

POISSONIAN AND BINOMIAL MODELS IN RADIONUCLIDE METROLOGY BY LIQUID SCINTILLATION COUNTING

por:

A. Grau Malonda

CENTRO DE INVESTIGACIONES

ENERGETICAS, MEDIOAMBIENTALES Y TECNOLOGICAS

MADRID, 1990

440102 SCINTILLATION COUNTING EFFICIENCY COINCIDENCE METHODS PHOTOMULTIPLIERS DISTRIBUTION FUNCTIONS MATHEMATICAL MODELS COMPARATIVE EVALUATIONS

1

CLASIFICACION DOE Y DESCRIPTORES

Toda correspondencia en relación con este trabajo debe dirigirse al Servicio de Información y Documentación, Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, Ciudad Universitaria, 28040-MADRID, ESPAÑA.

Las solicitudes de ejemplares deben dirigirse a este mismo Servicio.

Los descriptores se han seleccionado del Thesauro del DOE para describir las materias que contiene este informe con vistas a su recuperación. La catalogación se ha hecho utilizando el documento DOE/TIC-4602 (Rev. 1) Descriptive Cataloguing On-Line, y la clasificación de acuerdo con el documento DOE/TIC.4584-R7 Subject Categories and Scope publicados por el Office of Scientific and Technical Information del Departamento de Energía de los Estados Unidos.

Se autoriza la reproducción de los resúmenes analíticos que aparecen en esta publicación.

Este trabajo se ha recibido para su impresión en Mayo de 1991

Depósito Legal nº M-18969-1991 ISBN 84-7834-106-4 ISSN 614-087-X NIPO 238-91-004-4

IMPRIME CIEMAT

CONTENTS

1.	Introduction	7
2.	Three photomultipliers detectors	8
3.	Poissonian model	9
4.	Binomial model	10
5.	Equivalence of both models	13
6.	System with two photomultipliers	13
7.	System with three photomultipliers	16
8.	Conclusions	22
9.	References	23

1. INTRODUCTION

It is a well - know fact that the use of a coincidence system reduce profoundly the contribution of the thermoionic noise to the counting rate.

A negative aspect of the coincidence system is the decrease of the counting efficiency as the energy of the interesting particles is low, lower than 100 keV, and the number of the photomultipliers used is high.

In 1961, in a pioneer work, Horrocks and Studier [1] proposed the idea that the ratio between the counting efficiencies calculated for two photomultipliers in coincidence and for only one photomultiplier is related to the counting efficiency in such a way that if it is extrapolated to rate 1, the extrapolated counting efficiency will be of 100% and the extrapolated counting rate will coincide with the activity expressed in the same time units. Although the idea and procedure described are correct, the difficulty of getting the real counting rate in an isolated photomultiplier, due to the interference of the thermoionic noise, limits the applicability of the procedure to radionuclides with beta-ray emisssion higher than 100 keV.

It seems immediate to think that the Horrocks and Student procedure may be applied to a detector system with three photomultipliers and use the counting efficiencies for triple and double coincidences.

The main difficulty was the fact of the quantum efficiency from the photomultipliers being very reduced, especially in the case of 3 H (lower than 10%), which meant a high uncertainty in the determination of the specimen activity.

The development of photomultipliers with high quantum efficiency changed radically the situation to the extent that, already in 1966. Schwerdtel [2] assambled the first system based on three photomultipliers. In 1979, Pochwalski and al. [4] assembled a system of triple and double coincidences, much more alaborate, in order to obtain directly the activity of a radiactive specimen.

In this paper it is described and analyzed the way a system with three photomultipliers work. The two models developed to calculate the triple to double coincidence ratio and the counting efficiencies are: the model based in the distribution of Poisson, developed by Grau Malonda and Coursey [5] for beta-ray emitters and Grau Carlés and Grau Malonda [6] for electron capture decay; and the model based on a binomial distribution, Broda and al. [7] for

beta-ray emitters. It is shown in this paper that, in spite of the different hypothesis and statistics applied, the two models lead to identic results for monoenergetic electron emission.

2. THREE PHOTOMULTIPLIERS DETECTORS

When the detector system of the liquid scintillation spectrometer is constituted by three photomultipliers, the liquid scintillation emission is distributed among the three tubes. If the three photomultipliers are placed around the vial on a plane normal to the axis of the vial and making an angle of 120° between each of them, the light received through every photomultiplier will be, as an average, the same. If, in addition, the photomultipliers have the same characteristics, the output signals will be similar.

A system with three photomultipliers A_1 , A_2 and A_3 may work in three different ways: without any coincidence, or S mode; with two photomultipliers working in coincidence, or D mode; with three photomultipliers in coincidence, or T mode.

Different forms of logic pulse additions may be associated to each mode:

 S_1 , individual signal from A_1 , A_2 or A_3 .

 S_2 , signal being the addition of two photomultipliers, $A_1 + A_2$, $A_1 + A_3$ or $A_2 + A_3$.

