REACTOR POWER DECAY-HEAT CALCULATIONS:

IDENTIFYING FISSION PRODUCTS TO BENEFIT FROM STUDY BY MEANS OF TOTAL ABSORPTION GAMMA-RAY SPECTROSCOPY

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1974-2001: UKAEA and AEA Technology,
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2001-2009: IAEA Nuclear Data Section, Austria
2009-2011: University of Surrey, UK (Visiting Researcher);
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35+ years of evaluating decay data
- actinides and heavy elements
- activation products
- fission products
- standards/calibrants

Assembly of nuclear data libraries
- fission and fusion reactor applications
- fuel reprocessing and waste management
- spectral analyses

BUT NOT A DECAY HEAT SPECIALIST

NUCLEAR DATA FOR DECAY HEAT CALCULATIONS - THERMAL AND FAST NEUTRON FISSION

$$H_\alpha(t) = \sum_{i=1}^{M} \lambda_i^T \, N_i(t) \, E_\alpha^i$$

$$H_\beta(t) = \sum_{i=1}^{M} \lambda_i^T \, N_i(t) \, E_\beta^i$$

$$H_\gamma(t) = \sum_{i=1}^{M} \lambda_i^T \, N_i(t) \, E_\gamma^i$$

Derivation of $N_i(t)$ through inventory codes

- wide range of radionuclides and stable nuclides
- σ: total neutron absorption, (n,γ) and (n,2n) cross sections of actinides

NUCLEAR DATA FOR DECAY HEAT CALCULATIONS

$\sigma^F_{a,k}$ - effective group-averaged fission cross section of actinide *a* in the k^{th} neutron group,

$\sigma^A_{i,j}$ - total neutron absorption cross section of fission product *i*,

$\sigma^{(n,\gamma)}_{i,j}$ - (n, γ) cross section of fission product *i*,

$\sigma^{(n,2n)}_{i,j}$ - (n, 2n) cross section of fission product *i*,

$Y^i_{a,k}$ - independent yields for fission product *i*,

λ_i - decay constant(s) of fission product *i*,

$k_\alpha, k_{\beta^-}, k_{\beta^+}$ - branching fractions for α, β- and β+ decay to nuclide Z, A,

$E^i_\alpha, E^i_\beta, E^i_\gamma$ - mean alpha, beta and gamma energy releases per disintegration of nuclide *i*.

Decay Heat: Points of Note

- Fission products (~1040 nuclides)
  - fission yields
  - decay data
  - cross sections
- Actinides – maximum ~ 20-30% of total decay heat up to ~ 10^8 s cooling time [~ 3 years], and then beyond
- Activation products – extremely minor contribution to thermal systems

ACTINIDE DECAY

20 - 30% contribution:

^{239}U + ^{239}Np for cooling times < 10^6 sec

^{242}Cm for cooling times from 10^6 to 10^8 sec

also ^{237}U

^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu

^{241}Am

^{244}Cm

Data are reasonably well-defined (but also need to consider consequences of high burn-up strategies)

ACTIVATION PRODUCTS

Fast-neutron induced activation:

~10% contribution at 5 years cooling arises from ^{60}Co

$^{58}Ni(n, p)^{58}Co(n, \gamma)^{59}Co(n, \gamma)^{60}Co$

Sodium coolant:

