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ANL Activities in Support of the IAEA-CRP on "Updated Decay Data Library for Actinides"

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2008 USNDP Meeting, BNL, November 5-7, 2008

3rd IAEA-CRP Meeting

❑ October 8-10, 2008, IAEA, Vienna

Program Officer – **Mark A. Kellett, *IAEA***

M.-M. Be (*France*)

V.P. Chechev (*Russian Federation*)

X. Huang (*PR China*)

F.G. Kondev (*USA*)

A. Luca (*Romania*)

G. Mukherjee (*India*)

A.L. Nichols (*IAEA*)

A.K. Pearce (*UK*)

D.H. Abriola (*IAEA*) (observer)

IAEA

What was discussed at the meeting

- status of evaluations carried out by IAEA-CRP members
  - ✓ presentations were given by all participants
- review of evaluation procedures & rules
- allocation & re-allocation of nuclei
- final report & publications
- list of actions

Current status (October 2008)

Participant	Actinides	Decay daughters
A. Luca	^{234}Th, ^{236}U	^{211}Bi, ^{211}Po, ^{228}Ra
A. L. Nichols	^{228}Th, $^{242, 242m, 244, 244m}$Am	^{208}Tl, ^{212}Pb, $^{212, 215}$Bi, $^{212, 216}$Po, $^{211, 219}$At, ^{219,220}Rn, ^{224}Ra
A. Pearce	^{232}Th, ^{231}Pa, ^{232}U	^{228}Ac, ^{223}Ra
F. G. Kondev	$^{243, 245, 246}$Cm	^{206}Hg, $^{206, 207, 209}$Tl, $^{209, 211}$Pb
G. Mukherjee	^{229}Th, ^{233}U	
M.-M. Bé	^{243}Am, $^{234, 238}$U, ^{252}Cf	^{210}Tl, $^{210, 214}$Pb, $^{210, 214}$Bi, $^{210, 214, 218}$Po, ^{218}At, ^{218,222}Rn, ^{226}Ra
V. P. Chechev	^{233}Th, ^{233}Pa, $^{237, 239}$U, $^{236, 236m, 237, 238, 239}$Np, $^{238, 239, 240, 241, 242}$Pu, ^{241}Am, $^{242, 244}$Cm	^{227}Ac
Huang Xiaolong	^{231}Th, $^{234, 234m}$Pa, ^{235}U	$^{221, 223}$Fr, ^{217}At, ^{217}Rn, ^{213}Bi, ^{213}Po, ^{225}Ra, ^{225}Ac
Unallocated		^{215}Po, ^{215}At

19 completed
17 in progress
40

18 completed
16 in progress
45

Outline of the FY08 ANL activities

- What has been done following discussions held at the last IAEA-CRP meeting:

  - **Measurements:**

    ✓ lifetime measurements for long-lived MA nuclides
    ✓ α–decay & γ–ray measurements of ^{243}Cm
    ✓ γ–ray measurements of ^{233}Pa and ^{237}Np

  - **Evaluations:**

    ✓ Status of assigned evaluations to ANL

- What we plan to do in the (near) future

Half-lives of long-lived MA nuclides

- using mass-separated sources of long-lived Cm and Cf isotopes; the in-growth technique in conjunction with α– decay spectroscopy

$$^{244}Cm \xrightarrow[18\ y]{\alpha} {}^{240}Pu \xrightarrow[6545\ y]{\alpha} {}^{236}U$$

I. Ahmad et al., NIM **A579** (2007) 458

R(^{240}Pu/^{244}Cm)=0.6860 (11) %

$T_{1/2}$(^{244}Cm)=18.11 (3) y

$T_{1/2}$(^{240}Pu) = 6545 (19) y

$$^{249}Cf \xrightarrow[351\ y]{\alpha} {}^{245}Cm \xrightarrow[8245\ y]{\alpha} {}^{241}Pu$$

F.G. Kondev et al., Proc. ND2007 p. 93

R(^{245}Cm/^{249}Cf) = 0.2474 (20)%

$T_{1/2}$(^{249}Cf) = 350.6(21) y

$T_{1/2}$(^{245}Cm) = 8245(70) y

$$^{250}Cf \xrightarrow[13\ y]{\alpha} {}^{246}Cm \xrightarrow[4706\ y]{\alpha} {}^{242}Pu$$

