CIEMAT/NIST STANDARDIZATION METHOD EXTENDED TO ANODE OUTPUTS FOR BETA AND ELECTRON-CAPTURE NUCLIDES

J. F. ORTIZ

Departmento de Mecánica, Escuela Técnica Superior de Ingenieros Industriales (ETSII), Universidad Nacional de Educación a Distancia (UNED), E-28040 Madrid, Spain

J. M. LOS ARCOS and AGUSTIN GRAU MALONDA

Instituto de Investigación Básica, Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT), E-28040 Madrid, Spain

ABSTRACT. We present here the application of the CIEMAT/NIST method, extended with computed anodic efficiency, to the standardization of beta emitters or electron-capture nuclides, for a liquid scintillation counting system system with two photomultiplier tubes (PMT) working in summed-coincidence mode. We assume that electron amplification through all photocathode and dynode PMT stages is governed by the Poisson distribution. We tested the procedure with several samples of ^{45}Ca and ^{55}Fe radioactive solutions, quenched with CCl_4, in Optiphase HiSafe™ II scintillator. The efficiency tracing was performed with a set of ^{3}H standard reference samples, prepared with the same scintillator, with efficiencies varying between 15% and 50%. The final computed efficiencies for the problem samples are between 90% and 95% for ^{45}Ca, or between 45% and 50% for ^{55}Fe, and agree with self-standardized experimental values within 0.6%. The results of the CIEMAT/NIST extended method, compared to the conventional method, show small discrepancies of <0.4% for the full range of quenching.

INTRODUCTION

The Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas/National Institute of Standards and Technology (CIEMAT/NIST) efficiency-tracing method (Grau & García-Toraño 1981; García-Toraño & Grau 1985) permits calculation of liquid scintillation counting (LSC) efficiency of beta emitters as a function of a figure of merit (FM) at the photocathode level. Researchers (Breitenberger 1955; Lombard & Martin 1971; Gale and Gibson 1966; Prescott 1966; Ballini 1974; Ortiz *et al.* 1992a,b) showed that the electronic signal amplification along the dynodic stages of a photomultiplier tube (PMT) involves an efficiency loss related to the probability of non-detection. Ortiz, Grau and Los Arcos (1991) described a model to account for PMT losses and evaluated subsequent dynode efficiencies with two sets of standard reference samples of ^{3}H and ^{14}C.

We use the same model here to compute the efficiency at the anode level of the PMTs, and apply the anode-extended CIEMAT/NIST method to the standardization of two sets of problem samples of ^{45}Ca and ^{55}Fe. We compare the results to those obtained by the conventional procedure, when no dynode amplification is considered.

EVALUATION OF ANODE EFFICIENCY

In an LSC system with two phototubes working in summed-coincidence mode, the anode efficiency can be described by three parameters: 1) the FM of the scintillator-photocathode system (Grau & García-Toraño 1981; García-Toraño & Grau 1985); 2) the gain of the first dynode, δ_1; and 3) the gain of the other dynodes, δ, usually $<\delta_1$ (Ortiz, Grau & Los Arcos 1991; Ortiz *et al.* 1992a,b). The FM represents the energy needed to produce a photoelectron, whereas dynode gain affects the response when a single electron reaches it.

When a radioactive emission occurs and an energy, E, is deposited in the scintillator cocktail, the mean number of electrons generated at the cathode is:

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$$n(E)=\frac{EQ(E)}{M} \tag{1}$$

where Q(E) is the ionization quenching factor (Birks 1964; Los Arcos & Borras 1990). Assuming a Poisson response for the cathode, the probability of obtaining r electrons at the output of the photocathode is

$$P(r,E)=\frac{n(E)^r e^{n(E)}}{r!} \quad . \tag{2}$$

If we also assume that dynodes have a Poisson response, each group of r electrons that leaves the cathode is amplified through the K dynodes of the PMT, thus generating in the anode an output signal corresponding to the deposited energy, E. The final detection efficiency will be lower than the photocathode efficiency, due to the non-detection probability of each dynode stage. For a cascaded process, with gains, δ_k, the probability of having no electrons at the kth dynode stage is given by Ortiz *et al.* (1992a,b):

$$\begin{aligned} P_k(r,0)&=e^{\delta_k r(P_{k+1}(1,0)-1)}; \quad k<K \\ P_k(r,0)&=e^{-\delta_k r}; \qquad k=K \quad . \end{aligned} \tag{3}$$