^{22}Na and ^{24}Na

Data are well defined

SENSITIVITY STUDIES

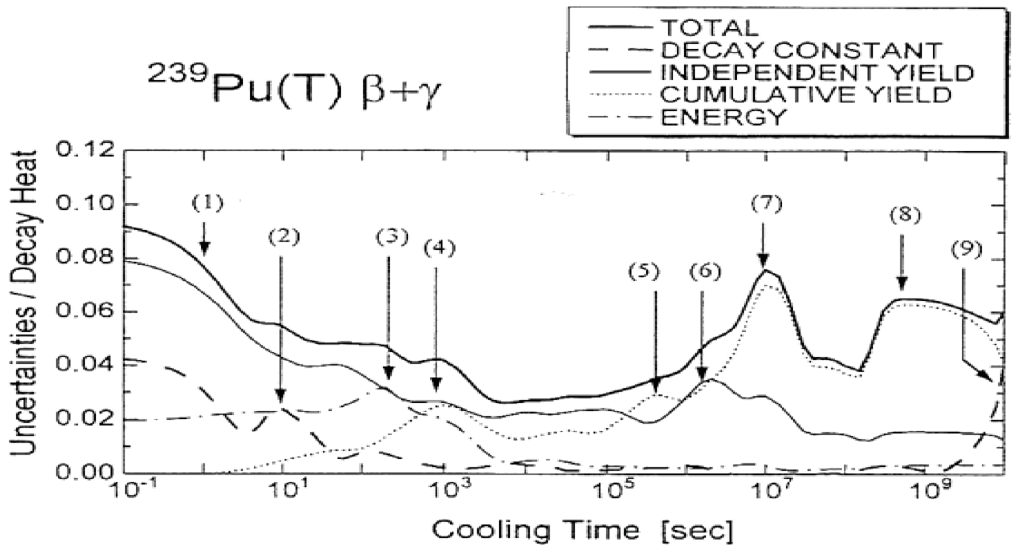
- Schmittroth (1976)
- Schmittroth and Schenter (1977)
- Oyamatsu *et al.* (1997)

Analyses in terms of :

decay energies

fission yields

half-lives



Uncertainties in total decay heat for ^{239}Pu(T) without taking correlation effects into account.

Main parameters contributing to the uncertainties in total decay heat for ^{239}Pu(T).

No.	t [sec]	Parameter	Nuclide
(1)	0~1	Independent Yield	^{97}Sr
			^{97m}Y
			^{102}Nb
		Decay Constant	^{101}Zr
			^{104m}Nb
(2)	10	Decay Constant	^{101}Zr
(3)	10^{2}~	Decay Energy	^{103}Mo
			^{103}Tc
(4)	10^{3}	Cumulative Yield	^{102}Nb
			^{102m}Nb
(5)	~10^{6}	Cumulative Yield	^{99}Zr
			^{132}Sb
			^{140}Cs
(6)	10^{6}~	Independent Yield	^{140}Ba
(7)	10^{7}	Cumulative Yield	^{95}Sr
			^{95}Y
			^{103}Mo
(8)	10^{8}~	Cumulative Yield	^{90}Rb
			^{90m}Rb
(9)	~10^{10}	Decay Constant	^{126}Sn
			^{137}Cs

(1997)

FISSION YIELDS

^{97}Sr
$^{97}Y^{m}$
^{102}Nb
} 0-1 sec

^{102}Nb
$^{102}Nb^{m}$
} 10^{3} sec

^{134}Te
^{138}Xe
^{142}Ba
} 10^{4} sec

^{90}Rb
$^{90}Rb^{m}$
^{137}Xe
} 10^{9} sec

DECAY CONSTANTS

$^{104}Nb^{m}$ 0-1 sec

^{101}Zr 0-10 sec

^{137}Cs 10^{10} sec

^{126}Sn 10^{10} sec

DECAY ENERGIES

^{103}Mo
^{103}Tc
} 10^{2} sec

FISSION YIELDS: STATUS 2008

(1) International network of co-operation between fission yield experts :

- clean-up of data bases
- model development
- share evaluation procedures

(2) Empirical models (Z_p and A'_p)

- reliability from thermal to 15-MeV neutrons

(3) Computer programs :

- correlations and covariance matrices

(4) Complete fission yield data sets

(5) IAEA-TECDOC-1168 (2000), and IAEA-STI/PUB/1286 (2008)

FISSION YIELDS: PROBLEMS 2008

(1) Models and systematics incapable of generating accurate and reliable fission yield estimates at different neutron energies

(2) Isomeric yields - inadequate model, and insufficient experimental data

More experimental data required

DECAY DATA

λ_i	decay constants ($\lambda = \frac{\ln 2}{t_{1/2}}$)
k_α , k_{β^-} , k_{β^+}	branching fractions for α, β^- and β^+ decay
E^i_α, E^i_β, E^i_γ	mean alpha, beta and gamma energy releases of nuclide

DECAY DATA FILES

- fission products
- actinides and decay chains

Half-lives
Isomeric branching ratios
Mean energies (can be derived from discrete decay data)

