

LS Spectrometry

(or maybe just counting)

via CIEMAT-NIST efficiency tracing (CNET)

Two amusing quotes

“NIST -- one of premier experimental LS groups in the world”

- ◆ CIEMAT/NIST method co-originator
- ◆ cocktail composition effects

“A principal and powerful tool (“workhorse”) of the Radioactivity Group”

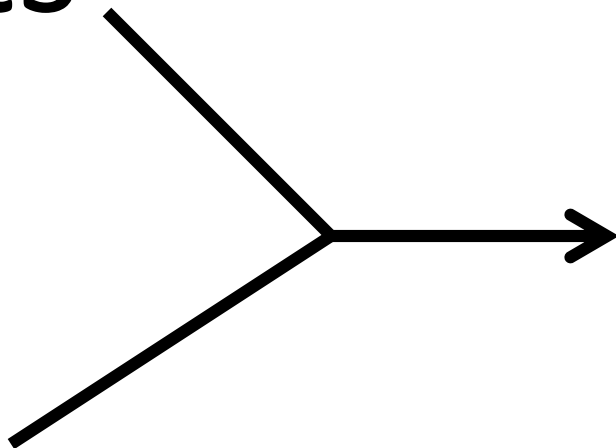
R. Collé

NIST Radioactivity Group TALK

10 July 2013

counts

time



Bq

Measure of radioactivity

Number (dN) of spontaneous nuclear transformations per unit time interval (dt)

$$-dN/dt = N\lambda$$

$$[\lambda = \ln(2)/T_{1/2}]$$

SI unit

$$s^{-1} = \text{Bq}$$

A counting process (of emitted radiations)

- ◆ “absolute”; primary; fundamental
- ◆ relative; secondary

detection efficiency concept in both

Typical measurement model

$$R(t) = C/T$$

$$= R_B + A_0 (m/M) \varepsilon \Gamma G(t) f_i f_j \dots + A_I \varepsilon_I \dots$$



Scintillation Counting

1

Liquid Scintillation Cocktail

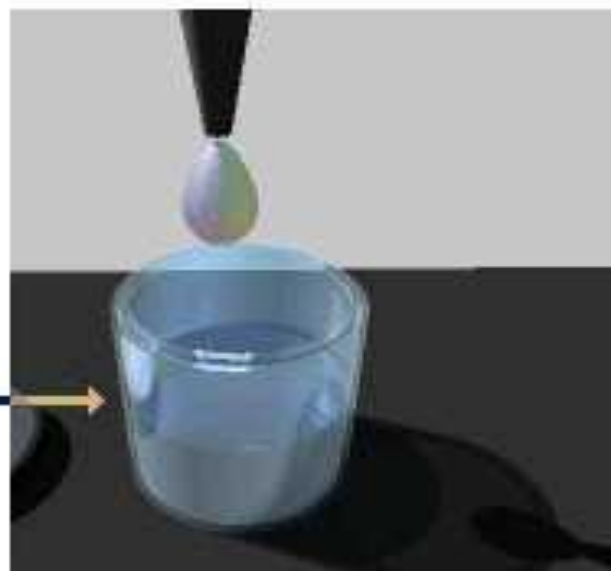
Components:

Solvent: Typically toluene, xylene, pseudocumene, or an alkyl benzene type solvent.

Emulsifier: A detergent type molecule that ensures proper mixing of aqueous samples.

Fluor: A fluorescent solute.

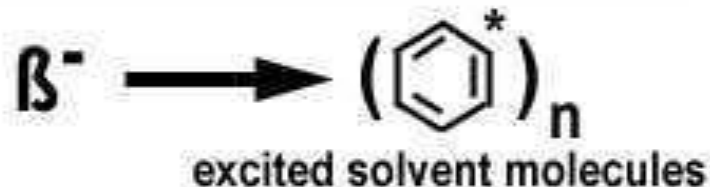
Process: Radioactive Sample is added to scintillation cocktail.



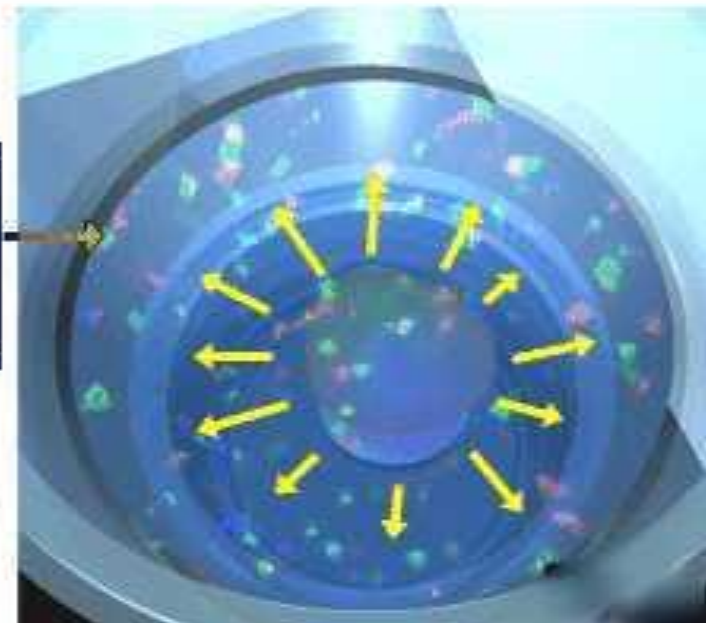
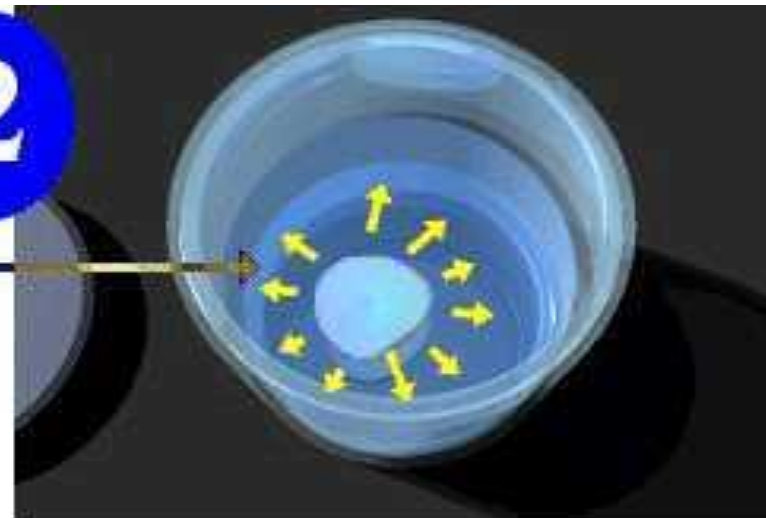
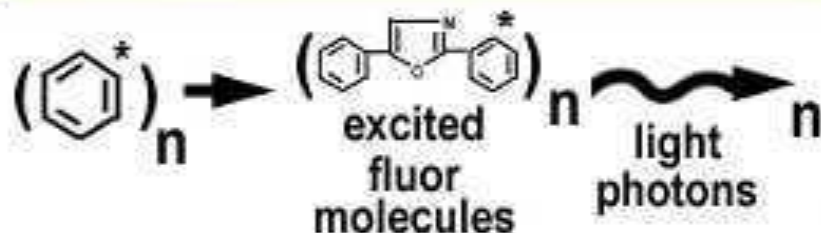
Scintillation Counting