Thus, in a single PMT, the counting efficiency at the photocathode level, related to the deposited energy, E, is

$$\varepsilon_f=\sum_{r=1}^{\infty} P(r,E) \tag{4}$$

and the efficiency at the anode is then

$$\varepsilon_a(E)= \varepsilon_f(E)-\sum_{r=1}^{\infty} P(r,E)P_1(r,0) \quad . \tag{5}$$

For two PMTs working in summed-coincidence, the counting efficiency is

$$\varepsilon(E)= [\varepsilon_a(E)]^2 \quad . \tag{6}$$

Thus, for a β emitter with Fermi spectrum, N(E), and end energy, E_m, the total efficiency ε_β is

$$\varepsilon_\beta= \int_0^{E_m} N(E)\varepsilon(E)dE \quad . \tag{7}$$

In the same way, for an electron-capture nuclide that deposits an energy, E_i, with probability, $W(E_i)$, in the ith atomic rearrangement, the total efficiency is

$$\varepsilon_{ec}= \sum_{i=1}^{N_v} W(E_i)\varepsilon(E_i) \tag{8}$$

where N_v is the total number of atomic rearrangements that follow the electron capture.

METHODS[1]

The experimental measurements were performed with an LKB Rackbeta 1219 Spectral liquid scintillation counter with two EMI 9829QB PMTs working in summed-coincidence and a ^{226}Ra source for quench correction by the external standard method. The PMTs have a quartz window and are linear-focused, with 12 dynodes. The dynodic gains, δ_1, δ, estimated from manufacturer-supplied specification sheets for BeCu surfaces and measurements of inter-electrode voltages, are 4.8 and 2.6, respectively. The samples were prepared in glass vials, with low potassium content, filled with 15 ml of Optiphase HiSafe™ II scintillator for ^{45}Ca and Insta-Gel® for ^{55}Fe. Two stable solutions of (HDEHP)$_n$^{45}Ca and (HDEHP)$_n$^{55}Fe in xylene, with 0.4 $\mu g\ \mu l^{-1}$ of Ca^{2+} and 0.11 $\mu g\ \mu l^{-1}$ of Fe^{3+}, were used to prepare 7 samples of ^{45}Ca and 5 samples of ^{55}Fe with different degrees of quench. The experimental curve for quench calibration was obtained with samples from a standard reference solution of ^{3}H-labeled n-hexadecane. In all cases, CCl_4 was used as quench agent and the radioactive materials were weighed with a Sartorius 1712MP8 electronic scale.

RESULTS

The activity concentration of the ^{45}Ca and ^{55}Fe solutions was determined from

$$a = \frac{N}{\varepsilon\ m} \tag{9}$$

where a is the activity concentration, N is the total count rate, ε is the counting efficiency and m is the mass of the radioactive solution.

The counting efficiency ε can be determined by the CIEMAT/NIST efficiency-tracing method (Grau & García-Toraño 1981), which requires an experimental efficiency-quench curve for ^{3}H samples and the computed curves of efficiency *vs.* FM for ^{3}H, ^{45}Ca and ^{55}Fe. To investigate the influence of PMT losses on the accuracy of the CIEMAT/NIST method, the efficiencies have been computed at both photocathode and anode levels, giving two efficiency estimates, ε_c and ε_a, and consequently two methods for estimating the efficiency.

Calculation of the conventional photocathode efficiency of the β emitters, ^{3}H and ^{45}Ca ($\varepsilon_c(^{3}H)$ and $\varepsilon_c(^{45}Ca)$), were performed with the EFFY program (García-Toraño & Grau 1985); those corresponding to $\varepsilon_c(^{55}Fe)$ were carried out with the VIASKL code (Los Arcos, Grau & Fernandez 1987), specially designed for electron-capture nuclides. Both codes were modified to account for the 12 dynode stages in each PMT to compute the anode efficiencies of ^{45}Ca and ^{55}Fe, ($\varepsilon_a(^{45}Ca)$ and $\varepsilon_a(^{55}Fe)$). Figure 1A shows the experimental ^{3}H efficiency *vs.* the quenching parameter, and Figure 1B, the computed ^{3}H efficiencies as a function of the FM; the values at the cathode and the anode differing <1.8% for FMs between 1 and 5.