F.G. Kondev et al., Appl. Rad. & Isot. **65** (2007) 335

R(^{246}Cm/^{250}Cf)=0.9359 (17) %

$T_{1/2}$(^{250}Cf)=13.08 (9) y

$T_{1/2}$(^{246}Cm) = 4706 (40) years

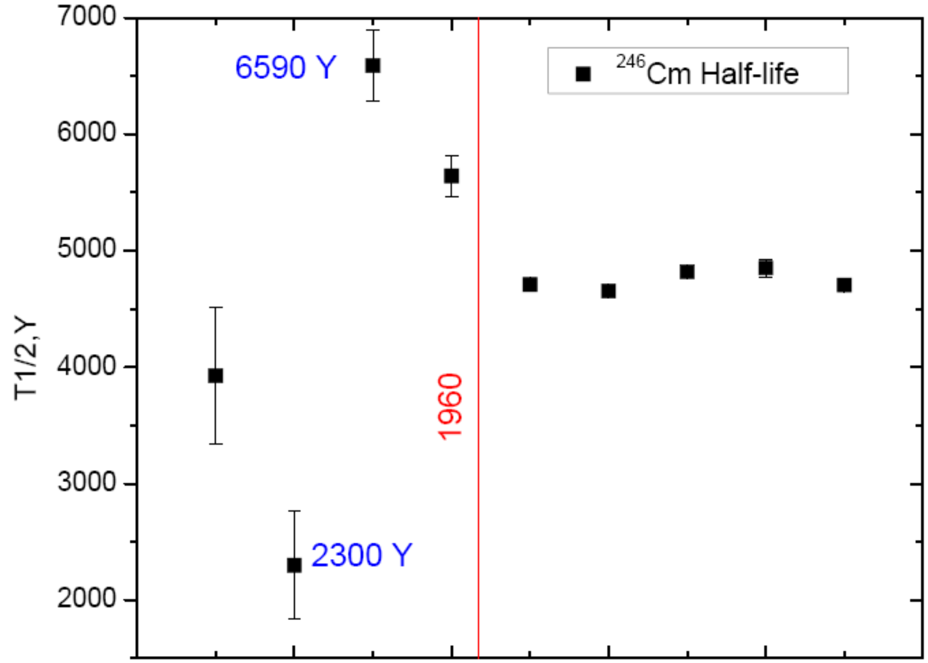
What is the impact: ^{246}Cm example

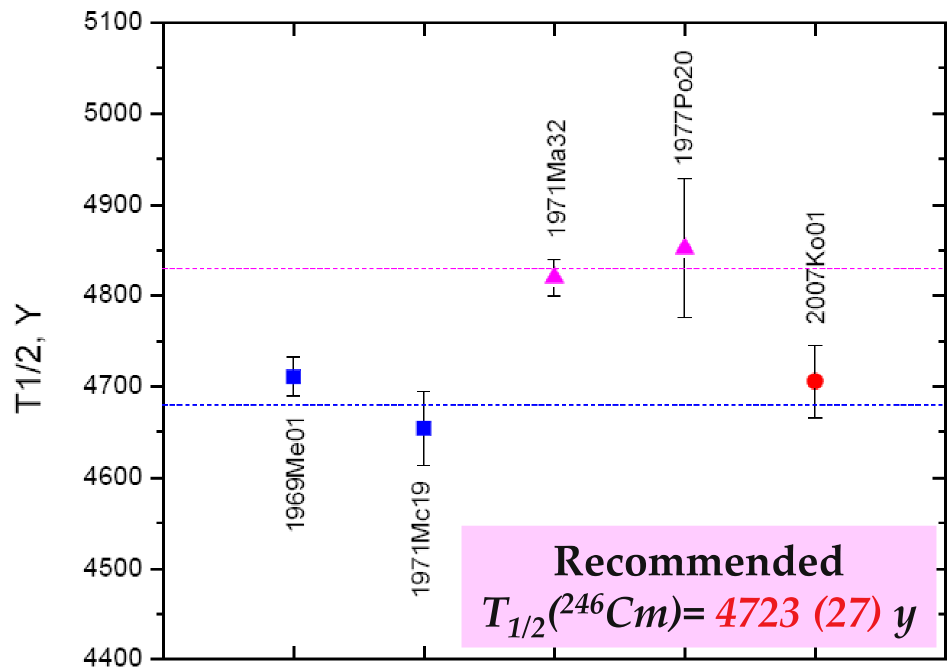
Decay data: review of measurements, evaluations and compilations

A.L. Nichols*

Applied Radiation and Isotopes 55 (2001) 23–70

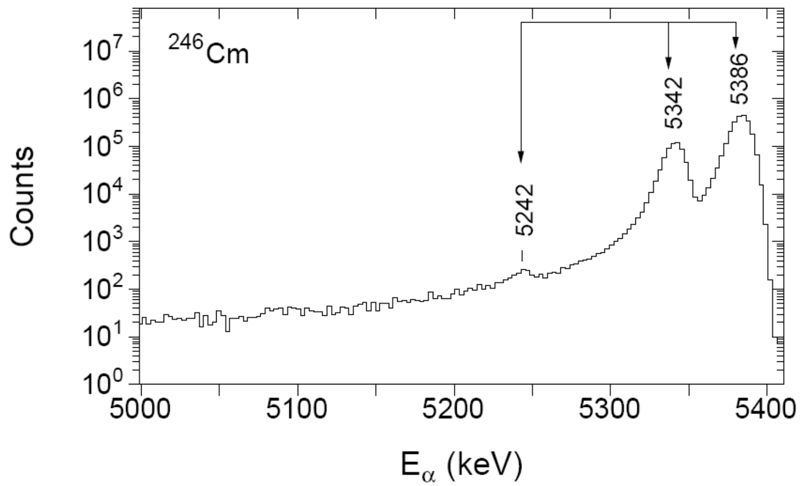
need to know $T_{1/2}(^{246}Cm)$ with accuracy better than 1%



