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ACCURATE DISINTEGRATION-RATE MEASUREMENT OF ^{55}Fe BY LIQUID SCINTILLATION COUNTING

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Council for Scientific and Industrial Research

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SINOPSIS

'n Metode vir die akkurate meting van disintegrasietempo van ^{55}Fe deur middel van vloeistofsintillasietelling, word beskryf. Die metode is gebaseer op die gebruik van berekende doeltreffendheidsfunksies tesame met een van die kernsoorte ^{54}Mn of ^{51}Cr as interne standaard. Laasgenoemde maak dit moontlik om teldoeltreffendhede in gemengde bronne te meet met behulp van koïnsidensietelling. Die metode is onlangs deur die NVS gebruik tydens 'n internasionale vergelyking van radioaktiwiteitsmetings; die resultate van nege deelnemende laboratoriums toon 'n spreiding van etlike persent.

SYNOPSIS

A method involving liquid scintillation counting is described for the accurate measurement of disintegration rate of ^{55}Fe. The method is based on the use of calculated efficiency functions together with either of the nuclides ^{54}Mn and ^{51}Cr as internal standards for measurement of counting efficiencies by coincidence counting. The method was used by the NAC during a recent international intercomparison of radioactivity measurements, and a summary of the results obtained by nine participating laboratories is presented. A spread in results of several percent is evident

INTRODUCTION

Establishing a fundamental or direct method of radioactivity standardisation for radionuclides which decay by electron capture to ground state is inherently difficult. In the case of ^{55}Fe, determination of the activity of a source from counting-rate measurements alone without use of any other standards or calibrated equipment is certainly a challenge because of the relatively low energy of the radiations that are emitted from the electron shells.

Internal liquid-scintillation counting of soft radiations is a well-known technique of relative measurement, and this counting technique was applied in the direct method of disintegration-rate measurement of ^{55}Fe reported here. The method is based on the calculation of counting-efficiency functions of the liquid scintillation counter and the use of ^{54}Mn and/or ^{51}Cr as internal standards. Mixed sources of ^{55}Fe and either ^{54}Mn or ^{51}Cr as internal standard were prepared and counted in a 4π gamma-coincidence arrangement. The counting efficiency for the internal standard in the mixed source could be measured by coincidence counting as the ratio N_c/N_γ where N_c is the coincidence counting rate and N_γ the gamma counting rate.

Following Gibson and Marshall[1], counting efficiencies were calculated as function of the figure of merit. This established so-called efficiency functions, or the relationship between counting efficiencies for the abovementioned radionuclides. From these efficiency functions and the measured counting efficiency for the internal standard, the relevant counting efficiency for the ^{55}Fe component of the mixed source could be determined. The disintegration rate of the ^{55}Fe was then determined as:

$$N_o = \frac{\text{Counting rate of } {}^{55}\text{Fe}}{\text{Counting efficiency for } {}^{55}\text{Fe}}$$

This work is seen as an improvement on a previously-published method where efficiency functions were determined experimentally and the counting efficiency for ^{55}Fe was obtained by interpolation[2].

The method was recently used during an international intercomparison of radioactivity measurements[10].

CALCULATION OF EFFICIENCY FUNCTIONS

As shown by Gibson and Gale[3] and Gibson and Marshall[1] the counting efficiency for electrons of a discrete energy E can be calculated from the zero probability Z which in turn is dependent on the figure of merit, P (electrons keV^{-1}). On the assumption that all single-electron pulses are counted, the zero probability can be written:

$$Z = e^{-n} = e^{-P.E.F(E)}$$

where n is the mean number of photo-electrons at the photocathode, and F(E) is the relative scintillation efficiency which accounts for the fact that as the rate of energy deposition of an electron in the scintillator increases with decreasing energy, so the light output per energy input is decreased and the scintillation efficiency is reduced.

The escape of characteristic X-rays from the scintillator must also be taken into account, and the contribution to the counting efficiency of X-rays with energy E is then described by:

$$\epsilon = (1 - Z)[1 - J(E)]$$

where J(E) is the escape probability.