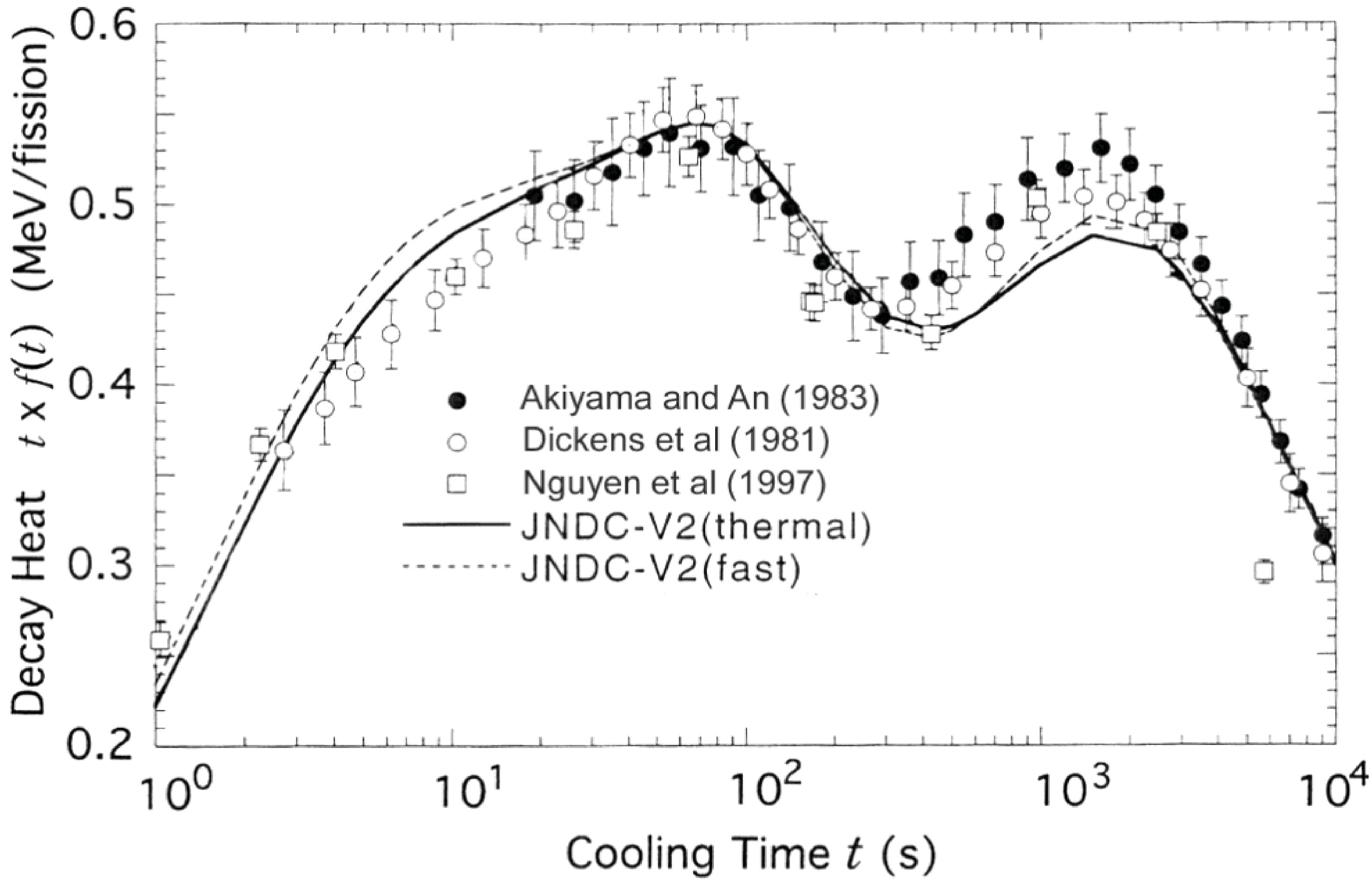
DECAY HEAT DISCREPANCIES

Yoshida *et al*. (1997 and 1999)

JNDC-FP-V2 library:

0 - 20 sec

300 - 3000 sec



Gamma-ray discrepancies seen in ^{239}Pu decay heat after a fission burst (Yoshida et al, 1999).