2

Beta particles are emitted, which cause solvent molecules to become excited.

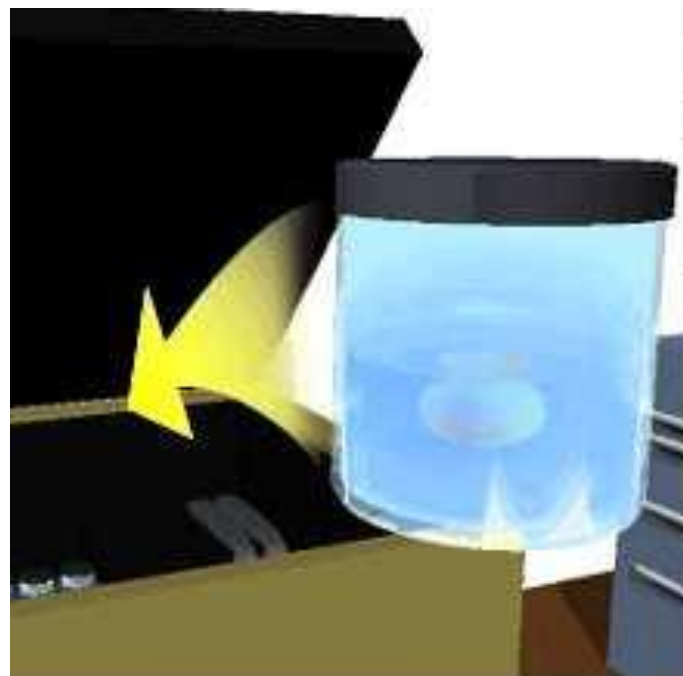


The energy of the solvent molecules is transferred to the fluor molecules, which in turn emit light.



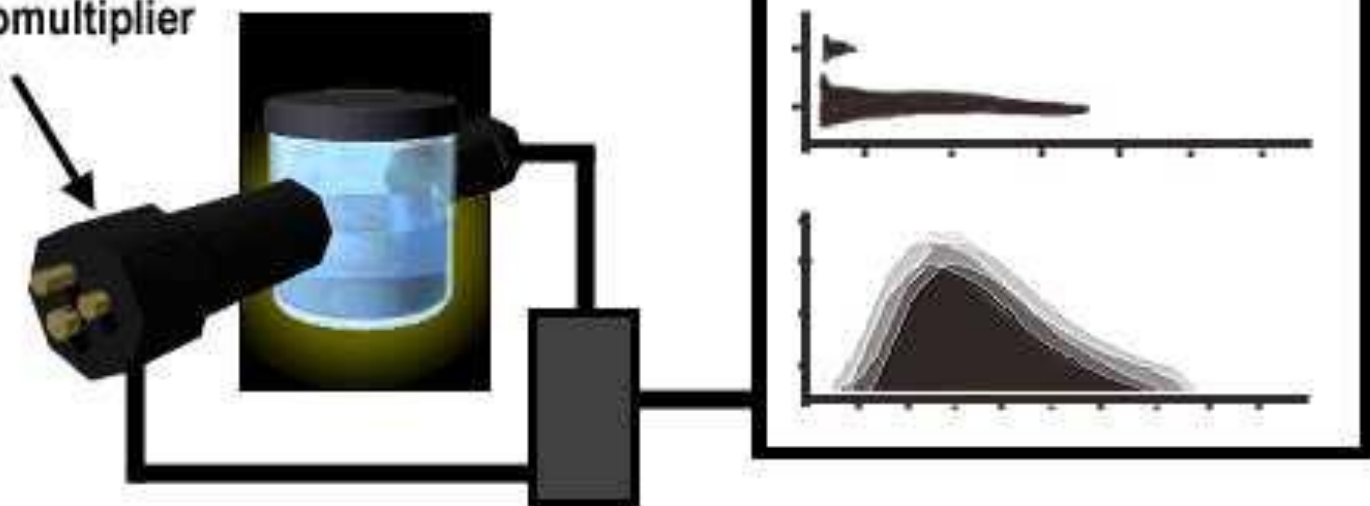
Scintillation Counting

3

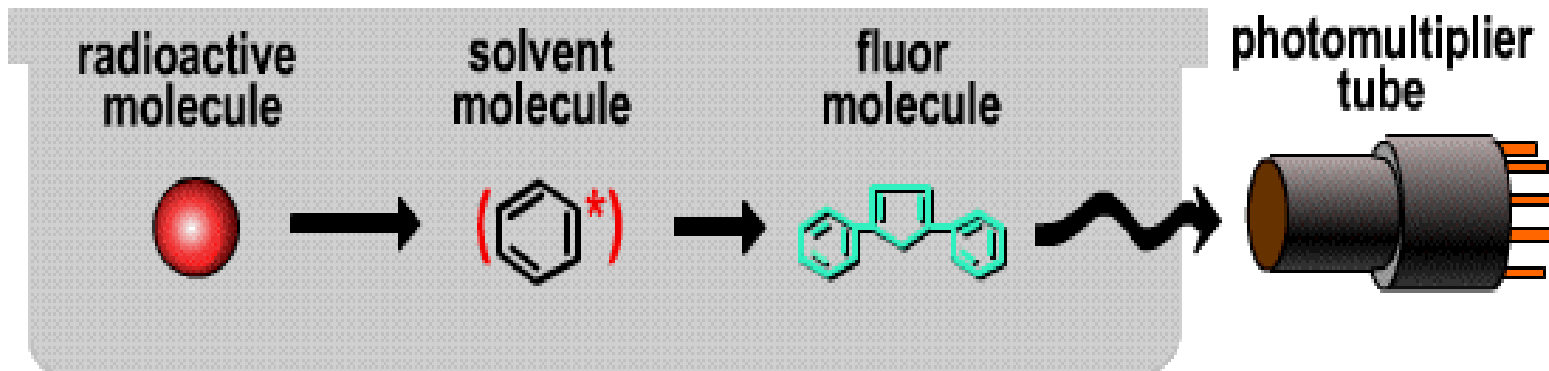


The jar is placed inside a scintillation counter, which captures and digitizes the light photons.

