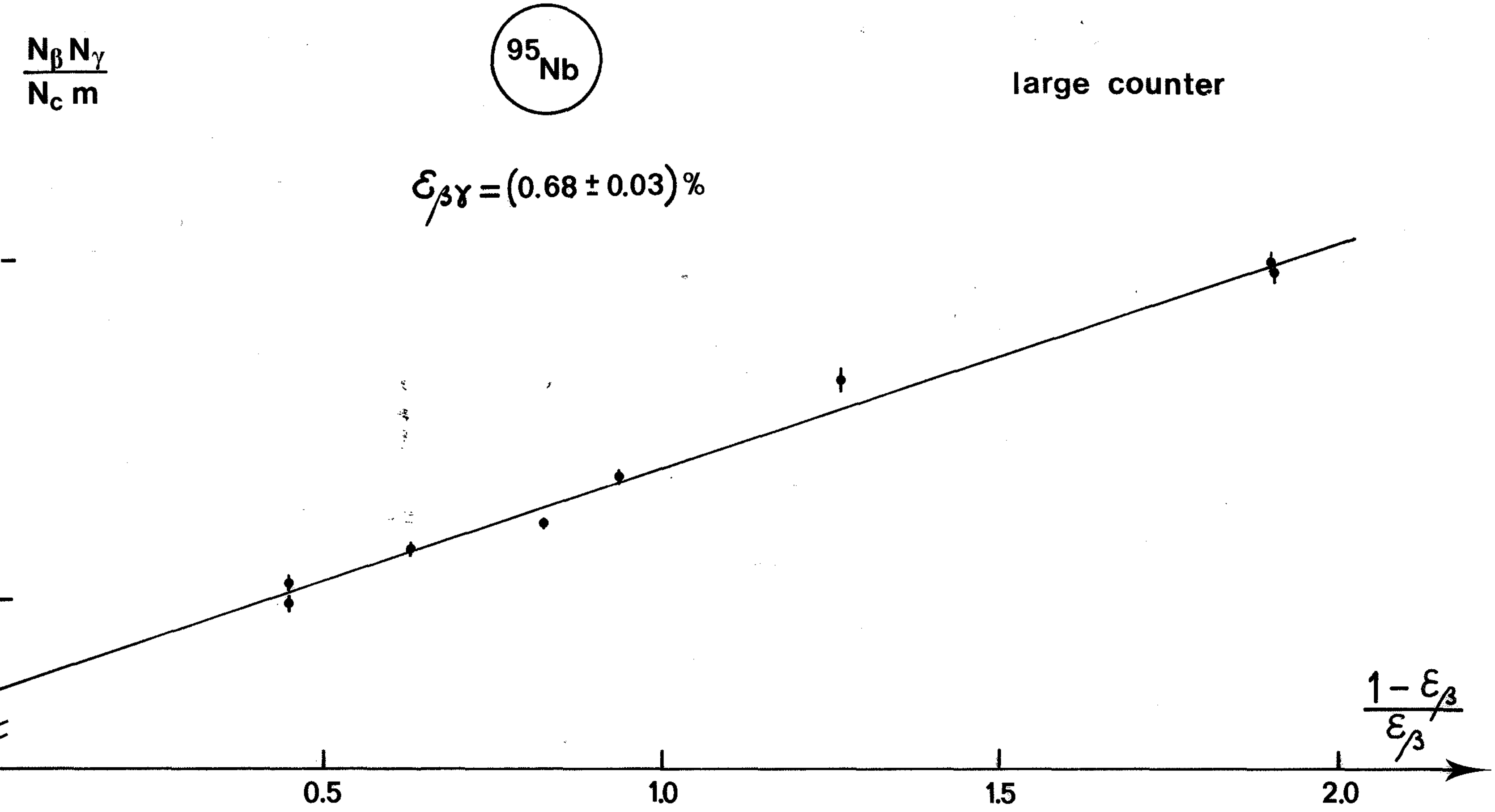

Figure 8 - $\varepsilon_{\beta\gamma}$ of the large counter for ^{95}Nb, linear adjustment, CH_4.