The ^{45}Ca efficiencies at the cathode and the anode *vs.* the FM are shown in Figure 2A, which shows very small efficiency losses (<0.2%). After application of the CIEMAT/NIST procedure using both efficiency estimates, the final efficiency *vs.* quench curves, $\varepsilon_c(^{45}Ca)$ and $\varepsilon_a(^{45}Ca)$ almost completely overlap (Fig. 2B).

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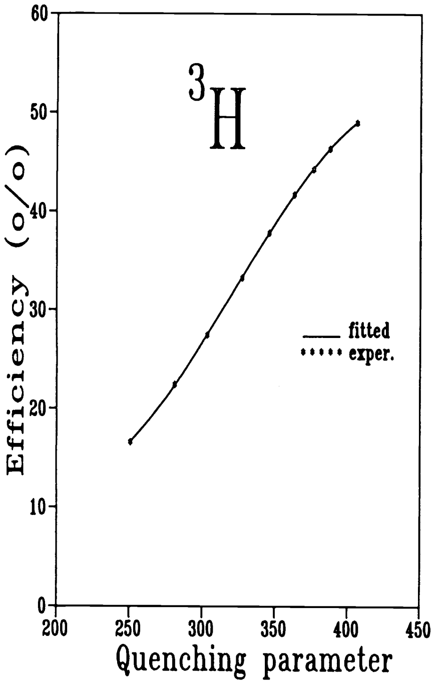

Fig. 1A. Experimental quench curve of ^{3}H efficiency

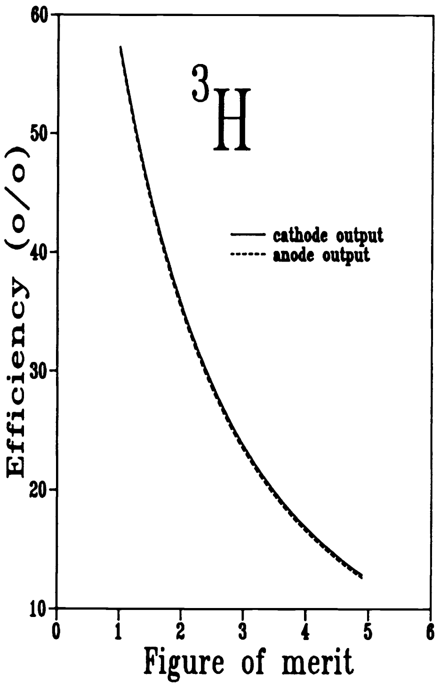

Fig. 1B. Computed curves (cathode, anode) of ^{3}H efficiency *vs.* the FM

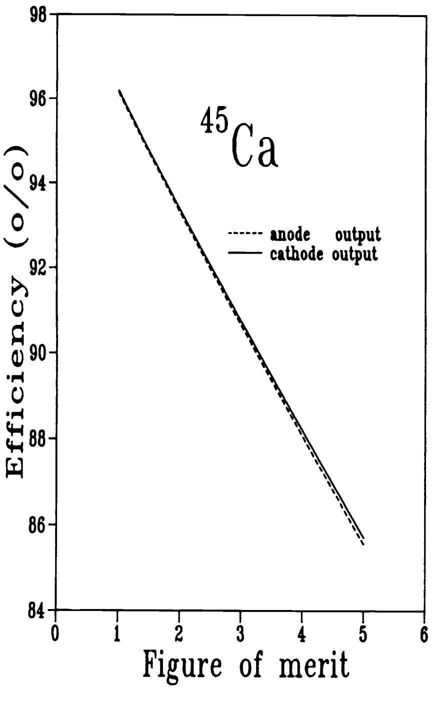

Fig. 2A. Computed curves (cathode, anode) of ^{45}Ca efficiency *vs.* the FM

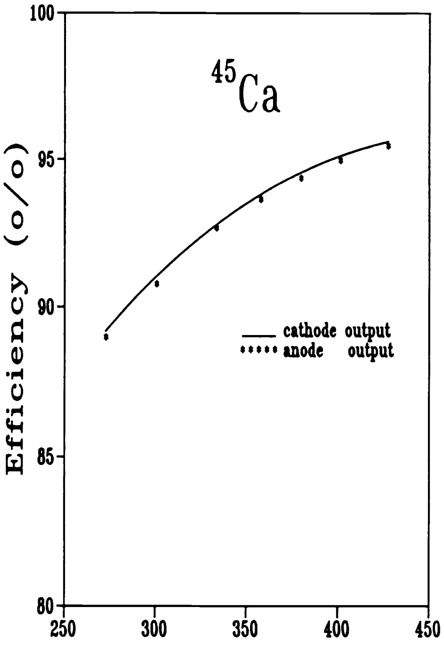

Fig. 2B. CIEMAT/NIST predicted values (cathode, anode) for the ^{45}Ca efficiency-quench curves