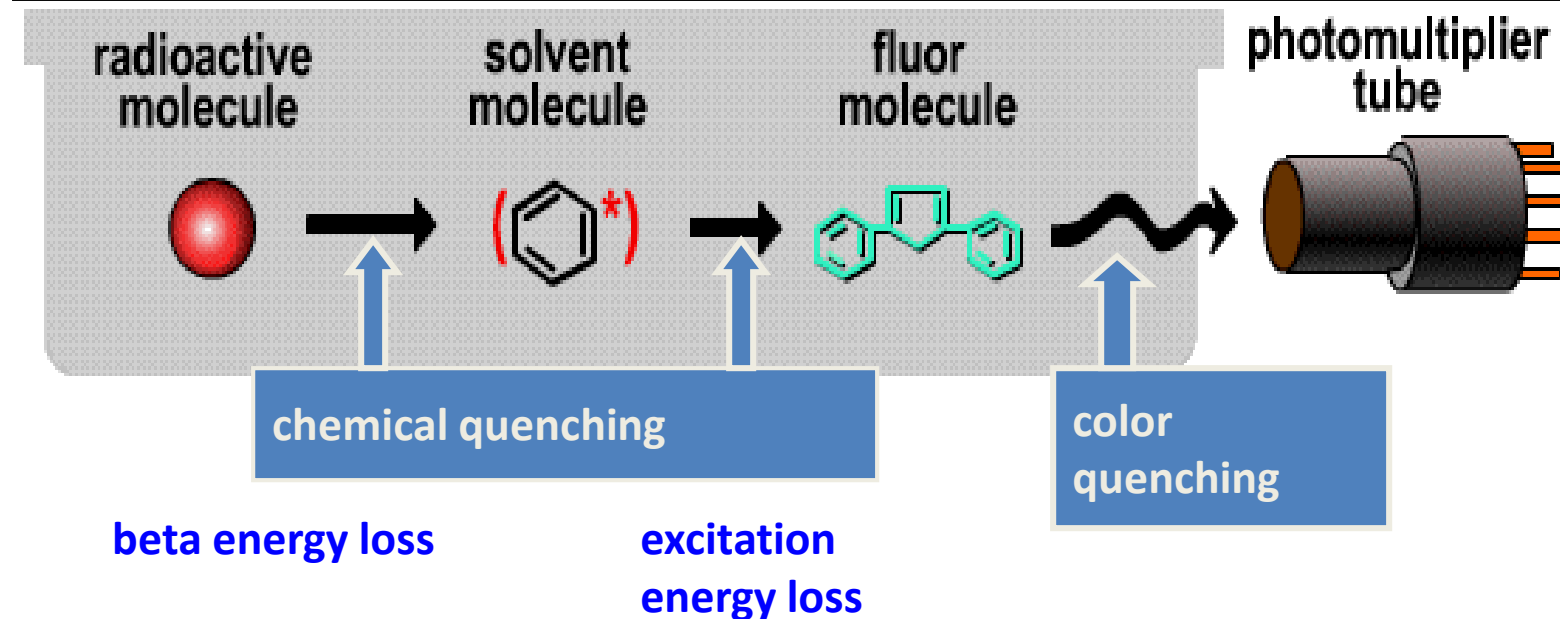
photomultiplier
tube



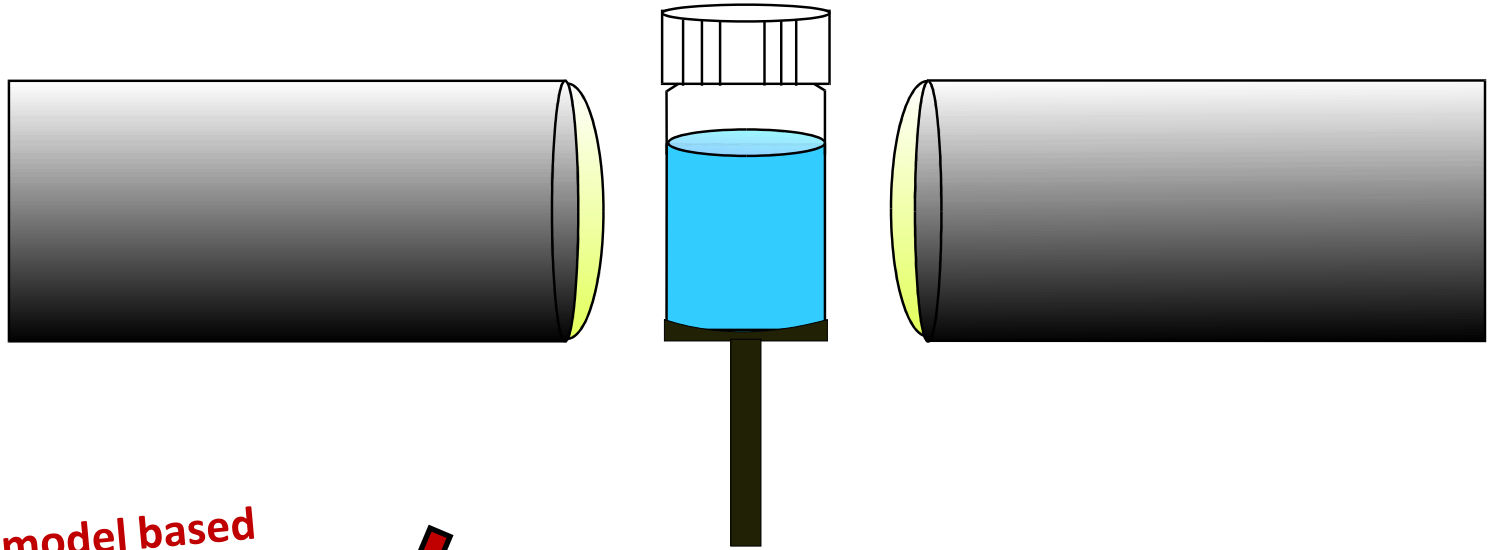
The Scintillation Process



The Quenching Process



Two-Phototube Coincidence Liquid Scintillation Counting with the CIEMAT/NIST Efficiency Tracing Method