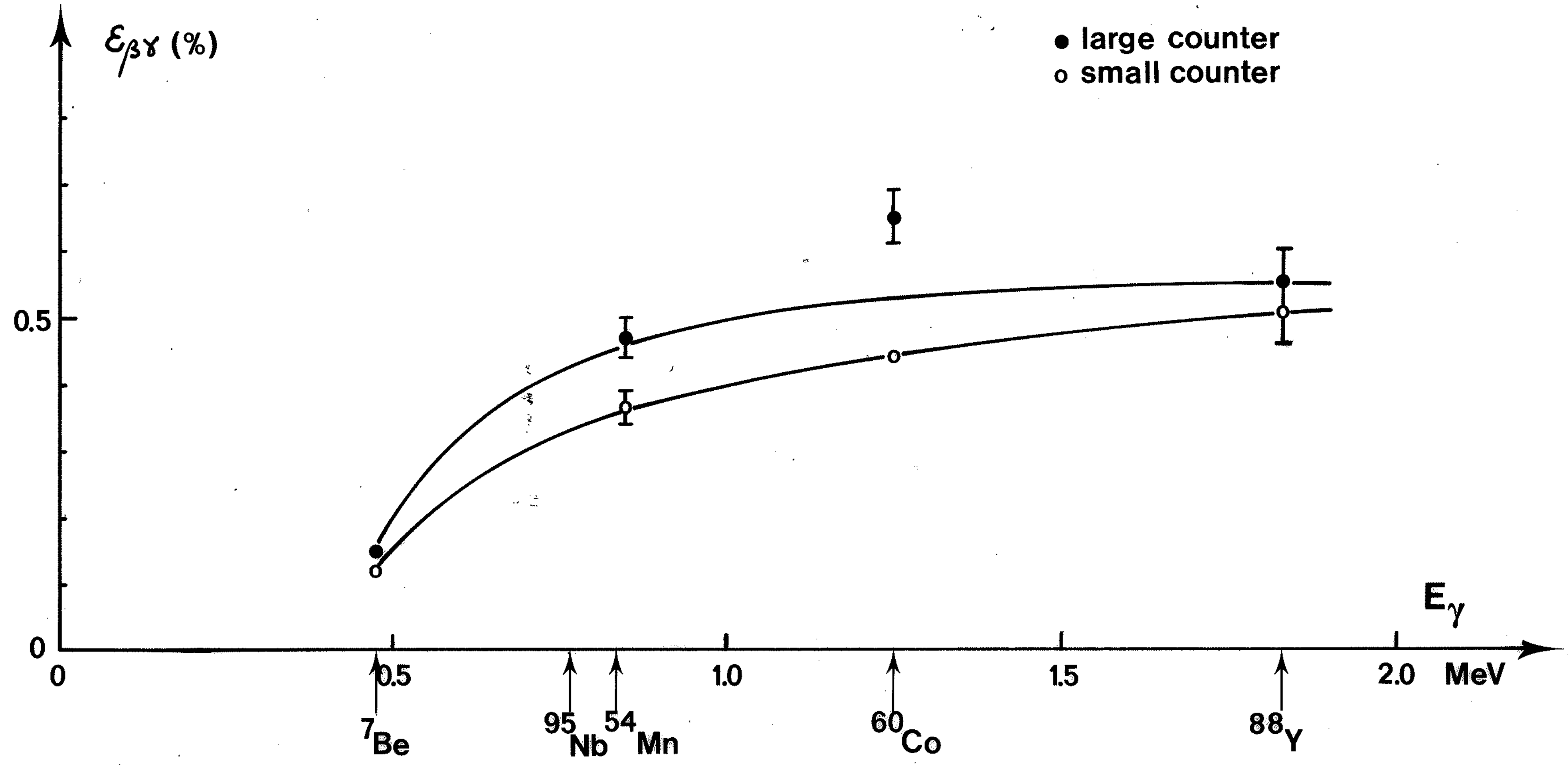

Figure 9 – Graphical representation of the results for $\varepsilon_{\beta\gamma}$ obtained with the two counters, using five different nuclides.