 S_3 , signal being the addition of the three photomultipliers, $A_1 + A_2 + A_3$

When the system works in a double coincidence mode, the following forms of logical addition may result:

D₁, coincident signal without any addition, A₁ A₂, A₂A₃ or A₁A₃

D₂, addition of a couple of coincident signals, A₁ A₂+ A₁A₃, A₁A₂+A₂A₃ or A₁A₃+A₂A₃

 D_3 , addition of the three double coincidences $A_1 A_2 + A_1 A_3 + A_2 A_3$

Finally, the mode of triple coincidence is:

T, coincidence of the three photomultipliers, $A_1A_2A_3$

Fig. 1 represents the block-diagram of a system with three photomultipliers and the electronic scheme to obtain the addition of the double coincidence D_3 and the triple T. The photomultiplier pulses go to four gates C of the "y" type. Three of them work as double coincidence gates and their outputs are logically summed in the "or" type gate that constitutes the first output for the system $(A_1A_2+A_1A_3+A_2A_3)$. The fourth C gate is a triple coincidence $A_1A_2A_3$

3. POISSONIAN MODEL

In this model it is assumed as a basic hypothesis that the photocathode answer is a discrete distribution of photoelectrons following the statistical distribution of Poisson, when a monoenergetic electron bean interacts with the liquid scintillator.

The Poisson distribution is given by:

$$P(n,\overline{n}) = \frac{\overline{n}^{n} e^{-\overline{n}}}{n!}$$
(1)

where \overline{n} is the average of the expected electron number and $P(n, \overline{n})$ the probability of being emitted exactly n electrons as the expected average is \overline{n} .

The second hypothesis implicitly admited is whenever in the photocathods is emitted at least one photoelectron a pulse will be produced at the output of the photomultiplier anode. Therefore, the counting efficiency for a single photomultiplier may be expressed as:

$$\varepsilon = \sum_{n=1}^{\infty} P(n, \overline{n})$$
 (2)

or

$$\varepsilon = 1 - P(o, \overline{n}) = 1 - e^{-\overline{n}}$$
(3)

In order to connect the initial energy from the incident particle with the average value of expected electrons it is defined a free parameter λ , such as

$$\lambda = \frac{E Q(E)}{m}$$
(4)

where Q(E) is a function taking into account the non-linear effects of change from the kinetic energy of the particle into scintillation emission and m is the average number of photoelectrons emitted by all the photomultipliers in the system. For one photomultipliers $m = \overline{n}$ and for p photomultipliers $m = p\overline{n}$.

It is assumed that the only non-linear effect is due to ionization quench. Following Birks [8] the conection factor due to ionization quench is:

$$Q(E) = \frac{1}{E} \int_{0}^{E} \frac{dE}{1 + kB \left(\frac{dE}{dx}\right)}$$
(5)

where k is a constant and $B\left(\frac{dE}{dx}\right)$ is the concentration of ionizing events.

Expressions [2] and [3] are valid for just one photomultiplier. The expressions are modified when the signals from several photomultipliers are taken to logic systems of coincidence and addition.

Tables 1 and 2 show the corresponding formulae allowing to obtain the counting efficiency for the different operation modes of the logic system, in the case of systems with 2 and 3 photomultipliers.

4. BINOMIAL MODEL

In this model it is assumed that when a beam of monoenergetic electrons interact with the liquid scintillator the number of emitted photons per electron reaching the photocathodes follows the Poisson distribution. The probability that a determined number f of photons arrives at the photomultipliers will be given by

$$P(f, \overline{f}) = \frac{\overline{f}^{f} e^{-f}}{f!}$$
(6)

where \overline{f} is the average number of photons.

_

It is beside supposed that the emission of photoelectrons by the photocathode is ruled by the binomial distribution:

$$P_{b}(k, f, \varepsilon_{q}) = \frac{f!}{k!(f-k)!} = \varepsilon_{q}^{k}(1-\varepsilon_{q})^{f-k}$$
(7)

where k is the number of photoelectrons emitted through the interaction of f photons, ϵ_q is the quantum efficiency of the photocathode.

When just f photons arrive at the photocathodes they will be distributed among the photomultipliers in such a way that the probability of a determined distribution being produced will be:

$$P_{d} = \frac{1}{Q^{f}} \frac{f !}{f_{1} ! f_{2} ! ... f_{Q} !}$$
(8)

where Q is the number of photomultipliers and

$$f = f_1 + f_2 + \dots + f_Q$$
 (9)

If X is the probability of at least a photoelectron being produced in each of the active photomultipliers, it allows to express the probability $P_r(f)$ of a fixed number of photons f producing a countable pulse:

$$P_r = P_d X \tag{10}$$

The additions of the probabilities P_r for all sets of numbers $f_1, f_2, ..., f_Q$ subjected to the condition (9) is:

$$P_{r}(f) = \frac{f !}{Q^{f}} \sum_{f_{1}=0}^{Y_{1}} \sum_{f_{2}=0}^{Y_{2}} \dots \sum_{f_{Q-1}=0}^{Y_{Q}} \frac{1}{f_{1} ! f_{2} ! \dots f_{Q-1} ! (f - f_{1} - \dots - f_{Q-1})!}$$
(11)

where

1

$$Y_1 = f$$

 $Y_2 = f - f_1$
.....
 $Y_{Q-1} = f - f_1 - f_2 - ... - f_{Q-2}$

When the system has two photomultipliers, Q=2, we shall have

$$P_{r}(f) = \frac{f!}{2^{f}} \sum_{a_{1=0}}^{f} \frac{X}{f_{1}!(f - f_{1})!}$$
(12)

