

Radioactivity Group Meeting

9 January 2008

Advances in the SRM Program + some metrology

Brought to you by

Ron Collé

Lizbeth Laureano-Perez

Includes major contributions by

Ryan Fitzgerald

lisa Outola

Brian Zimmerman

And some minor ones by

Dan Golas

Jerry LaRosa

Leticia Pibida

Evan Crawford

Ken Inn

Lynne King

Nicholas Perichon

Bruce Norman

ASK HOW YOU TOO CAN BE INCLUDED IN NEXT YEAR'S LIST !

Last time (29 march 2007) we talked about

new SRM production & standardization

^{60}Co ^{137}Cs ^{55}Fe ^{210}Pb $^{166\text{m}}\text{Ho}$ $^{90}\text{Sr}(2)$

new SRMS of others --nat'l matrix (2); ^{226}Ra -Rn (6)

re-certifications (w & w/o meas.)

ongoing & future SRMs

^{55}Fe std. + BIPM intercomparison (link to calorimetry)

^{209}Po / ^{210}Pb problems ← papers

^{209}Po half-life (Poland, France, NIST ?) ← died

^{63}Ni standardization & half-life --38 years (LNHB) ← paper

calorimetry / ^{14}C half-life / (Columbia Univ.) ← died

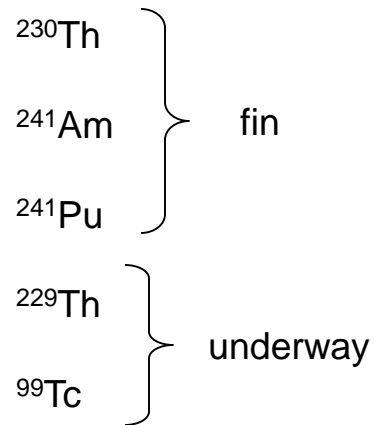
Si(Li) x-ray detection ← It's here – no time !

measurement comparison compilations ← lots more

completed
work

New stuff

Standardizations & SRMs



NEXT (approx. order)

^{228}Ra

^{239}Pu

^{209}Po

^{244}Cm

^{63}Ni

^{238}Pu

^{243}Am

Other projects:

LS α wall effect studies (w/ Ryan)

NPL / NIST ^{210}Pb std. comparison

Si(Li) detector system (w/ Brian)

cryogenic cal. α spect. (collaboration with Boulder / Los Alamos / LNHB)

measurement comparisons (w/ Ryan, Brian, lisa, others)

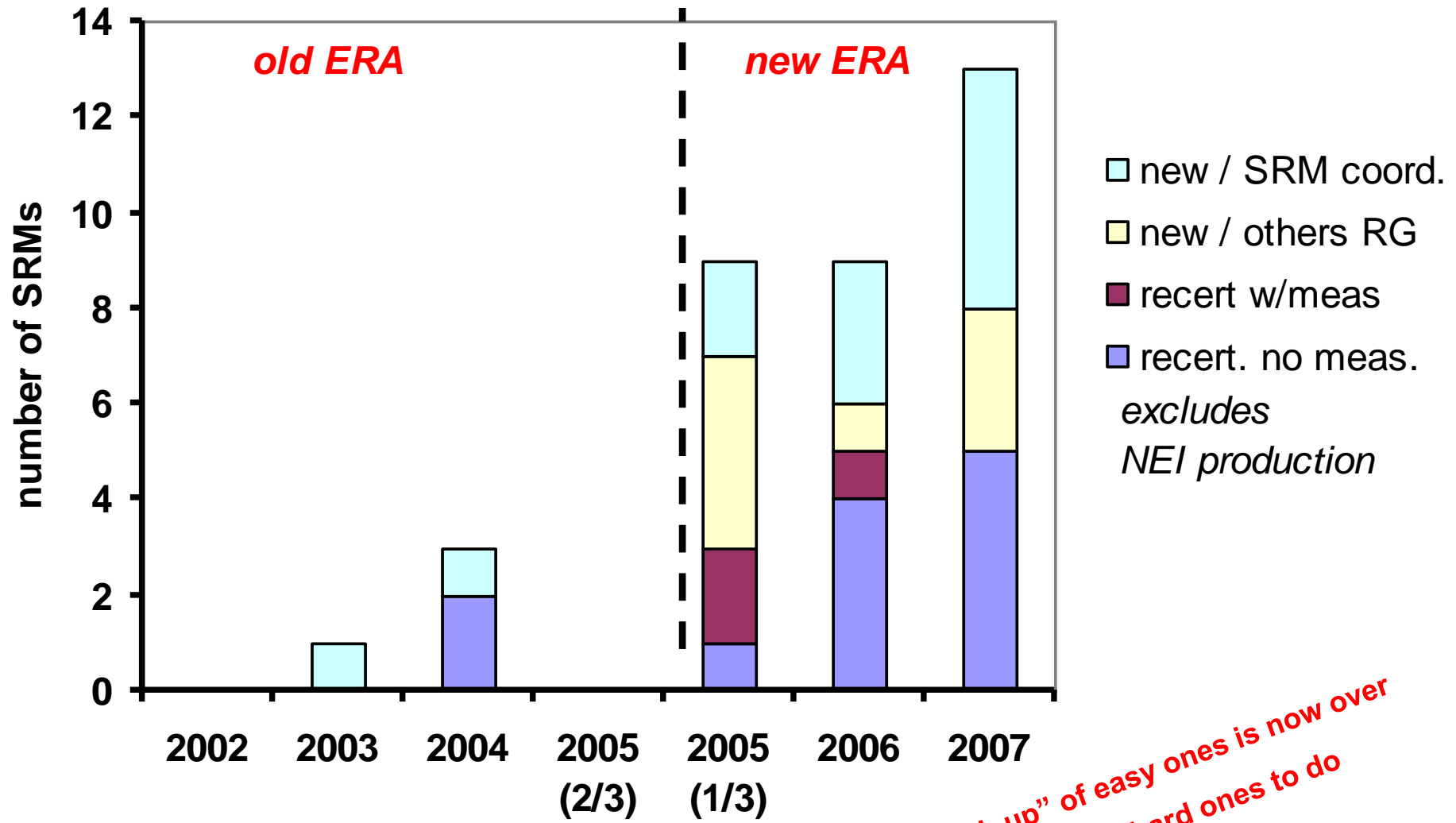
^{241}Am links – 4 std solutions – LS & anticon.

^{241}Pu LS CNET & TDCR / NIST & LNHB / ^{241}Am ingrowth

^{229}Th – LS – CNET & TDCR & anticon. – LNHB, Los alamos

^{99}Tc – CNET & TDCR & anticon.

SRMs to Inventory



**“catch up” of easy ones is now over
many hard ones to do**

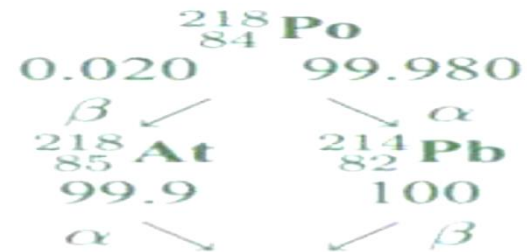
Lizabeth

^{230}Th

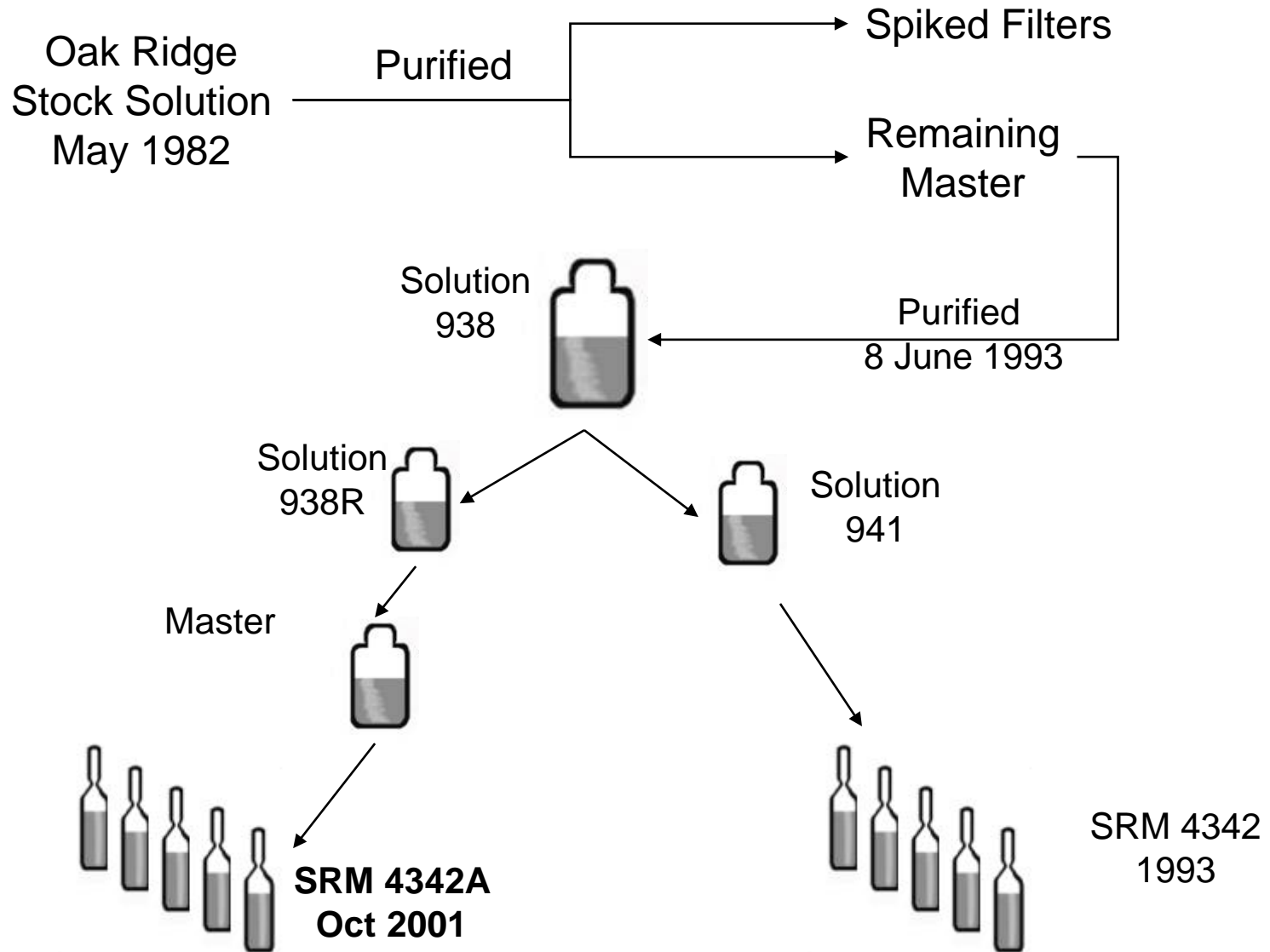
^{241}Am

^{241}Pu

Th-230



Thorium-230



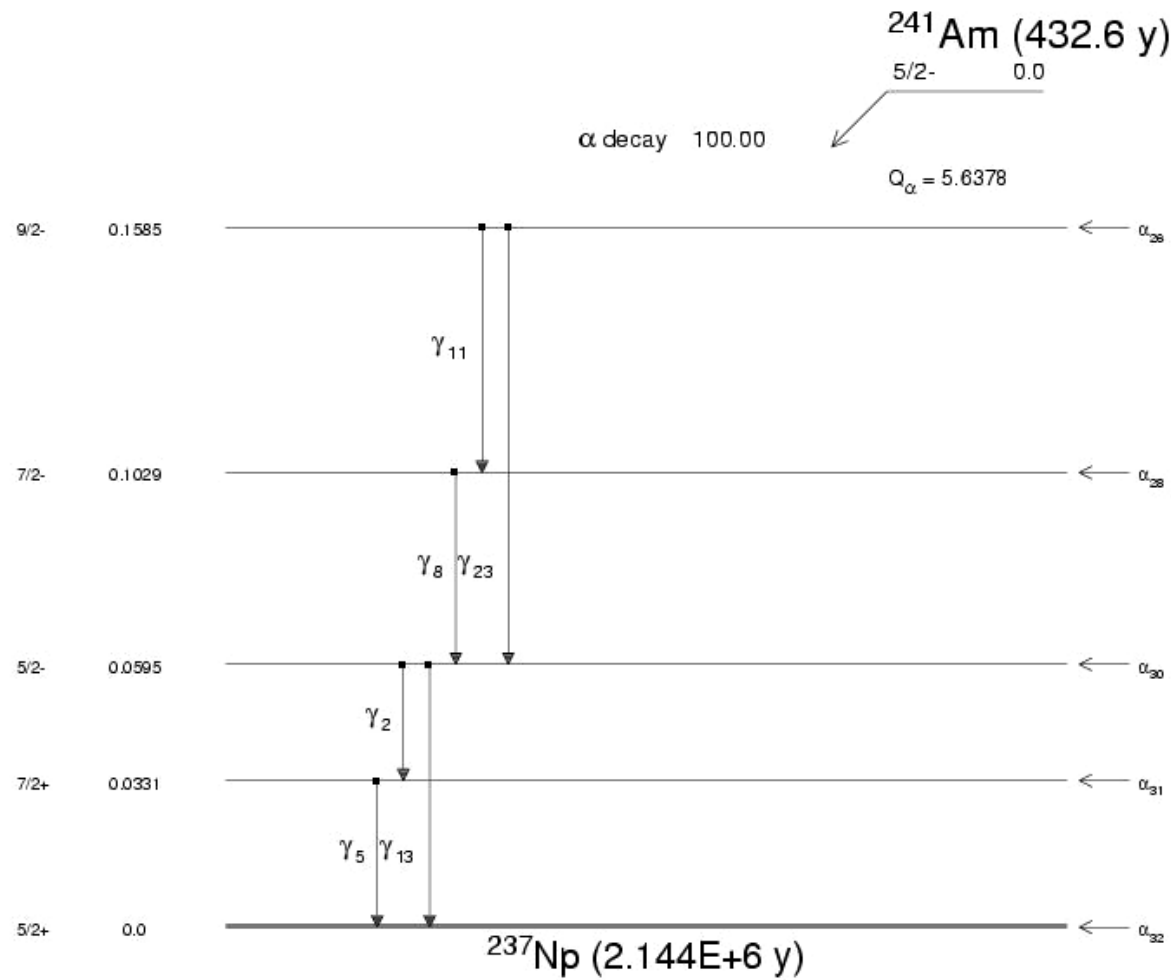
Th-230

- LS Measurement
 - Corrected for Ra-226 subseries ingrowth
- 69 determinations; variables include:
 - 3 counters
 - 1 composition
 - 3 sources per composition
 - 1 to 5 cycles / 60 to 360 minutes per measurement
 - 5 - 40 days of aging
- Impurities
 - Alpha Spectrometry

