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Status of IAEA-CRP on "Updated Decay Data Library for Actinides"

Filip G. Kondev

Outline

- Brief summary of discussions held at the 2nd IAEA-CRP meeting
- Status of IAEA-CRP (as of March 2007)
- Results from new measurements in support of evaluation activities

2nd IAEA-CRP Meeting

- 28 – 30 March 2007, INDC(NDS)-0508, IAEA, Vienna

participants

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*) - absent

G. Mukherjee (*India*)

A.L. Nichols (*IAEA*)

A.K. Pearce (*UK*)

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

What was discussed

- status of evaluations carried out by IAEA-CRP members
  - ✓ presentations were given by all participants
- status of measurements effort in support of IAEA-CRP
  - ✓ presentation from ANL
  - ✓ discussions for future measurements activities with participants from other laboratories (UK & India)
- review of evaluation procedures & rules
- allocation & re-allocation of nuclei
- list of actions

Presentations

Marie-Martine Bé, Vanessa Chisté, Christophe Dulieu
CEA Saclay /LNHB

Nuclide	Status
Cf-252	In progress
Am-243	
U-238	Done
U-234	Done
Ra-226	Review in progress
Rn-222	Review in progress
Po-218	Review in progress
*At-218	Review in progress
*Rn-218	Review in progress
Po-214	Review in progress
Bi-214	Review in progress
Pb-214	Review in progress
Po-210	
Bi-210	
Pb-210	Review in progress
*Tl-210	Review in progress

Presentations – cont.

Valery Chechev
V.G. Khlopin Radium Institute, 194021 Saint Petersburg, Russia

Nuclide	Status of evaluation
U-237	Completed, placed on the DDEP site
U-239	Completed, prepared for the DDEP site
Np-236	Completed, placed on the DDEP site
Np-237	In progress
Np-238	Completed, placed on the DDEP site
Np-239	Completed, minor update to be done
Pu-239	Completed, prepared for the DDEP site
Pu-241	Completed, placed on the DDEP site

discrepancies for ^{239}U and ^{236}Np were noted; also lack of confirmatory measurements for ^{237}U half-life

Presentations – cont.

- X. Huang
  - ✓ ^{213}Bi & ^{225}Ac
- F.G. Kondev
  - ✓ ^{206}Tl & ^{246}Cm
- G. Mukherjee
  - ✓ ^{229}Th & ^{233}U
- A.L. Nichols
  - ✓ ^{242}Am & ^{244}Am
- A. Pearce
  - ✓ ^{232}U & ^{228}Ac

IAEA

IAEA-CRP on
"Updated Decay Data Library for Actinides"

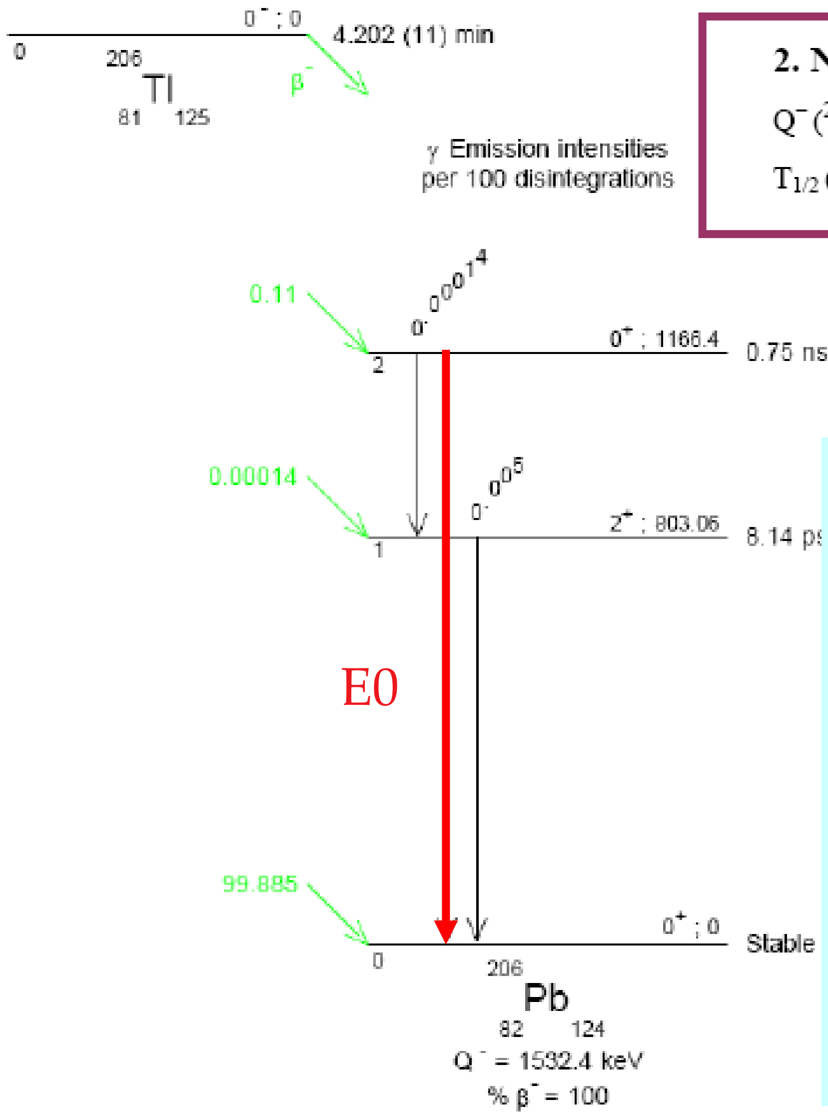
Current status (March 2007)