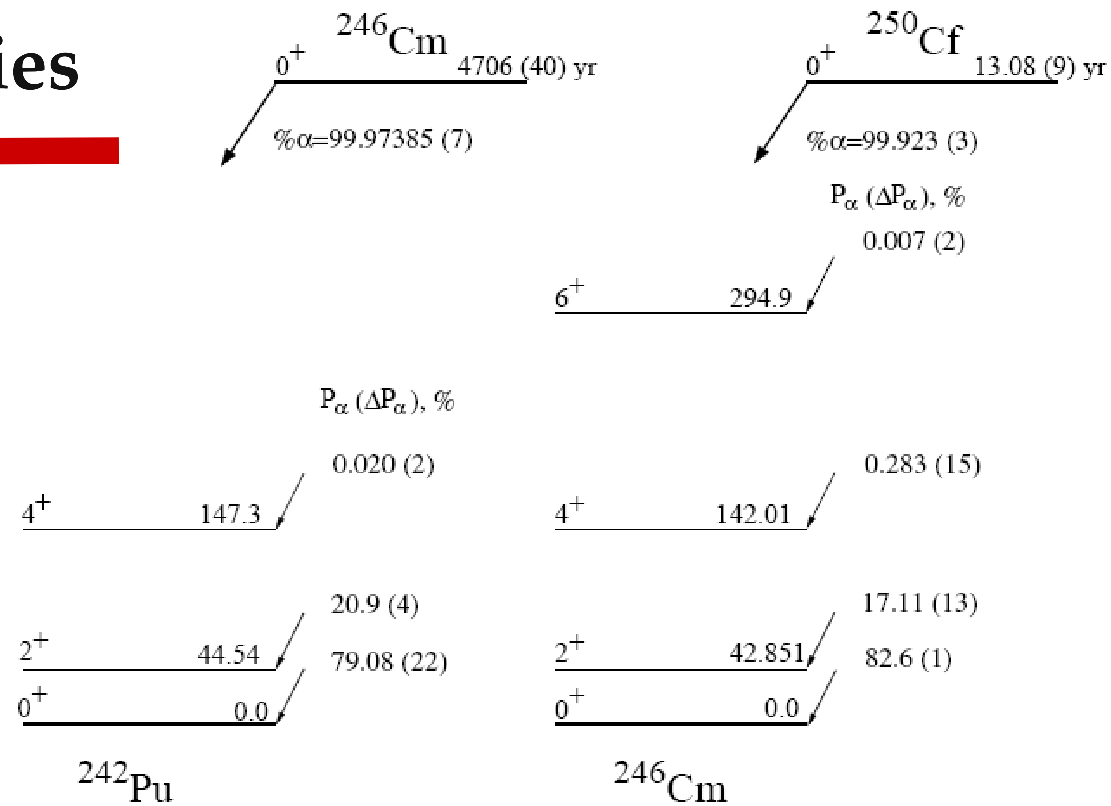


the goal has been achieved!

α–Emission Probabilities



F.G. Kondev et al., Appl. Rad. & Isot. **65** (2007) 335



	$\alpha_{0.0}$		$\alpha_{44.5}$		$\alpha_{147.3}$	
Author	E_α, keV	P_α, %	E_α, keV	P_α, %	E_α, keV	P_α, %
Belov et al. (1963)	5387	78	5345	22	–	–
Dzhelepov et al. (1963)	5387 (4)	78 (5)	5345 (5)	22 (5)	–	–
Baranov et al. (1966)	5385	79	5342	21	–	–
Shatinskii (1984)	5386.5 (10)	82.2 (12)	5343.5 (10)	17.8 (12)	–	–
Present work	5386 (3)	79.08 (22)	5342 (3)	20.9 (4)	5242 (3)	0.020 (2)

α–Emission probabilities – cont.

- there seems to be systematic differences in α–emission probabilities measured using semiconductor detectors (e.g. Si(LI) and PIPS) and magnetic spectrometers
  - ✓ I. Ahmad, NIM A223 (1984) 319; A. Koua Aka et al., NIM A369 (1996) 477; E. Garcia-Torano et al., NIM A550 (2005) 581 and others

- need to understand these discrepancies: α– and γ–ray decay spectroscopy studies of ^{243}Cm
  - ✓ α–emission probabilities – using both Passivated Implanted Planar Silicon (PIPS) detectors and double-focusing magnetic spectrometer
  - ✓ γ–emission probabilities – using large Ge and LEPS detectors, including conversion electrons studies

^{243}Cm α–Emission probabilities – cont

- **magnetic spectrometer:** data collected in mid 1970's using the Argonne double-focusing magnetic spectrometer

I. Ahmad et al., Nucl. Phys. A208 (1973) 287

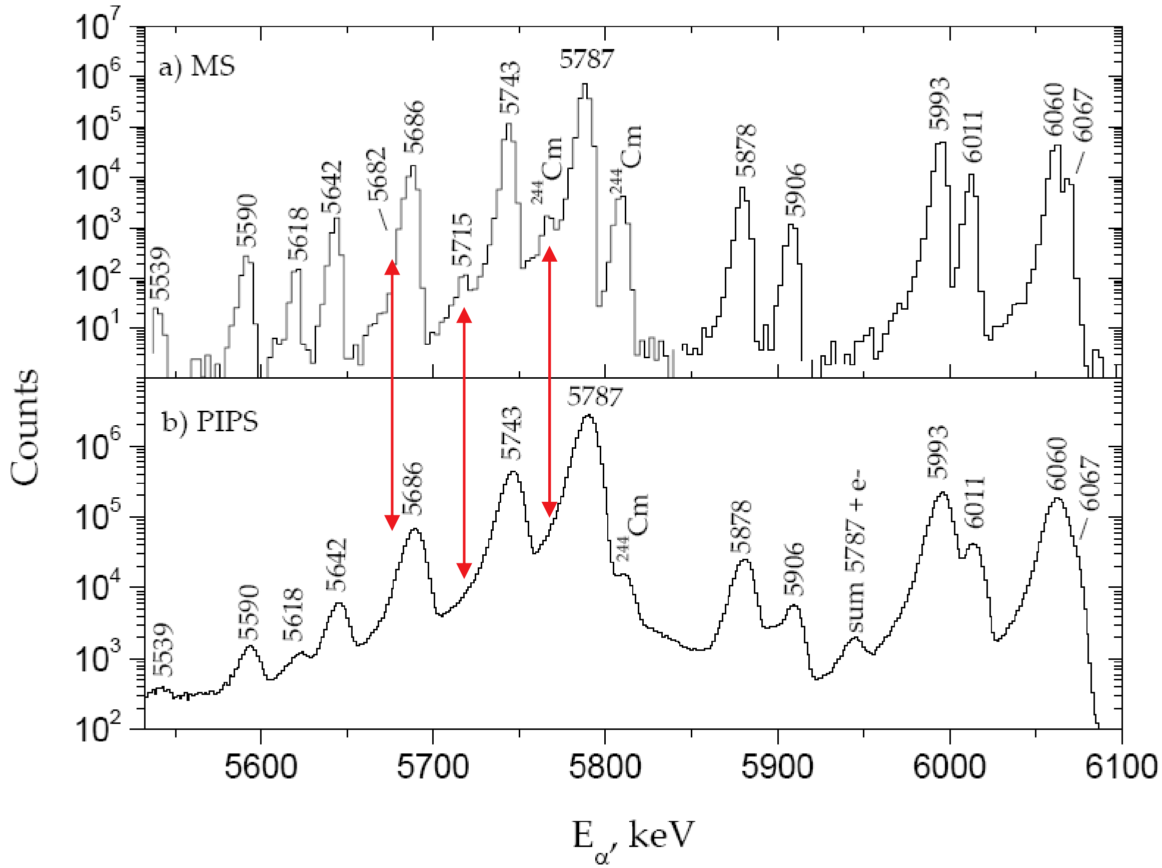
  - ✓ energy resolution (FWHM) of **5 keV**
  - ✓ transmission efficiency of Ω=0.1 % for 6 MeV α–particles