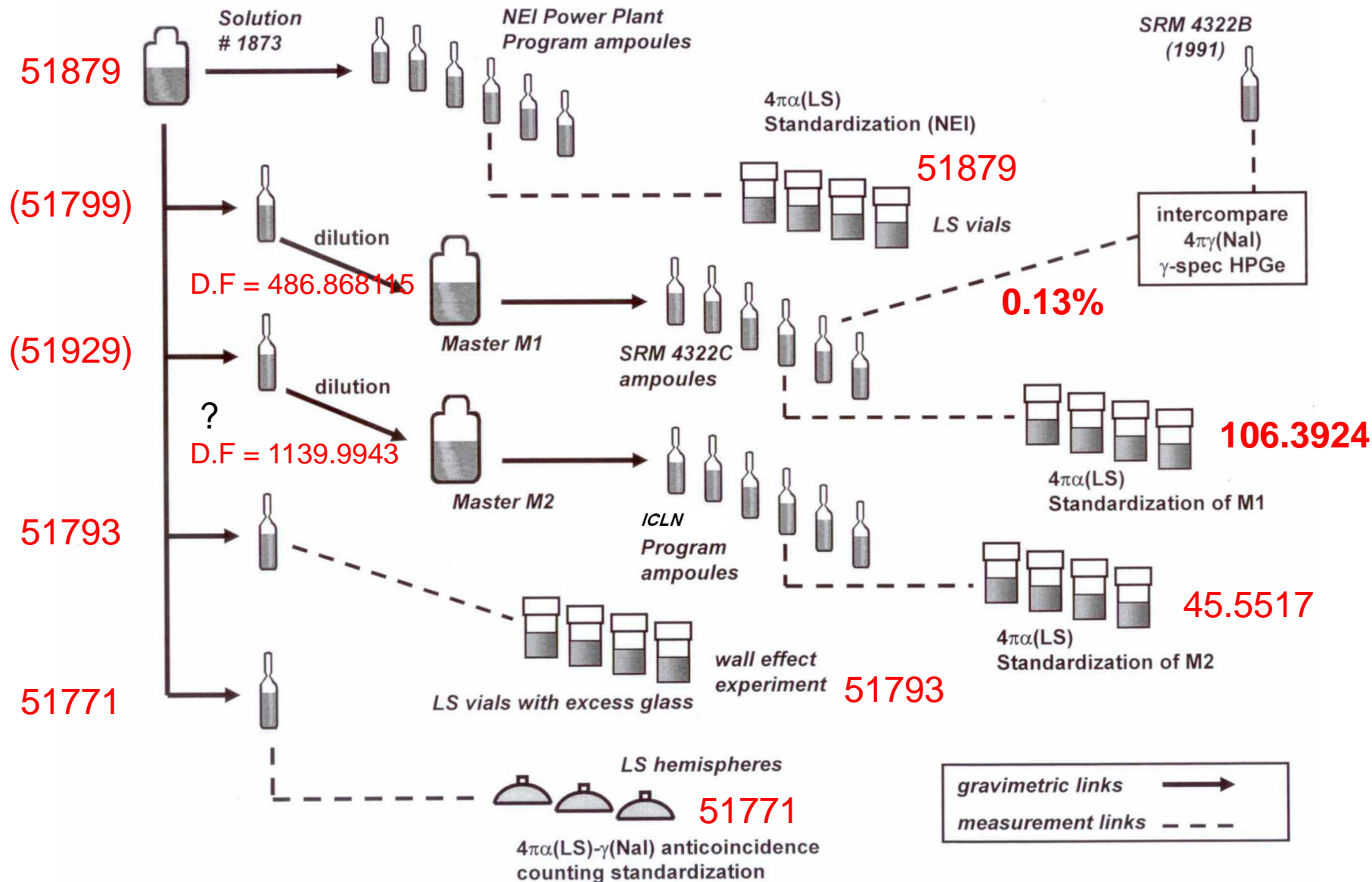
Th-230 Results

- Certified activity = $40.83 \pm 0.16 \text{ Bq}\cdot\text{g}^{-1}$ at 1200EST 1 April 2007
- Preliminary Measurement by Lucas, $A = 40.90 \text{ Bq}\cdot\text{g}^{-1}$ at 1200EST 1 April 2007
- Activity by α spectrometry = $41.0 \pm 1.2 \text{ Bq}\cdot\text{g}^{-1}$ – lisa
- Attempted confirmation by Ingrowth of Ra-226 with PIC measurements were 9% different -Peter

Am-241



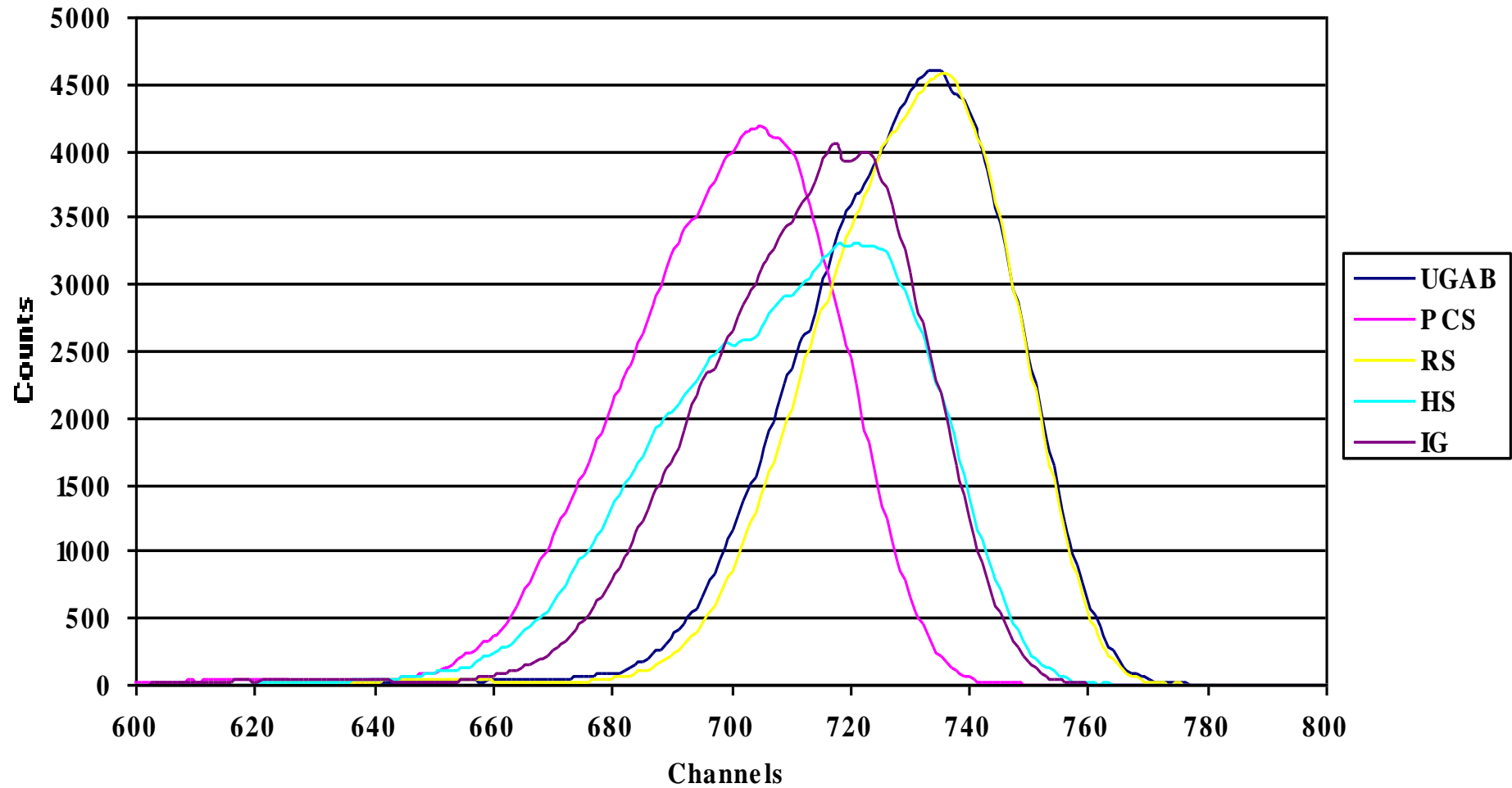
Am-241



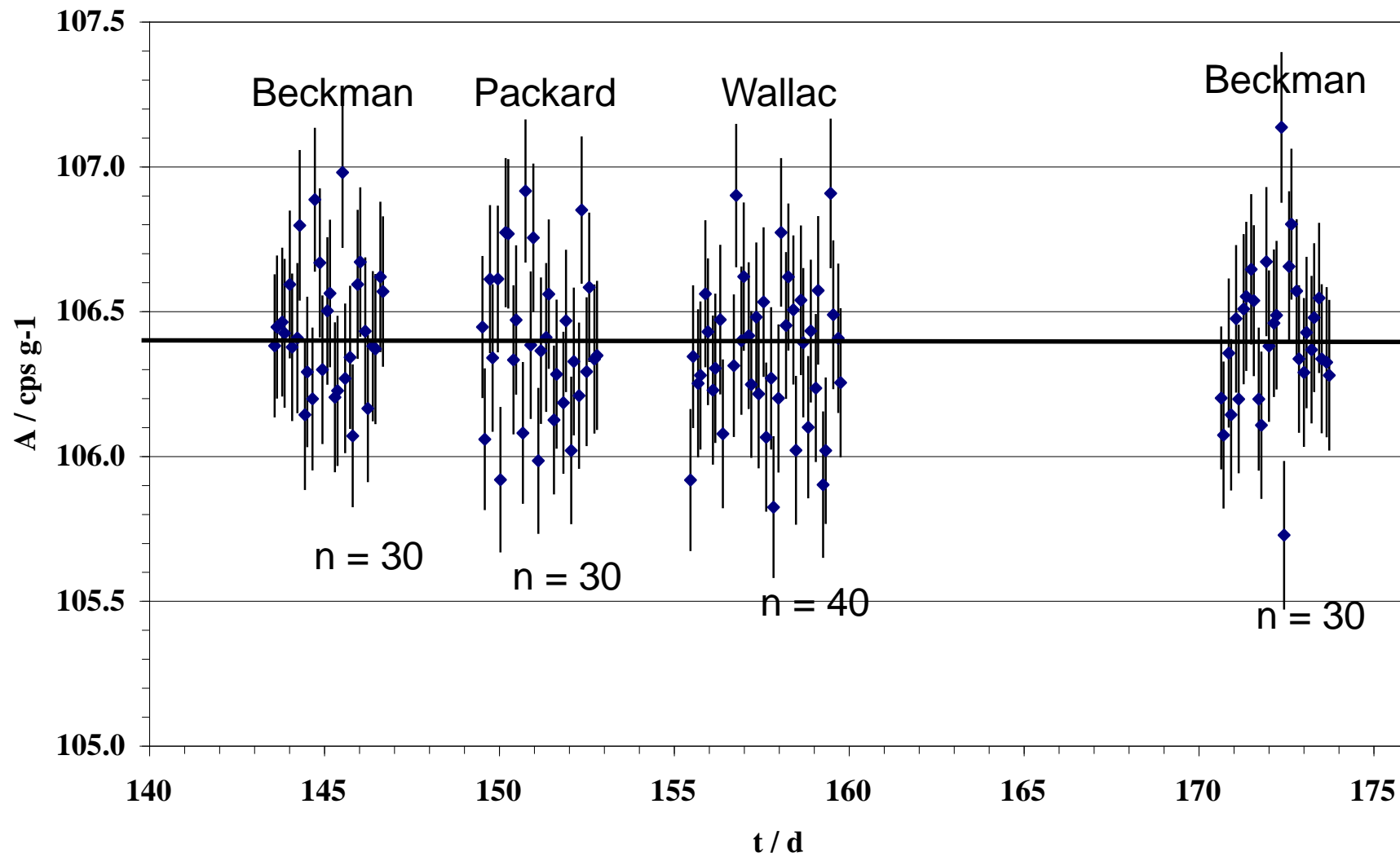
LS CNET

- 130 determinations; variables include:
 - 3 counters
 - 5 compositions
 - 5 scintillants
 - 2 sources per composition
 - 3 to 5 cycles / 100 minutes per measurement
 - 30 days of aging

Am-241 Spectra



Am-241



Pu-241

^{241}Pu (14.290 y)