Participant	Actinides	Decay daughters
A. Luca	^{234}Th, ^{236}U	^{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, ^{235}U	$^{221, 223}$Fr, ^{217}At, ^{217}Rn, ^{213}Bi, ^{213}Po, ^{225}Ra, ^{225}Ac
Unallocated		^{211}Bi, $^{211, 215}$Po, ^{215}At

17 completed
8 in progress
38

1 completed
14 in progress
45

^{206}Tl – ANL evaluation



2. NUCLEAR DATA

Q^- (^{206}Tl): 1532.4 (6) keV

$T_{1/2}$ (^{206}Tl): 4.202 (11) min

Produced by:

^{205}Tl (n,γ)

^{210}Bi α–decay

(d,p), (t,α) & (d,α)

- decays mainly (99.89%) to the 0+ GS of ^{206}Pb
- a weak (0.11%) branch to the 0+ excited state at 1166.4 keV
- E0 transition to the 0+ GS of ^{206}Pb – accurate knowledge of absolute X-ray intensities is important for applications involving γ–ray spectrometry

^{206}Tl γ–ray emission probabilities

Table 4 Experimental and evaluated γ–ray emission probabilities.

Authors	$P_{\gamma1,0}$, %	$P_{XK}(\gamma2,0)$ % [a]	$P_{\gamma2,1}$, %	Comment [b]
1968Zo02	0.0055 (5)			Not used
1970Zo02	0.0055 (4)			Expt.
1972CoYX	0.0041 (6)	0.08 (2)	<0.00026	Expt.
1972Gr01	0.004 (1)	0.10 (2)	<0.001	Expt.
Adopted	**0.0050 (3)**	**0.09 (1)**	**0.00013 (13)**	**Evaluated**

[a] Absolute KX-ray yield
[b] Expt. – experimental value used in the present evaluation. The 1968Zo02 value is superseded by 1970Zo02

γ2,0 is a pure E0 transition, so Pγ(γ2,0)=0.000!!!

$$P_{\gamma+ce}(\gamma 2,0) = P_{ce}(\gamma 2,0) = \left(P_{KX}(\gamma 2,0)/\omega_K\right)/(K/T) = 0.110(14)\%$$

Fluorescence yield: ω_K=0.963 (4)

K/T=0.85 (6) from K/L=5.7 (4) – weighted mean of 5.61 (38) (1990Tr01) and 6 (1) (1977Dr08) – note using electronics Ω_K(E0) and Ω_L(E0) factors from BrICC
K/T=0.855 (excellent agreement –independent test for BrICC!!!)

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^{206}Tl X–ray probabilities

5.1 X-Ray Emissions

The X-ray yield in β^- decay of ^{206}Tl is produced entirely in the decay of the 1166.4 keV (E0, $0^+ \to 0^+$) transition. Contributions from the much weaker 803.06 and 363.3 keV transitions can be neglected, since their X-ray yields are several orders of magnitude smaller than that of the 1166.4 keV transition.

For the 1166.4 keV E0 ($0^+ \to 0^+$) transition, the number of vacancies in the K-shell per 100 disintegrations was determined as:

$$N_K = P_{ceK} = P_{XK} / \omega_K = 0.090(10) / 0.963(4) = 0.093(11)\,.$$

The corresponding number of vacancies in the L shell per 100 disintegrations was then determined as:

$$N_L = P_{ceL} + n_{KL} \times N_K = 0.0163(22) + 0.811(5) \times 0.093(11) = 0.092(11)\%$$

where $P_{ceL} = P_{ceK} / (K / L) = 0.0163(22)\%$ with K/L=5.7 (4), a weighted mean of 5.61 (38) (1990Tr01) and 6 (1) (1977Dr08). The number of X-rays per 100 disintegrations was then calculated as:

$$P_{XK} = \omega_K \times N_K \text{ and } P_{XL} = \tilde{\omega}_L \times N_L$$

^{206}Tl X-ray data

	E_X, keV	P_X, per 100 disintegrations			
		CRP	ENDF/B-VI	ENDF/B-VII	JEFF3.1
X_K		0.090 (10)			
$K\alpha_2$	72.8049	0.026 (3)	0.022 (5)	1.26 (24) E-5	1.38 (14) E-5
$K\alpha_1$	74.97	0.044 (5)	0.037 (8)	2.1 (4) E-5	2.33 (23) E-5
$K\beta_3$	84.451			2.5 (5) E-6	2.7 (3) E-6
$K\beta_1$	84.937	0.0150 (17)	0.013 (3)	4.9 (9) E-6	5.2 (5) E-6
$K\beta_5$	85.47				
$K\beta_2$	87.238		0.0032 (7)	1.8 (3) E-6	2.4 (3) E-6
$K\beta_4$	87.58	0.0045 (6)			
$KO_{2,3}$	87.911				

similar discrepancies exist for electron (Auger & CE) data

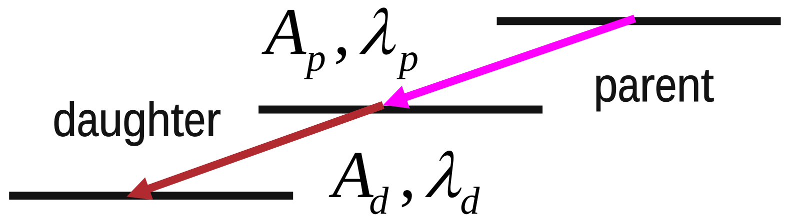
Measurements – discussed at 1st RCM

- lifetimes and emission probabilities for a number of Cm isotopes
  - ✓ ^{243}Cm, ^{244}Cm, ^{245}Cm and ^{246}Cm
- gamma-ray emission probabilities for ^{233}Pa
  - ✓ to resolve discrepancies between previous measurements for the 28.557 keV line
- future measurements - driven by evaluators' requests
  - ✓ a list of mass separated sources available at ANL
  - ✓ time & resources consuming

"DO NOT WASTE VALUABLE RESOURCES ON THE MEASUREMENT OF NUCLEAR PARAMETERS OF LITTLE OR NO CONCERN" **– A.L. Nichols**

Half-life of a very long-lived nuclide

$$T_{1/2} = \ln 2 \times \frac{N}{A}$$



- must know absolute efficiencies, and hence, the uncertainties are potentially large, especially systematics ones!