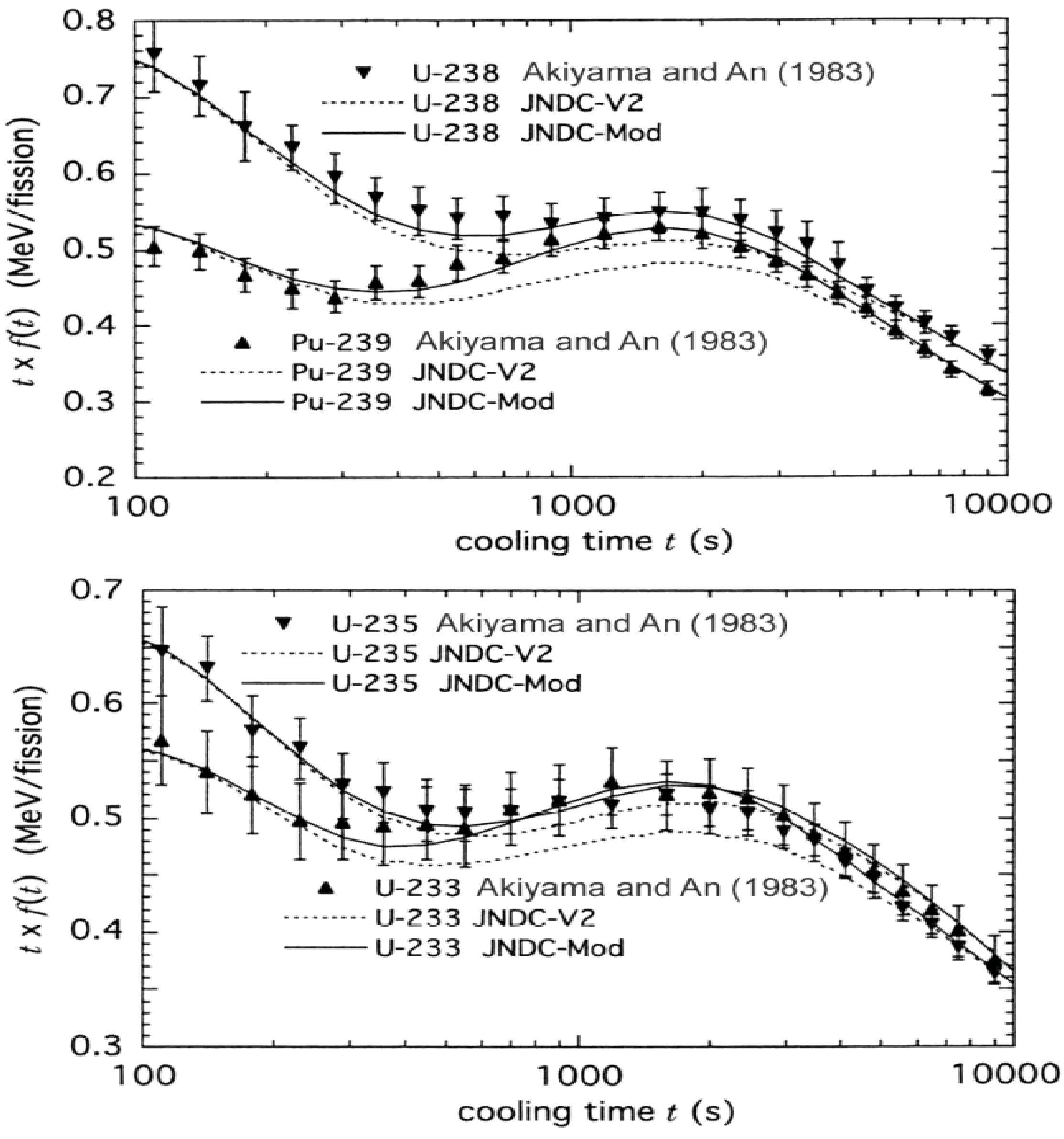
YOSHIDA *et al*. (1997 and 1999)

Introduced additional β^- decay chain of two radionuclides

Energy release of 1.5 MeV per decay

Discrepancy between 300 and 3000 sec reduced significantly (eliminated?) in various fissioning systems:

- candidates for additional energy release included ^{102}Tc, ^{104}Tc and ^{105}Tc



Removal of γ-ray discrepancy by including
1.5 MeV γ-ray emitter (Yoshida et al, 1999).