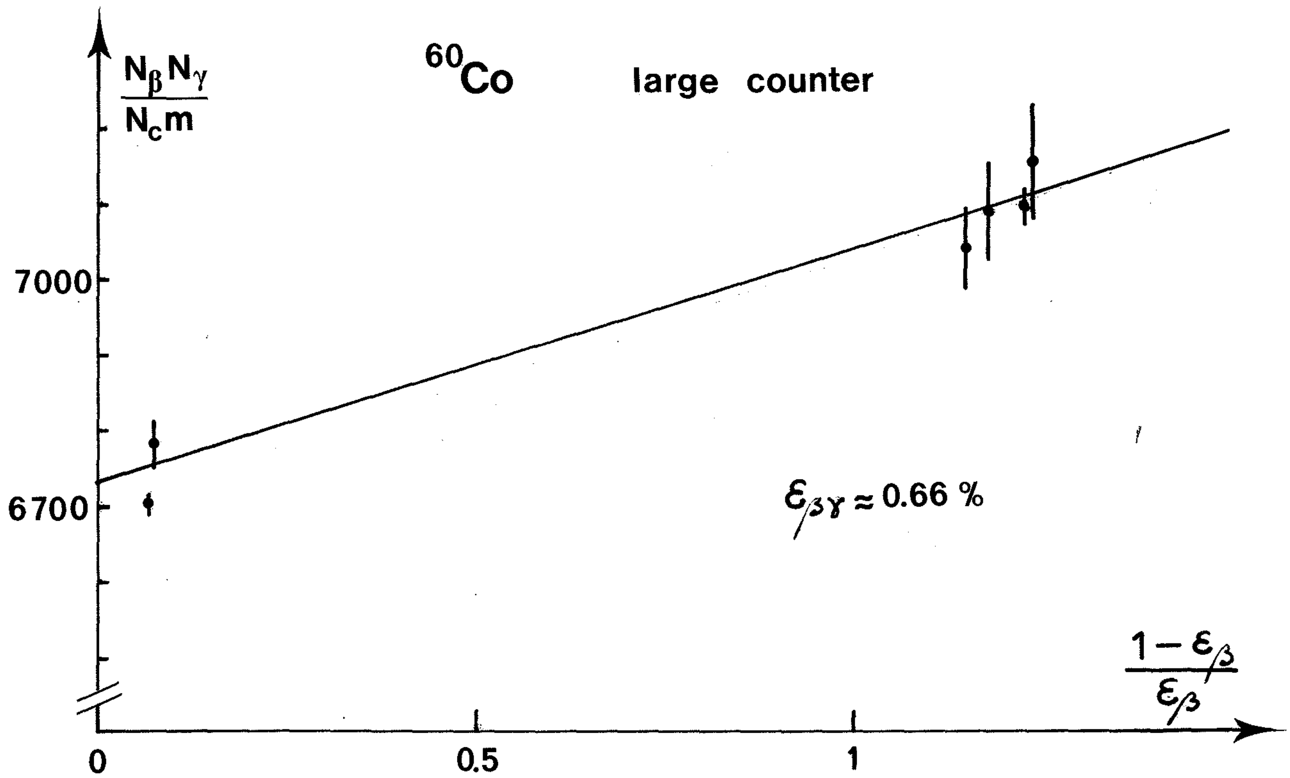

Figure 10 - Measurement of $\varepsilon_{\beta\gamma}$ of the large counter (4 sources of ^{60}Co); ε_β was varied by using either one half-counter or both halves in parallel.