$^{250}Cf \xrightarrow[13\ y]{\alpha} {}^{246}Cm \xrightarrow[4747\ y]{\alpha} {}^{242}Pu \xrightarrow[3.810^{5}\ y]{\alpha}$

$$T_{1/2}(n) = T_{1/2}(ref) \times \frac{A(ref)}{A(n)} \times \frac{N(n)}{N(ref)}$$

$$\frac{A_p(t)}{A_d(t)} = \frac{\lambda_d}{\lambda_d - \lambda_p}(1 - e^{-(\lambda_d - \lambda_p)t})$$

- usually relative methods have been used in order to avoid the accurate quantification of absolute efficiencies
- must know with a good accuracy $T_{1/2}$(ref) (many old values – need to recalibrate)
- must know $T_{1/2}$ of the parent nuclide with good accuracy
- ANL has access to many mass-separated sources of long-lived Pu, Am, Cm and Cf isotopes

^{240}Pu (test)

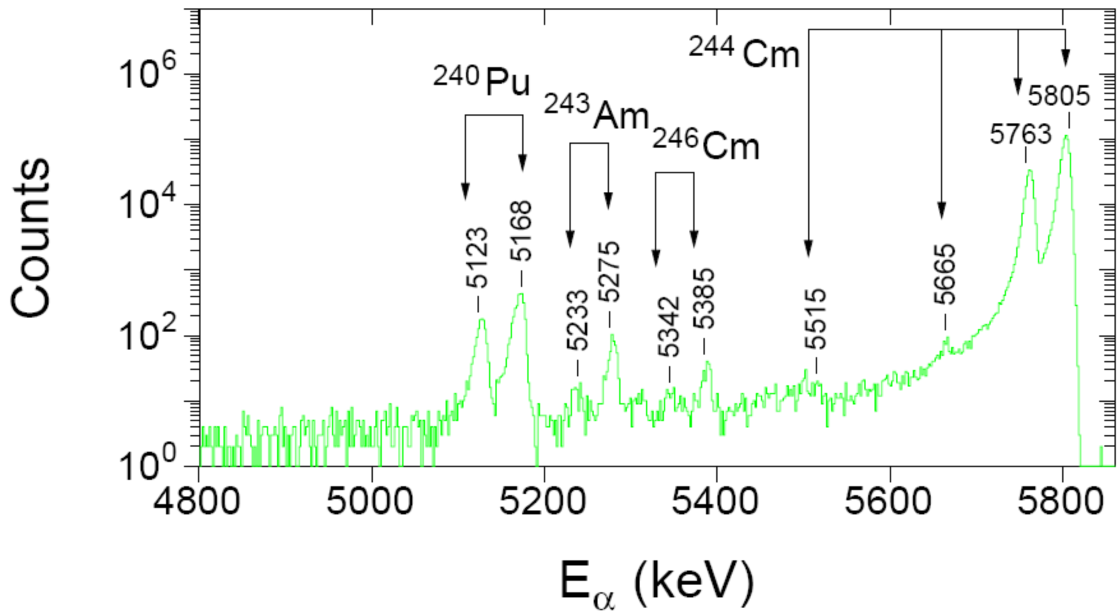
$^{244}Cm \xrightarrow[18\ y]{\alpha} {}^{240}Pu \xrightarrow[6545\ y]{\alpha} {}^{236}U$

t_{growth}=32.6 *y*
$t_{measure}$=52.6 *h* (Ω=0.73%)
$t_{measure}$=24.5 *d* (Ω=6.0%)

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



6561 (7) y
(ENSDF)

US Half-life Committee
6564 (11) *y*

Summary of previous measurements of ^{240}Pu half-life.

Half-life (years)[1)]	Method used for the measurement [2)]	Reference
6569±6	Specific activity (MS + α activity)	Jaffey *et al.*, 1978 [6]
6574±7	(Ingrowth of daughter ^{236}U)	Beckman *et al.*, 1984 [7]
6571±7	Specific activity (MS + α activity)	Steinkruger *et al.*, 1984 [8]
6552.2±2.0	Specific activity (MS + α activity)	Lucas and Noyce, 1984 [9]
6552.4±1.7	Calorimetric homogeneity	Rudy *et al.*, 1984 [10]

1) 1σ uncertainties are given.
2) MS=mass spectrometer.