CONFLICT

Short-lived fission products (half-lives less than ~ 1 min)

→ difficult to characterise discrete transitions

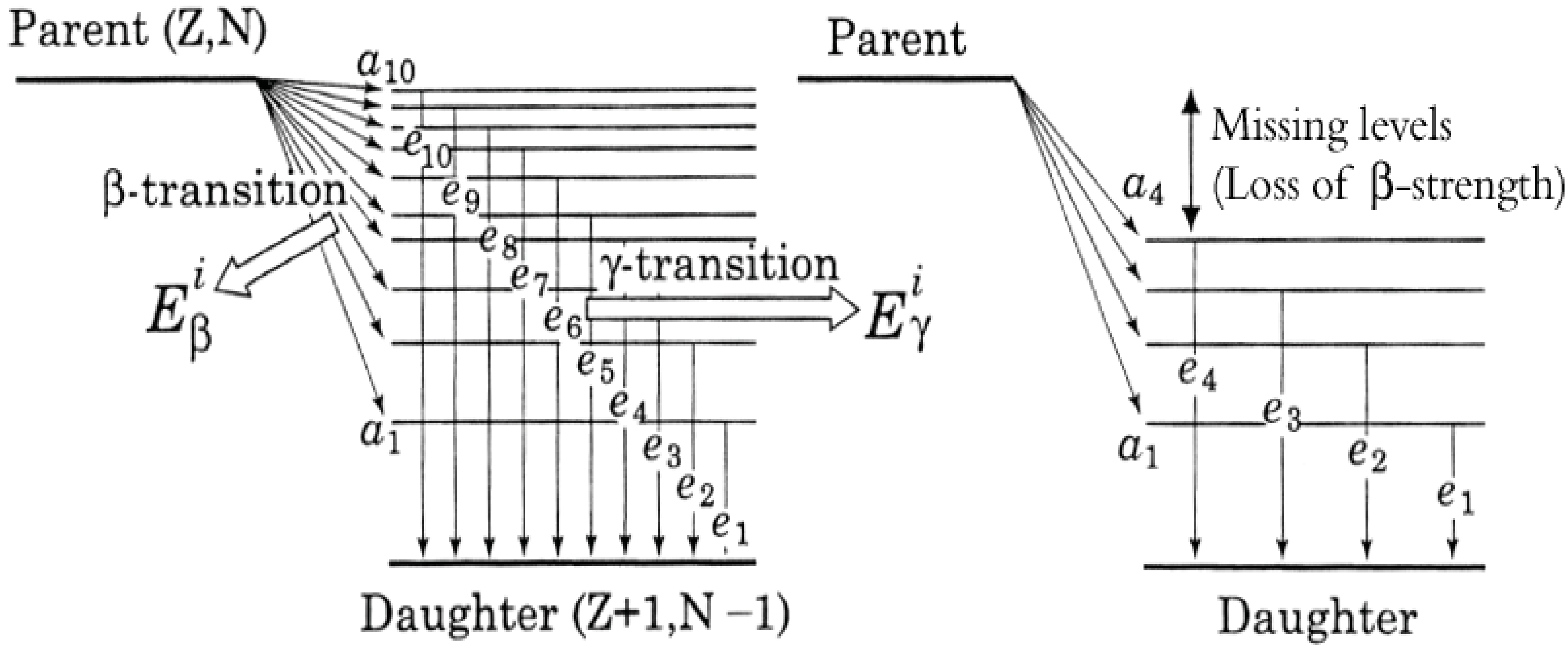
→ lack of credible decay data

Pandemonium effect (Hardy *et al.*, 1977)

PANDEMONIUM

1) Inability to detect low-intensity, high-energy gamma-ray transitions (Hardy *et al.*, 1977)

2) Impact on gamma and beta decay heat components:
loss of γ-strength lowers E_γ and $H_\gamma(t)$, and effectively increases E_β and $H_\beta(t)$



Effect of missing nuclear levels: loss of γ-ray energy release(E_γ^i).