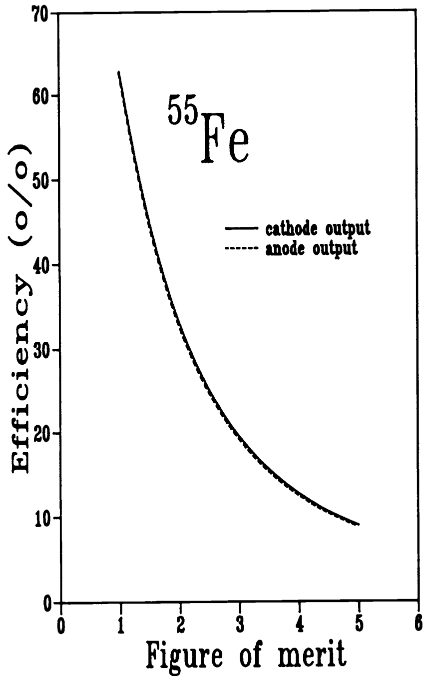

Fig. 3A. Computed curves (cathode, anode) of ^{55}Fe efficiency *vs.* the FM

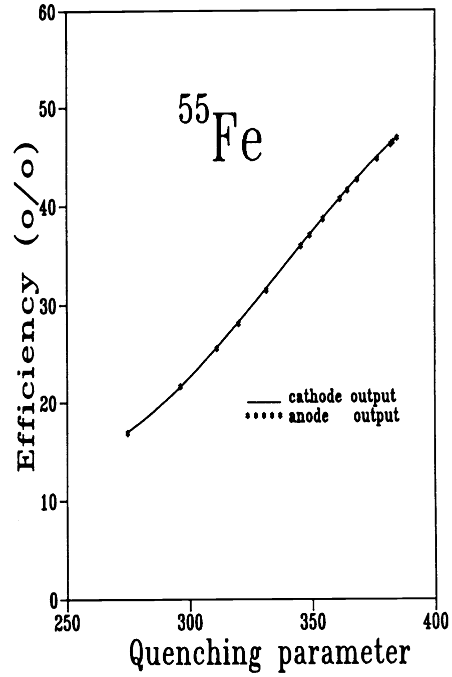

Fig. 3B. CIEMAT/NIST predicted values (cathode, anode) for the ^{55}Fe efficiency-quench curves

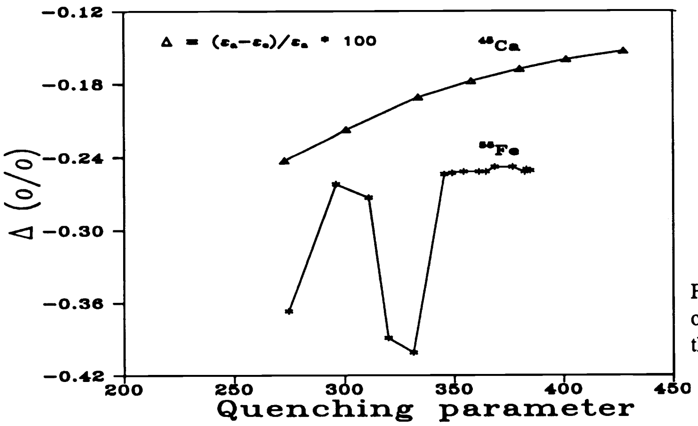

Fig. 4. Relative differences between the cathode and anode efficiencies obtained through the CIEMAT/NIST method

Figure 3A presents the ^{55}Fe efficiency values, computed as a function of the FM; Figure 3B shows the final efficiency-quench curves, obtained after application of the CIEMAT/NIST procedure. The relative differences can be better appreciated in Figure 4, which shows very good agreement of both conventional and anode-extended procedures, with differences <0.2% for ^{45}Ca and <0.4% for ^{55}Fe, respectively.