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

^{246}Cm

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Decay data: review of measurements, evaluations and compilations

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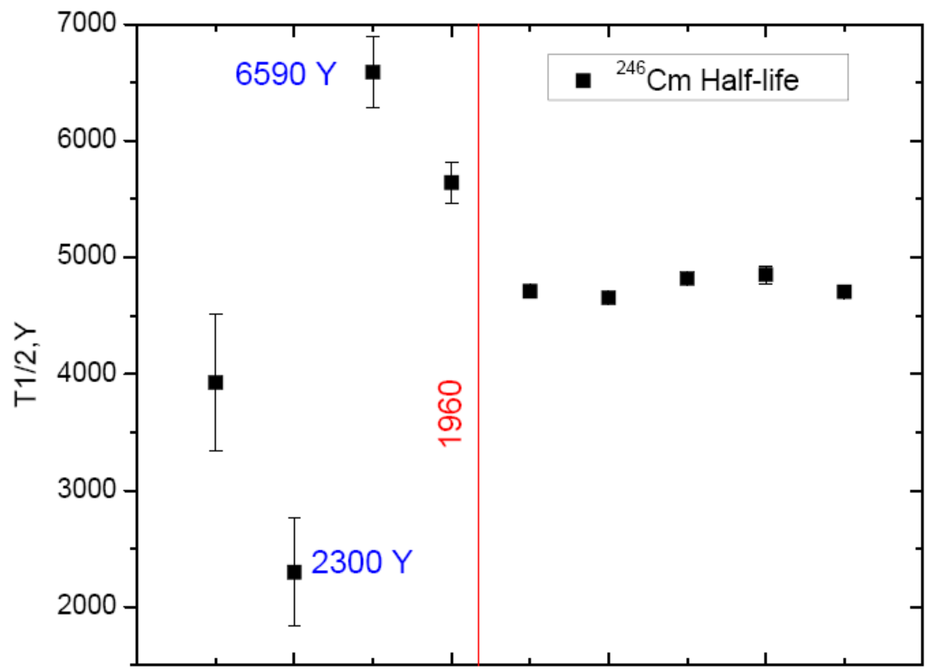
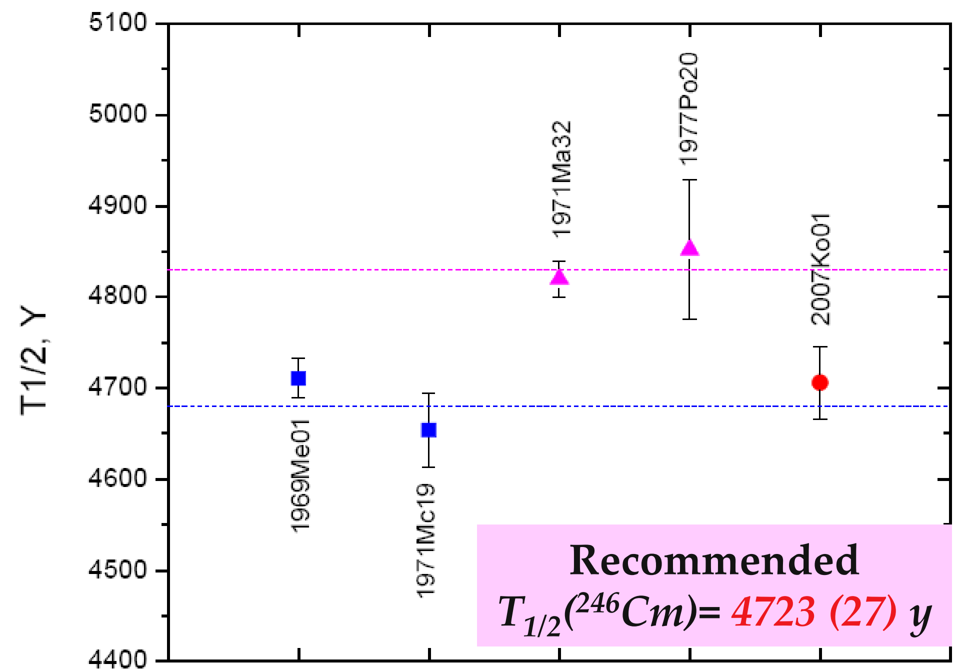
need to know $T_{1/2}(^{246}Cm)$ with accuracy better than 1%





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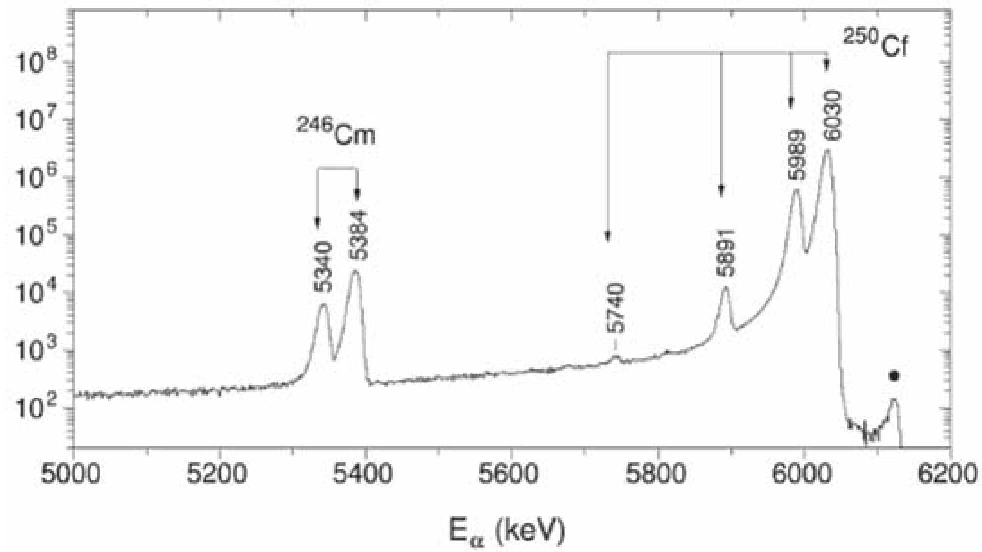
Measurements of the half-life of ^{246}Cm and the α-decay emission probabilities of ^{246}Cm and ^{250}Cf