$5/2+$

0.0

β^- decay 99.99

$Q_{\beta^-} = 0.0207$

β_1^-

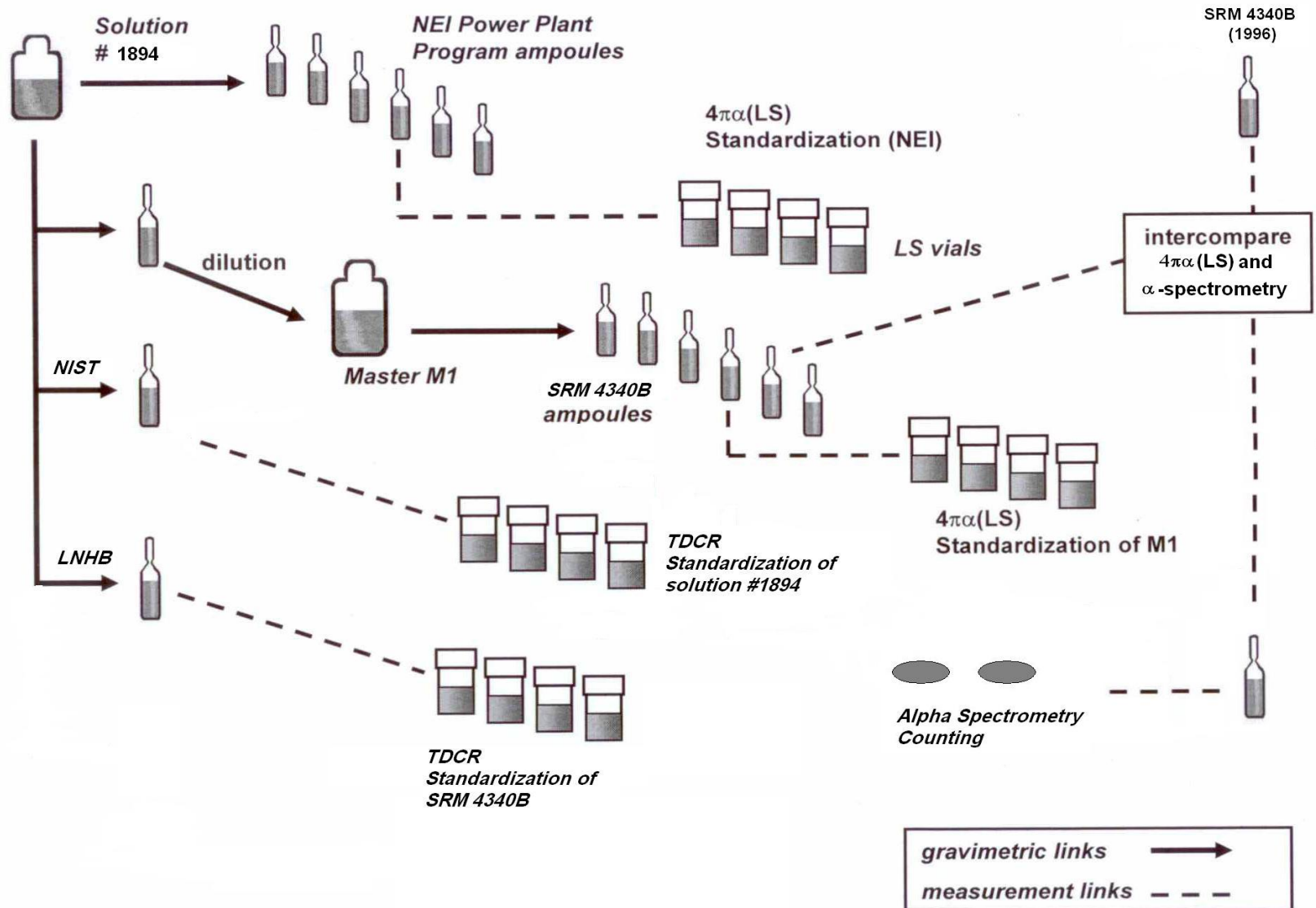


^{241}Am (432.6 y)

0.0

$5/2-$

Pu-241



Pu-241 Results

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

LS CNET

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

Pu-241 Results

- CNET and TDCR in serious disagreement (7.7%)
 - Presumably based on same model
 - NOT due to spectrum
 - EFFY and CN2003 codes small differences
 - CN2003 invariant of kB number
 - CN2003 agrees with LNHB tracer code

Pu-241 Results

- CNET results correct based on
 - Agreement with old certificate
 - Based on Am-241 Ingrowth
 - Confirmation based on Am-241 Ingrowth determined
 - LS CNET
 - Alpha Spectrometry
 - LS Spectra

	Activity (Bq•g ⁻¹)	Difference (%)
Old Certificate (SRM 4340A)	250.4	-
LS CNET	253.3	1.4
Alpha Spectrometry	258.0	3
LS Spectra	253.9	1.4

Ron

^{90}Sr -- history

^{210}Pb – NPL / NIST

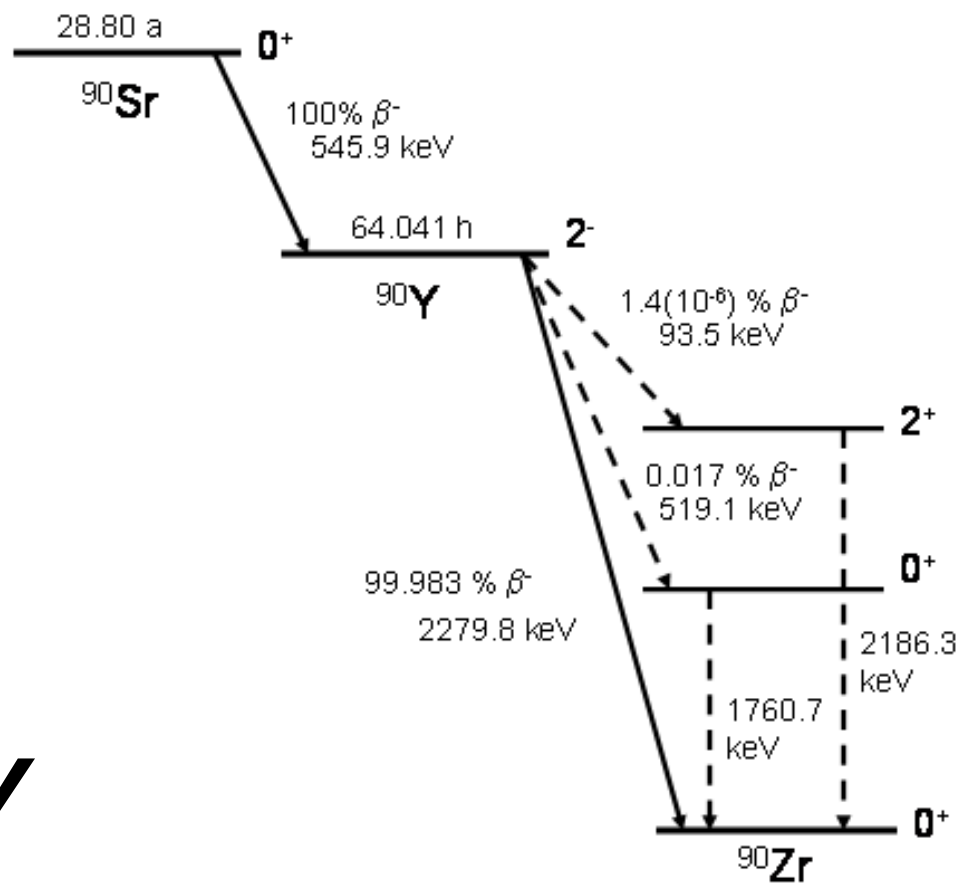
LS wall effect for α 's

Si(Li) detector

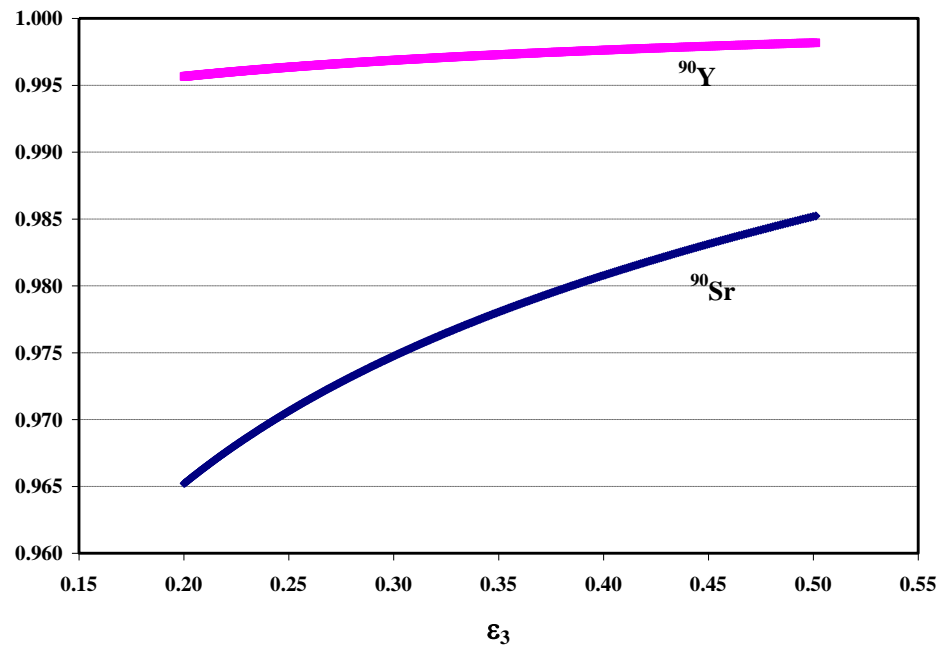
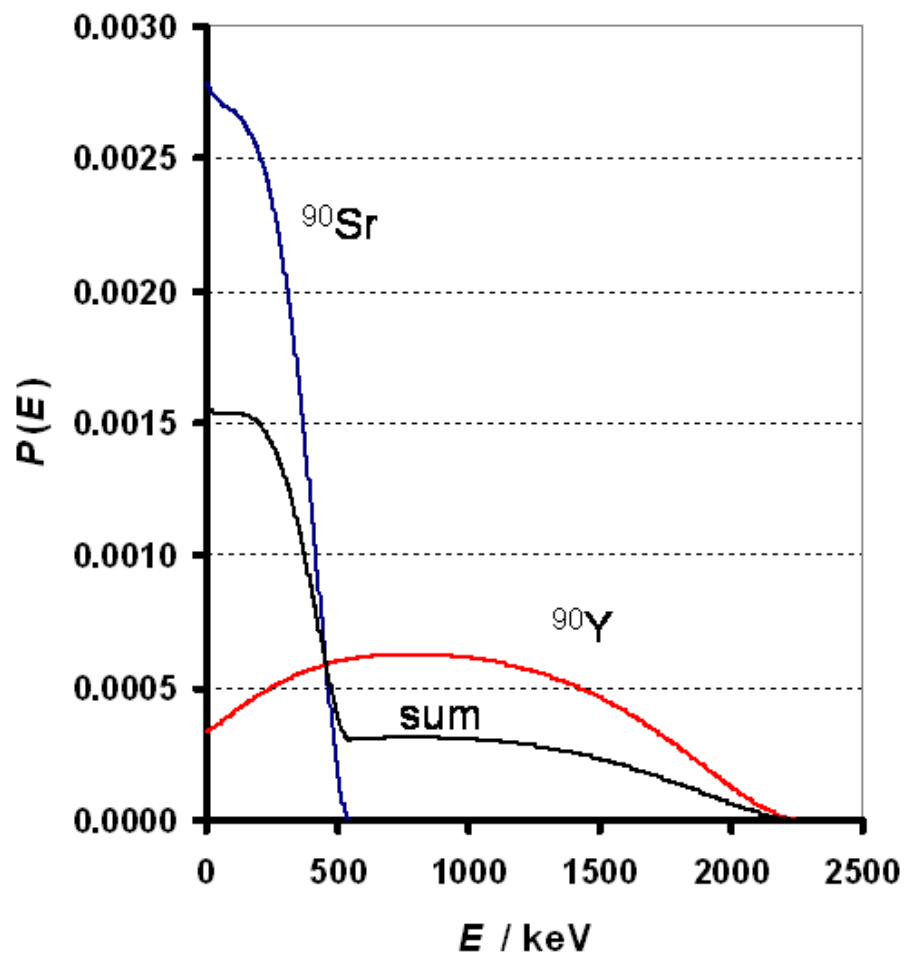
Cryogenic calorimeter (α spectrometry)

^{229}Th

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



fairly easy case



^{90}Sr

2 new SRMs (Dec 2006)

$$4919\text{I} = 4.261 \text{ kBq g}^{-1} \pm 0.48 \% (k = 2)$$

$$4239 = 31.79 \text{ kBq g}^{-1} \pm 0.46 \%$$

(LS CNET 3 cocktail compositions / 3 counters)

precision $< 0.1 \%$ ($\nu_{\text{eff}} = 293$); normal

Measurement comparison

$$\text{CNET/TDCR} = 1.00094 \pm 0.55 \% (k = 1)$$

A Half-Century of Radioactivity Solution Standards of ^{90}Sr - ^{90}Y

R. Collé, L. Laureano-Perez, and B.E. Zimmerman

Series	Instrument	Age / d	N d	ϵ_{H-3}	ϵ_{Sr-90}	ϵ_{Y-90}	Average / $kBq \cdot g^{-1}$	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