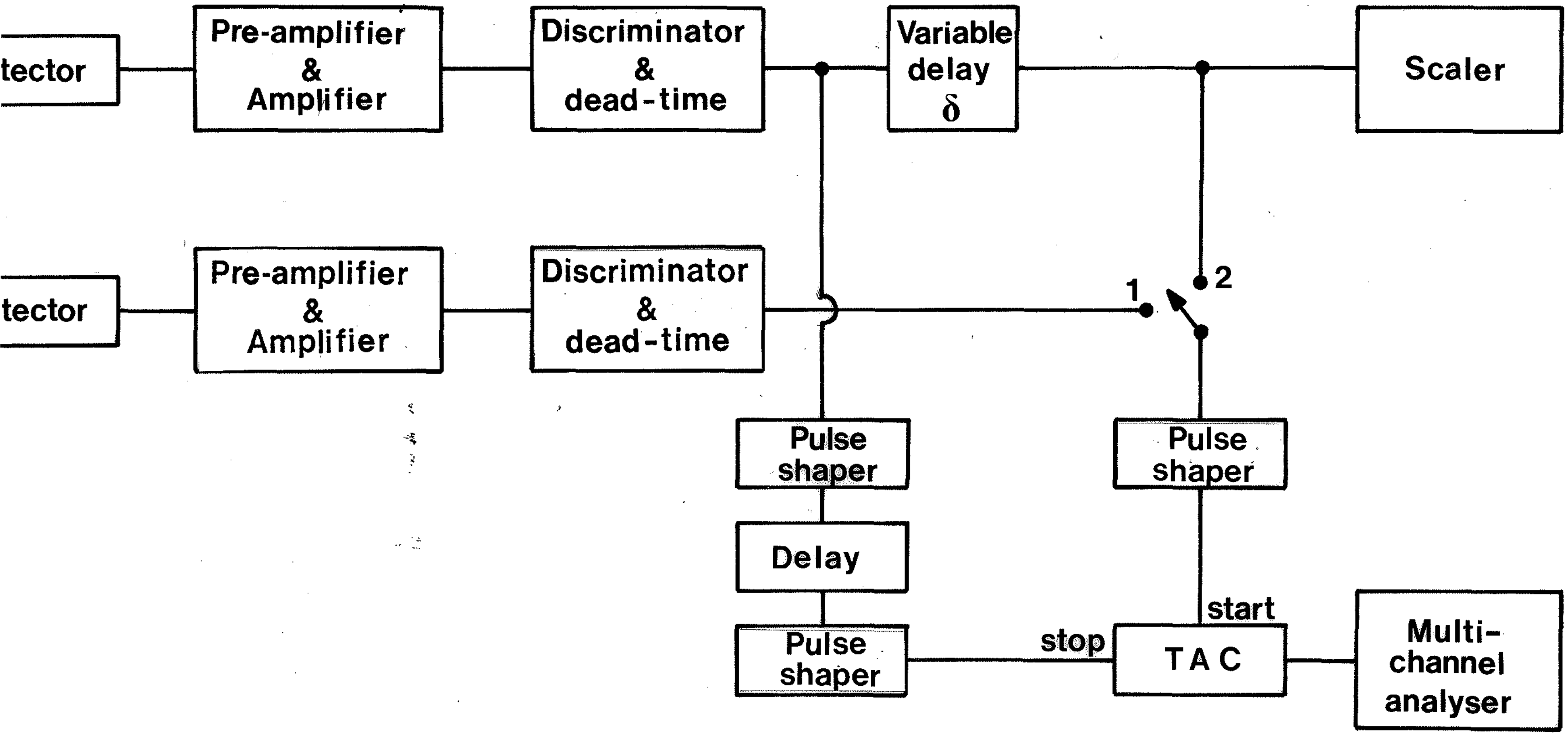

Figure 11 - Block diagram of the circuit for determining the time distribution of the γ pulses with respect to the correlated β pulses. TAC = time-to-amplitude converter.