- **semiconductor detectors:** Passivated Implanted Planar Silicon (PIPS) detector (25 mm^2 active area)

  - ✓ energy resolution (FWHM) of **12 keV**
  - ✓ small geometrical efficiency of Ω=0.225% in order to minimize α–e-coincidence summing effects

^{243}Cm α–Emission probabilities – cont

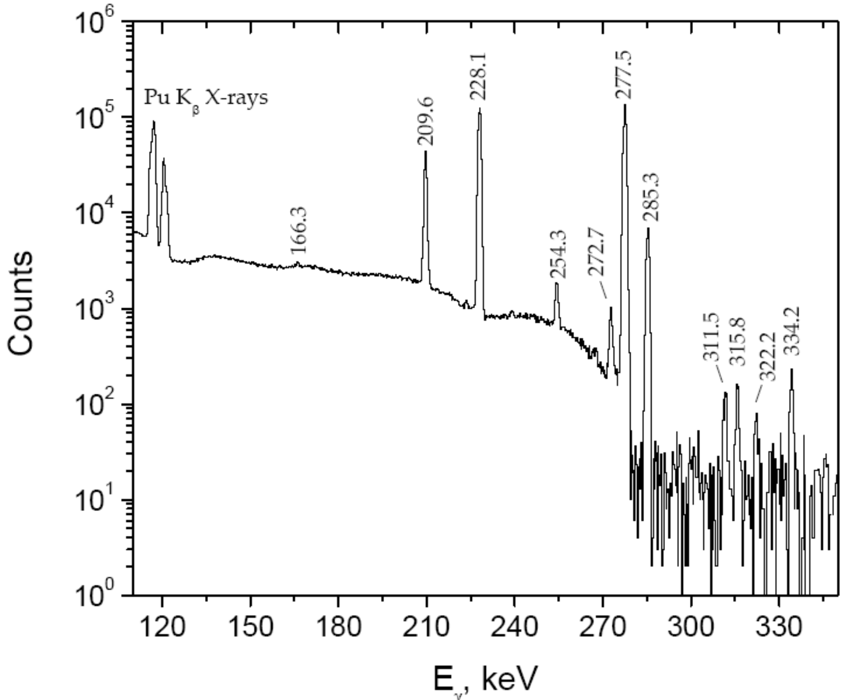
E_α (keV)	P_α (%)	
	MS	PIPS
5539 (1)	0.003 (1)	0.005 (2)
5590 (1)	0.029 (2)	0.034 (4)
5618 (1)	0.016 (2)	0.025 (4)
5642 (1)	0.125 (6)	0.17 (2)
5682 (2)	0.29 (2)	–
5686 (1)	1.52 (5)	1.8 (1)
5715 (2)	0.016 (2)	–
5743 (1)	11.1 (2)	11.6 (4)
5787 (1)	74.2 (8)	72.9 (12)
5878 (2)	0.64 (3)	0.69 (4)
5906 (1)	0.136 (6)	0.160 (15)
5993 (1)	5.6 (2)	5.8 (2)
6011 (1)	1.05 (5)	1.1 (2)
6060 (1)	4.3 (2)	4.5 (3)
6067 (1)	0.96 (5)	1.1 (1)



past discrepancies observed can most likely be attributed to differences in the data analysis procedures, rather than to the inherent applicability and nature of the two methods

Absolute γ–ray emission probabilities

Measurements of the absolute γ–ray intensities per 100 α decays of ^{243}Cm



❑ **α counting** – using a 450-mm² PIPS detector

✓absolute geometrical efficiency of Ω=0.5119 (17)%, determined using ^{243}Am and ^{249}Cf mass-separated sources of known activity

E_γ (keV)	P_γ (photons per 100 α decays of ^{243}Cm)
209.6	3.25 (8)
228.1	10.4 (2)
254.3	0.116 (5)
272.7	0.086 (4)
277.5	14.0 (2)
285.3	0.76 (2)

❑ **γ–ray counting**: using 2-cm³ LEPS (FWHM = 0.5 keV at 122.06 keV) and 25% Ge (FWHM = 1.8 keV at 1332.49 keV) detectors

✓ absolute efficiency using a calibrated mix source ^{57,60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs, ^{139}Ce, ^{203}Hg and ^{241}Am, and isotopically pure ^{152}Eu, ^{182}Ta and ^{243}Am

^{233}Pa γ–ray emission probabilities

- it has been of a special interest since the first IAEA-CRP (1977)
  - ✓ very high-precision measurement on P_γ (312 keV), e.g. =38.6 (5) % (Gehrke et al.), 38.5 (4)% (Schotzig et al.), 38.65 (39) % (Vaninbroukx et al.), 38.7 (4) % (Woods et al.), 37.80 (23) % (Luca et al.), BUT …