TOTALLY model based

$$\varepsilon = \int_0^{E_{\max}} \left\{ 1 - \exp \left[-M^{-1}EQ(E)W(E) \right] \right\}^2 P(Z,E) dE \times \left(\int_0^{E_{\max}} P(Z,E) dE \right)^{-1}$$

see Zimmerman & Colle, *J. Res. NIST*, **102**, 455 (1997)

overall efficiency

$$\varepsilon = \int_0^{E_{\max}} \left\{ 1 - \exp \left[-M^{-1} E \underline{Q(E)} \underline{W(E)} \right] \right\}^2 \underline{P(Z, E)} dE \times \left(\int_0^{E_{\max}} P(Z, E) dE \right)^{-1}$$

fractional energy losses
inside (Q) outside (W)

$Q = f(kB, dE/dx)$

Fermi function (normalized)

energy of decay event

M = "figure of merit" – once said to represent the "energy (in keV) required to produce one photoelectron at 1st dynode of PMT" -- used to develop the model

efficiency at one E integrated over entire spectrum

THIS ONLY APPLIES TO EVENTS ABOVE DETECTION THRESHOLD. TO INCLUDE EVENTS BELOW, MUST EXTRAPOLATE TO ZERO THRESHOLD – ACHIEVED WITH CNET

Comes from :

average number photoelectrons at 1st dynode
 $\check{n} = M^{-1} E Q W$

Poisson probability of detecting x from mean \check{n}
 $P(x, n) = (n^x / x!) / \exp(-\check{n})$

Probability of detecting zero (non-detection)
 $P(0, \check{n}) = \exp(-\check{n})$

two phototubes in coincidence
 $\check{\varepsilon}(E) = [1 - \exp(-\check{n})]^2$

Integrate over all energies to get ε

HOW TO DO

(experimentally)

Prepare series of ^3H
cocktails with known
activity (from
standard)

Prepare series of ^{00}X
cocktails

use perfectly
matched
compositions

quench matched cocktails with
imposed chemical agent to vary
efficiency over a "large" range

measure both
 ^3H and ^{00}X series
in LS counter
(few times)

Obtain

count rate

R_3

R_x

QIP

(quench
indicating
parameter)

Q_3

Q_x

do multiple compositions using
more than one scintillation fluid

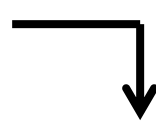
use multiple counters
(different operating characteristics)

some compatible; some not (see in
results)

final results for traced activity should be invariant of
experimental conditions: measurement occasion
(time), activity level, cocktail composition,
quenching, detection efficiency, LS counter, etc

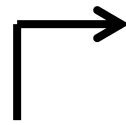
*Many codes
available for
different
applications*

nuclear data for
nuclide ^{00}X
decay scheme



la boîte noire

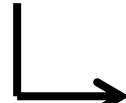
$$\varepsilon_x = f(M_x)$$



nuclear data for
 ^3H decay scheme



$$\varepsilon_3 = f(M_3)$$



leap of faith

$$M_x = M_3$$

DATA (discrete)

$$R_3 = f(Q_3)$$
$$R_x = f(Q_x)$$

CALCULATION (step sizes)

$$\varepsilon_3 = f(M_3)$$
$$\varepsilon_x = f(M_x)$$

GET
 $\varepsilon_3 = R_3 / A_3(\text{std})$

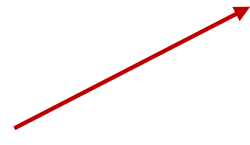
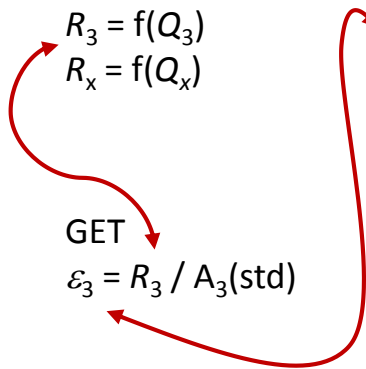
ELIMINATE ε_3 VARIABLE
to obtain
 $M_3 = f(Q_3)$