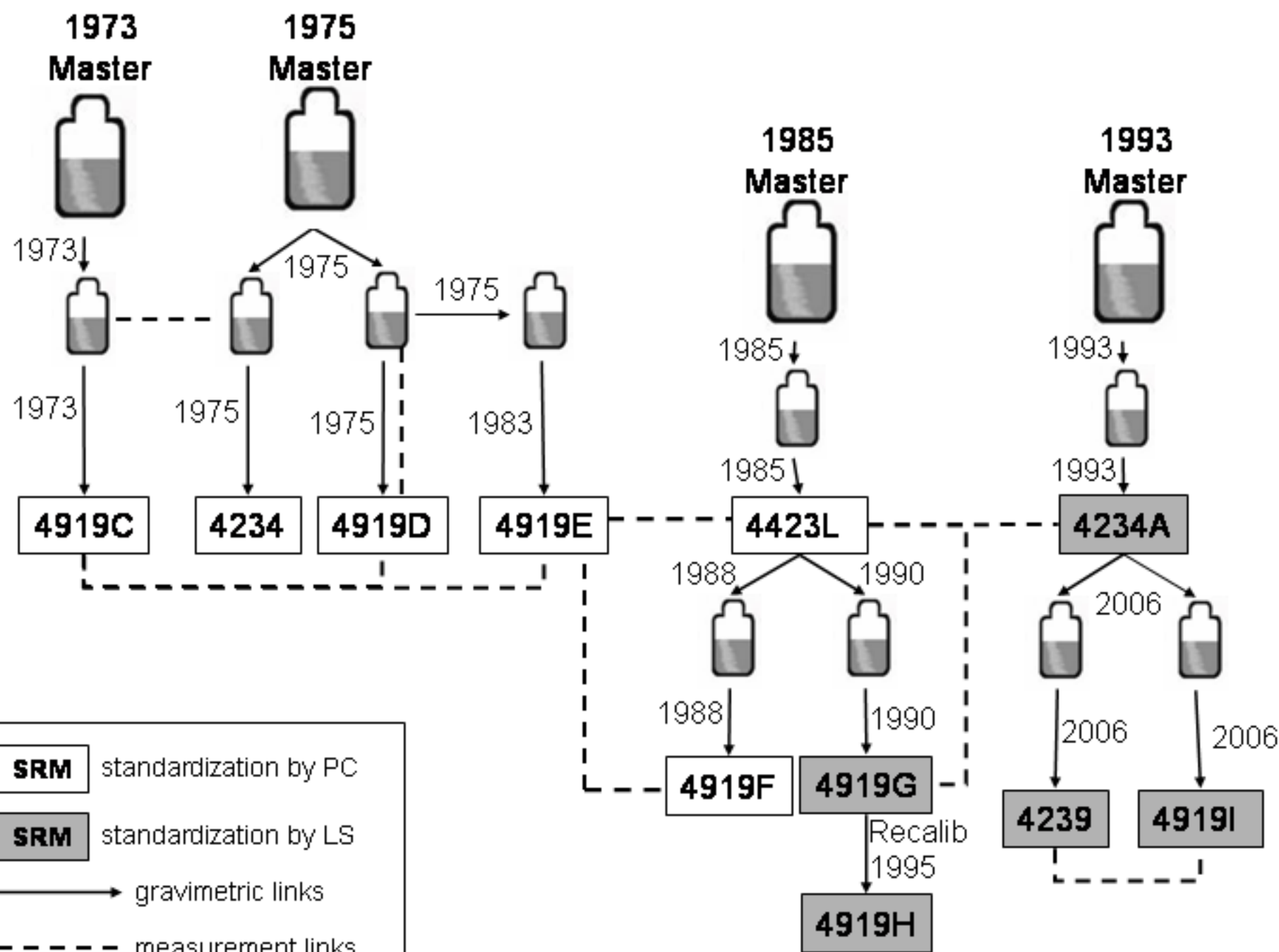
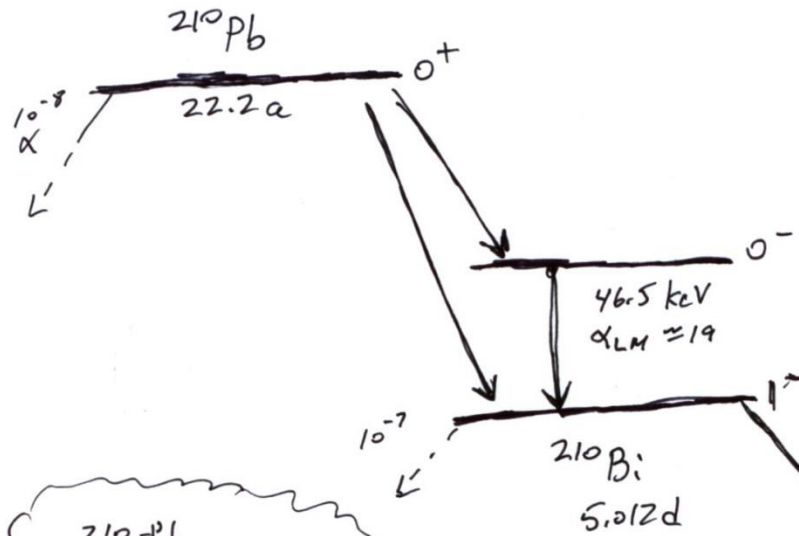


Table 1. Solution standards of ^{90}Sr - ^{90}Y disseminated by NBS/NIST from c.1950 to 2007.

SRM	Reference time	Primary standardization method	Solution ^(a) $\frac{^{90}\text{Sr}/\text{Sr}}{^{90}\text{Y}/\text{Y}}$	^{90}Sr massic activity $\text{kBq}\cdot\text{g}^{-1}$	Reported uncertainty	Uncertainty method
4919	(c. 1950)	?	?	? ^(b)	?	?
4919A	(1953)	4 π PC	$\frac{(10^{-4})}{(10^{-7})}$	≈ 5 ^(b)	($\approx 2\%$)	? ^(c)
4919B	(1957)	4 π PC	$\frac{(10^{-4})}{(10^{-7})}$	≈ 5 ^(b)	($\approx 2\%$)	? ^(c)
4919C	1973 19 march	4 π PC	$\frac{1.3(10^{-6})}{2.9(10^{-10})}$	0.1120 ^(b)	2.0 %	LC ^(d)
4919D	1975 26 april	4 π PC	$\frac{7.7(10^{-6})}{2.0(10^{-9})}$	2.017 ^(b)	2.1 %	LC ^(d)
4234	1975 4 august	4 π PC	$\frac{1.6(10^{-3})}{4.2(10^{-7})}$	626.0 ^(b)	1.47 %	LC ^(d)
4919E	1983 1 may	4 π PC	$\frac{1.3(10^{-5})}{3.3(10^{-9})}$	3.375	1.77 %	LC2 ^(e)
4423L	1985 16 november	4 π PC	$\frac{4.1(10^{-3})}{1.0(10^{-6})}$	4403	1.05 %	3 \times QC ^(f)
4919F	1988 1 may	4 π PC	$\frac{4.7(10^{-5})}{1.3(10^{-8})}$	4.094	1.16 %	3 \times QC ^(f)
4919G	1990 1 august	4 π LS	$\frac{9.1(10^{-6})}{2.3(10^{-9})}$	4.514	1.05 %	3 \times QC ^(f)
4234A	1995 13 march	4 π LS	$\frac{1.7(10^{-3})}{4.5(10^{-7})}$	2494.	0.56 %	2 \times QC ^(g)
4919H	1995 1 july	4 π LS	$\frac{6.9(10^{-6})}{2.2(10^{-9})}$	4.010	0.74 %	2 \times QC ^(g)
4919I	2006 25 december	4 π LS	$\frac{4.1(10^{-5})}{9.1(10^{-9})}$	4.261	0.48 %	2 \times QC ^(g)
4239	2006 25 december	4 π LS	$\frac{2.5(10^{-4})}{6.0(10^{-8})}$	31.79	0.46 %	2 \times QC ^(g)

^{210}Pb

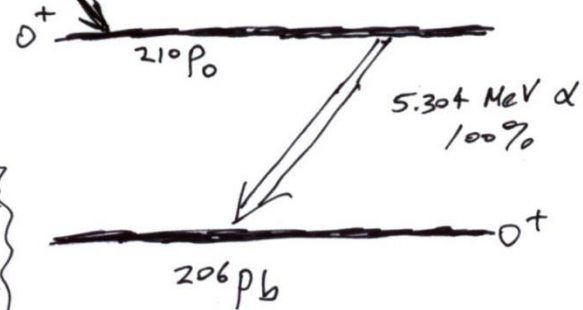


$$E_{LS} = E_{\beta} + E_{x\text{ce}} + E_{B_i} + E_{p_0}$$

(30%) \downarrow \downarrow
 90% 99% 100%

^{210}Pb
 $E_{\beta(\text{ave})} = 4.16\text{ keV}$ (84%)
 $E_{\beta(\text{ave})} = 16.16\text{ keV}$ (16%)
 both 1st forbidden, non-unique

^{210}Bi
 $E_{\beta(\text{ave})} = 389\text{ keV}$ (100%)
 1st forbidden, non-unique
 ce-L 30 keV (60%)
 ce-M 43 keV (30%)



^{210}Pb

Liz, et alia, *ARI* **65**, 1368 (2007)

Old stuff

$$\text{SRM 4337} = 9.037 \text{ Bq g}^{-1} \pm 1.2 \% (k=1)$$

Compare to CNET

anticoïn.	= + 0.7 %
^{210}Po α Spect	= - 3.0 % (^{209}Po 102 a)
	= - 1.3 % (128 a)
HPGe	= + 4.7 %

new stuff

NPL Standard (333 Bq g^{-1}) – based on dilution of PTB std.

PTB did ^{210}Po α Spect

NPL confirm with ^{210}Bi ingrowth by Cerenkov (CNET for ^{210}Bi efficiency)

NPL / NIST ^{210}Pb



Cert. = $0.037484 \pm 1.5 \%$ ($k = 1$)

$4\pi\alpha\beta$ LS = $0.037542 \pm 0.17 \%$ (+ 0.15 %)

$4\pi\gamma(\text{NaI})$ = $0.037373 \pm 0.30 \%$ (- 0.30 %)

^{210}Po assay = $0.03736 \pm 0.7 \%$ (- 0.30 %)

HPGe = $0.03754 \pm 0.7 \%$ (+ 0.15 %)

Si(Li) = $0.0374 \pm > 1 \%$ *incomplete*



LS WALL EFFECT

My friend

54

P Cassette

Says 0.2 %

Based on ...

Pu-238

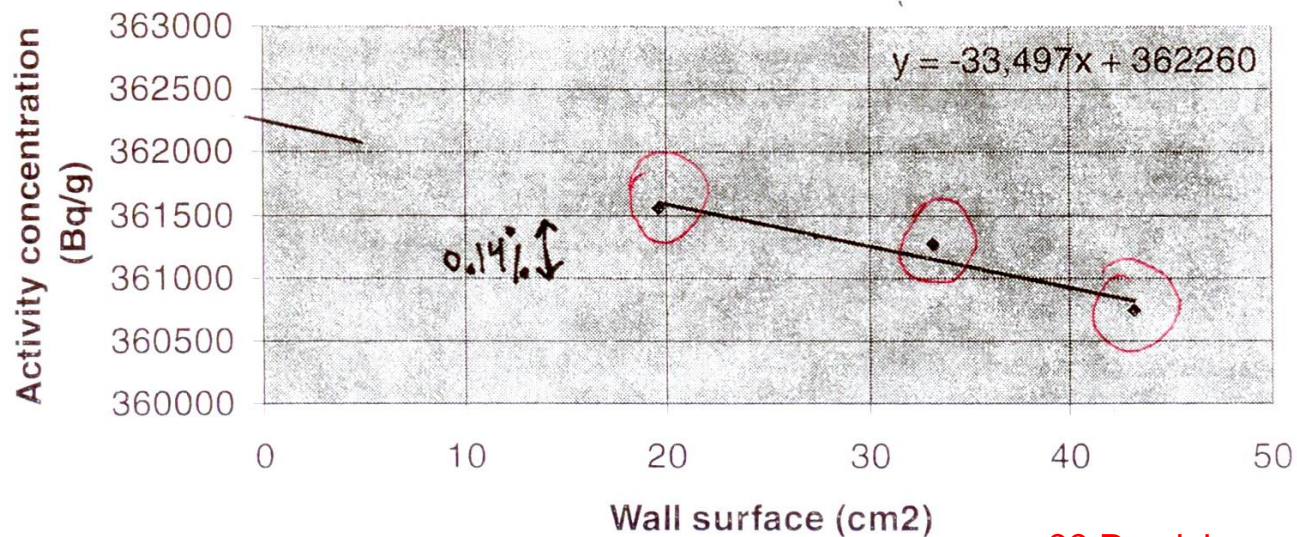


Figure 7 Counting rate of a ^{238}Pu source with different wall surfaces

?? Precision uncertainty

?? Replications

Can not reproduce effect (magnitude) using ^{210}Po & ^{241}Am

3 experiments

Vial – 27.3 cm²

Glass beads 34.1 – 54.4 cm² + area / volume ratios

Sleeves 34.4 – 39.2 cm²

Rods & tubes 36.9 – 45.5 cm²

Lots of screwy results

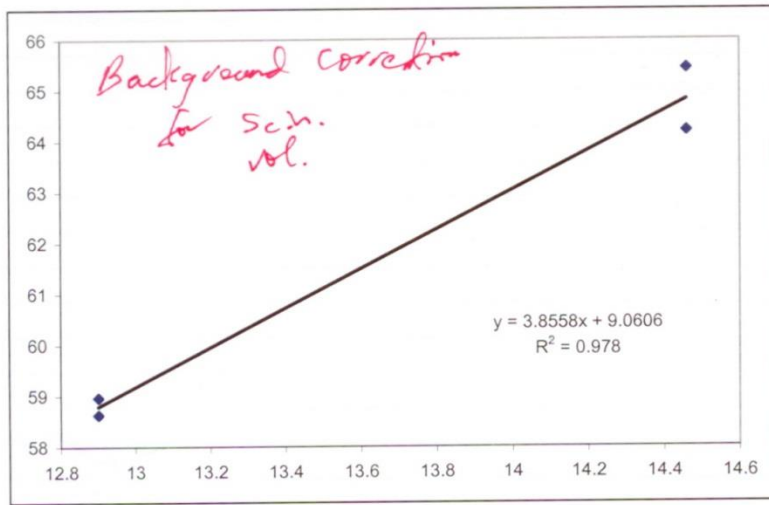
LS imprecision – 0.2 to 0.3 % typical

funny light effects – reflections from beads

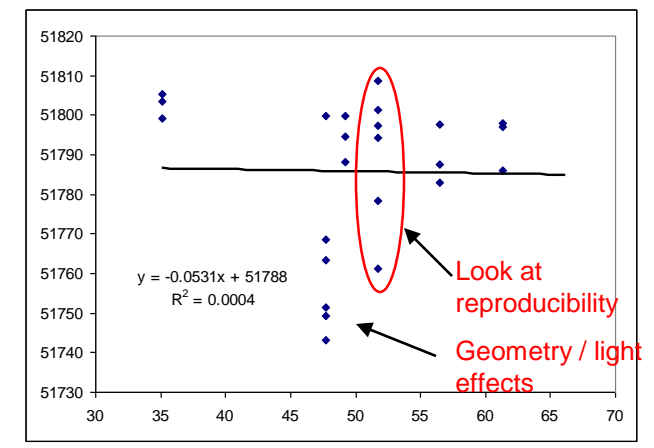
tilting rods & sleeves – indeterminate volumes