The function F(E) varies significantly in the energy range of interest here[4] and this enforces the separate consideration of all the relevant rays and energies (regardless of whether they are coincident in time or not) associated with the rearrangement of the electronic shells following the electron capture.

If the counting cell is viewed by two similar multiplier phototubes in coincidence, a general expression for the counting efficiency can be written:

$$\epsilon = \sum_j \emptyset_j [1 - e^{-n_j}]^2$$

with

$$n_j = P \sum_i E_i . F(E_i)$$

where the summation over i is for the different rays associated with a certain

TABLE 1. ELECTRON ENERGIES USED FOR CALCULATION OF EFFICIENCY FUNCTIONS

Interaction	^{55}Fe		^{54}Mn		^{51}Cr	
	Electron energy, keV[5]	F(E)[4]	Electron energy, keV[5]	F(E)[4]	Electron energy, keV[5]	F(E)[4]
K X-ray escape with L_{II}/L_{III} Auger production.						
L Auger electron	0.639	0.378	0.576	0.365	0.513	0.356
M Auger electron	0.004	0	0.002	0	0.002	0
N Auger electron	0.004	0	0.002	0	0.002	0
K X-ray absorption with L_{II}/L_{III} Auger production.						
K X-ray-produced electron	5.612	0.650	5.129	0.635	4.668	0.623
L Auger electron	0.639	0.378	0.576	0.365	0.513	0.356
Carbon K Auger electron	0.284	0.307	0.284	0.307	0.284	0.307
K Auger emission with L_{II}/L_{III} X-ray absorption and L_{II}/L_{III} Auger production						
K Auger electron	5.246	0.641	4.830	0.625	4.432	0.615
L Auger electron	0.639	0.378	0.576	0.365	0.513	0.356
L X-ray produced electron	0.362	0.326	0.297	0.314	0.234	0.294
Carbon K Auger electron	0.284	0.307	0.284	0.307	0.284	0.307
K Auger emission with production of 2 Auger electrons.						
K Auger electron	5.246	0.641	4.830	0.625	4.432	0.615
L Auger electron	0.639	0.378	0.576	0.365	0.513	0.356
L Auger electron	0.639	0.378	0.576	0.365	0.513	0.356
L_I Auger emission.						
L_I Auger electron	0.671	0.384	0.610	0.372	0.552	0.362

TABLE 2. PARAMETERS USED IN CALCULATIONS OF ϕ_j

Parameter	^{55}Fe	^{54}Mn	^{51}Cr
K-capture probability, S	0.887 (6)	0.889 (6)	0.890 (6)
K-fluorescence yield, W_K	0.313 (7)	0.283 (7)	0.253 (7)
L-fluorescence yield, W_L	0.0029 (7)	0.0026 (7)	0.0023 (7)
X-ray escape probabilities, J_K	0.041 (1)	0.037 (1)	0.032 (1)

Interaction	j	ϕ_j		
		^{55}Fe	^{54}Mn	^{51}Cr
K X-ray escape with L_{II}/L_{III} Auger electron production. $S.W_K(1 - W_L)$	1	0.0114	0.0092	0.0071
K X-ray absorption with L_{II}/L_{III} Auger electron production. $S.W_K(1 - J_K)(1 - W_L)$	2	0.2655	0.2417	0.2175
K Auger emission with L_{II}/L_{III} X-ray absorption and L_{II}/L_{III} Auger production $S(1-W_K)(1-W_L)$	3	0.0036	0.0033	0.0031
K Auger emission with production of 2 Auger electrons. $S(1 - W_K)(1 - W_L)(1 - W_L)$	4	0.6060	0.6341	0.6617
L_I Auger emission. $(1 - S)(1 - W_L)$	5	0.1124	0.1107	0.1097