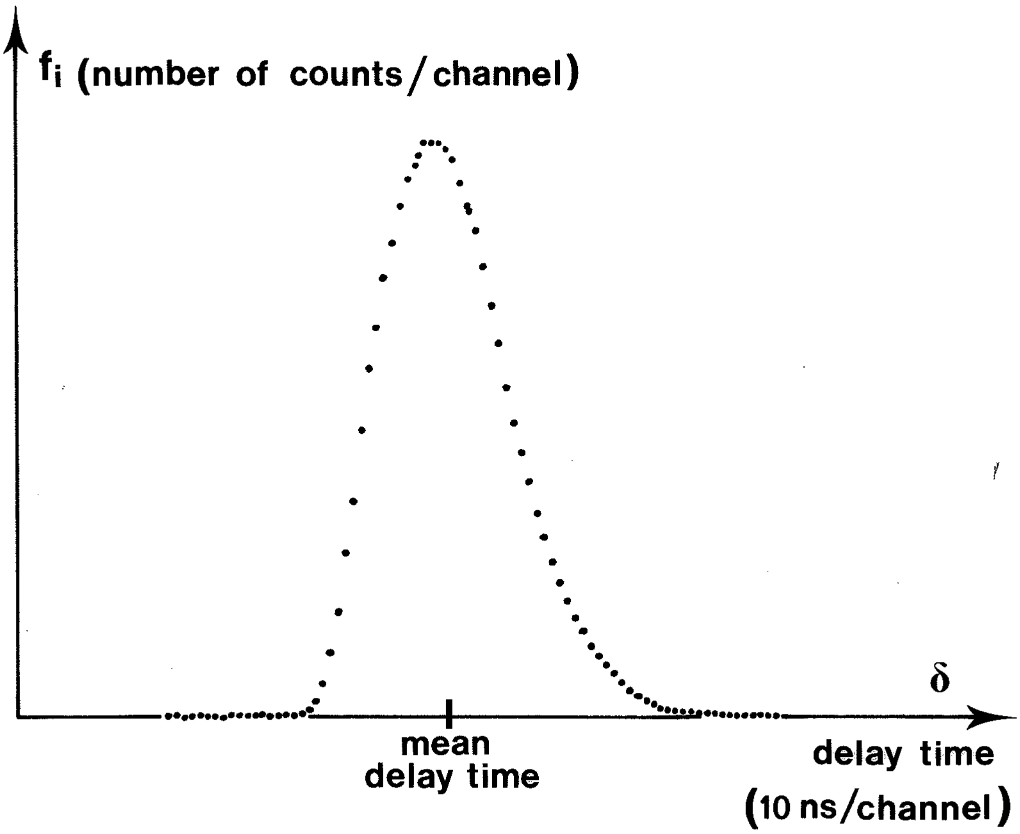

Figure 12 - Time distribution of the γ pulses with respect to the correlated β pulses for a ^{60}Co source.