WITH $M_x = M_3$
have $M_x = f(Q_x)$

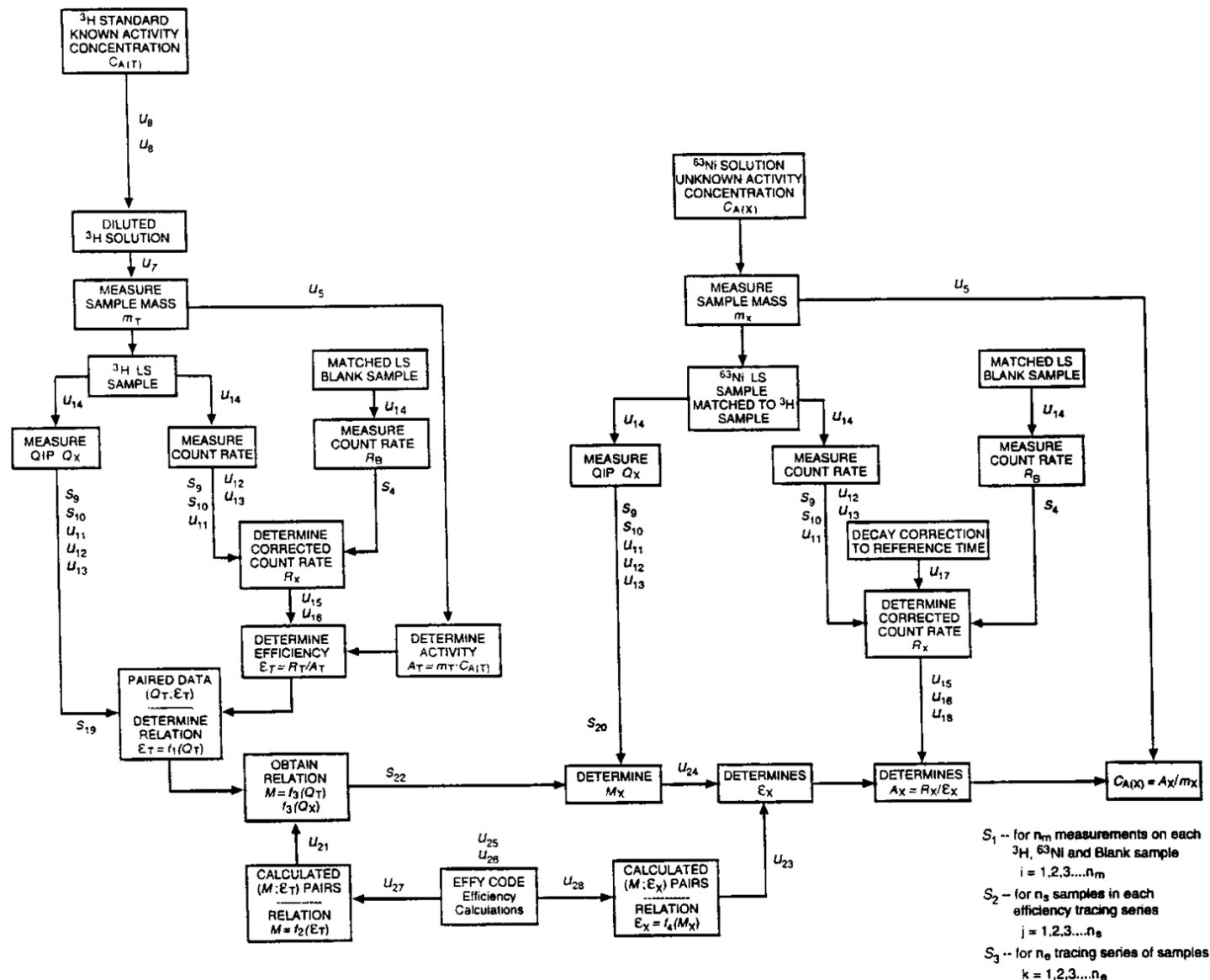
WITH M_x & R_x
get A_x

voilà

measured

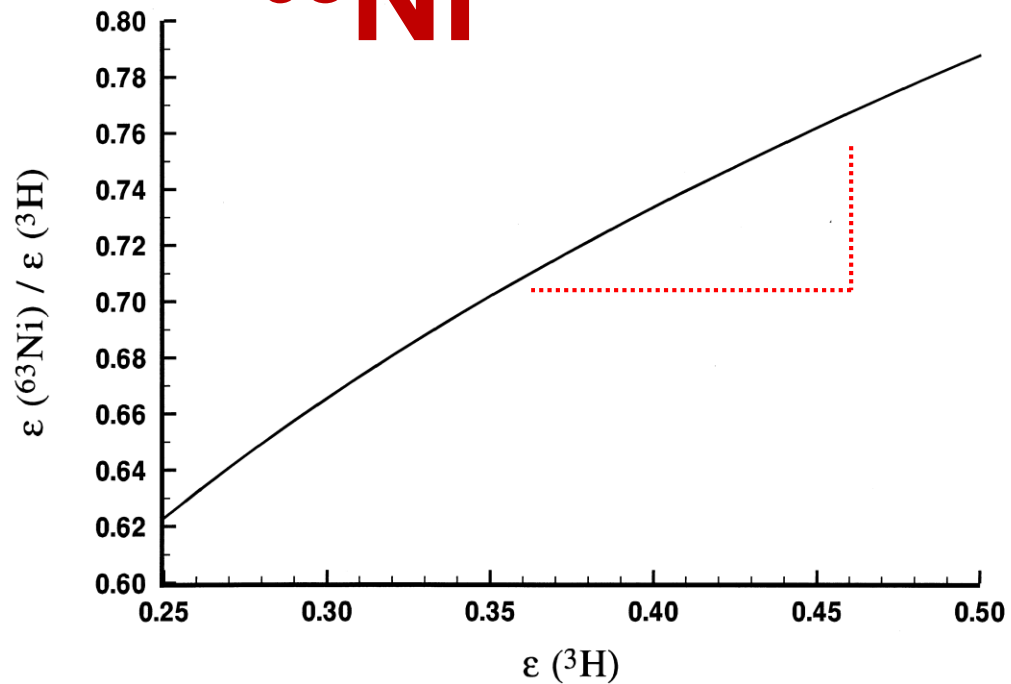
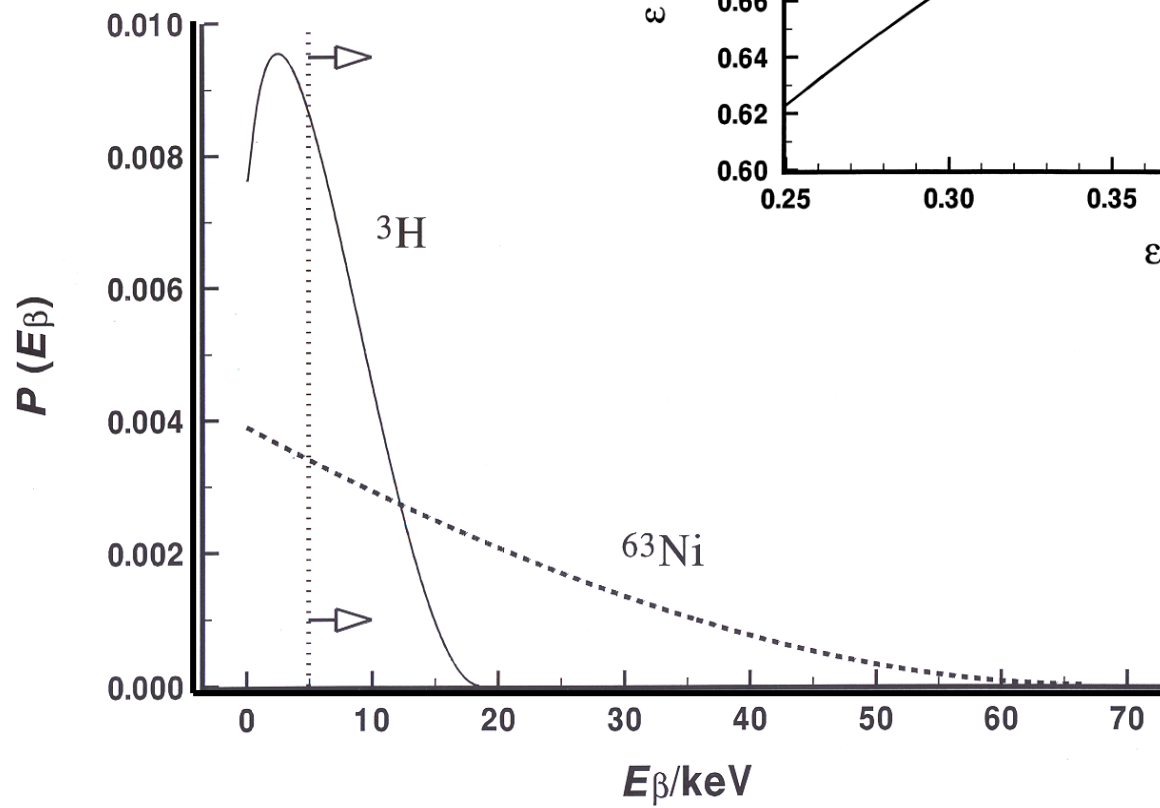