F.G. Kondev[a,*], I. Ahmad[b], J.P. Greene[b], M.A. Kellett[c], A.L. Nichols[c]

t_{growth}= 27.8532 y

$t_{measure}$= 23.0 *h* (Ω=0.06%)

$t_{measure}$= 88.9 *h* (Ω=0.23 %)



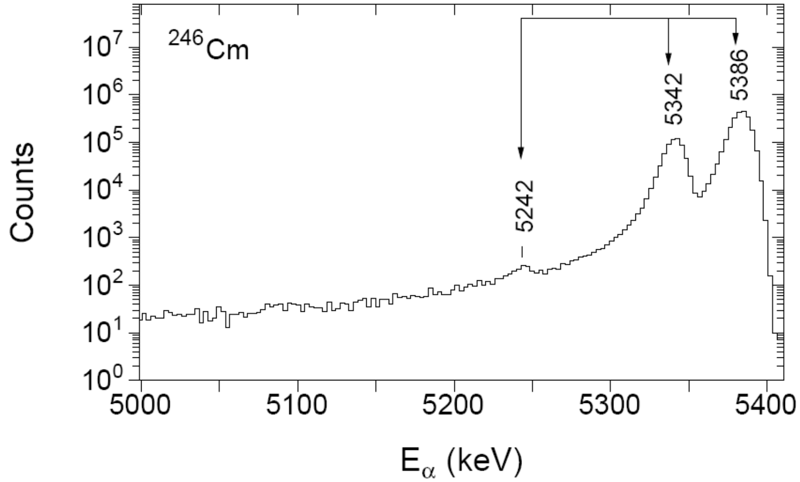
R(^{246}Cm/^{250}Cf)=0.9359 (17) %

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

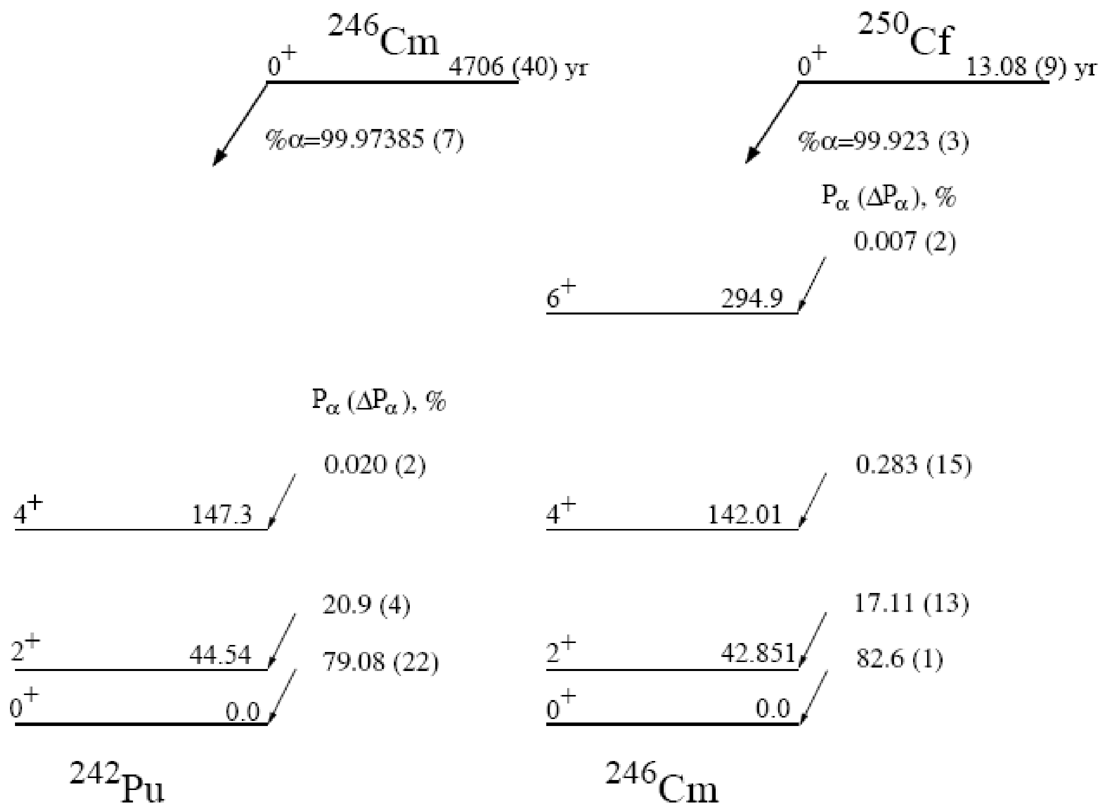
$T_{1/2}$(^{246}Cm)=4706 (40) years

$^{250}Cf \xrightarrow[13\ y]{\alpha} {}^{246}Cm \xrightarrow[4747\ y]{\alpha} {}^{242}Pu \xrightarrow[3.8 10^5\ y]{\alpha}$

Emission Probabilities



$t_{measure}$= 23.0 *h* (Ω=0.06%)



	$\alpha_{0.0}$		$\alpha_{44.5}$		$\alpha_{147.3}$	
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)