TABLE 3. CALCULATED COUNTING EFFICIENCIES

P	^{55}Fe	^{54}Mn	^{51}Cr
0.06	0.0379	0.0311	0.0256
0.08	0.0625	0.0517	0.0429
0.10	0.0908	0.0756	0.0631
0.12	0.1216	0.1021	0.0857
0.14	0.1542	0.1303	0.1101
0.16	0.1877	0.1597	0.1358
0.18	0.2217	0.1899	0.1624
0.20	0.2557	0.2204	0.1896
0.22	0.2893	0.2509	0.2170
0.24	0.3222	0.2812	0.2445
0.26	0.3543	0.3109	0.2718
0.28	0.3853	0.3401	0.2988
0.30	0.4151	0.3684	0.3253
0.32	0.4438	0.3959	0.3512
0.34	0.4711	0.4224	0.3765
0.36	0.4972	0.4480	0.4011
0.38	0.5219	0.4725	0.4249
0.40	0.5453	0.4960	0.4479
0.42	0.5675	0.5184	0.4701
0.44	0.5884	0.5397	0.4914
0.46	0.6080	0.5600	0.5119
0.48	0.6266	0.5794	0.5315
0.50	0.6439	0.5977	0.5503

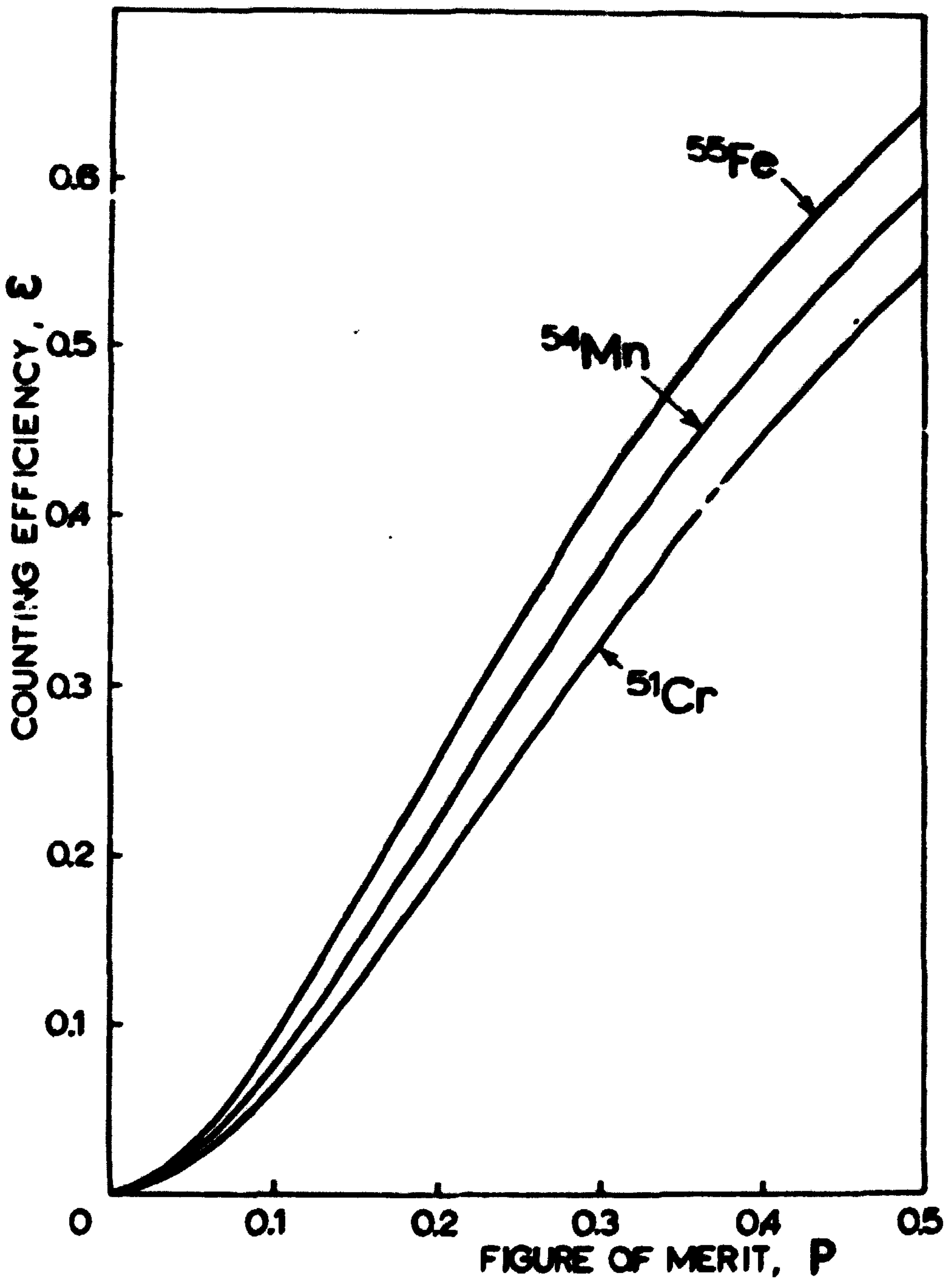

FIGURE 1: Counting efficiencies as function of the figure of merit, P.

mode of interaction, and the summation over j is for the different modes of interaction that can follow the electron capture (see Table 1). The factors ϕ_j comprise electron-capture probabilities (see Table 2). The situation is complicated, and Gibson and Marshall made some simplifying assumptions which were also followed in this work.

The counting efficiency, ϵ, as function of P was calculated in this way for the radionuclides ^{55}Fe, ^{54}Mn and ^{51}Cr as is shown in Figure 1. An appreciable number of constants and parameters from the literature were necessary for the calculations. They are listed in Tables 1 and 2. Table 3 contains some calculated counting efficiencies.

The efficiency functions, $\epsilon_{Fe}(\epsilon_{Mn})$ and $\epsilon_{Fe}(\epsilon_{Cr})$ were found to be described adequately by second degree polynomials:

$$\epsilon_{Fe} = a_o + a_1 \epsilon_{Is} + a_2(\epsilon_{Is})^2$$

where ϵ_{Is} stands for either ϵ_{Mn} or ϵ_{Cr} and where the constants a_o, a_1 and a_2 have different values in different ranges of ϵ as is shown in Table 4.

EXPERIMENTAL