    41.6 (9) % (Harada et al. J. Nucl. Sci. and Techn. 43 (2006) 1289)
- at the last two meetings inconsistencies for P_γ(28.6 keV) were pointed out

E_γ/keV	P_γ (%)									
	Albridge et al. (1961)	Valkeapaa et al. (1973)[a]	Gehrke et al. (1979)	Vaninbroukx et al. (1984)	Kouassi et al. (1990)[a]	Luca et al. (2000)	Schotzig et al. (2000)	Woods et al. (2000)	Luca et al. (2002)	Shchukin et al. (2004)
28.559(10)		0.070(8)		0.15(1)	0.075(8)	0.034(10)			0.034(10)	0.019(2)

Why is important? – 28.6 keV (M1+E2) transition determines the β– feeding to the 5/2+, 340 keV level:

$$P_{\gamma,tot}(28.6\ \text{keV})=P_\gamma(28.6\ \text{keV})*(1+\alpha_T)=P_\gamma*\mathbf{311!}$$

- there are differences between various measurements
- there are differences between various evaluations, e.g. ENSDF vs. DDEP
- there has been a lot of effort in the past, but the decay scheme is still "discrepant"

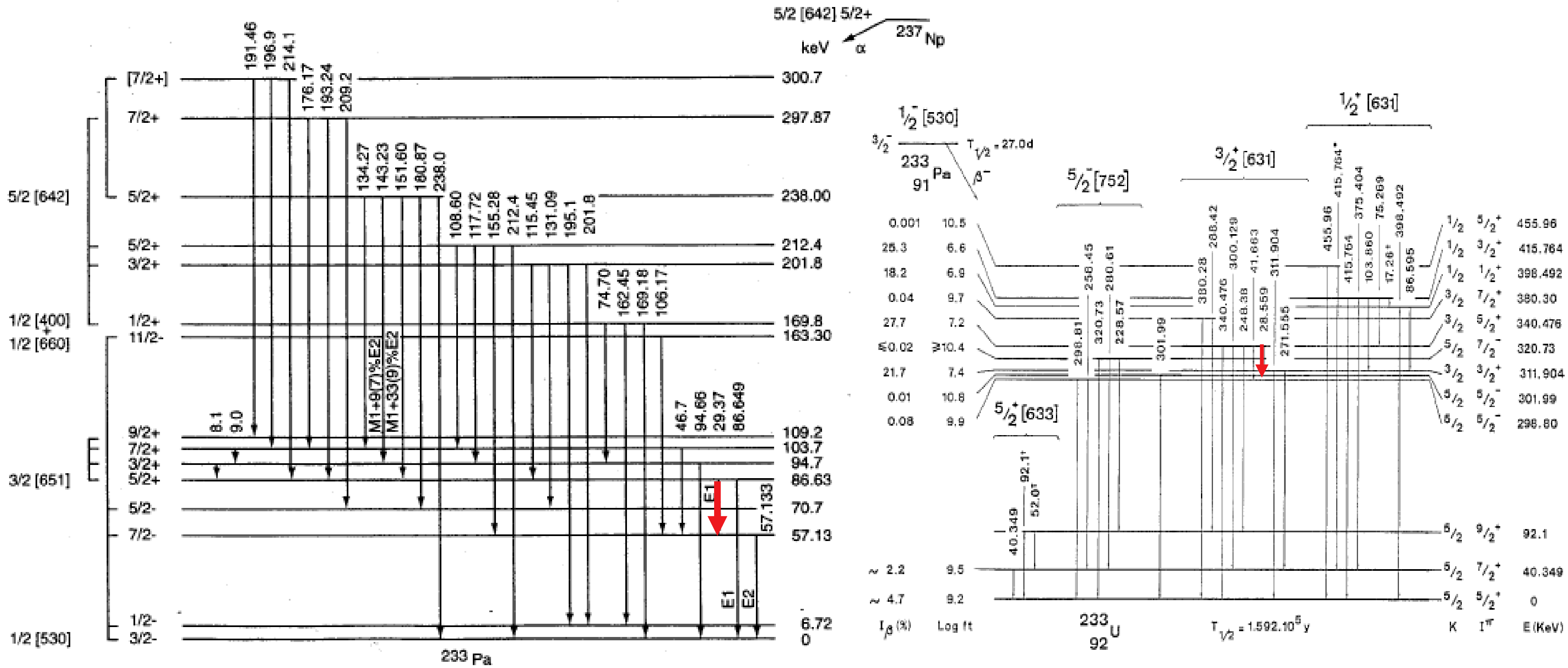
^{233}Pa γ–ray emission probabilities - cont

- There are two ways to produce ^{233}Pa:
  - ✓ ^{232}Th(n,γ)^{233}Th(β–)^{233}Pa(β–)^{233}U
  - ✓ ^{237}Np(α)^{233}Pa(β–)^{233}U

E_γ/keV	P_γ (%)									
		Th		Np	Th	Np			Np	Np
	Albridge et al. (1961)	Valkeapaa et al. (1973)[a]	Gehrke et al. (1979)	Vaninbroukx et al. (1984)	Kouassi et al. (1990)[a]	Luca et al. (2000)	Schotzig et al. (2000)	Woods et al. (2000)	Luca et al. (2002)	Shchukin et al. (2004)
28.559(10)		0.070(8)		0.15(1)	0.075(8)	0.034(10)			0.034(10)	0.019(2)

^{233}Pa γ–ray emission probabilities - cont

- ^{237}Np/^{233}Pa in equilibrium – potential problems
  - ✓ 29.37 keV E1 (^{237}Np) & 28.557 keV (M1+E2) (^{233}Pa)