And if for three photomultipliers, Q = 3, it results:

$$P_{R}(f) = \frac{f!}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{X}{f_{1} ! f_{2} ! (f - f_{1} - f_{2})!}$$
(13)

The counting probability for \overline{f} photons may be calculated through the expression

$$P_{e}(\bar{f}) = P(f, \bar{f}) P_{R}(f)$$
(14)

And so, the counting efficiency will be given by the expression

$$\varepsilon = P_E(\overline{f}) = \sum_{f=1}^{\infty} P(f, \overline{f}) P_R(f)$$
 (15)

Then the counting efficiency for a system with two photomultipliers is given by

$$\varepsilon = P_{E}(\bar{f}) = e^{-\bar{f}} \sum_{f=f_{1=0}}^{\infty} \sum_{f=f_{1=0}}^{f} \left(\frac{\bar{f}}{2}\right)^{f} \frac{X}{f_{1}!(f-f_{1})!}$$
 (16)

as well as for three photomultipliers it will be

$$\varepsilon = P_{E}(\bar{f}) = e^{-\bar{f}} \sum_{f=1}^{\infty} \sum_{a_{1=0}}^{f} \sum_{a_{2=0}}^{f-\bar{f}_{1}} \left(\frac{\bar{f}}{3}\right)^{f} \frac{X}{f_{1} ! f_{2} ! (f - f_{1} - f_{2}) !}$$
(17)

Tables 3 and 4 give the equations for X for the systems with 2 and 3 photomultipliers. The meaning of the different functions is as follows:

$$P(f_{1}) = 1 - (1 - \varepsilon_{q})^{f_{1}}$$
(18)

$$P (f - f_1) = 1 - (1 - \varepsilon_q)^{f - f_1}$$
(19)

$$P (f - f_1 - f_2) = 1 - (1 - \varepsilon_q)^{f - f_1 - f_2}$$
(20)

5. EQUIVALENCE OF BOTH MODELS

We shall proof that both models the binomial and Poissonian lead to the same values of the counting efficiency. Instead of carrying out the demostration in its most general form, for a number p of photomultipliers when q of them are active and for complex conbinations of coincidences and logic additions, we shall consider the arrangements shown in Tables 1 to 4.

The procedure followed in every case will consist in starting for the counting efficiency formula for the binomial model, Tables 3 and 4. Through mathematical transformations it will be reached the corresponding counting efficiency expression in the Poissonian model. To make easier the deduction procedure it will be obtained the formula designed as non-detection mode in Tables 1 and 2.

6. SYSTEM WITH TWO PHOTOMULTIPLIERS

As the detector system has two photomultipliers, the way of being treated the signals shows three modes: only one active photomultiplier, U; two photomultipliers in addition, S; and two photomultipliers in coincidence, D. We shall analyze each of them beginning with the last one.

6.1 COINCIDENT SIGNALS $\rm A_1~A_2$

From the development of the expression for X in Table 3 it may be obtained:

$$X = 1 - (1 - \varepsilon_q)^{f_1} - (1 - \varepsilon_q)^{f - f_1} + (1 - \varepsilon_q)^{f}$$
(21)

Taking this expression to the formula (16) it will result

$$\varepsilon = \sum_{f=1}^{\infty} \sum_{f_{1}=0}^{f} \frac{P(f, \overline{f})}{2^{f}} \begin{pmatrix} f \\ f_{1} \end{pmatrix} \left\{ 1 - (1 - \varepsilon_{q})^{f_{1}} - (1 - \varepsilon_{q})^{f - f_{1}} + (1 - \varepsilon_{q})^{f} \right\}$$
(22)

By developping the right hand member of (22) we have

$$\sum_{\mathfrak{f}_{1=0}}^{\mathfrak{f}} \begin{pmatrix} \mathfrak{f} \\ \mathfrak{f}_{1} \end{pmatrix} = 2^{\mathfrak{f}}$$
(23)

$$\sum_{f_1=0}^{f} {\binom{f}{f_1}} (1 - \varepsilon_q)^{f_1} = 2^{f} \left(1 - \frac{\varepsilon_q}{2}\right)^{f}$$
(24)

$$\sum_{f_1=0}^{f} {\binom{f}{f_1}} (1 - \varepsilon_q)^{f - f_1} = 2^{f} \left(1 - \frac{\varepsilon_q}{2}\right)^{f}$$
(25)

$$\sum_{f_{1}=0}^{f} {\binom{f}{f_{1}}} (1 - \varepsilon_{q})^{f} = 2^{f} (1 - \varepsilon_{q})^{f}$$
(26)

And so

$$\varepsilon = \sum_{f=1}^{\infty} P(f, \bar{f}) \left\{ 1 - 2 \left(1 - \frac{\varepsilon_q}{2} \right)^f + (1 - \varepsilon_q)^f \right\} =$$

$$= \sum_{f=1}^{\infty} \left\{ P(f, \bar{f}) - 2 P\left[f, \bar{f} \left(1 - \frac{\varepsilon_q}{2} \right) \right] e^{-\bar{f} - \varepsilon_{q/2}} + \right.$$

$$+ P\left[f, \bar{f} \left(1 - \varepsilon_q \right) \right] e^{-\bar{f} - \varepsilon_q} \left\} = (1 - e^{-\bar{n}})^2$$
(27)

Taking into account that

$$\overline{f} \varepsilon_q = 2 \overline{n}$$
 (28)

because \overline{n} is the number of photoelectrons emitted by only one photomultiplier.