Apart from the phototubes the counting equipment was essentially the same as has been described in previous reports(8), (9). Two RCA 6850 multiplier phototubes were used in coincidence for viewing the counting cell which contained 12.25 ml of 'Insta-Gel' liquid scintillator. The gamma counter was a conventional 76 cm x 76 cm NaI crystal. The electronic amplification in the 4π channel was set to have the single-electron peak at about 5.0 Volt, and the bias levels in the two pulse-height selectors were then set at 0.5 Volt. Under these circumstances the satellite pulses for a source of ^{55}Fe amounted to 0.14% at highest counting efficiency, to about 1.5% for ^{54}Mn and to about 0.2% for ^{51}Cr.

In order that the ratio N_c/N_γ shall be a good measure of counting efficiency, care must be taken to eliminate the Compton spectrum in the gamma-counting channel. A window discriminator was therefore used to isolate and count the upper half of the gamma-ray peak only.

A suitable sample of the internal standard nuclide was first counted at a

TABLE 4. CONSTANTS OF THE SECOND DEGREE POLYNOMIAL

$$\epsilon_{Fe} = a_o + a_1 \cdot \epsilon_{Mn} + a_2 (\epsilon_{Mn})^2$$

Efficiency range	a_o	a_1	a_2
0.05 - 0.15	0.0005	1.2129	-0.2557
0.15 - 0.50	0.0015	1.1981	-0.2050

$$\epsilon_{Fe} = a_o + a_1 \cdot \epsilon_{Cr} + a_2 (\epsilon_{Cr})^2$$

Efficiency range	a_o	a_1	a_2
0.03 - 0.10	0.0008	1.4682	-0.6882
0.10 - 0.20	0.0025	1.4340	-0.5209
0.15 - 0.30	0.0043	1.4140	-0.4653
0.30 - 0.45	0.0068	1.3957	-0.4318

number of different counting efficiencies, including the highest attainable. Lower counting efficiencies were obtained by inserting light absorbers in-between the counting cell and one or both of the phototubes. This procedure established the 4π-counting rate as function of counting efficiency for the internal standard. The counting efficiencies for ^{54}Mn were varied between about 0.08 and 0.53 and for ^{51}Cr between about 0.05 and 0.50. Subsequently the ^{55}Fe sample was accurately weighed out into the counting cell, thoroughly mixed and the counting was repeated. Addition of the ^{55}Fe sample significantly lowered the counting efficiencies. Variation of counting efficiency for the mixed source was then again brought about by use of light absorbers between photocathode and counting cell.