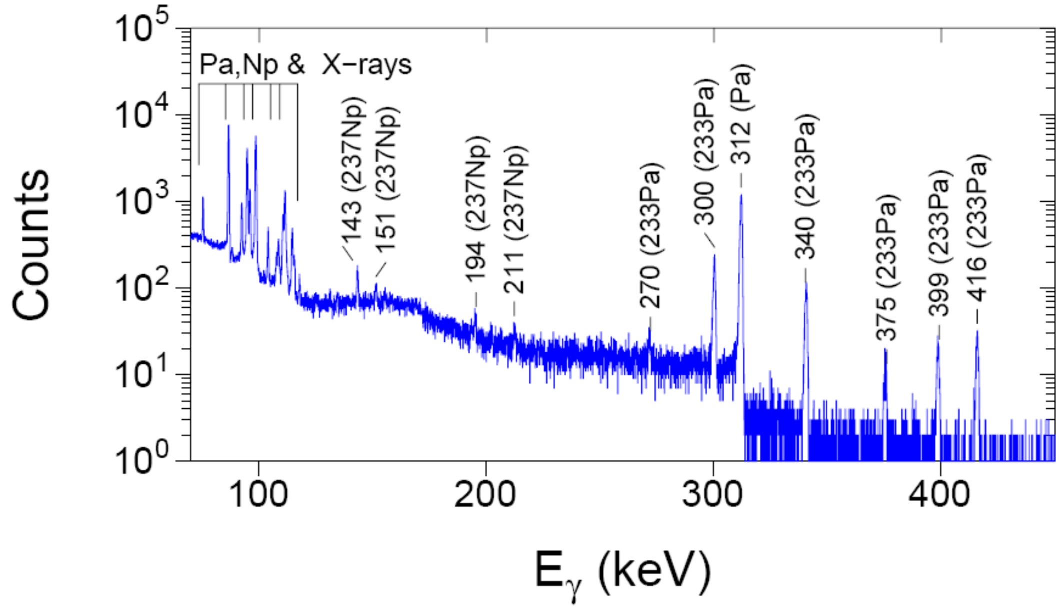


^{233}Pa γ–ray emission probabilities - cont

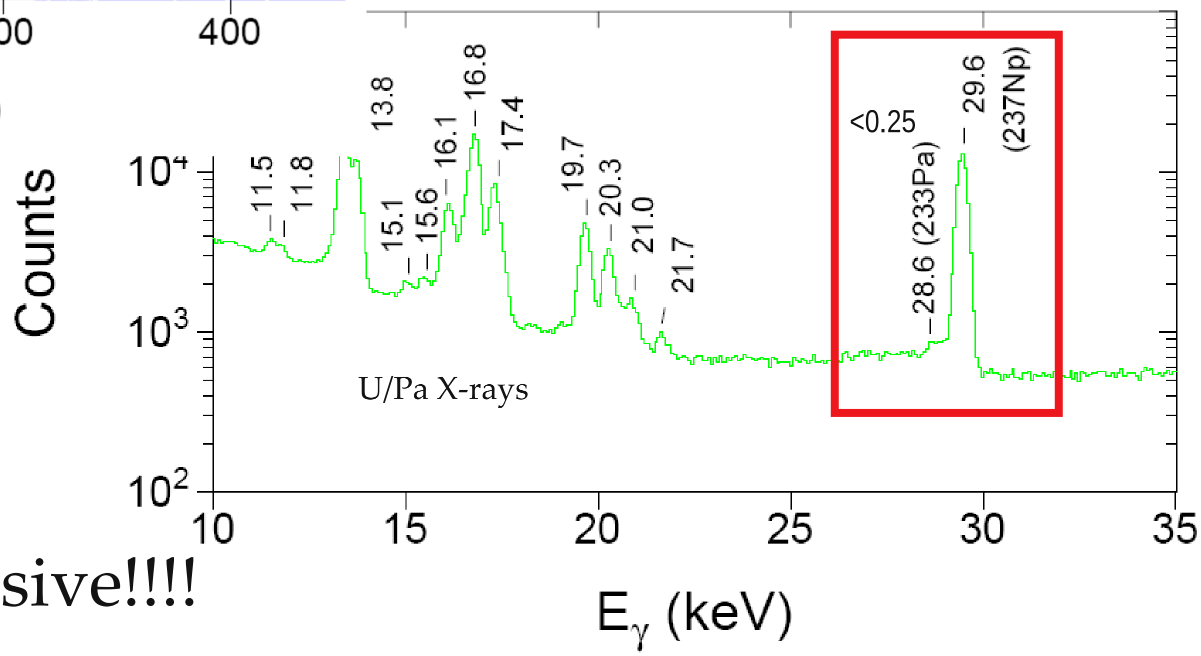
initial measurements at ANL (2006)

- using a mass separated ^{237}Np source – 4 nCi (~5μg)
- several measurements in an underground laboratory
  - ✓ 2cm^2 x 1cm^2 LEPS (0.5 keV @122.06 keV)
  - ✓ 25% and 110% Ge detectors (1.2 keV @1132.51 keV)