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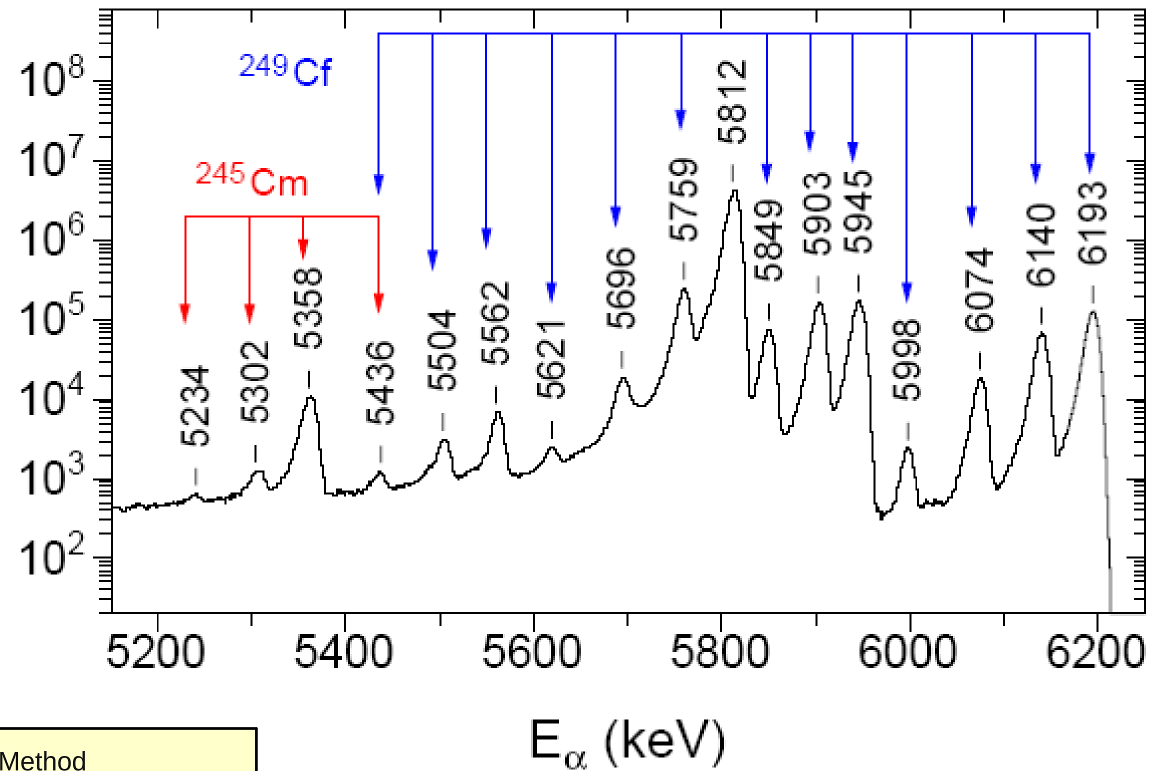
^{245}Cm

8500 (100) y ENSDF
8480 (60) y Holden (1989)
8500 (200) y IAEA TD-261



T1/2,Y	ΔT1/2,Y	Reference	Method
20000		1954Hu50	
11500	5000	1954Fr19	Rel. activity to T1/2(244Cm)
14300	2900	1955Br02	Alpha counting
7500	1900	1957Hu76	
9320	280	1961Ca01	Rel. activity to T1/2(244Cm)
8265	180	1969Me01	Rel. activity to T1/2(252Cf)
8532	53	1971Ma32	Rel. activity to T1/2(244Cm)
8445	200	1982Po14	Several methods, T1/2(244Cm)

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

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

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

^{233}Pa

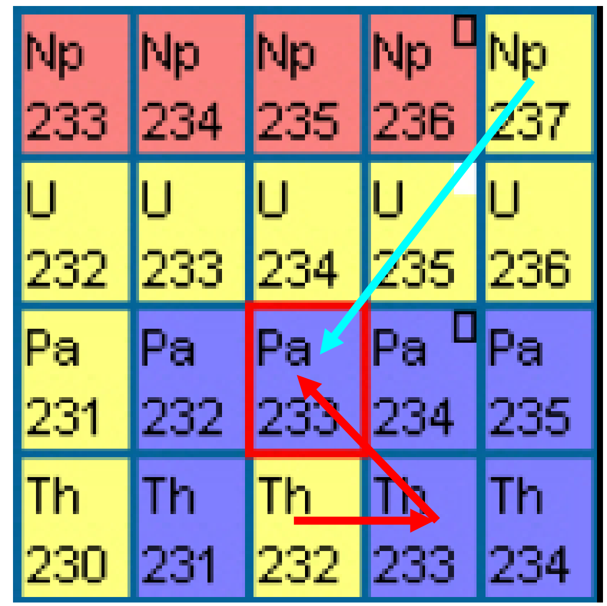
- special interest to first IAEA-CRP (1977)
  high-precision absolute measurements on P_γ (312 keV)
  - ✓ 38.6 (5) % (Gehrke, Helmer, Reich, 1979)
  - ✓ 38.6 (15) % (Poenitz, Smith, 1978)
  - ✓ 41.6 (9) % (Harada et al, 2006)
- inconsistencies in P_γ(28.6 keV) were pointed out at 1st RCM meeting

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)