Examples of counting rates for representative sources at maximum counting efficiency are given in Table 5. The ^{54}Mn contribution to the combined counting rates of a mixed source was about 25% and when ^{51}Cr was used as internal standard, the contribution to the 4π-counting rate was about 40%. Because of the relatively low gamma-ray intensity (0.095) more ^{51}Cr had to be added.

The ^{55}Fe component of the combined counting rate was found by appropriate subtraction of the component due to the internal standard. This subtraction also eliminated the background and the satellite impulses associated with the internal standard. The apparent disintegration rate of the ^{55}Fe component was then found as:

$$N_o = \frac{\text{Combined counting rate} - \text{internal-standard counting rate}}{\epsilon_{Fe}}$$

where ϵ_{Fe} was found from the measured ratio N_c/N_γ and the appropriate efficiency function.

RESULTS AND DISCUSSION

Figures 2 and 3 show the apparent disintegration rate of representative ^{55}Fe sources as function of counting efficiency. The vertical scatter of the data points appears to be larger than could be explained by purely statistical considerations. The apparent disintegration rate drops at counting efficiencies < 0.2 irrespective of which internal standard was used, but appears to be independent of counting efficiency at the higher

TABLE 5. COUNTING RATES

		^{54}Mn (s^{-1})	$^{55}Fe+^{54}Mn$ (s^{-1})	^{51}Cr (s^{-1})	$^{55}Fe+^{51}Cr$ (s^{-1})
Background	4π	16.50	16.50	16.50	16.50
	gamma	2.61	2.61	3.36	3.36
	coincidence	0.04	0.04	0.09	0.09
Representative sources	4π	5000	20800	16600	41500
	gamma	184	179	188	180
	coincidence	98	90	94	83

TABLE 6. ^{55}Fe STANDARDIZATION

Internal standard	^{54}Mn	^{51}Cr
Radioactivity concentration of ^{55}Fe solution, (MBq, g^{-1})	3.977	3.950
Standard error of mean (MBq.g^{-1})	0.004	0.007
Number of degrees of freedom	8	8
Systematic uncertainty, (%)	0.85	0.85

counting efficiencies.

The question arises how faithfully does the apparent disintegration rate at higher counting efficiencies reflect the true disintegration rate. The results were therefore also treated in an alternative way as follows. A linear relationship was found between the 4π-counting rate and the counting efficiency which on extrapolation to $\epsilon_{Fe} = 1$ yielded, whithin statistical uncertainties, the same results as before (see Figure 4.). It was therefore concluded that measurement at one relatively high counting efficiency would be sufficiently accurate for determination of the disintegration rate of the ^{55}Fe component of the mixed source.

Table 6 contains the results of measurements on the radioactivity concentration of an ^{55}Fe solution obtained with use of both the internal standards under discussion. These results were obtained during participation in an international intercomparison of radioactivity measurement organized by the NPL in the first quarter of 1979. There is a significant difference of 0.67% between the two results. A systematic error can of course be made when equating N_c/N_γ with the counting efficiency, but the magnitude of such an error ought to be different for different internal standards. The difference found was therefore taken as a measure of the abovementioned systematic error.

An appreciable number of constants and parameters from the literature were used in calculation of the efficiency functions. It should be noted, however, that the final result is not directly dependent on the absolute values of the calculated counting efficiencies. It is the relationship between calculated efficiencies that is used, and it can be argued that this relationship is probably more accurate than the values of the efficiencies themselves. The accuracy of the final result is, however, directly dependent on the accuracy of the measured counting efficiency for the tracer nuclide. The ratio N_c/N_γ must therefore be measured with care and a sufficient quantity of internal standard nuclide must be used to ensure adequate gamma- and coincidence-counting rates.