^{233}Pa γ–ray emission probabilities - cont



- ❑ high isotopic purity



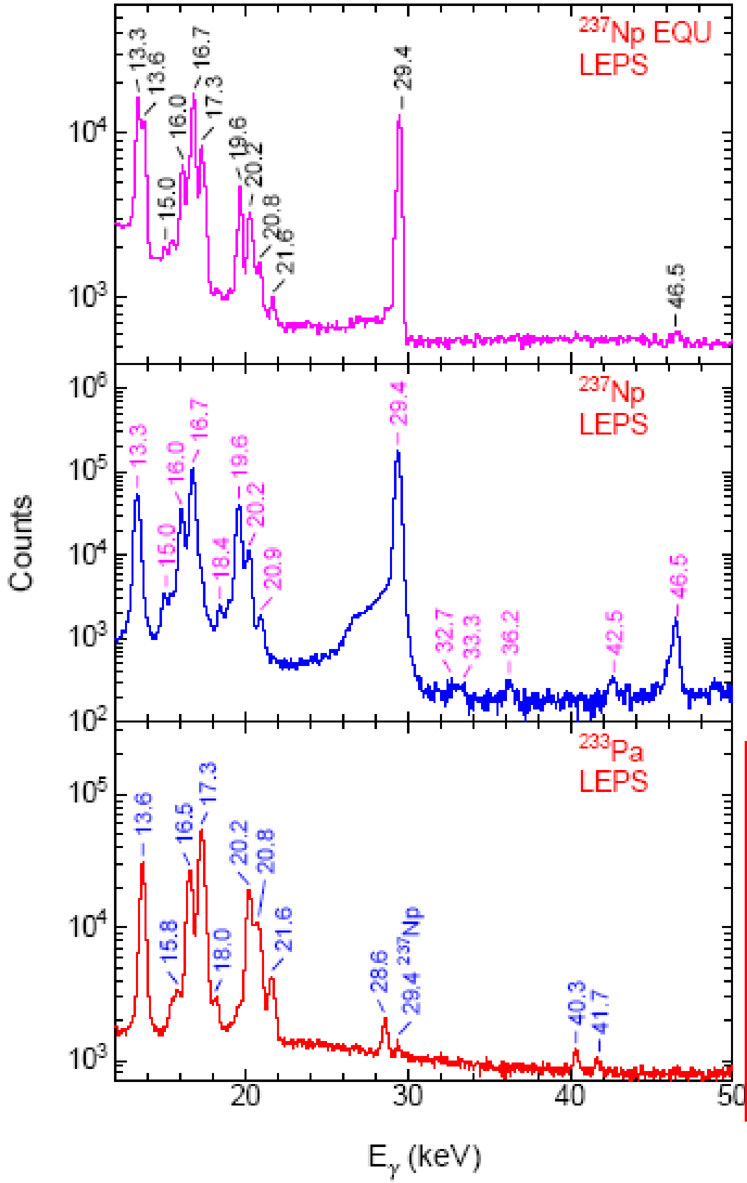
- ❑ … but, it is inconclusive!!!!

^{233}Pa γ–ray emission probabilities - cont

New measurements at ANL – (August/September 2008)

- using a chemical separation to extract ^{233}Pa from ^{237}Np
  - ✓ procedure was similar to that used by Gehrke et al. – dissolved ^{237}Np material (in equilibrium with ^{233}Pa) in HNO_3 – transferred the solution to a beaker and dry it – use 4 M of HNO_3 to dissolve ^{237}Np (but not ^{233}Pa) – repeat the procedure several times to achieve the desired purity
  - ✓ several sources were produced and measured with 3 cm^3 LEPS & 25% Ge detectors – efficiency calibration determined using a mix source ^{57,60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs, ^{139}Ce, ^{203}Hg and ^{241}Am, and isotopically pure ^{243}Am source – accuracy ~1% for low- and high-energy photons