62. SIGNALS ADDITITON $A_1 + A_2$

~

The developed expression for X in this situation is

$$X = 1 - (1 - \varepsilon_q)^{f}$$
⁽²⁹⁾

Therefore the counting efficiency may be expressed as:

$$\varepsilon = \sum_{f=1}^{\infty} \sum_{f_{1=0}}^{f} \frac{P(f, \bar{f})}{2^{f}} \begin{pmatrix} f \\ f_{1} \end{pmatrix} \left[1 - (1 - \varepsilon_{q})^{f} \right] =$$
$$= \sum_{f=1}^{\infty} P(f, \bar{f}) \left[1 - (1 - \varepsilon_{q})^{f} \right]$$
(30)

It results by simplification

$$\varepsilon = 1 - \varepsilon^{\overline{f} \varepsilon_{q}} = 1 - e^{-2 \overline{n}}$$
(31)

As we wanted to demostrate

6.3 ONLY ONE ACTIVE PHOTOMULTIPLIER A1

In this case

$$X = 1 - (1 - \varepsilon_q)^{f}$$
⁽³²⁾

and the counting efficiency is:

$$\varepsilon = \sum_{f=1}^{\infty} \sum_{f_{1}=0}^{f} \frac{P(f, \bar{f})}{2^{f}} \begin{pmatrix} f \\ f_{1} \end{pmatrix} \begin{bmatrix} 1 - (1 - \varepsilon_{q})^{f_{1}} \end{bmatrix} =$$
$$= \sum_{f=1}^{\infty} P(f, \bar{f}) \begin{bmatrix} 1 - (1 - \varepsilon_{q})^{f} \end{bmatrix}$$
(33)

Applying the property that the Poisson distribution is normalized to the unit and simplifying, we have:

$$\varepsilon = 1 - \varepsilon^{-\overline{\mathbf{f}}} \varepsilon_{\mathbf{q}}^{2} = 1 - e^{-\overline{\mathbf{n}}}$$
(34)

7. SYSTEM WITH THREE PHOTOMULTIPLIERS

As it may be observed in Table 2 the number of ways to deal with the signal is much higher that in the case of the two photomultipliers.

First of all it will be studied the case where the three photomultiplier work in coincidence.

7.1 TRIPLE COINCIDENCE A1A2A3

By developing the formula for X in Table 2, it is obtained

$$X = 1 - (1 - \varepsilon_{q})^{f_{1}} - (1 - \varepsilon_{q})^{f_{2}} + (1 - \varepsilon_{q})^{f_{1} + f_{2}} - (1 - \varepsilon_{q})^{f - f_{1}} - (1 - \varepsilon_{q})^{f - f_{1}} - (1 - \varepsilon_{q})^{f}$$

$$(35)$$

And taking this expression to the formula (17) it results:

$$\varepsilon = \sum_{f=1}^{\infty} \frac{P(f, \bar{f})}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f!}{f_{1}!f_{2}!(f-f_{1}-f_{2})!} \left\{ 1 - (1 - \varepsilon_{q})^{f_{1}} - (1 - \varepsilon_{q})^{f_{2}} + (1 - \varepsilon_{q})^{f_{1}+f_{2}} - (1 - \varepsilon_{q})^{f-f_{1}} - f_{2} + (1 - \varepsilon_{q})^{f-f_{2}} + (1 - \varepsilon_{q})^{f-f_{1}} - (1 - \varepsilon_{q})^{f} \right\}$$
(36)

By developing each of the members in the right side of (36) and carrying out several mathematical simplifications we reach

$$\sum_{f_1=0}^{f} \sum_{f_2=0}^{f-f_1} \frac{f_1}{f_1 + f_2 + (f - f_1 - f_2)} = 3^{f}$$
(37)

$$\sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f_{1}}{f_{1} ! f_{2} ! (f - f_{1} - f_{2}) !} (1 - \varepsilon_{q})^{f_{1}} = 3^{f} \left(1 - \frac{\varepsilon_{q}}{3}\right)^{f}$$
(38)

$$\sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f_{1}}{f_{1} + f_{2} + (f - f_{1} - f_{2})} \left(1 - \varepsilon_{q}\right)^{f_{2}} = 3^{f} \left(1 - \frac{\varepsilon_{q}}{3}\right)^{f}$$
(39)

$$\sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f_{1}}{f_{1} ! f_{2} ! (f - f_{1} - f_{2}) !} (1 - \varepsilon_{q})^{f_{1} + f_{2}} = 3^{f} \left(1 - 2 \frac{\varepsilon_{q}}{3}\right)^{f}$$
(40)

$$\sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f_{1}}{f_{1} + f_{2} + (f - f_{1} - f_{2})} \left(1 - \varepsilon_{q}\right)^{f-f_{1} - f_{2}} = 3^{f} \left(1 - \frac{\varepsilon_{q}}{3}\right)^{f}$$
(41)

$$\sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f_{1}}{f_{1} \mid f_{2} \mid (f-f_{1}-f_{2}) \mid} (1-\varepsilon_{q})^{f-f_{2}} = 3^{f} \left(1-2\frac{\varepsilon_{q}}{3}\right)^{f}$$
(42)

$$\sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f_{1}}{f_{1} + f_{2} + (f - f_{1} - f_{2}) + (f - f_{1} - f_{2})} (1 - \varepsilon_{q})^{f-f_{1}} = 3^{f} \left(1 - 2 \frac{\varepsilon_{q}}{3}\right)^{f}$$
(43)

$$\sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f_{1}}{f_{1} + f_{2} + (f - f_{1} - f_{2})} \left(1 - \varepsilon_{q}\right)^{f} = 3^{f} \left(1 - \varepsilon_{q}\right)^{f}$$
(44)