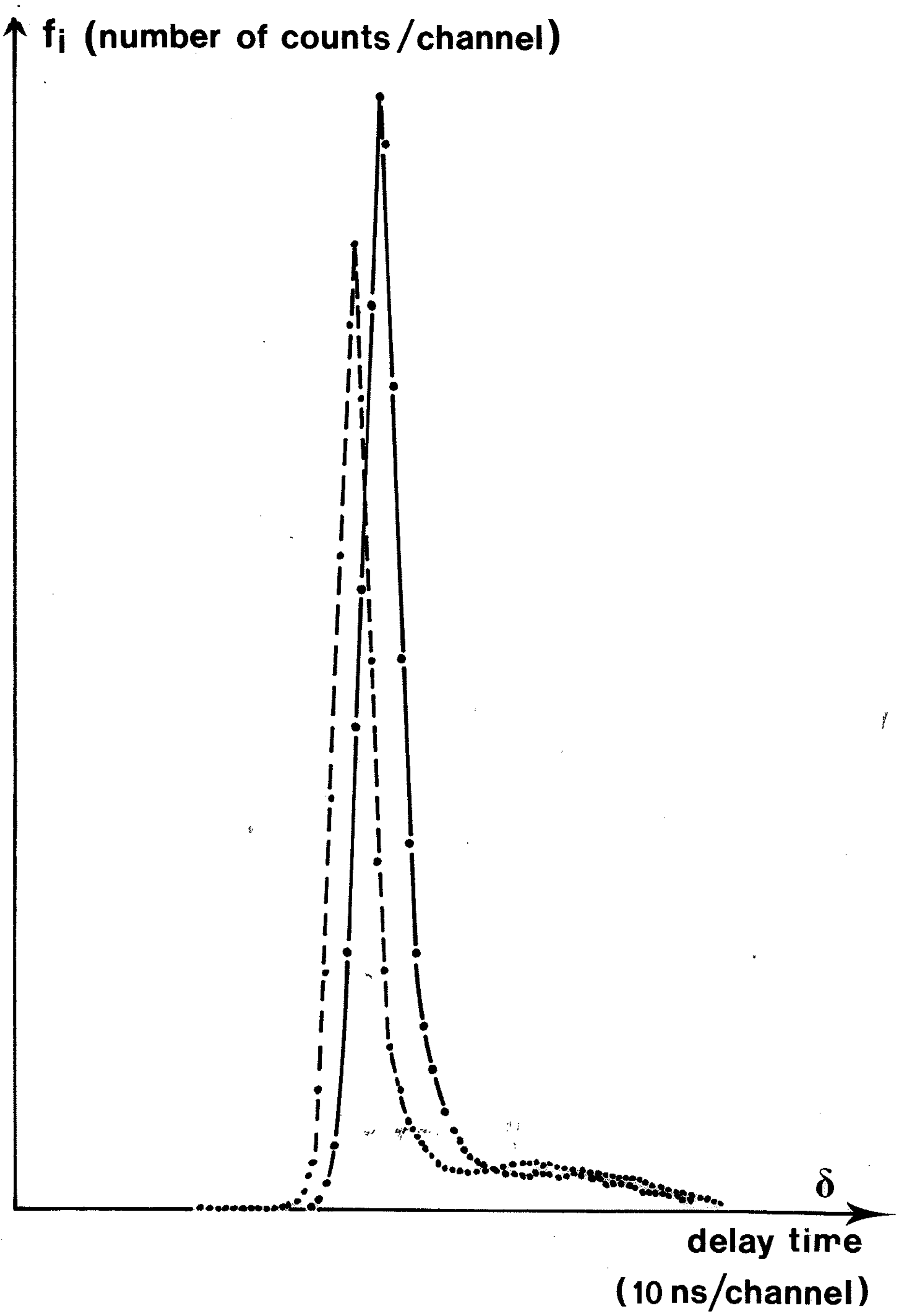

Figure 13 - Time distribution of the γ pulses with respect to the correlated β pulses for a ^{54}Mn source. The curve on the left was obtained with an anode voltage of 3 800 V, that on the right with 3 700 V, in CH_4.