sample masses

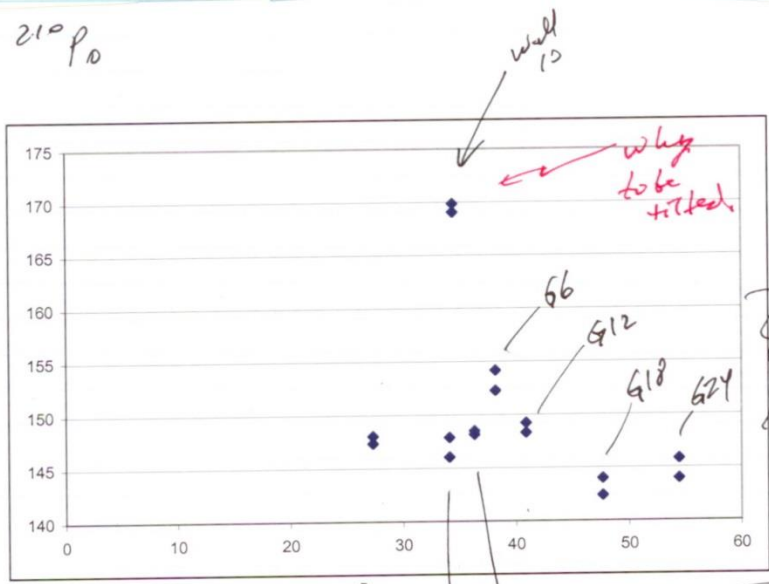
cocktail instability



²⁴¹Am



²¹⁰Po



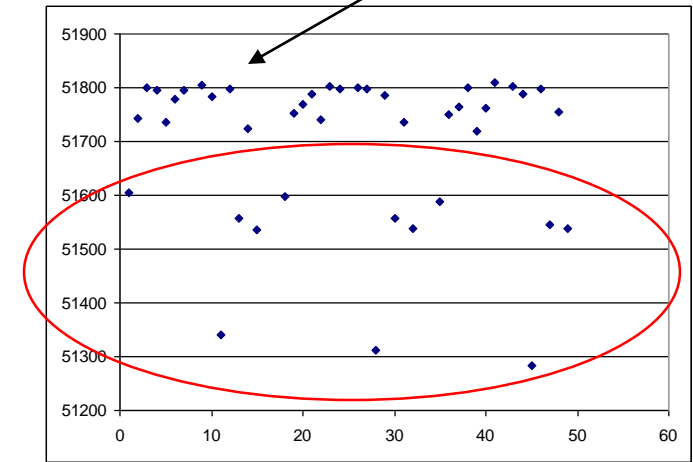
Good
have
1.3 lit
effect
#?!

↑
v. vol.
12 14

well
effect

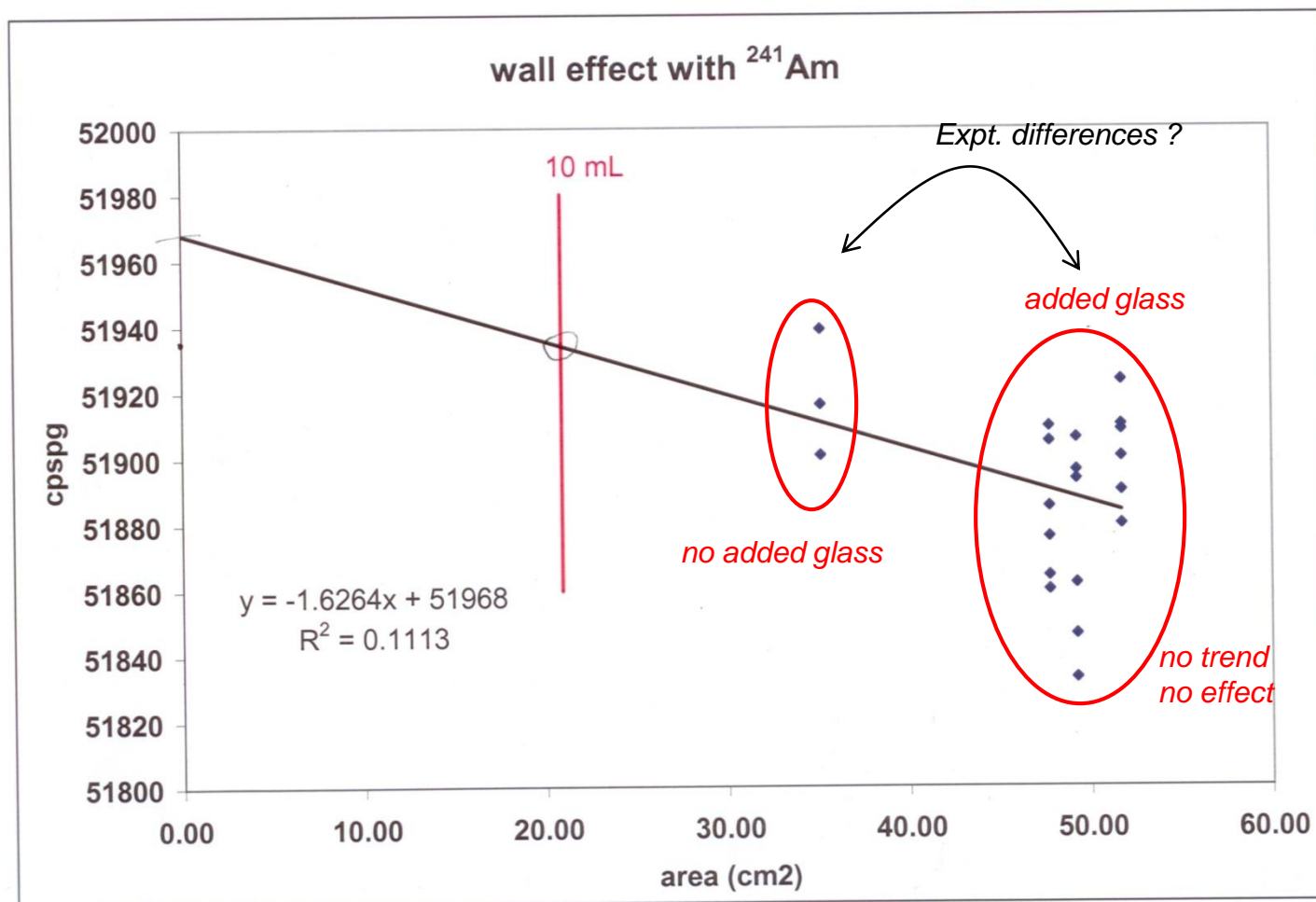
No apparent
effect with area

Sequential samples



cleanest expt !

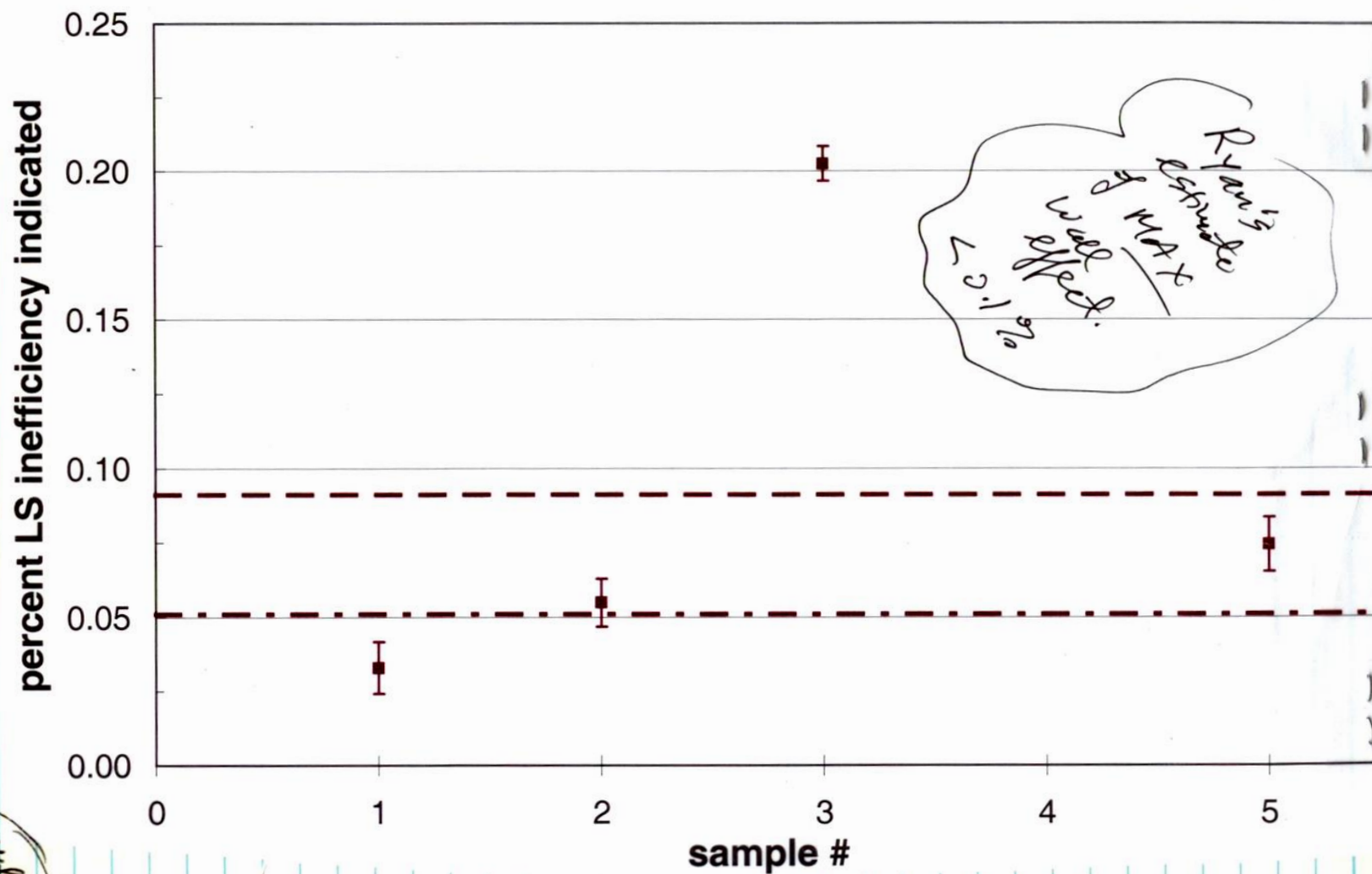
< 0.06 % effect



from Ryan

9/5/2004
2004

LS inefficiency for 4-mL hemispheres - γ -ray gate



wall effect

With Brian

Si(Li) detector

Set up this past summer

Windowless chamber

Pump problems

We have to move !