CIEMAT/NIST method -- measurement & uncertainty model

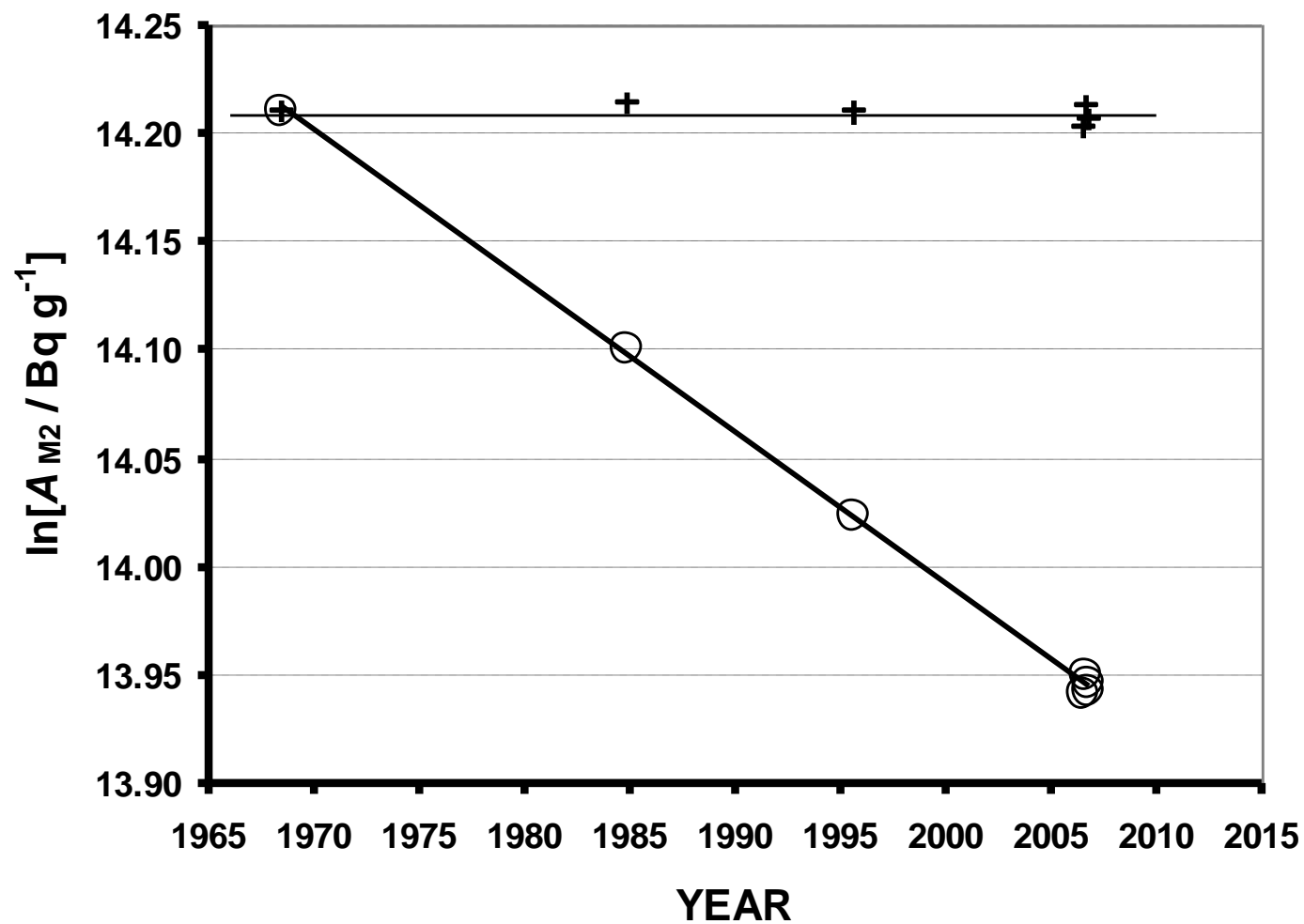


^{63}Ni

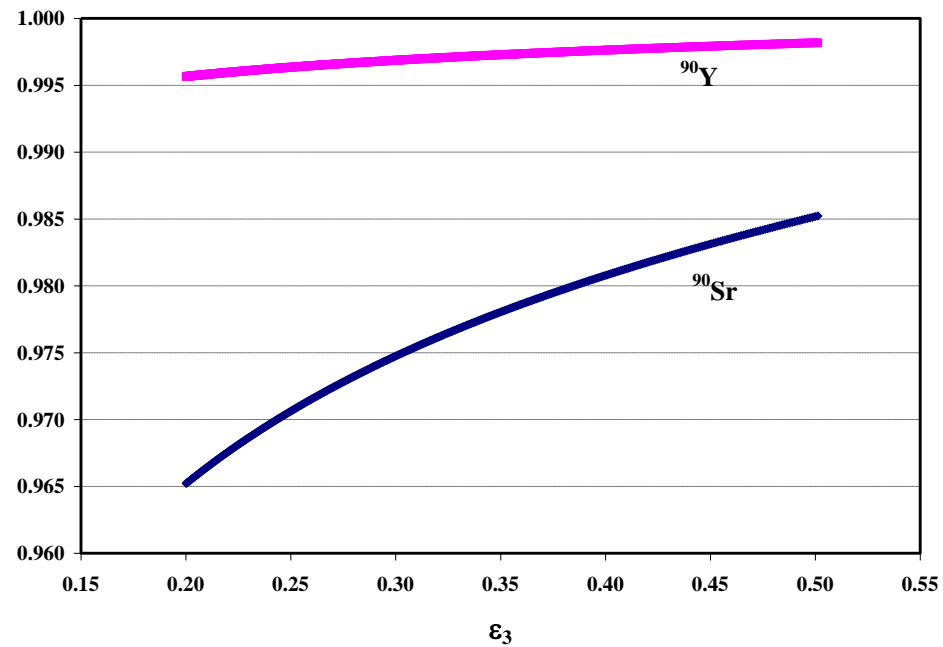
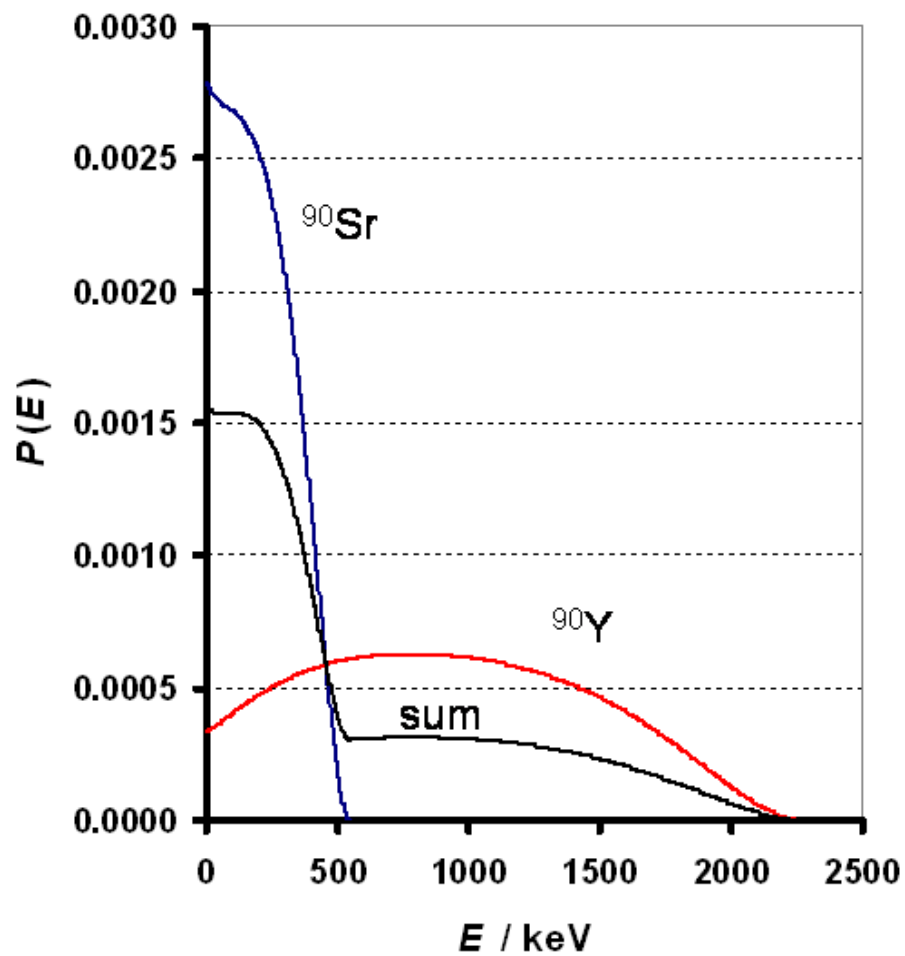
5 keV



^{63}Ni



fairly easy case



$^{90}\text{Sr} - ^{90}\text{Y}$

Series	Instrument	Age / d	Nd	ϵ_{H-3}	ϵ_{Sr-90}	ϵ_{Y-90}	Average/ kBq•g ⁻¹	s / %
I	B	2	35	0.29 - 0.43	0.974 - 0.983	0.997 - 0.998	31.82	0.02
	P	6	21	0.23 - 0.35	0.968 - 0.979	0.996 - 0.997	31.77	0.02
	W	9	21	0.23 - 0.35	0.968 - 0.979	0.996 - 0.998	31.78	0.02
	B	14	21	0.30 - 0.43	0.974 - 0.982	0.997 - 0.998	31.83	0.02
II	B	2	35	0.28 - 0.41	0.973 - 0.981	0.997 - 0.998	31.82	0.03
	P	6	21	0.21 - 0.33	0.966 - 0.977	0.996 - 0.997	31.78	0.03
	W	9	21	0.21 - 0.33	0.966 - 0.977	0.996 - 0.998	31.78	0.04
	B	14	21	0.28 - 0.40	0.972 - 0.981	0.997 - 0.998	31.81	0.04
III	B	2	35	0.28 - 0.42	0.973 - 0.982	0.997 - 0.998	31.79	0.04
	P	6	21	0.21 - 0.34	0.966 - 0.977	0.996 - 0.997	31.73	0.01
	W	9	21	0.21 - 0.34	0.966 - 0.977	0.996 - 0.998	31.75	0.05
	B	14	21	0.28 - 0.41	0.972 - 0.982	0.997 - 0.998	31.81	0.02