SPECTRAL MEASUREMENTS

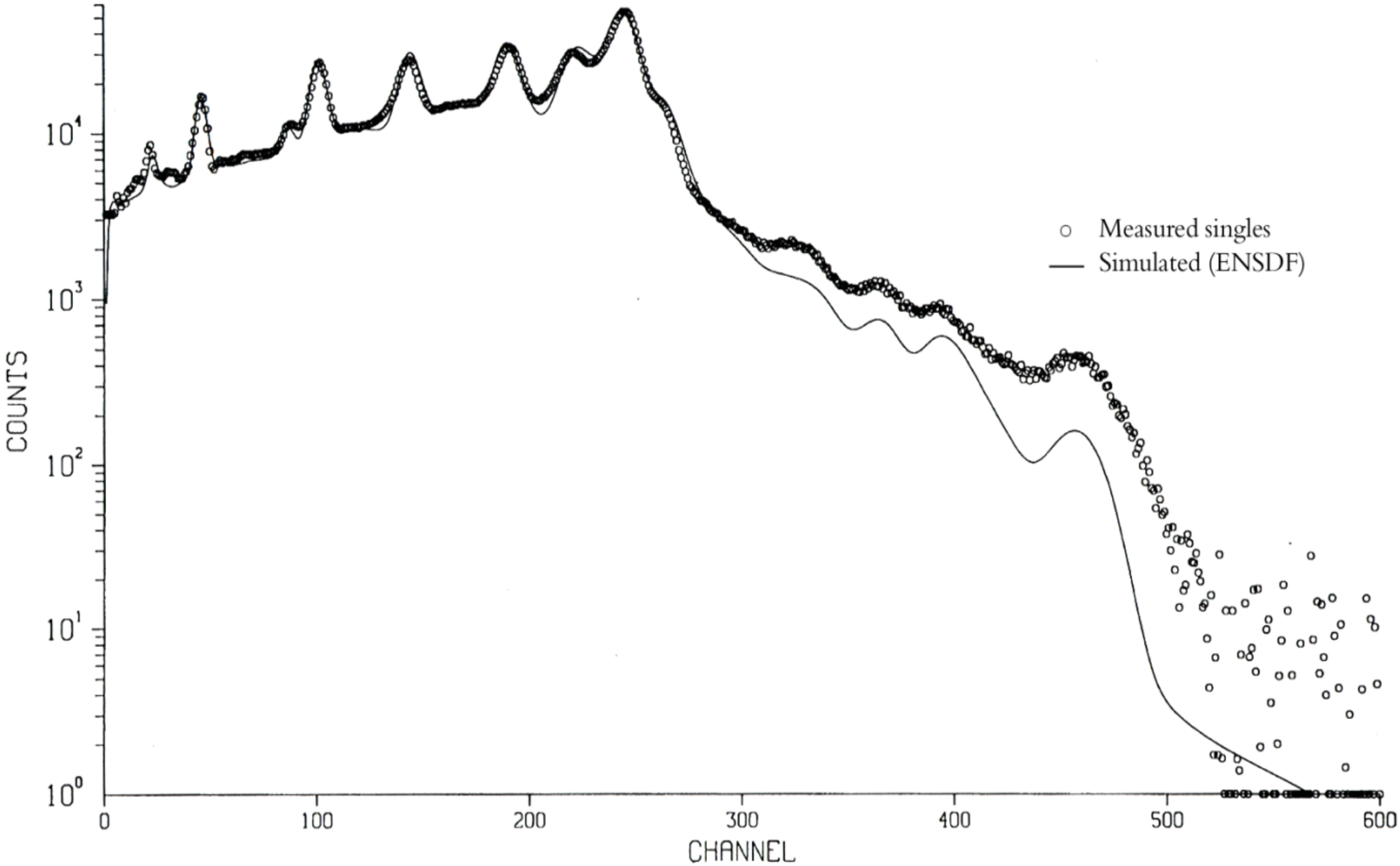
Greenwood *et al*. (INEL: 1994, 1996 and 1997)

He gas-jet transport to mass separator
Rapid collection, transport and measurement