^{233}Pa γ–ray emission probabilities - cont

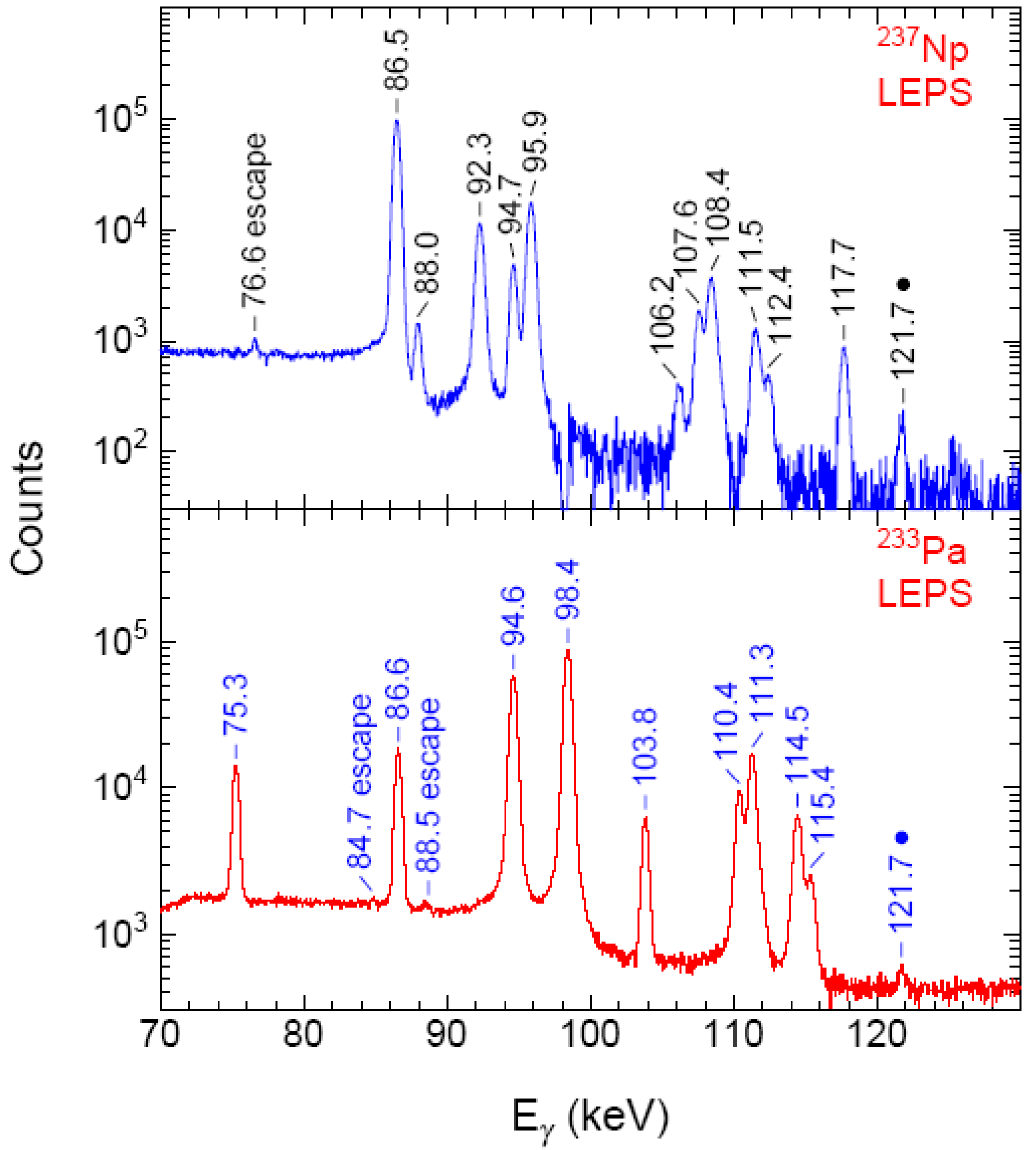


- ✓ 29.4 keV ^{237}Np line dominates – high Compton tail that masks 28.6 keV (^{233}Pa)
- ✓ Compton background associated with much stronger high-energy γ–rays of ^{233}Pa
- ✓ $P_\gamma(29.4/^{237}Np)/P_\gamma(75.3/^{233}Pa) = 10.6$ (1)

- ✓ no ^{233}Pa – 75.3 keV line (^{233}Pa) is gone – pure Pa X-rays (from decay of ^{237}Np)
- ✓ Compton background associated with the high-energy γ–rays of ^{233}Pa is reduced

- ✓ 29.4 keV ^{237}Np line is significantly reduced, e.g. $P_\gamma(29.4/^{237}Np)/P_\gamma(75.3/^{233}Pa) = 0.060$ (17)
- ✓ pure U X-rays
- ✓ S(28.6 keV)=9000 counts – statistical uncertainty of about 1%

^{233}Pa γ–ray emission probabilities - cont



- ✓ resolve 86.6 keV (^{233}Pa) and 86.5 keV (^{237}Np) lines
- ✓ resolve 94.6 keV (^{233}Pa) and 94.7 keV (^{237}Np) lines
- ✓ resolve 111.3 keV (^{233}Pa) and 111.5 keV (^{237}Np) lines

^{233}Pa γ–ray emission probabilities - cont

E_γ/keV	P_γ (%) Albridge et al. (1961)	Valkeapaa et al. (1973)[a]	Gehrke et al. (1979)	Vaninbroukx et al. (1984)	Kouassi et al. (1990)[a]	Luca et al. (2000)	Schotzig et al. (2000)	Woods et al. (2000)	Luca et al. (2002)	Shchukin et al. (2004)
28.559(10)		0.070(8)		0.15(1)	0.075(8)	0.034(10)			0.034(10)	0.019(2)

Eγ, keV	present	Valkeapaa73	Kouassi90	Gehrke79	Vanin.84	Woods88
28.57	0.076 (3)	0.068(8)	0.074 (8)		0.15 (1)	0.068 (9)
29.37	0.0169 (15)					
39.77	0.0034 (9)					
40.33	0.0228 (14)	0.039 (8)	0.024 (4)			
41.65	0.0121 (10)	0.013 (4)	0.014 (3)			
75.26	1.27 (3)	1.25 (8)	1.25 (9)	1.39 (8)	1.30 (4)	1.25 (9)
86.57	2.00 (4)	1.87 (23)	1.93 (11)	1.97 (12)		1.87 (25)
94.64	8.51 (17)					
98.42	13.70 (27)					
103.84	0.85 (2)	0.73 (8)	0.847 (6)	0.87 (3)	0.87 (3)	0.73 (9)
110.41	1.64 (3)					
111.30	3.23 (7)					
114.48	1.31 (3)					
115.38	0.423 (9)					
271.57	0.361 (12)	0.30 (3)	0.334 (17)	0.33 (1)	0.32 (1)	
300.16	6.41 (13)	6.57 (31)	6.76 (6)	6.62 (10)	6.64 (6)	6.57 (46)
311.94	38.6 (5)			38.6 (5)	38.65 (39)	

(preliminary)

^{233}Pa X-ray emission probabilities

Energy, keV	Present work		DeVries, 2008		Chechev, 2006		Kouassi, 1973	
XL								
Lη	0.25	0.01			0.282	0.013		
Lβ	18.73	0.38	26.27	0.34	18.6	0.5		
Lγ	4.03	0.08	5.21	0.08	4.34	0.12		
XK								
94.64	8.51	0.17	8.50	0.11	9.09	0.25	8.57	0.42
98.42	13.70	0.27	14.02	0.19	14.6	0.4	13.50	0.65
110.41	1.64	0.03	1.694	0.026	1.73	0.07	1.62	0.08
111.30	3.23	0.07	3.24	0.05	3.52	0.15	3.29	0.25
114.48	1.31	0.03	1.317	0.021	1.37	0.06	1.31	0.08
115.38	0.423	0.009	0.406	0.011	0.416	0.018	0.39	0.02

(preliminary)

DeVries & Griffin (Appl. Rad. & Isot. 66, 2008) – seems to be discrepant at lower energies.

Evaluation activities

- **Completed Evaluations:**
  - ✓ ^{246}Cm & ^{206}Tl: reviewed & completed
  - ✓ ^{206}Hg: completed – reviewed by E. Browne (LBNL)
- **Work in progress:**
  - ✓ ^{243}Cm and ^{245}Cm
  - ✓ ^{209}Tl & ^{209}Pb – together with G. Mukherjee (India)
- **Work not initiated, yet:**
  - ✓ ^{207}Tl & ^{211}Pb