- 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 - source production

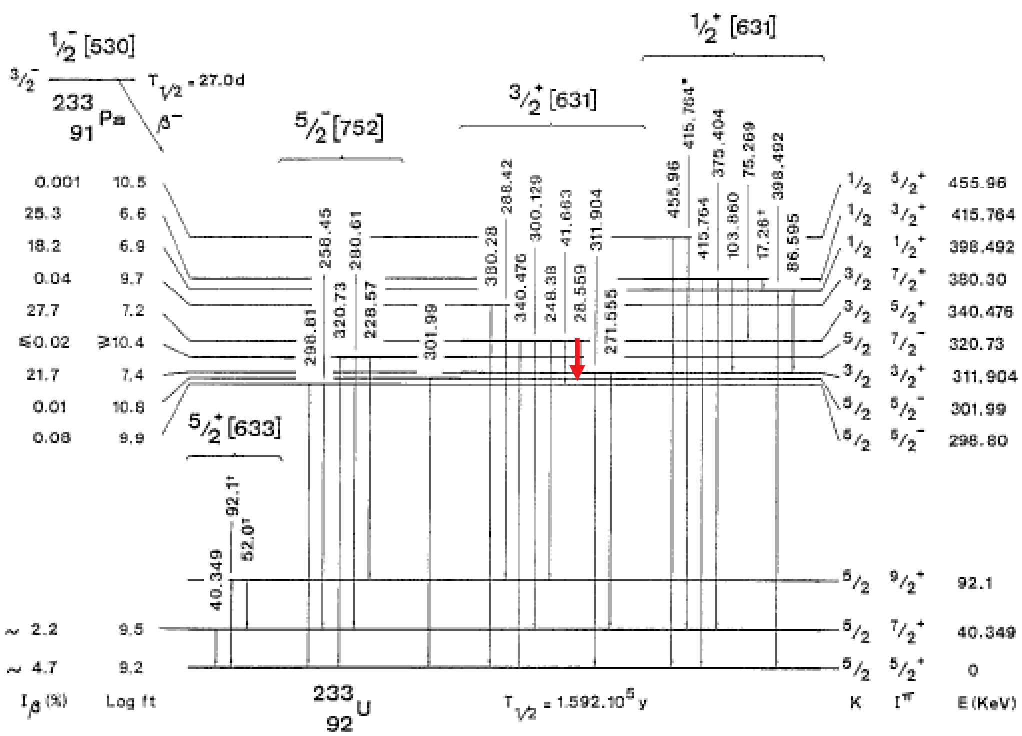
- There are two ways to produce ^{233}Pa
  - a) $^{232}Th(n,\gamma)^{233}Th(\beta^-)^{233}Pa(\beta^-)^{233}U$
  - b) $^{237}Np(\alpha)^{233}Pa(\beta^-)^{233}U$

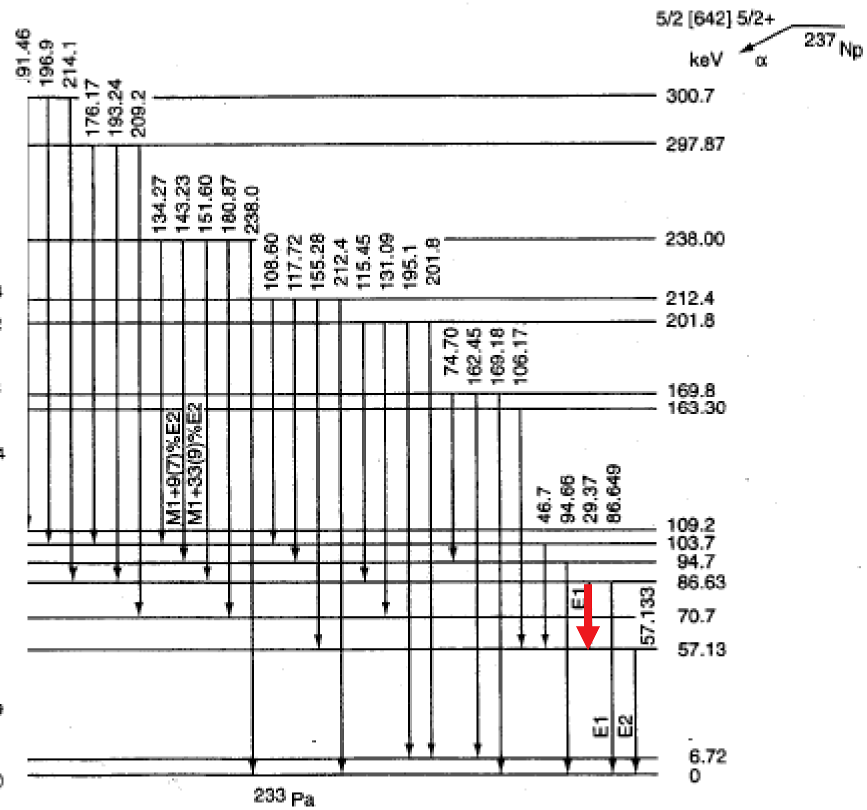


E_γ/keV	P_γ (%)	Th		Np	Th	Np			Np	Np
	Albridge et al. (1961)	Valkeapaa et al. (1973)[a]	Gehrke et al. (1979)	Vaninbroukx et al. (1984)	Kouassi et al. (1990)[a]	Luca et al. (2000)	Schotzig et al. (2000)	Woods et al. (2000)	Luca et al. (2002)	Shchukin et al. (2004)
28.559(10)		0.070(8)		0.15(1)	0.075(8)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