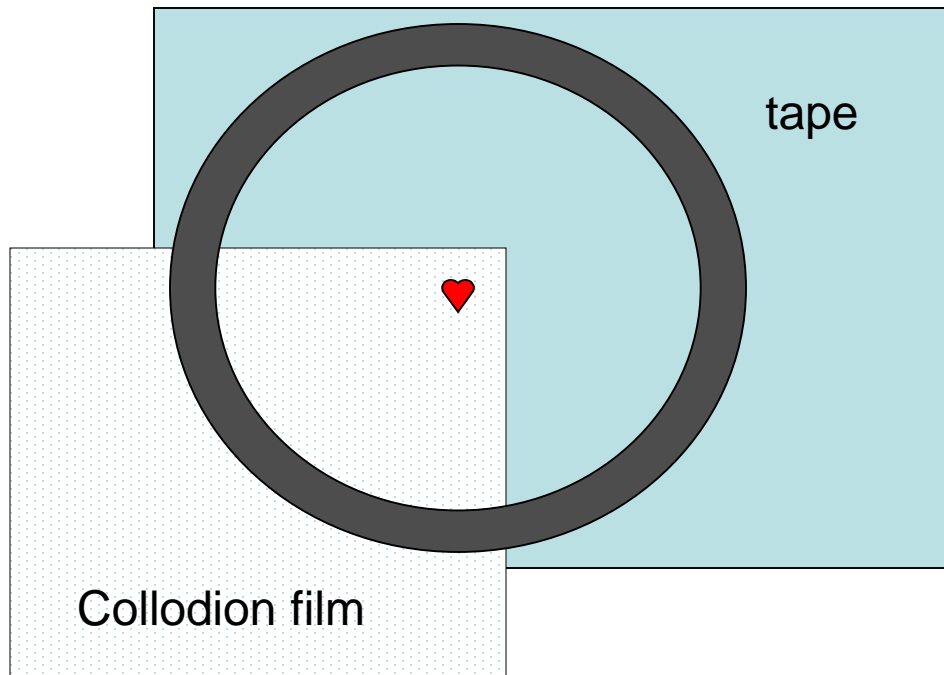
No time to work on

Can see down to Al K x rays – 1.5 keV

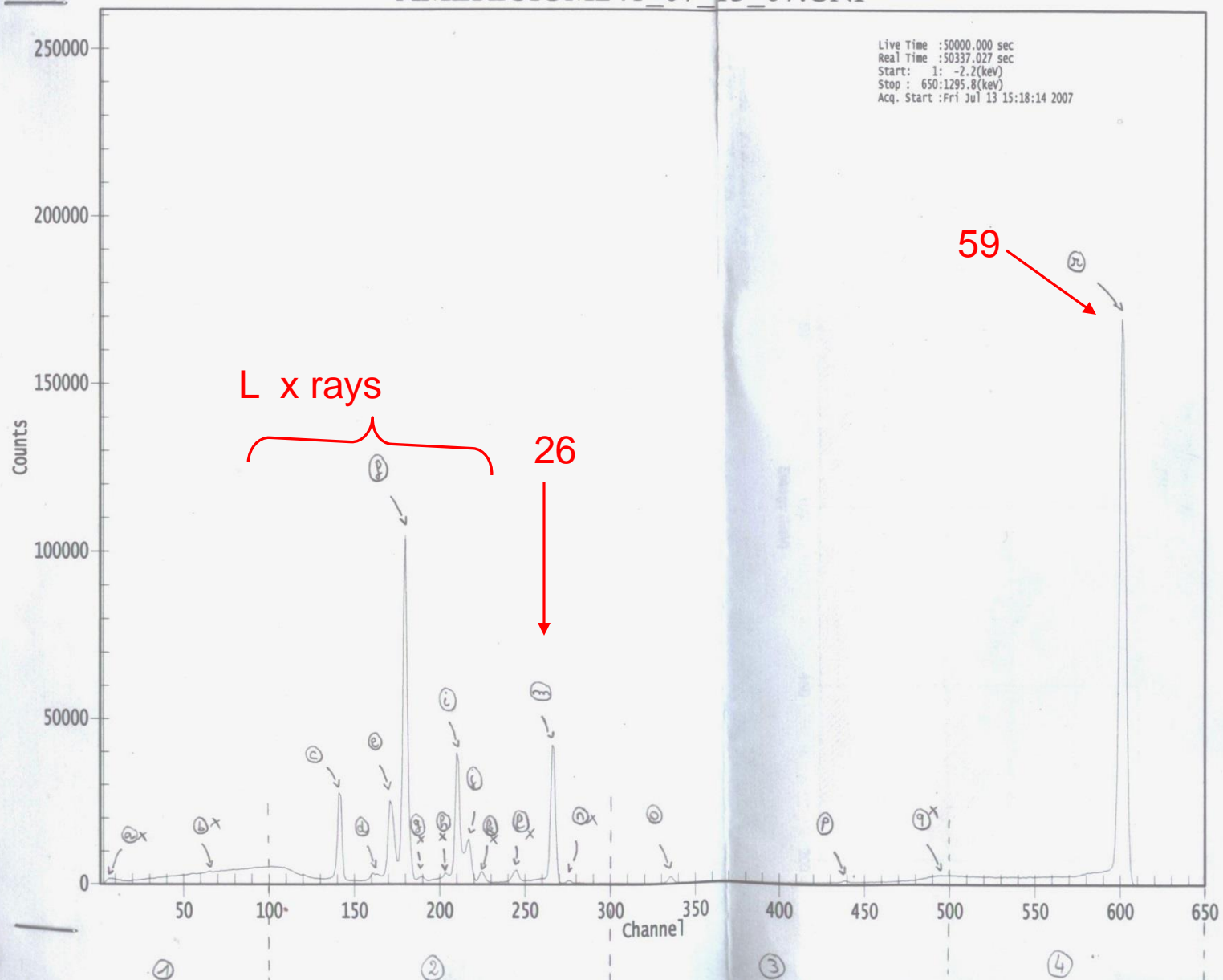
Can measure in ampoules

down to ^{241}Am L x rays (> 12 keV)

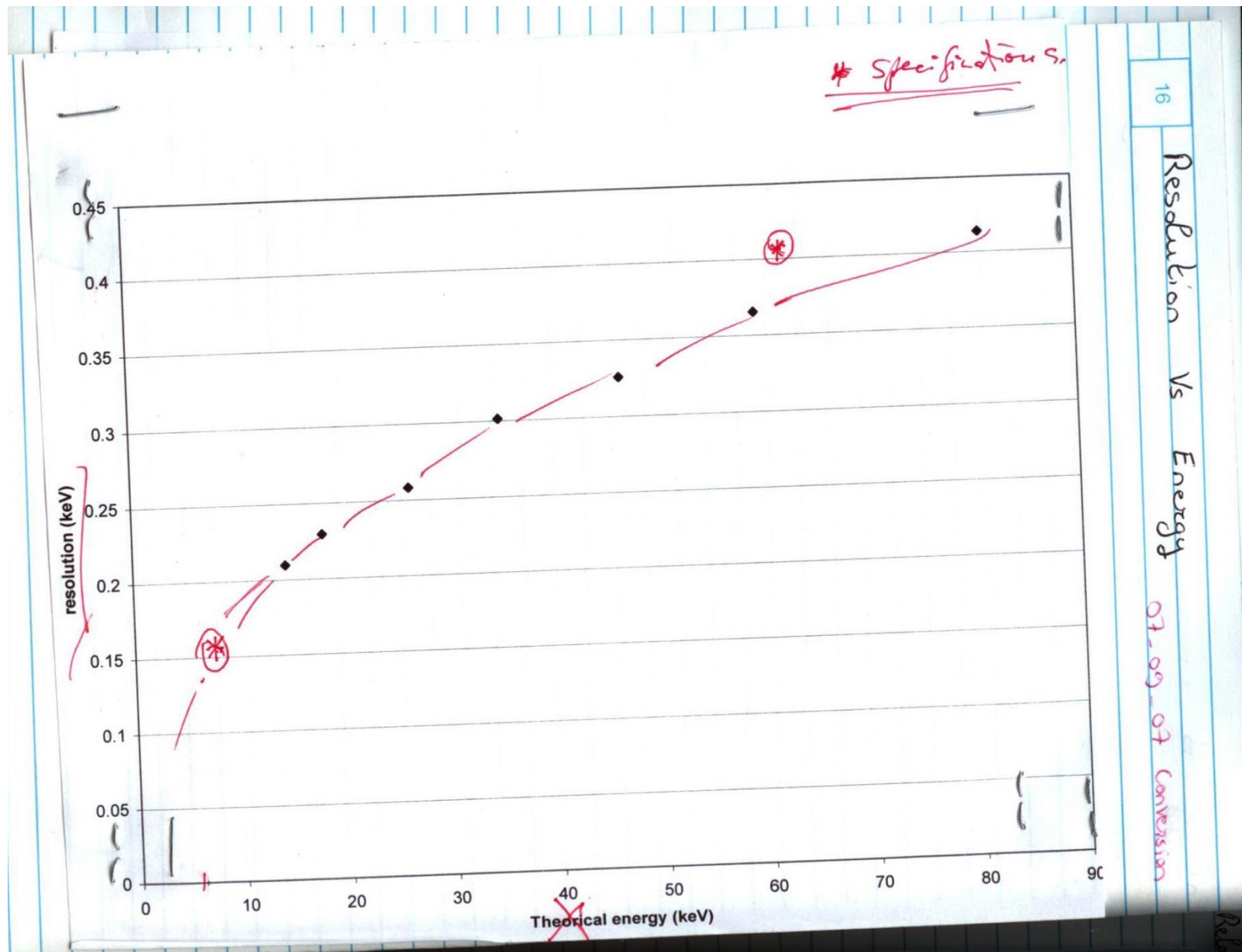
Point sources in range of few keV to > 100 keV