INTERNATIONAL INTERCOMPARISON, ^{55}Fe

Results obtained by nine participating laboratories are summarised in graphical form in Figure 5. With the exclusion of one result, a total

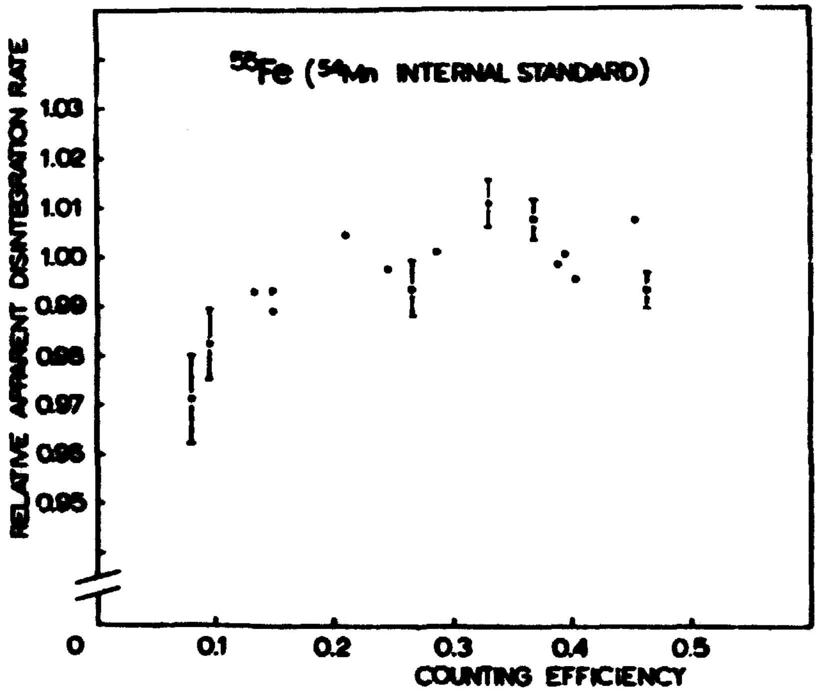

FIGURE 2: Apparent disintegration rate of ^{55}Fe source as function of counting efficiency, obtained with ^{54}Mn as internal standard.

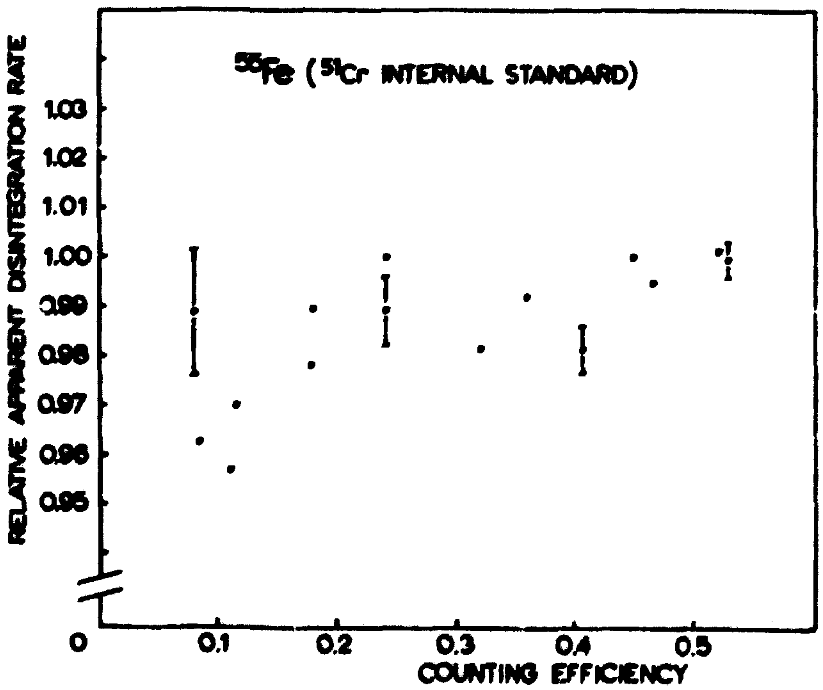

FIGURE 3: Apparent disintegration rate of ^{55}Fe source as function of counting efficiency, obtained with ^{51}Cr as internal standard.

spread in results of about 4.8% is evident. The systematic uncertainties quoted tend to be high, reaching almost 9% in one case. It is therefore clear that the last word about the accurate measurement of the radioactivity of ^{55}Fe sources has not yet been written.