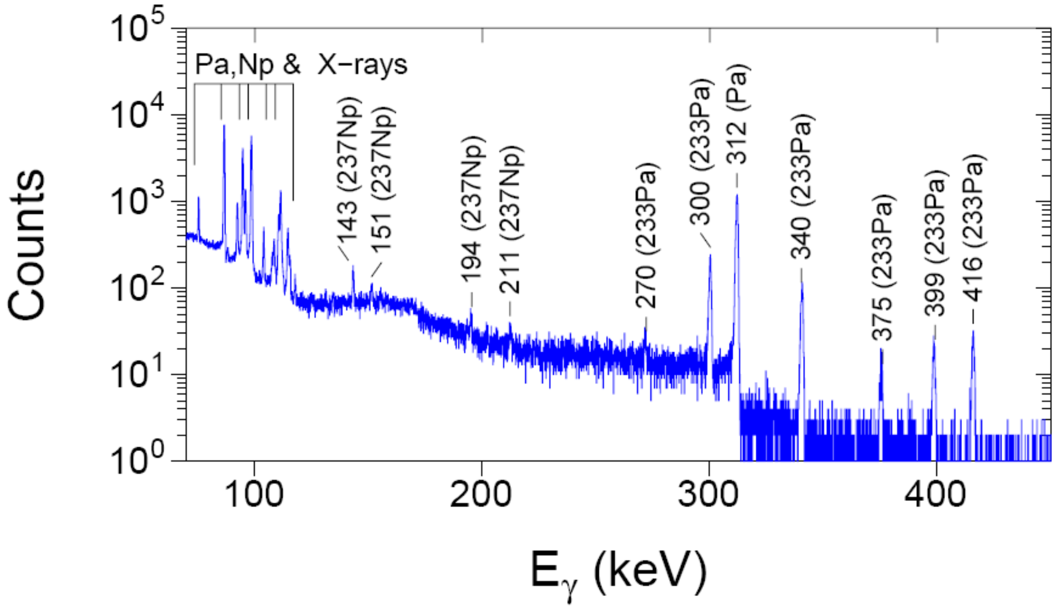
^{233}Pa - source production

- why ^{237}Np/^{233}Pa in equilibrium is NOT a good choice
  - ✓ 29.37 keV E1 (^{237}Np) & 28.557 keV (M1+E2) (^{233}Pa)

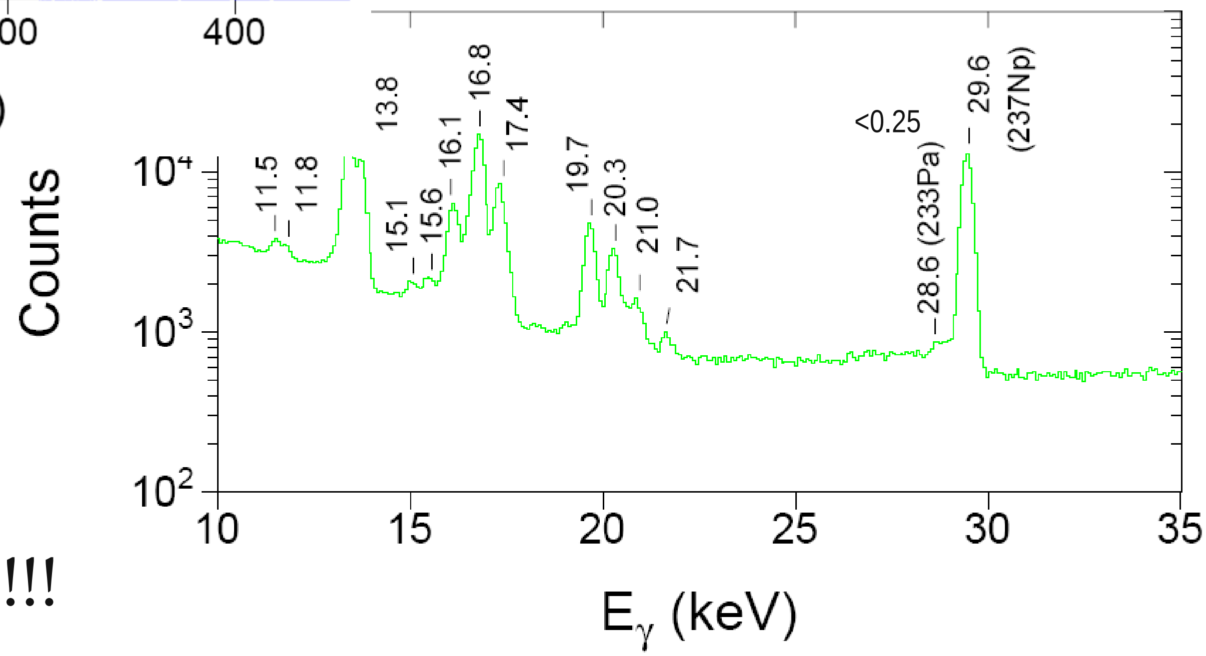




Some preliminary results



- ❑ high isotopic purity



- ❑ … but inconclusive!!!!

We have not given up!

- Conclusion: wrong way to go!
- Possible solution: milk ^{233}Pa from ^{237}Np and use radiochemistry to separate them
- Likely the quantification of 28.557 keV gamma-ray emission probability won't solve the inconsistency!
  - ✓ conversion electron studies
  - ✓ LEPS-LEPS, CE-γ coincidences

<u>Also need to re-assess old data</u>

✓ δ frequently from ICC and sub-shell ratios, but …
α_T(M1)(28.6γ)=31.7(5) by Woods et al. , while BrICC gives
α_T(M1)(28.6γ)=29.8 (5), e.g. 6% difference

Other work in progress

- Complete decay spectroscopy of ^{243}Cm & ^{245}Cm
  - ✓ α–emission probabilities –using PIPS and magnetic spectrometer
  - ✓ γ–emission probabilities – using large Ge and LEPS, including conversion electron studies
- Systematic differences in α–emission probabilities collected using a) semiconductor detectors (e.g. Si(Li) and PIPS) and b) magnetic spectrographs
  - ✓ 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 many others
  - ✓ we must understand these discrepancies – systematics investigation using MA sources at ANL and comprehensive assessment of early measurements using magnetic spectrograph at ANL

Acknowledgments

- I. Ahmad and J.P. Greene (measurements)

  ANL

- A.L. Nichols & M.A. Kellett

  IAEA, Vienna, Austria

- M.-M. Be, V.P. Chechev, X. Huang, A. Luca, G. Mukherjee & A.K. Pearce

  IAEA-CRP participants

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