If the expressions (37) to (44) are substituted in the equation (36) and some mathematical simplifications are carryed out, it results:

$$\varepsilon = 1 - 3 \ e^{-\tilde{f} \ \varepsilon_q/3} + 3 \ e^{-2\tilde{f} \ \varepsilon_q/3} - e^{-\tilde{f} \ \varepsilon_q} = (1 - e^{-\tilde{n}})^3$$
(45)

7.1 ONLY ONE ACTIVE PHOTOMULTIPLIER A1

In this case

$$X = 1 - (1 - \varepsilon_{\alpha})^{f}$$
 (46)

and the counting efficiency is

$$\varepsilon = \sum_{f=1}^{\infty} \frac{P(f, \tilde{f})}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f!}{f_{1}!f_{2}!(f-f_{1}-f_{2})!} \left[1-(1-\varepsilon_{q})^{f_{1}}\right]$$
(47)

Taking into account the results (37) and (38) we may write:

$$\varepsilon = \sum_{f=1}^{\infty} P(f, \bar{f}) \left[1 - \left(1 - \frac{\varepsilon_q}{3} \right)^f \right]$$
(48)

By simplifying after some operations we obtain

$$\varepsilon = 1 - e^{-\overline{f} \varepsilon_q/3} = 1 - e^{-\overline{n}}$$
(49)

As we intended to demostrate.

7.2 TWO PHOTOMULTIPLIERS IN ADDITION $A_1 + A_2$

The function X is

$$X = 1 - (1 - \varepsilon_q)^{f} 1^{+f} 2$$
(50)

The formula for the counting efficiency may be expressed as

$$\varepsilon = \sum_{f=1}^{\infty} \frac{P(f, \bar{f})}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f!}{f_{1}!f_{2}!(f-f_{1}-f_{2})!} \left[1-(1-\varepsilon_{q})^{f_{1}+f_{2}}\right]$$
(51)

and taking into consideration the formulae (37) and (40) we may write

$$\varepsilon = \sum_{f=0}^{\infty} P(f, \bar{f}) \left[1 - \left(1 - 2\frac{\varepsilon_q}{3} \right)^f \right]$$
(52)

which after a few simplifications is reduced to:

$$\varepsilon = 1 - e^{-2\tilde{t} \varepsilon_q/3} = 1 - e^{-2\bar{n}}$$
 (53)

as we wanted to demostrate.

7.3 THREE PHOTOMULTIPLIERS IN ADDITION A₁ + A₂ + A₃

After several simplifications the expression for X is reduced to:

$$X = 1 - (1 - \varepsilon_q)^{f}$$
(54)

that carried to (17) gives

$$\varepsilon = \sum_{f=1}^{\infty} \frac{P(f, \bar{f})}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f!}{f_{1}!f_{2}!(f-f_{1}-f_{2})!} \left[1-(1-\varepsilon_{q})^{f}\right]$$
(55)

and applying (37) and (44) yield

$$\varepsilon = \sum_{f=1}^{\infty} P(f, \bar{f}) \left[1 - (1 - \varepsilon_q)^f \right]$$
(56)

it is to say

$$\varepsilon = 1 - e^{-\tilde{f} \varepsilon_q} = 1 - e^{-3 \bar{n}}$$
(57)

7.4 TWO PHOTOMULTIPLIERS IN COINCIDENCE $\rm A_1~A_2$

From Table 4 it is obtained by operation

$$X = 1 - (1 - \varepsilon_q)^{f_1} - (1 - \varepsilon_q)^{f_2} + (1 - \varepsilon_q)^{f_1 + f_2}$$
(58)

The counting efficiency will be expressed as

$$\varepsilon = \sum_{f=1}^{\infty} \frac{P(f, \bar{f})}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f!}{f_{1}!f_{2}!(f-f_{1}-f_{2})!} \left\{ 1 - (1 - \varepsilon_{q})^{f_{1}} - (1 - \varepsilon_{q})^{f_{1}} - (1 - \varepsilon_{q})^{f_{2}} + (1 - \varepsilon_{q})^{f_{1}} + f_{2} \right\}$$
(59)

and applying the results (37) to (40) we have:

$$\varepsilon = \sum_{f=1}^{\infty} P(f, \bar{f}) \quad \left\{ 1 - 2 \left(1 - \frac{\varepsilon_q}{3} \right)^f + \left(1 - 2 \frac{\varepsilon_q}{3} \right)^f \right\}$$
(60)

that after a few simple transformations gives

$$\varepsilon = 1 - 2 e^{-\overline{i} \varepsilon_q/3} + e^{-2 \overline{i} \varepsilon_q/3} = (1 - e^{-\overline{n}})^2$$
 (61)

as we wanted to demostrate.