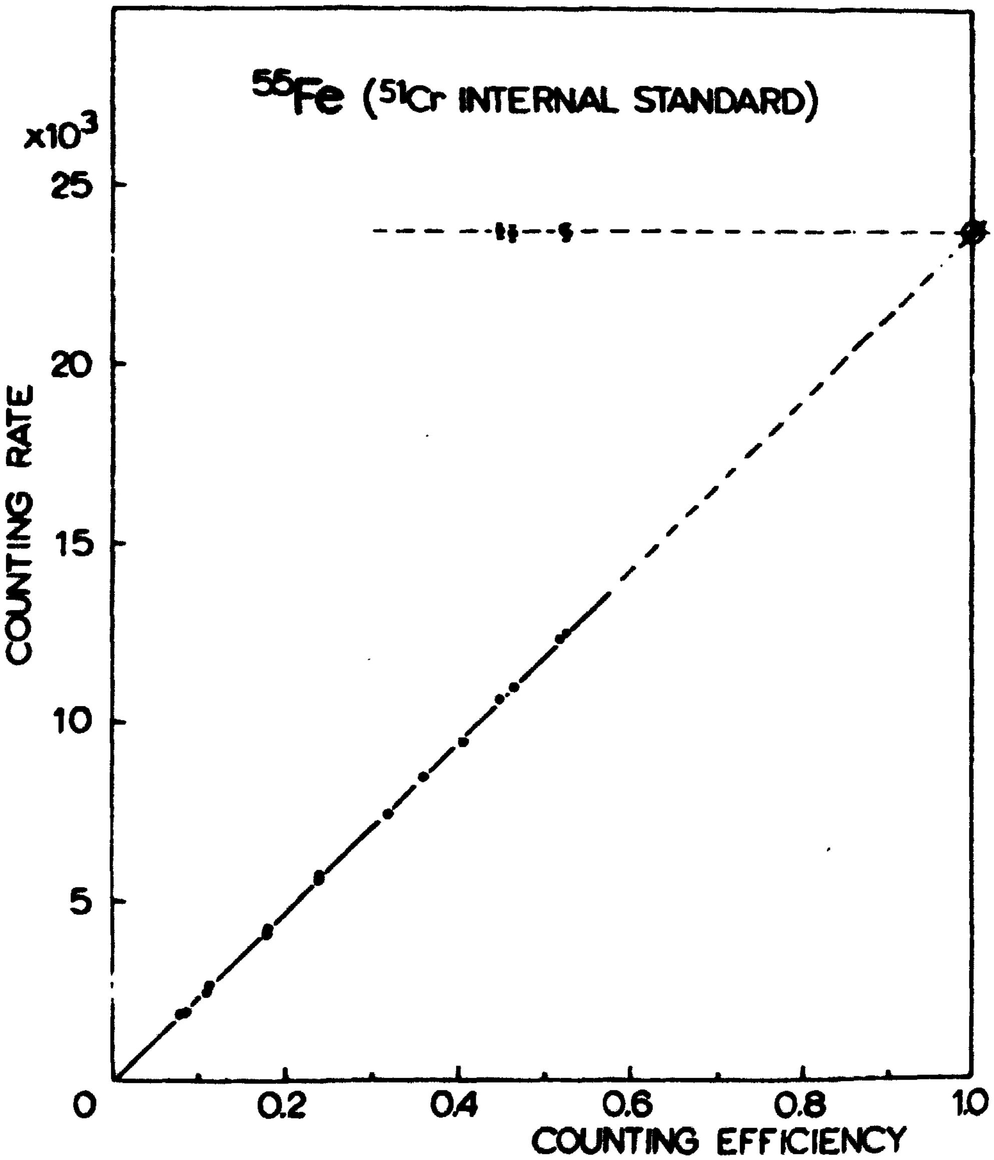

FIGURE 4: Linear relationship between 4π counting rate and counting efficiency for ^{55}Fe source obtained with ^{51}Cr as internal standard. Correlation coeficient, $r = 0.99991$. Some values of the ratio: 4π-counting rate/ϵ are also shown.