All	Unweighted grand mean						31.79	--
All	relative standard deviation of mean (n=12)						--	0.10

Comparison of the NIST and NPL ^{210}Pb standards by five measurement methods.

CNET 

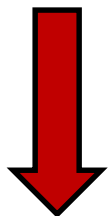
method	NPL / NIST ratio	relative standard uncertainty
NPL and NIST certified values from primary standardizations	0.037484	1.5 %
$4\pi\gamma(\text{NaI})$	0.037373	0.56 %
HPGe spectrometry.	0.036542	0.71 %
$4\pi\alpha\beta(\text{LS})$	0.037249	0.17 %
^{210}Po assay ($2\pi\alpha$ spect.)	0.03736	0.75 %
Si(Li) low-energy spectrometry	0.0381	1.9 %

^{210}Pb

-- harder (1^{st} time)

Pu-241 Results (21 keV β ; ^{241}Am α daughter)

details



LS CNET

Measurement Method	Activity (Bq•g ⁻¹)	Uncertainty (%)
LS CNET	258.5	0.5+
TDCR (NIST)	239.6	2.1
TDCR (LNHB)	240.1	1.3

- 8 %

312 determinations; variables include:

- 3 counters
- 3 compositions
- 5 to 6 sources per composition
- 2 activity levels/solutions
- 2 to 10 cycles / 60 to 100 minutes per measurement
- 69 days of aging