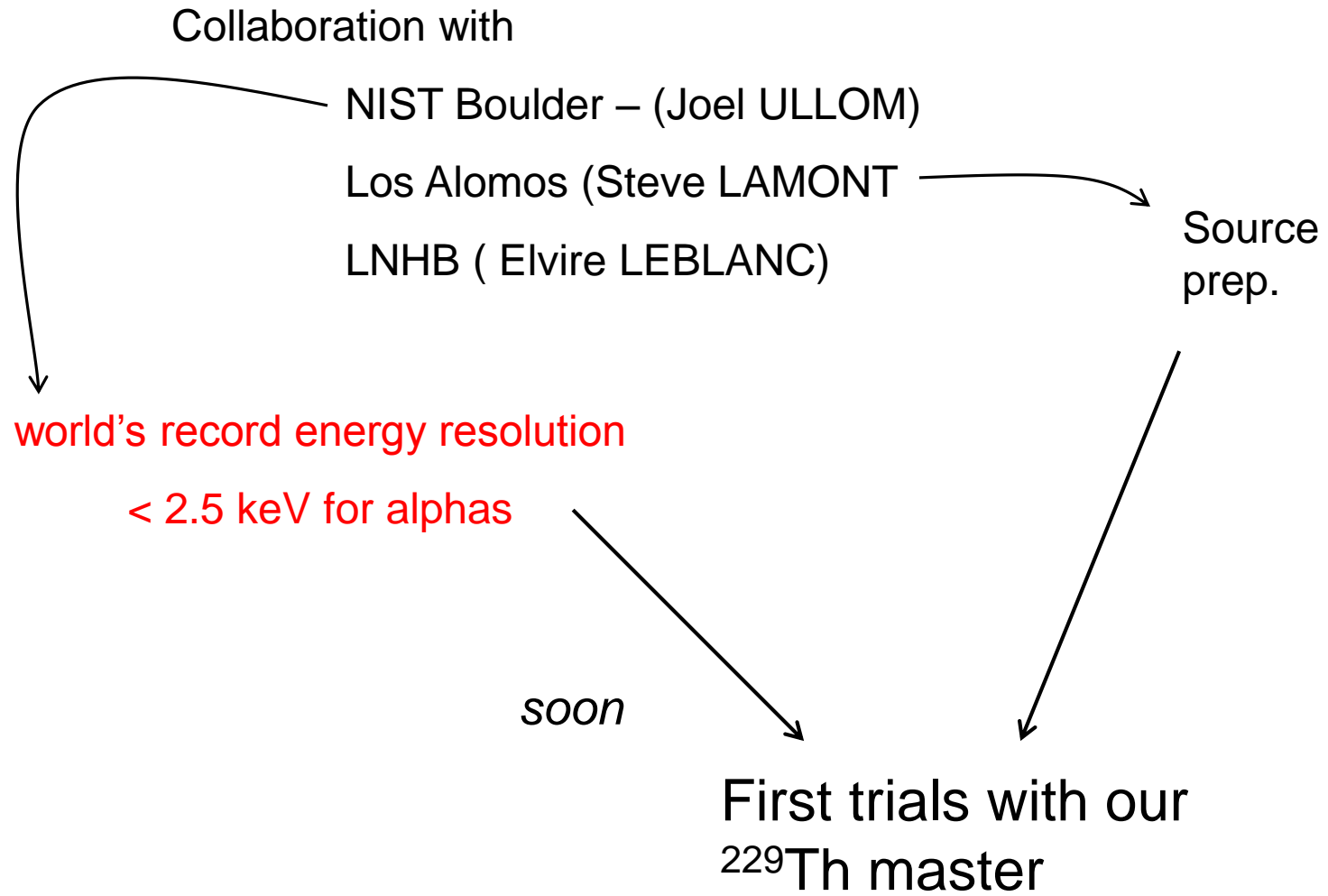
AMERICIUM241_07_13_07.CNF



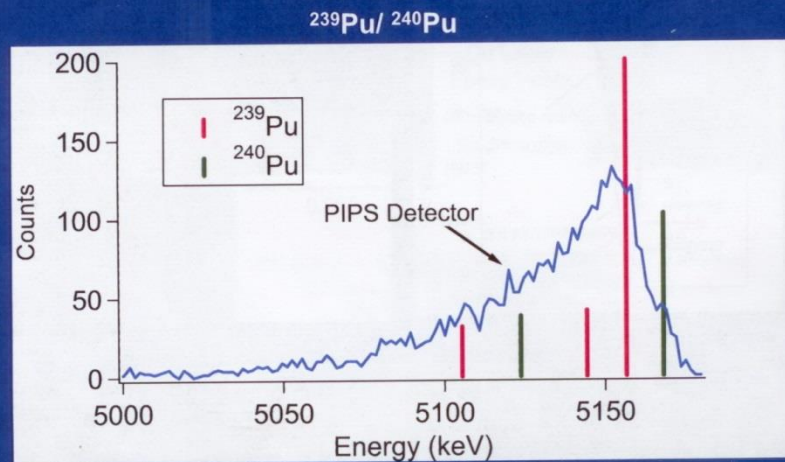
Detector resolution -- to specs



Cryogenic microcalorimeter for alpha spectroscopy



Measured response to mixed Pu isotopes

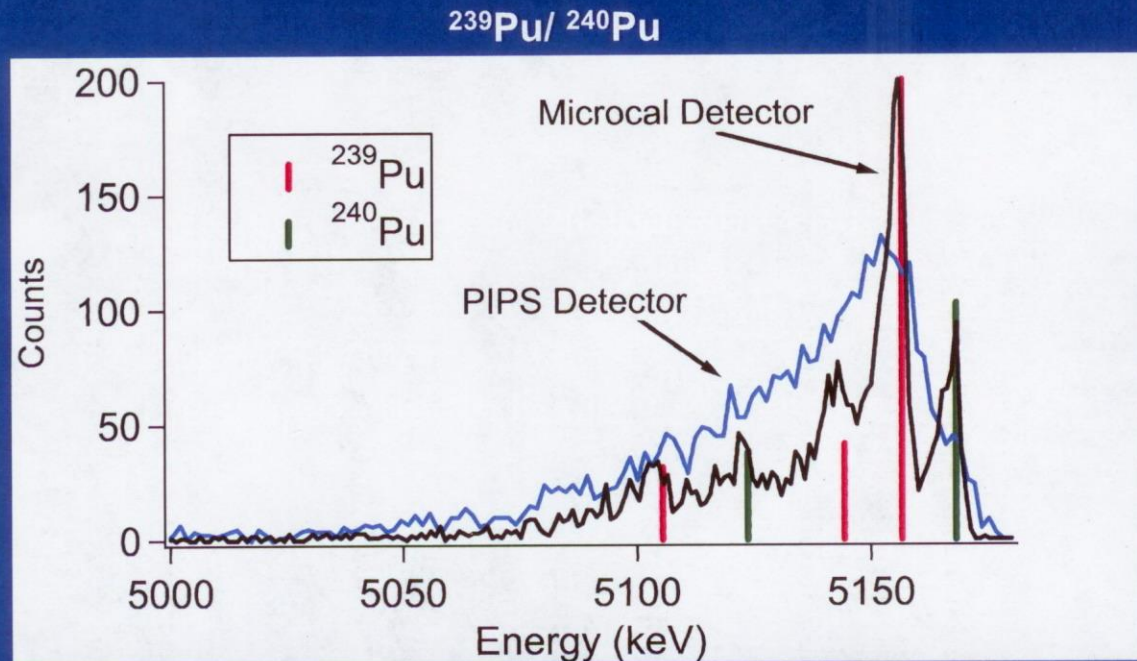


← Si detector for 5 unresolved lines

Compare with
cal response

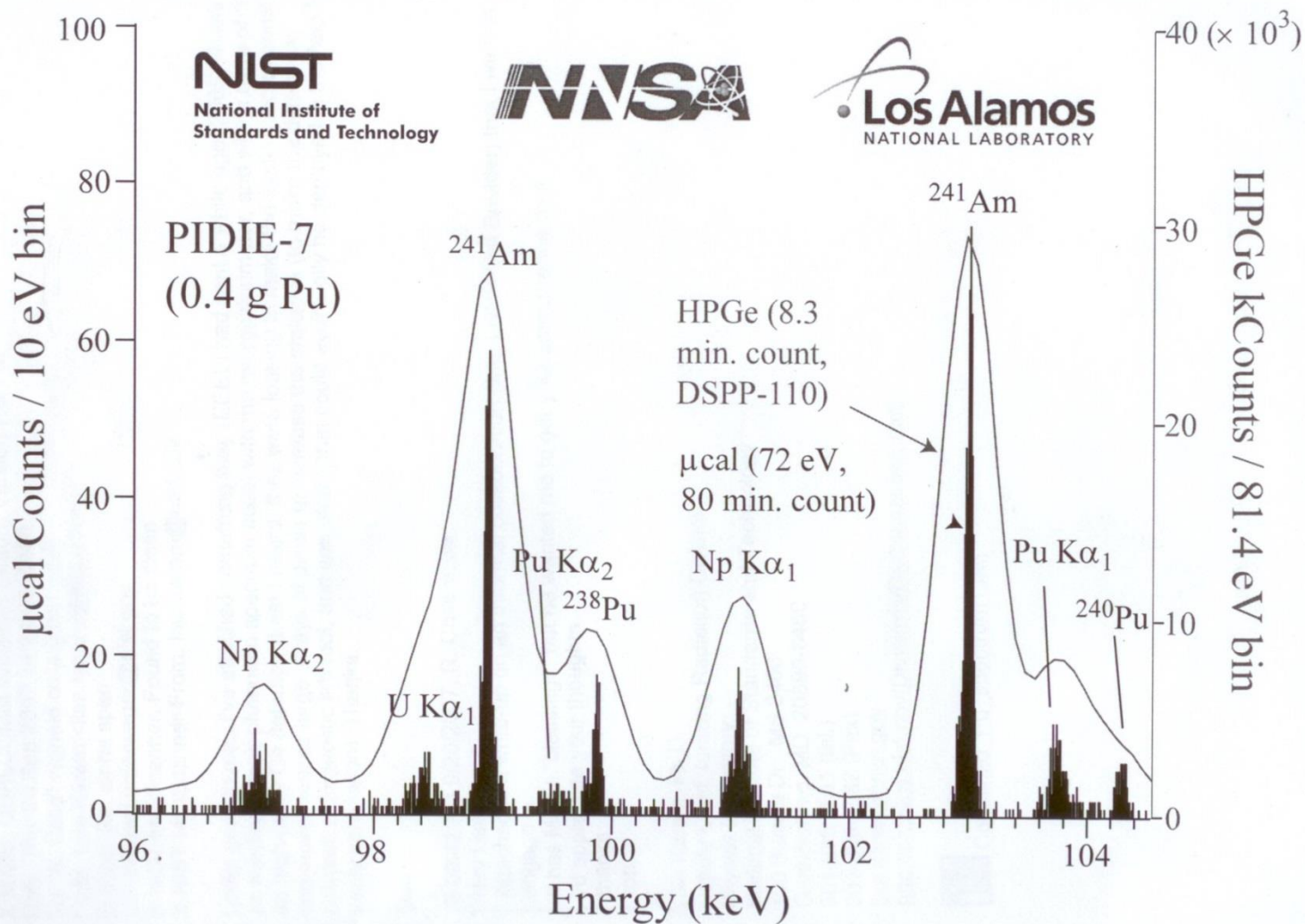


Measured response to mixed Pu isotopes



results for
photons


Pu spectra with both microcalorimeter and HP Ge detector





^{229}Th

\$ 75K + SRM (fin Dec 2008)

Difficult chain + impurities

➡ Ken – made master solution from Oak Ridge 

➡ Liz & I -- made first batch of 280 SRM ampoules (5 year supply !) 

➡ Brian -- used TDCR to look at effect of resolving time on LS doubles rate 

➡ Started LS CNET vs ^3H for betas --

3 cocktail compositions

counting done



many calculations still to do

➡ Ryan – primary comparison with anticoincidence counting


➡ Brian – TDCR maybe

➡ Comparison with LNHB

➡ Comparison with Los Alamos – maybe ?

+ cal sources for Boulder

➡ Lynne – made first cut $2\pi\alpha$ pc measurements 

➡ lisa – looked at Si spectrum 

➡ Ryan & I – will redo $2\pi\alpha$ pc with known resolving times

➡ will need impurity analyses (probably with chemistry)

^{229}Th 7340 a

$E_\alpha = (4.7-5.0)\text{ MeV}$
 $E_\gamma = 86$ $P_\gamma = 4\%$
 $E_\gamma = 193\text{ keV}$ $P_\gamma = 4.3\%$
other γ 's $< 1\%$
X-rays 80, 100 keV
CS 2, 6, 12 keV

$\alpha: 100\%$

^{225}Ra 14.9 ± 0.2 d

1st γ 's $E_\beta = 320\text{ keV}$ $\bar{E}_\beta = 93.4\text{ keV}$ $P_\beta = 69.5\%$
to g.s. $E_\beta = 371\text{ keV}$ $\bar{E}_\beta = 105\text{ keV}$ $P_\beta = 30.5\%$
 $E_\gamma = 40.0\text{ keV}$ $P_\gamma = 30.0\%$ $P_{\text{other}} = 1.31\%$

$\alpha: (0.026 \pm 0.006)\%$

$\beta: 100\%$

^{225}Ac 10.0 ± 0.1 d

$E_\alpha = (5.0-5.8)\text{ MeV}$
 $E_\alpha = (5.6-5.8)$ Mostly
 $E_\gamma = 100\text{ keV}$ $P_\gamma = 1.7\%$
Not many γ 's

$\alpha: 100\%$

^{221}Fr 4.9 ± 0.2 m

$E_\alpha = 6.1\text{ MeV}$ $P_\alpha = 15\%$
to g.s. $E_\alpha = 6.3\text{ MeV}$ $P_\alpha = 84\%$
 $E_\gamma = 218\text{ keV}$ $P_\gamma = 11.2\%$
not much else. (8 keV γ 's...)

$\alpha: 100\%$

^{217}At 32.4 ± 0.4 ms

to g.s. $E_\alpha = 7.067\text{ MeV}$ $P_\alpha = 99.9\%$

$\alpha: 100\%$

^{213}Bi 45.59 ± 0.06 m

to 2nd γ 's $E_\beta = 983\text{ keV}$ $\bar{E}_\beta = 320$ $P_\beta = 30.8\%$
to g.s. $E_\beta = 1420\text{ keV}$ $\bar{E}_\beta = 492$ $P_\beta = 65.9\%$
 $E_\gamma = 440\text{ keV}$ $P_\gamma = 25.9\%$

$\alpha: 2.09 \pm 0.03\%$

$\beta: 97.85 \pm 0.1\%$

^{209}Tl 2.20 ± 0.07 m

$E_\alpha = 5549$ $P_\alpha = 7.4\%$
 $E_\alpha = 5869$ $P_\alpha = 92.6\%$ to g.s.
 $E_\gamma = 324\text{ keV}$ $P_\gamma = 8\%$

$\beta: 100\%$

$\alpha: 100\%$

^{213}Po 4.2 ± 0.8 μs

to g.s. $E_\alpha = 8376\text{ keV}$
 $P_\alpha = 100\%$

^{209}Pb 3.253 ± 0.014 h

to g.s. $E_\beta = 644.6\text{ keV}$ $\bar{E}_\beta = 197.5\text{ keV}$ $P_\beta = 100\%$

$\beta: 100\%$

^{209}Bi 10¹⁸ a

Effect of resolve time on LS doubles rate

$$Y = a + b / (1 - e^{-\lambda(c+x)})$$

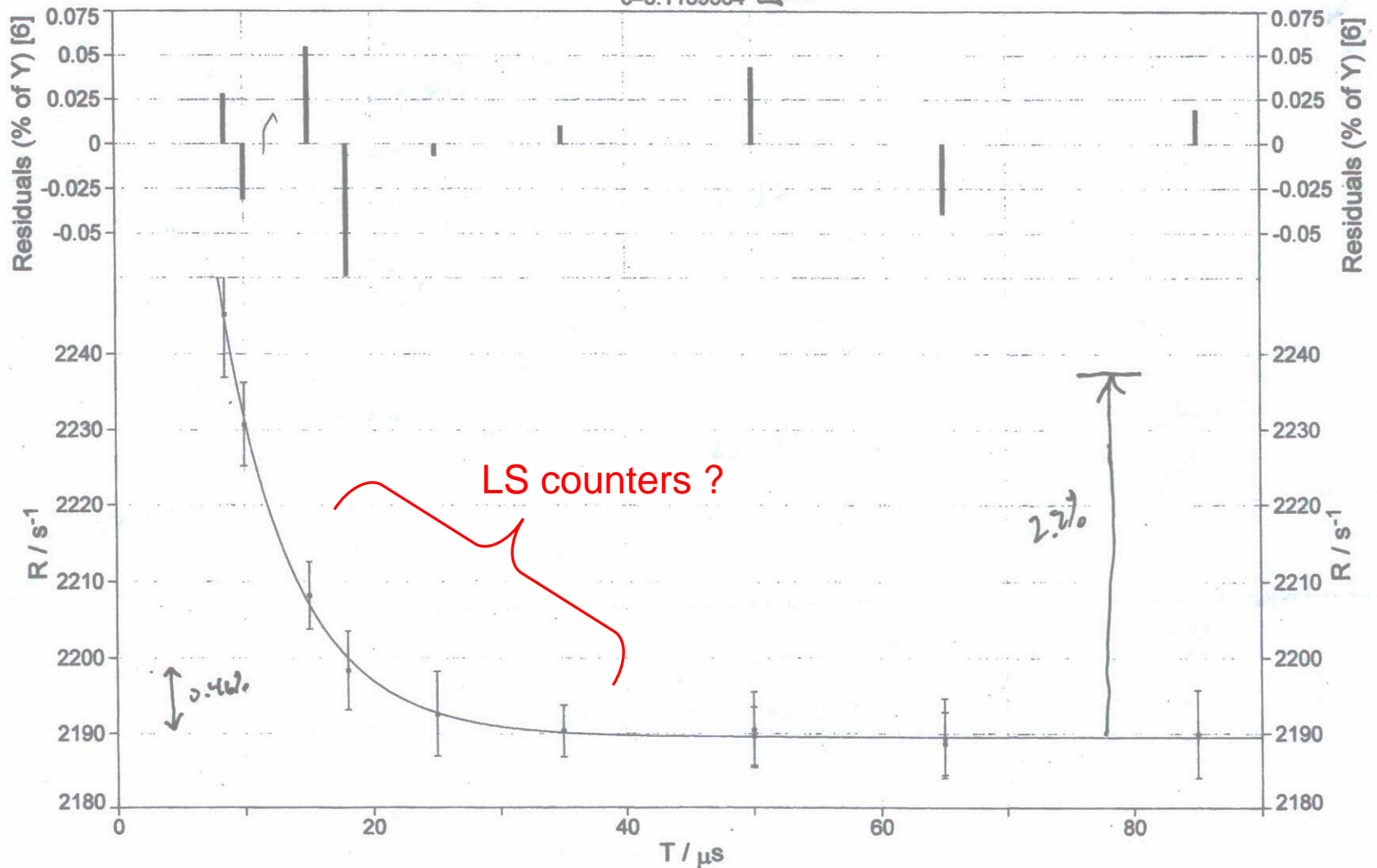
Th-229 deadtime Chan AB fit data

Rank 1 Eqn 8001 [UDF 1] y=Deadtime(a,b,c)