The problem samples were self-standardized by evaluating the activity concentration from each quenched vial, according to Equation 9, and estimating the activity concentration of the radioactive solution as the average value of all samples. With this average value and the measured count rates,

TABLE 1. Computed and Experimental Efficiencies and Activities Estimated from ^{45}Ca-Quenched Samples

	Photocathode				Anode			
Q	ε_{comp} (%)	a_c* Bq mg^{-1}	ε_{exp} (%)	Δ** (%)	ε_{comp} (%)	a_a† Bq mg^{-1}	ε_{exp} (%)	Δ** (%)
427.5	95.622	35.04	95.246	-0.39	95.476	35.09	95.068	-0.43
401.4	95.126	35.17	95.094	-0.03	94.973	35.22	94.916	-0.06
380.0	94.530	35.21	94.617	0.09	94.372	35.27	94.440	0.07
357.9	93.827	35.16	93.792	-0.04	93.661	35.23	93.661	-0.05
333.6	92.866	35.26	93.092	0.24	92.689	35.33	92.918	0.25
300.7	90.976	35.17	90.952	-0.03	90.779	35.25	90.782	0.00
272.7	89.205	35.23	89.344	0.16	88.989	35.32	89.177	0.21

*a_c = 35.18 Bq mg^{-1} ± 0.20 %
**$\Delta=(\varepsilon_{comp}-\varepsilon_{exp})/\varepsilon_{comp}$
†a_a = 35.24 Bq mg^{-1} ± 0.22 %

TABLE 2. Computed and Experimental Efficiencies and Activities Estimated from ^{55}Fe-Quenched Samples

	Photocathode				Anode			
Q	ε_{comp} (%)	a_c* Bq mg^{-1}	ε_{exp} (%)	Δ** (%)	ε_{comp} (%)	a_a† Bq mg^{-1}	ε_{exp} (%)	Δ** (%)
384.7	46.957	98.35	46.843	-0.24	46.839	98.59	46.736	-0.27
383.0	46.531	99.08	46.762	0.50	46.415	99.33	46.632	0.47
382.1	46.309	99.11	46.555	0.53	46.192	99.36	46.425	0.50
376.6	44.848	98.94	45.007	0.35	44.737	99.18	44.882	0.32
368.4	42.737	99.20	43.003	0.62	42.631	99.44	42.883	0.59
364.4	41.633	98.04	41.402	-0.55	41.528	98.29	41.287	-0.58
361.3	40.769	97.52	40.326	-1.09	40.666	97.76	40.214	-1.11
354.3	38.741	99.05	38.923	0.47	38.643	99.30	38.815	0.44
349.0	37.115	98.67	37.146	0.08	37.021	98.92	37.043	0.06
345.4	36.016	98.96	36.151	0.38	35.924	99.21	36.051	0.35
331.3	31.583	98.36	31.511	-0.23	31.457	98.76	31.423	-0.11
319.9	28.190	98.07	28.043	-0.52	28.080	98.45	27.964	-0.41
311.0	25.603	98.17	25.494	-0.43	25.537	98.42	25.423	-0.45
296.1	21.727	98.96	21.810	0.38	21.670	99.22	21.749	0.36
274.6	16.943	98.33	16.900	-0.26	16.881	98.69	16.853	-0.17

*a_c = 98.59 Bq mg^{-1} ± 0.51 %
**$\Delta=(\varepsilon_{comp}-\varepsilon_{exp})/\varepsilon_{comp}$
†a_a = 98.86 Bq mg^{-1} ± 0.50 %

the experimental efficiency of each sample was determined. Table 1 presents predicted efficiencies, activity concentration, experimental efficiency and its discrepancy with the predicted values at both the photocathode and the anode level for each ^{45}Ca sample. The average activities estimated from both methods agree within 0.2%, with efficiency discrepancies always <0.25%. Table 2 presents results obtained for the ^{55}Fe samples, the average activities differing by 0.3%, with efficiency discrepancies <0.6%.

CONCLUSIONS

The results obtained with two typical radionuclides, a pure β emitter, ^{45}Ca, and a pure electron-capture nuclide, ^{55}Fe, allow us to conclude that the CIEMAT/NIST method is essentially self-consistent, regardless of the PMT photocathode or anode stage at which the efficiency is evaluated. Although the efficiency associated with a given FM is evaluated more realistically at the anode level, the inclusion of the statistical fluctuations of up to 12 dynodes does not add any significant correction to the final values of the efficiency-quench curve for the problem nuclide. The differences between both methods are well below the effect of other uncertainty factors, such as count-rate statistics, ^{3}H standards calibration or the nuclear constants of the radionuclide, even though the anode-based procedure is very time-consuming.

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