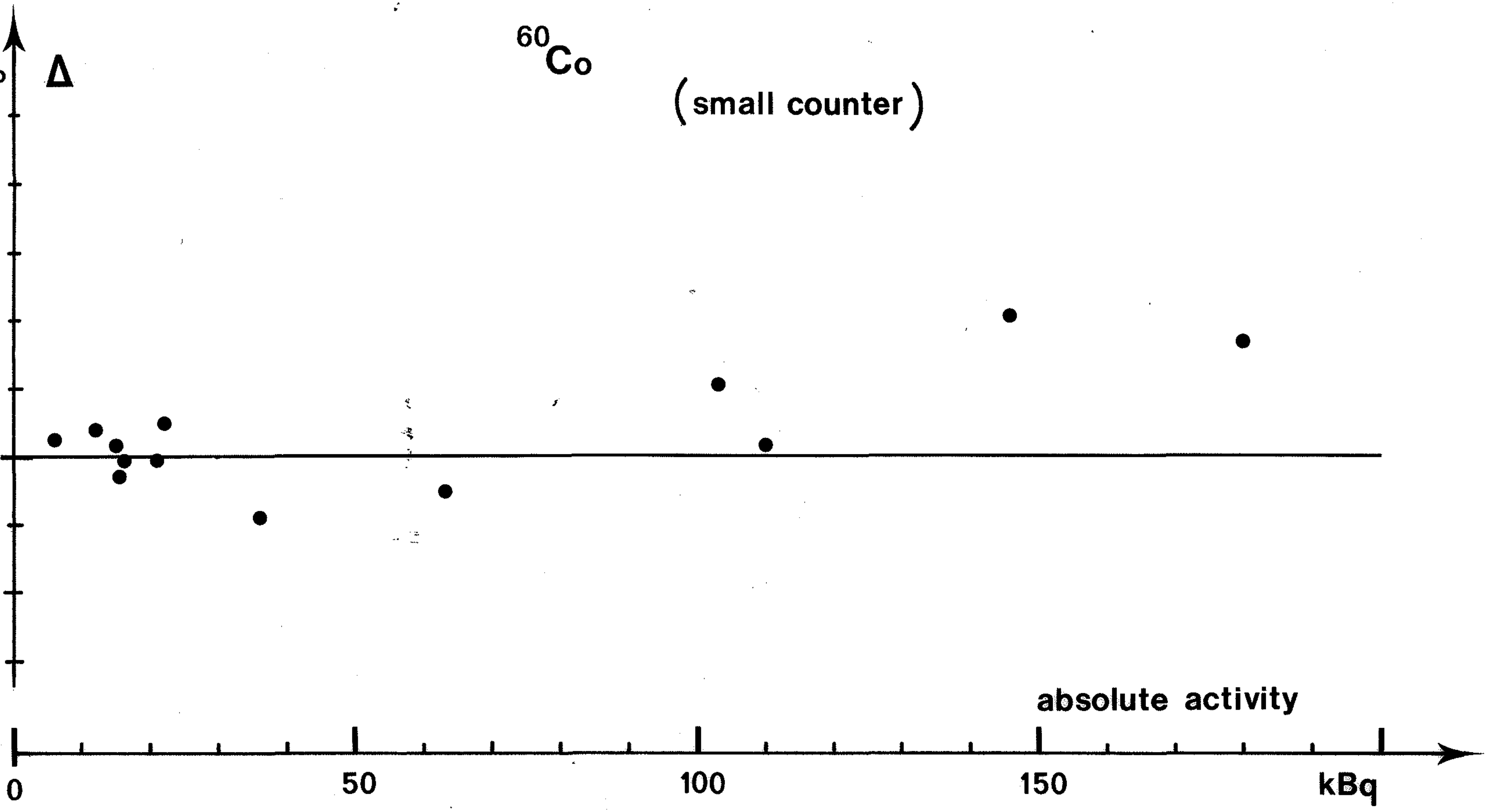

Figure 14 - Results of a high-count-rate experiment with ^{60}Co sources.
Δ = relative difference of measured and expected values.