$r^2=0.99700798$ DF Adj $r^2=0.99572569$ FitStdErr=0.90111686 Fstat=1332.8916

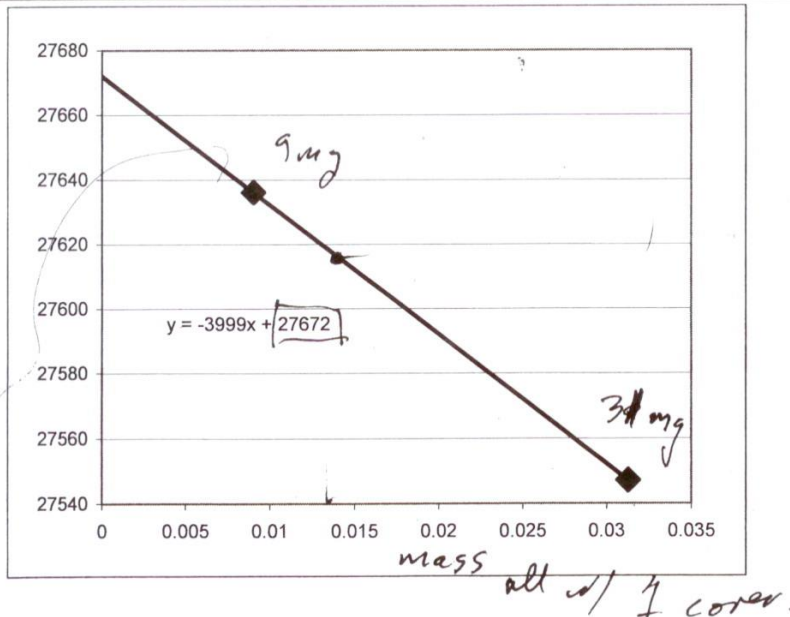
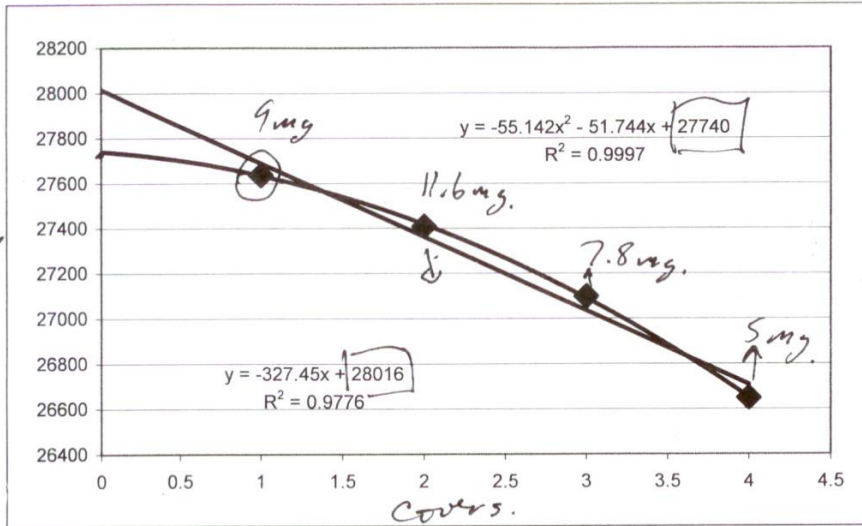
a=1723.7678 b=465.73607

c=5.1189534



a1	1	5.391246	0.009005	248.8624	27636.04
a2	2	5.370708	0.011554	316.6593	27407.98
a3	3	5.362959	0.007757	210.1835	27096.85
a4	4	5.357839	0.005125	136.5754	26648.23
b1	1	5.333601	0.024262	x	0
c1	1	5.302373	0.031259	861.0993	27547.04

2π pc



Rough approx.

Extrapolates to 27.7 cps/g

Assume 2π eff.

No impurities or side branches

Don't know resolve time yet

If 5 alphas (see all ^{213}Po)

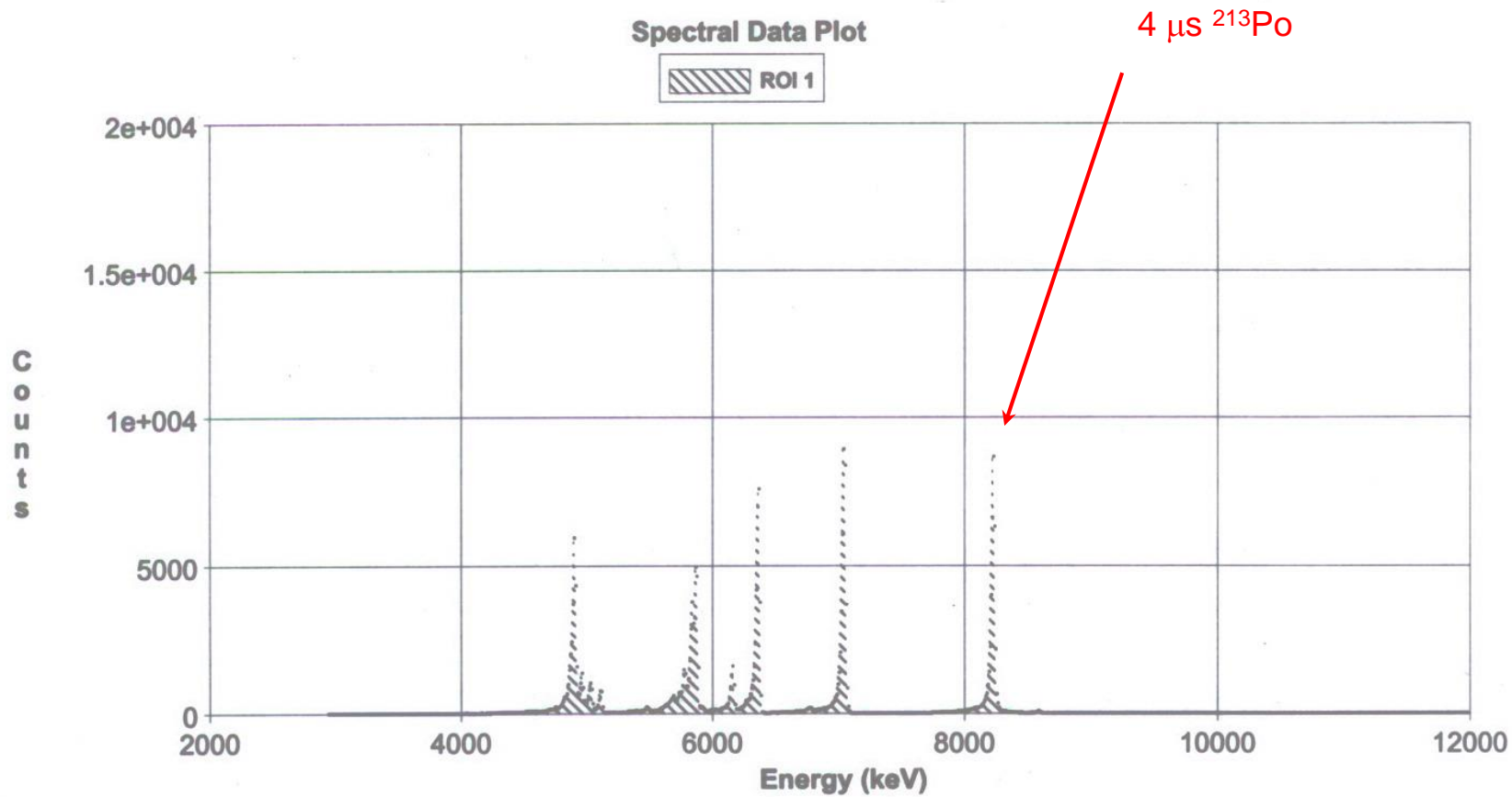
$$2(27.7)/5 = \mathbf{11.1} \text{ Bq/g}$$

If 4 alphas (see no ^{213}Po)

$$2(27.7)/4 = \mathbf{13.9} \text{ Bq/g}$$

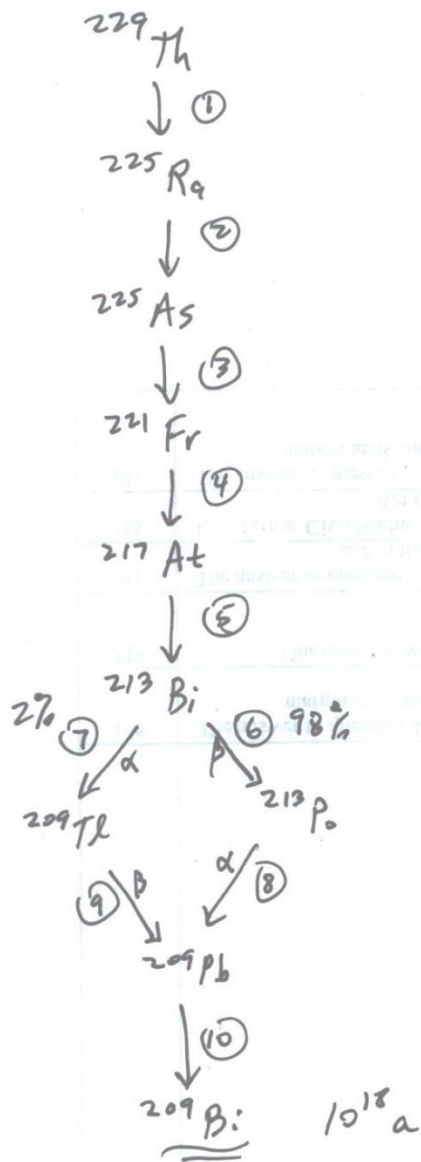
12/21/07

Th spectra



Datasource: RON TH DET10 DEC2107.CNF
Live Time: 11350 sec
Real Time: 11355 sec
Acq. Start: 12/21/2007 9:54:55 AM
Start: 1 : 2938.99 (keV)
Stop: 1024 : 11963.72 (keV)

LS EFF.



$$\epsilon_1 = 1 \quad \alpha$$

$$\epsilon_2 = 0.85 \quad \beta \text{ guess}$$

$$\epsilon_3 = 1 \quad \alpha$$

$$\epsilon_4 = 1 \quad \alpha$$

$$\epsilon_5 = 1 \quad \alpha \leftarrow 32.4 \text{ ms } \alpha \text{ loss at } 50 \mu\text{sec}$$

$$\epsilon_6 = 0.98(1) \quad \beta \text{ new } 15\%$$

$$\epsilon_7 = 0.02(1) \quad \alpha$$

$$\epsilon_8 = 0.98(?) \leftarrow \alpha \text{ see } 19\% \text{ at } 10 \mu\text{sec} \leftarrow 4.2 \mu\text{s} \leftarrow \text{all lost at } 50 \mu\text{s}$$

$$\epsilon_9 = 0.02(0.95) \quad \beta \text{ guess}$$

$$\epsilon_{10} = 0.98 \quad \beta \text{ like } 90\%$$

$$\epsilon_{\text{LS}} = 6.85 + \epsilon(4.2 \mu\text{s} \alpha)$$

Th-229

10 min
Cycles

fw

	cps/g	sd %	Counter	10 min Cycles	fw
SAB1	77336.22	0.165257	Packard	3	
UGAB2	77268.92	0.190814	Packard	3	
PCS	77190.91	0.189676	Packard	3	
UGAB1	79106.91	0.223437	Wallac	3	
UGAB2	78903.73	0.146673	Wallac	3	
PCS	78966.62	0.170525	Wallac	3	
UGAB1	79150.64	0.220805	Wallac	3	
UGAB2	78779.36	0.191218	Wallac	3	
PCS	78884.99	0.225582	Wallac	3	
UGAB1	77130.63	0.147437	Beckman	3	
UGAB2	76943.44	0.166699	Beckman	3	
PCS	77015.76	0.177814	Beckman	3	
UGAB1	77365.2	0.184959	Packard	5	
UGAB2	77036.89	0.213024	Packard	5	
PCS	77172.07	0.188474	Packard	5	

H-3			
PCS	15251.53	16.78472	Packard
	15172.27	16.74483	Wallac
	15242.99	16.08561	Wallac
	20638.06	14.53723	Beckman
	15027.26	16.59624	Packard

conclude independent of composition
(expect for α 's & h_i β 's)

• difference between counters due to resolve time differences (not thresholds - see ^{213}Po data)

2.2 % diff.

same

	Wallac	Packard	Beckman
PCS	78.97	77.19	77.02
	78.88	77.17	
UGAB1	79.11	77.34	77.13
UGAB2	79.15	77.37	
UGAB3	78.90	77.27	76.94
UGAB4	78.78	77.04	

if assume Packard & Beckman see ^{213}Po

then

$$\frac{77.14}{6.85} = 11.3 \text{ Bq/g}$$

LS first crude cut

Difference of 10 & 30 μs could account for observed 2.2 % counter difference

15.17
15.24

15.25
15.02

20.6

PCS
214

Ken's dilution of Oak Ridge activity → **11.25** Bq g⁻¹

LS eff guess & use Pack & Beck data
only assumes resolve time > 40 μs → **11.3**

Ryan's
first cut →  → **11.2**

PC / pt sources
approximation
if see all ²¹³Po → **11.1**

Far, far away from being
done

No corrections yet