7.5 ADDITION OF TWO COINCIDENCES A_1 A_2 + A_1 A_3

From Table 4 it is obtained

$$X = 1 + (1 - \varepsilon_q)^{f_1} - (1 - \varepsilon_q)^{f - f_1} + (1 - \varepsilon_q)^{f}$$
(62)

and the counting efficiency may be written as

$$\varepsilon = \sum_{f=1}^{\infty} \frac{P(f, \bar{f})}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f!}{f_{1}!f_{2}!(f-f_{1}-f_{2})!} \left\{ 1 - (1 - \varepsilon_{q})^{f_{1}} - (1 - \varepsilon_{q})^{f_{1}} - (1 - \varepsilon_{q})^{f-f_{1}} + (1 - \varepsilon_{q})^{f} \right\}$$

$$(63)$$

and introducing the results (37), (38), (43) and (44) it is obtained:

$$\varepsilon = \sum_{f=1}^{\infty} P_{-}(f, \bar{f}) \quad \left\{ 1 - \left(1 - \frac{\varepsilon_{q}}{3} \right)^{f} - \left(1 - 2\frac{\varepsilon_{q}}{3} \right)^{f} + (1 - \varepsilon_{q}) \right\}$$
(64)

and simplifying, it results:

$$\varepsilon = 1 - e^{-\bar{t} \varepsilon_{q}/3} - e^{-2\bar{t} \varepsilon_{q}/3} + e^{-\bar{t}\varepsilon_{q}} =$$

= 1 - e^{-\bar{n}} - e^{-2\bar{n}} + e^{-3\bar{n}} (65)

as we wanted to demostrate.

7.6 ADDITION OF THREE COINCIDENCES $A_1 A_2 + A_2 A_3 + A_3 A_1$

After a few simplifications it is obtained

$$X = 1 - (1 - \varepsilon_q)^{f_1 + f_2} - (1 - \varepsilon_q)^{f - f_1} - (1 - \varepsilon_q)^{f - f_2} + 2(1 - \varepsilon_q)^{f}$$
(66)

taking this expression to (17) it results

$$\varepsilon = \sum_{f=1}^{\infty} \frac{P(f, \bar{f})}{3^{f}} \sum_{f_{1}=0}^{f} \sum_{f_{2}=0}^{f-f_{1}} \frac{f!}{f_{1}!f_{2}!(f-f_{1}-f_{2})!} \left\{ 1 - (1 - \varepsilon_{q})^{f_{1}+f_{2}} - (1 - \varepsilon_{q})^{f-f_{1}} - (1 - \varepsilon_{q})^{f-f_{2}} + 2(1 - \varepsilon_{q})^{f} \right\}$$
(67)

Taking into account the formulae (37), (40), (42), (43) and (44) we get to

$$\varepsilon = \sum_{f=1}^{\infty} P(f, \bar{f}) \left\{ 1 - 3 \left(1 - 2 \frac{\varepsilon_q}{3} \right)^f + 2 \left(1 - \varepsilon_q \right)^f \right\}$$
(68)

and operating, it results

$$\varepsilon = 1 - 3 \ e^{-2\overline{i} \ \varepsilon_q/3} + 2 \ e^{-\overline{i} \ \varepsilon_q} = 1 - 3 \ e^{-2 \ \overline{n}} + 2 \ e^{-3 \ \overline{n}}$$
(69)

as we wanted to demostrate.

8. CONCLUSIONS

~

The main conclusion from the present work is that the Poissonian and binomial models are equivalent and that the counting efficiencies calculated through either of both models must be the same.

Therefore discrepancies between counting efficiencies for pure beta-ray emitter such as 3 H and 14 C as they are presented in the works (5) and (7) must correspond to differences in the formulae used for calculating the beta-ray espectra or in other case to the error introduced by limiting the sumation from one to infinity in the binomial model.

9. REFERENCES

- [1] HORROCKS, D.L., and STUDIER, M.H. Anal. Chem. <u>33 (1961)</u> 615
- [2] SCHWERDEL, E. Atomkernenergie <u>11</u>(1966) 324
- [3] POCHWALSKI, K. and RADOSZEWSKI, T. Report of Institute Of Nuclear Research, Warsow 1848 /OPIDI/E/A (1979)
- [4] POCHWALSKI, K., BRODA, R. and RADOSZEWSKI, T. Appl. Radiat. Isot. <u>39</u> (1988)165
- [5] GRAU MALONDA, A. and COURSEY, B.M., Appl. Radiat Isot. <u>39</u> (1988) 1191
- [6] GRAU CARLES, A. and GRAU MALONDA, A., Anales de Física B, <u>85</u> (1989) 160
- [7] BRODA, R., POCHWALSKI, K., and RADOSZEWSKI, T., Appl. Radiat. lsot. <u>39</u> (1988) 159
- [8] BIRKS, J.B., "The Theory and Practice of Scintillation Counting". (Pergamon Press, Oxford 1965) p. 185



Fig 1.- Different kinds of coincidence pulses in three photomultiplies system.