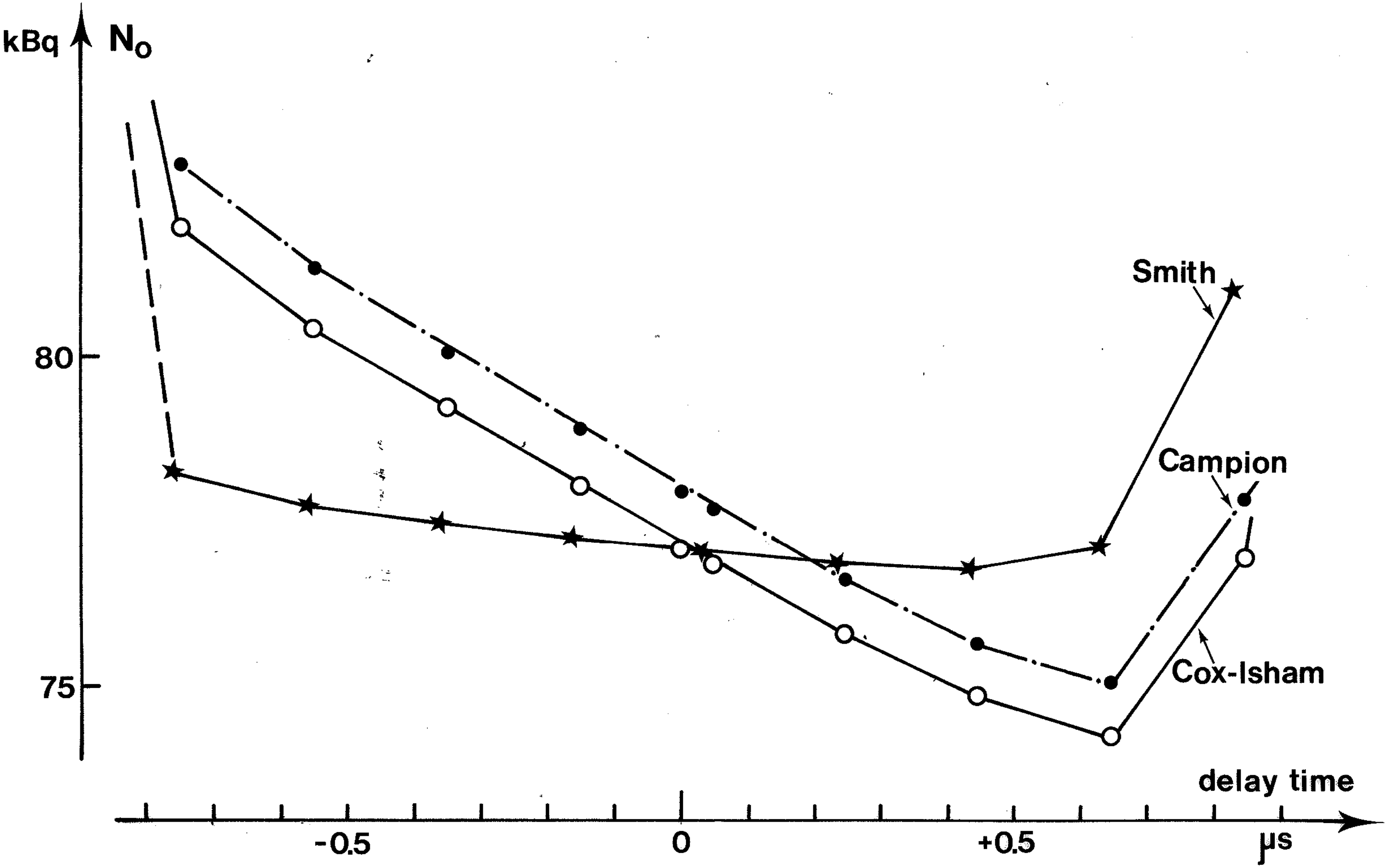

Figure 15 – Results of activity calculations from measurements of a strong ^{60}Co source, using different values of the delay δ .

References

[1] P.J. Campion, The standardization of radioisotopes by the beta-gamma coincidence method using high efficiency detectors, IJARI 4, 232-248 (1959)

[2] A. Williams and P.J. Campion, The measurement of the gamma sensitivity of a 4π β counter, IJARI 14, 533-540 (1963)

[3] J.S. Merritt and J.G.V. Taylor, The response of 4π proportional counters to γ-rays, Standardization of radionuclides, IAEA, Vienna, SM-79/66, 147-152 (1967)

[4] D.F. Urquhart, A new method for the measurement of the gamma counting efficiency of 4π beta gas counters, Standardization of radionuclides, IAEA, Vienna, SM-79/27, 167-181 (1967)

[5] Y. Kawada, Extended applications and improvement of the 4π β-γ coincidence method in the standardization of radionuclides, Researches of the Electrotechnical Laboratory, no. 730, 100 p. (1972)

[6] D. Smith, Improved correction formulae for coincidence counting, Nucl. Instr. and Meth. 152, 505-519 (1978)

[7] D. Smith, A. Williams and M.J. Woods, Intercomparison of high-count-rate ^{60}Co sources, Rapport BIPM-77/7, 65 p. (1977). A French translation "Comparaison internationale de sources de ^{60}Co à taux de comptage élevé" appeared in CCEMRI, Section II, 4e réunion-1977, Annexe R(II)3, BIPM (1977)

[8] D. Smith, Improved correction formulae for coincidence counting - continued, Draft NPL, Teddington (1979), 16 p.; see also: E. Funck, Modification of the Cox-Isham formula for 4πβ-γ-coincidence counting by the "Gandy"-effect and its automatic correction by electronic circuits, personal communication, PTB, Braunschweig (1978), 16 p.

[9] D.R. Cox and V. Isham, A bivariate point process with electronic counters, Proc. R. Soc. Lond. A 356, 149-160 (1977).