241Pu

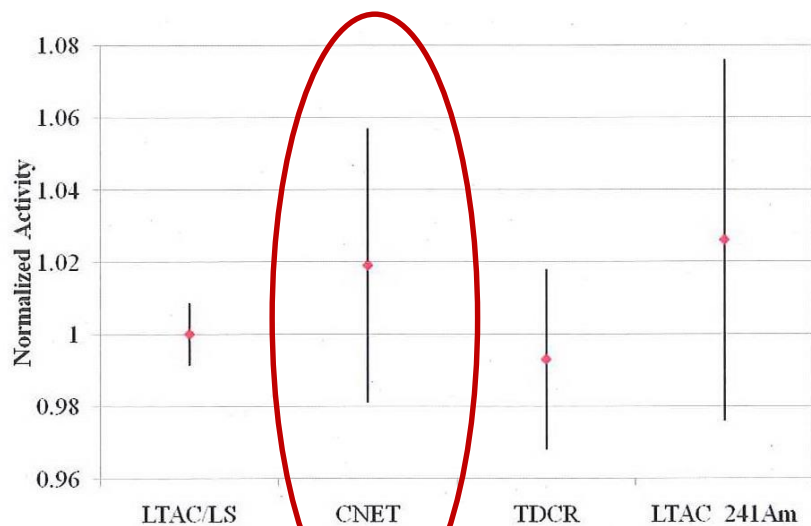
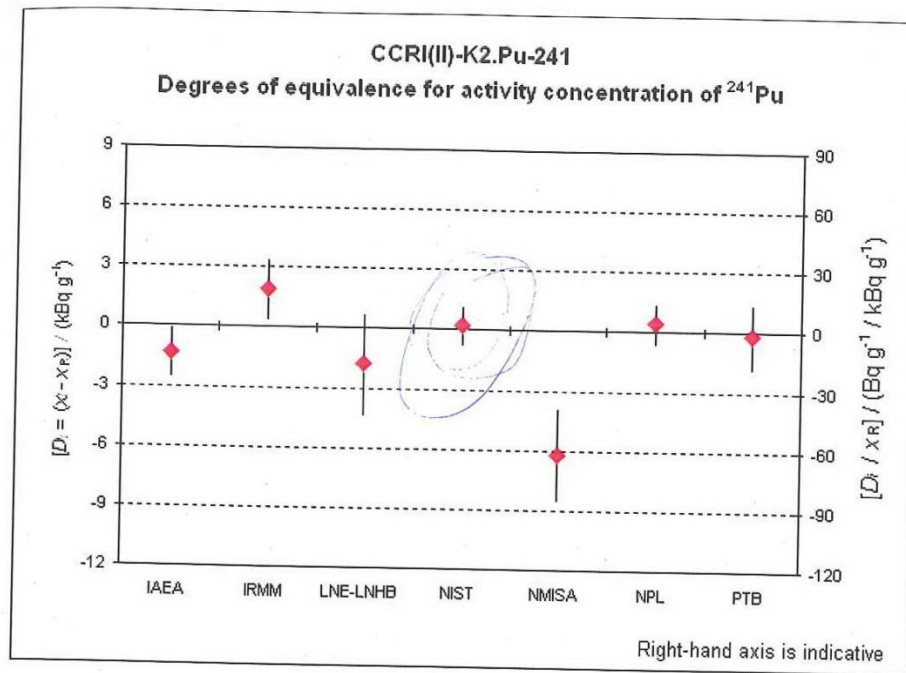


Figure 3 Normalized ^{241}Pu activity in the NPL solution and its respective uncertainty as determined by each method.

Figure 7. Graph of degrees of equivalence with the KCRV for ^{241}Pu (as it appears in the KCDB of the CIPM MRA)



Summary of some recent NIST primary standardizations and comparison to confirmatory measurements.

Nuclide	Method	relative standard uncertainty	Confirmatory Measurement	Difference (%)
⁶³ Ni	4 π LS TDCR (NIST)	0.16 %	4 $\pi\beta$ LS TDCR (LNHB) 4 $\pi\beta$ LS CNET (NIST)	-0.31 -0.77
⁵⁵ Fe (NIST)	4 π calorimetry (linked by LS)	0.39 %	4 π LS TDCR (Polatom) 4 π LS TDCR (LNHB)	-0.87 -0.43
⁵⁵ Fe (BIPM)	4 π calorimetry (linked by LS)	0.39 %	weighted mean value of 15 NMI labs	-0.37
²¹⁰ Pb	4 $\pi\alpha\beta$ LS CNET	1.2 %	4 $\pi\alpha\beta$ (LS)- γ (NaI) anticoin. counting 210Po α spect. (102 a ²⁰⁹ Po tracer) 210Po α spect. (128 a ²⁰⁹ Po tracer) HPGe photon spect.	+0.7 -3.0 -1.3 +4.7
²⁴¹ Pu	4 $\pi\beta$ LS CNET	1.9 %	LS (²⁴¹ Am ingrowth) 4 $\pi\beta$ LS TDCR (NIST) 4 $\pi\beta$ LS TDCR (LNHB)	+1.2 -7.9 * -7.7 *
²¹⁰ Pb	4 $\pi\alpha\beta$ LS CNET	1.2 %	compare to NPL standard (5 methods) see Table2	-0.3
⁹⁰ Sr	4 $\pi\beta$ LS TDCR	0.51 %	4 $\pi\beta$ LS CNET	+ 0.09
²⁴¹ Am	4 $\pi\alpha$ LS	0.22 %	4 $\pi\alpha$ LS (independent) 4 $\pi\alpha$ LS (independent)	-0.05 -0.01 -0.15
²²⁹ Th	4 $\pi\alpha\beta$ (LS)- γ (NaI) anticoincidence counting	0.28 %	4 $\pi\alpha\beta$ LS CNET 4 $\pi\alpha\beta$ LS TDCR 2 π α proportional counting HPGe photon spectrometry	-0.09 -1.7 -0.09 +2.1

* Values are discrepant, and not considered to have confirmed.

In “good” practice:

match cocktail compositions

so that both nuclide and ^3H are in as identical of an environment for chemical quenching as possible

use more than one LS cocktail composition, including at least two different scintillation fluids

to obviate (or account) for chemical composition effects

use two or more different counters

having differing characteristics: log vs. linear amplification; detection thresholds; dead times; etc.

use a wide quenching / efficiency range

so extrapolated result is efficiency independent

measure on different measurement occasions over a range of cocktail ages

try to evaluate dependence of chosen code on the result, as well as effect of the input parameters and assumptions