Т	A	В	L	Е	1
---	---	---	---	---	---

Counting efficiency formulae for a two photomultipliers system

Signal	Symbol	Total events	Non-detection mode
A or A 2	U	$\sum_{n=1}^{\infty} P(n,m) \ (1-2^{-n})$	1 – exp (– n)
A ₁ +A ₂	S	$\sum_{n=1}^{\infty} P (n, m)$	1 – exp (–2īī)
A ₁ A ₂	к	$\sum_{n=2}^{\infty} P(n, m) (1 - 2^{1-n})$	$[1 - \exp(- \overline{n})]^2$

ΤA	В	LE	2
----	---	----	---

Counting efficiency formulae for a three photomultiplier system

Signal	Symbol	Total events	Non-detection mode
A _i	U	$\sum_{n=1}^{\infty} P(n,m) (1 - 3^{-n} 2^{n})$	1 - exp (- n)
A _j + A _j	s ₂	$\sum_{n=1}^{\infty} P(n,m) (1 - 3^{-n})$	1 - exp (- 2 n)
A1+A2+A3	S ₃	$\sum_{n=1}^{\infty} P(n,m)$	1 - exp(-3īn)
A _i A _j D		$\sum_{n=2}^{\infty} P(n,m) \left[1 - 3^{-n} (2^{n+1} - 1)\right]$	[1 - exp(- n)]2
A _A +AA i j i k	D ₂	$\sum_{n=2}^{\infty} P(n,m) \left[1 - 3^{-n} (2^{n} + 1)\right]$	$1 - \exp(-\overline{n}) - \exp(-2\overline{n}) + \exp(3\overline{n})$
A A + A A	+		
+ ^{+A} 3 ^A 1	D ₃	$\sum_{n=2}^{\infty} P(n,m) (1 - 3^{1-n})$	1−3 exp(−2π) + +2 exp (−3π)
A1A2A3	т	$\sum_{n=3}^{\infty} P(n,m) \left[1 - 3^{1-n} (2^n - 1)\right]$	[1 - exp (- n)]3

TABLE 3

Probability	Х	for a	two	photomultip	liers s	ystem
-------------	---	-------	-----	-------------	---------	-------

Signal	Symbol	X
A ₁ or A ₂	U	P (f ₁)
A ₁ + A ₂	S	$P(f_1) + P(f - f_1) - P(f_1) P(f - f_1)$
A ₁ A ₂	D	P (f ₁) P (f - f ₁)

TABLE 4

Probability X for a three photomultipliers system

Signal	Symbol	X
A _i	U	P (f ₁)
A _i + A _j	S ₂	$P(f_1) + P(f_2) - P(f_1) P(f_2)$
A ₁ + A ₂ +A ₃	s ₃	$\begin{split} & P(\mathfrak{f}_1) + P(\mathfrak{f}_2) + P(\mathfrak{f}_1 - \mathfrak{f}_2) - P(\mathfrak{f}_1) P(\mathfrak{f}_2) - \\ & - P(\mathfrak{f}_2) (P(\mathfrak{f}_1 - \mathfrak{f}_2) - P(\mathfrak{f}_1) P(\mathfrak{f}_1 - \mathfrak{f}_2) + \\ & + P(\mathfrak{f}_1) P(\mathfrak{f}_2) P(\mathfrak{f}_1 - \mathfrak{f}_2) \end{split}$
A _i A _j	D	P(f ₁)P(f ₂)
A _i A _j +A _i A _k	D ₂	$P(f_1) P(f_2)+P(f_1)P(f_1-f_2)-P(f_1)P(f_2)$ $P(f-f_1-f_2)$
A ₁ A ₂ +A ₂ A ₃ +A	A ₃ A ₁ D ₃	$\begin{array}{l} P(\mathfrak{f}_{1}) \;\; P(\mathfrak{f}_{2}) + P(\mathfrak{f}_{2}) P(\mathfrak{f}_{1} - \mathfrak{f}_{2}) + \\ + P(\mathfrak{f}_{1} - \mathfrak{f}_{2}) P(\mathfrak{f}_{1}) - 2 \; P(\mathfrak{f}_{1}) P(\mathfrak{f}_{2}) P(\mathfrak{f}_{1} - \mathfrak{f}_{2}) \end{array}$
$A_1 A_2 A_3$	Т	$P(f_1)P(f_2)P(f_1-f_2)$