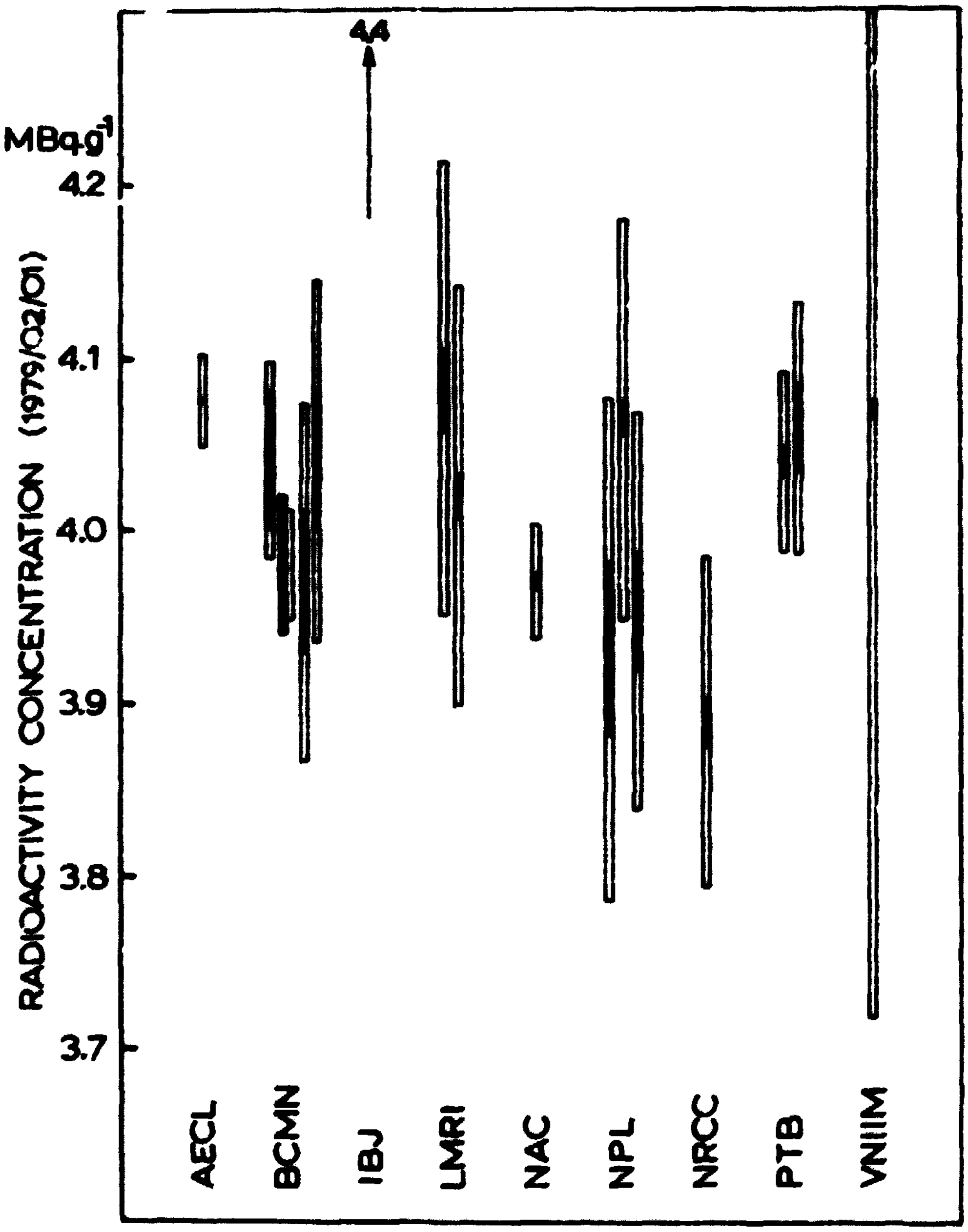

FIGURE 5: Graphical presentation of the results of the international comparison of radioactivity measurements: ^{55}Fe, February 1979(10). These results are preliminary and may be subject to modification.

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