CTEMAT-677 CIEMAT-677 Centro de Investigaciones Energéticas, Medicambientales y Tecnológicas. Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas. Instituto de Investigación Básica, Madrid. Instituto de Investigación Básica, Madrid. "Models Poissoniano y binomial en metrología de radio-"Models Poissoniano y binomial en metrología de radionucleídos por centelleo líquido". nucleídos por centelleo líquido". GRAU MALONDA, A. (1991) 23 pp., 1 fig., 4 tabl., 3 refs. En el presente trabajo se analizan los modelos poissoniano y binomial desarrollados GRAU MALONDA, A. (1991) 23 pp., 1 fig., 4 tabl., 3 refs. para calcular la eficiencia de recuento a partir de un parametro libre. Se demuestra En el presente trabajo se analizan los modelos poissoniano y binomial desarrollados matemáticamente que ambos modelos son equivalentes y que las efficiencias de recuento para calcular la eficiencia de recuento i partir de un parametro libre. Se demuestra calculadas a partir de uno u otro modelo son idénticas. matemáticamente que ambos modelos son equivalentes y que las eficiencias le recuento calculadas a partir de uno u otro modelo son idénticas. CLASIFICACION DOE Y DESCRIPTORES: 440102. Jointillation Counting. Efficiency. Coincidence Methods. Photomultipliers. distribution Functions. Mathematical Models. Compara CLASIFICACION DOE Y DESCRIPTORES: 440102. Scintillation Counting. Efficiency. Coincitive Evaluations. ience Methods. Photomultipliers. distribution Functions. Mathematical Models. Compara tive Evaluations. CIEMAT-677 CIEMAT-677 Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas. Centro de Investigaciones Energéticas, Medicambientales y Tecnológicas. Instituto de Investigación Básica, Madrid. Instituto de Investigación Básica. Madrid. "Models Poissoniano y binomial en metrología de radio-"Models Poissoniano y binomial en metrología de radionucleídos por centelleo líquido". nucleídos por centelleo líquido". GRAU MALONDA, A. (1991) 23 pp., ! fig., 4 tapl., 3 refs. GRAU MALONDA, A. (1991) 23 pp., 1 fig., 4 tabl., 3 refs. En el presente trabajo se analizan los modelos poissoniano y binomial desarrollados En el presente trabajo se analizan los modelos poissoniano y binomial desarrollados para calcular la eficiencia de recuento a partir de un parámetro libre. Se demuestra para calcular la eficiencia de recuento a partir de un parámetro libre. Se demuestra matemáticamente que ambos modelos son equivalentes y que las eficiencias de recuento matemáticamente que ambos modelos son equivalentes γ que las eficiencias de recuento calculadas a partir de uno u otro modelo son idénticas. calculadas a partir de uno u otro modelo son idénticas. CLASIFICACION DOE / DESCRIPTORES: 440102. Scintillation Counting. Efficiency. Coinci-CLASIFICACION DOE Y DESCRIPTORES: 440102. Scintillation Counting. Efficiency. Coincidence Methods. Photomultipliers. distribution Functions. Mathematical Models. Compara dence Methods. Photomultipliers, distribution Functions, Mathematical Models, Compara tive Evaluations. tive Evaluations.

	\ \
CIEMAT-677 Centro le Investigaciones Energéticas, Medioambientales y Tecnológicas. Instituto le Investigación Básica. Madrid. "Poissonian and binomial models in radionuclide metro- logy by liquid scintillation counting".	CIEMAT-677 Centro de Investigaciones Energéticas, Medioampientales y Tecnológicas. Instituto de Investigación Básica. Madrid. "Poissonian and binomial models in radionuclide metro- logy by liquid scintillation counting". ,
 GRAU MALONDA, A. (1991) 23 pp., 1 fig., 4 tabl., 3 refs. Binomial and Poissonian models developed for calculating the counting efficiency from a free parameter is analysed in this paper. This model have been applied to liquid scintillator counting systems with two or three photomultipliers. It is mathematically demostrated that both models are equivalent and that the rounting efficiencies calculated either from one or the other model are identical. DOE CLASSIFICATION AND DESCRIPTORS: 440102. Scintillation Counting Efficiency. Coincidence Methods. Photomultipliers. Distribution Functions. Mathematical Models. Comparative Evaluations. 	 GRAU MALONDA, A. (1991) 23 pp., 1 fig., 4 tabl., 3 refs. Binomial and Poissonian models developed for calculating the counting efficiency from a free parameter is inalysed in this paper. This model have been upplied to liquid scintillator counting systems with two or three photomultipliers. It is mathematically demostrated that both models are equivalent and that the counting efficiencies calculated either from one or the other model are identical. DOE CLASSIFICATION AND DESCRIPTORS: 440102. Scintillation Counting. Efficiency. Coincidence Methods. Photomultipliers. Distribution Punctions. Mathematical Models. Comparative Evaluations.
CIEMAT-677 Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas. Instituto de Investigación Básica. Madrid. "Poissonian and binomial models in radionuclide metro- logy by liquid scintillation counting".	CIEMAT-677 Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas. Instituto de Investigación Básica. Madrid. "Poissonian and binomial models in radionuclide metro- logy by liquid scintillation counting".
 GRAU MALONDA, A. (1991) 23 pp., 1 fig., 4 tabl., 3 refs. Binomial and Poissonian models developed for calculating the counting efficiency from a free parameter is analysed in this paper. This model have been applied to liquid scintillator counting systems with two or three photomultipliers. It is mathematically demostrated that both models are equivalent and that the counting efficiencies calculated either from one or the other model are identical. DOE CLASSIFICATION AND DESCRIPTORS: 440102. Scintillation Counting. Efficiency. Coincidence Methods. Photomultipliers. Distribution Functions. Mathematical Models. Comparative Evaluations. 	 GRAU MALONDA, A. (1991) 23 pp., 1 fig., 4 tabl., 8 refs. Binomial and Poissonian models developed for calculating the counting efficiency from a free parameter is analysed in this paper. This model have been applied to liquid scintillator counting systems with two or three photomultipliers. It is mathematically demostrated that both models are equivalent and that the counting efficiencies calculated either from one or the other model are identical. DOE CLASSIFICATION AND DESCRIPTORS: 440102. Scintillation Counting. Efficiency. Coincidence Methods. Photomultipliers. Distribution Functions. Mathematical Models.

_