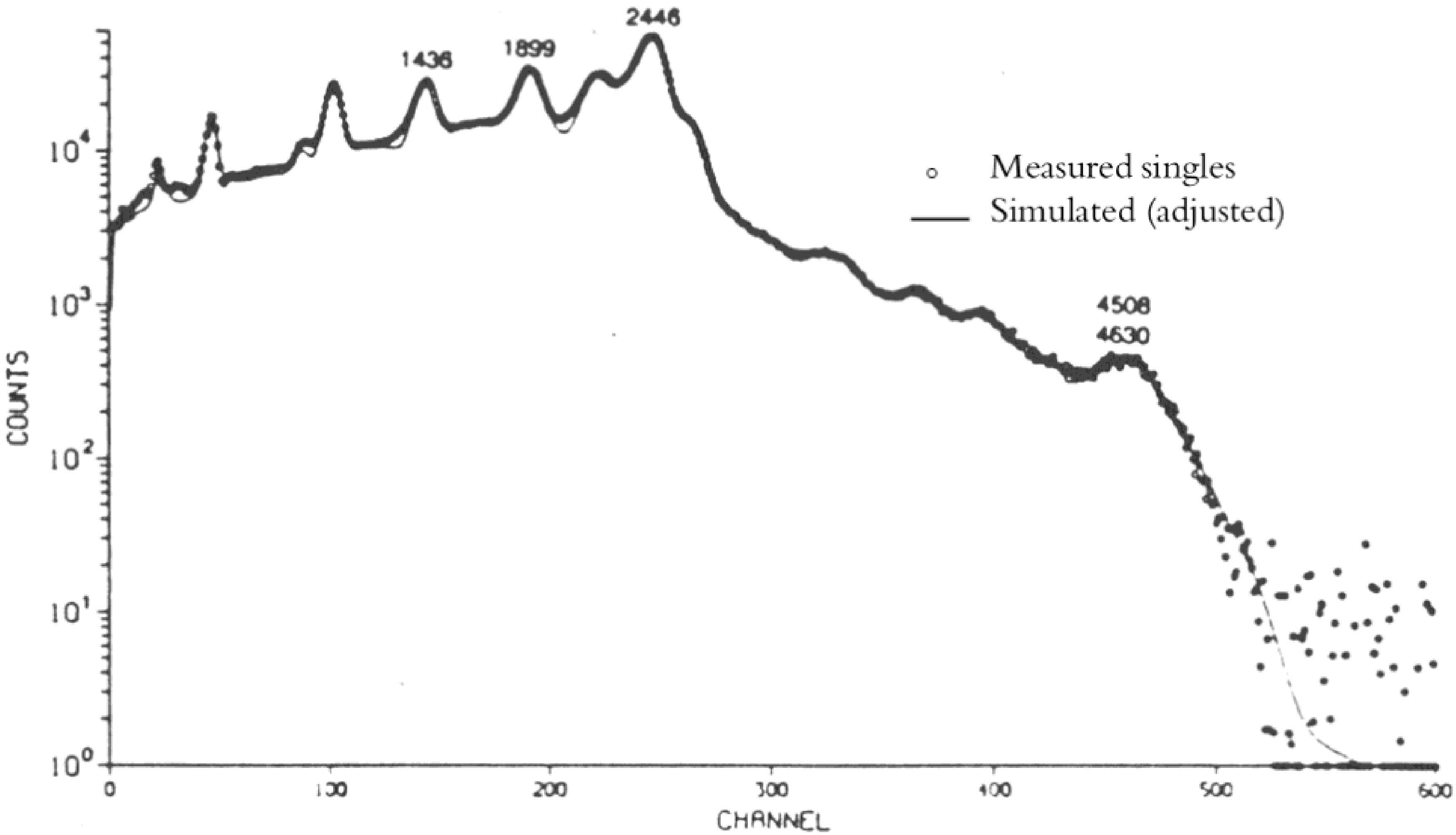
TAGS - total absorption gamma-ray spectroscopy on numerous fission products:

NaI(Tl) detector with deep-axial counting well for complete summing of γ-ray cascades;
Si detector located in the counting well

Operated in $4\pi\gamma-\beta^-$ coincidence mode to determine β^- branch to ground state of daughter



Comparison of measured singles spectrum for ^{138g}Cs with the simulated spectrum for the evaluated decay scheme (Greenwood et al, 1994).



Comparison of measured singles spectrum for ^{138g}Cs with the simulated spectrum for the re-adjusted decay scheme (Greenwood et al, 1994).

^{138g}Cs β^- emission probabilities, P_β (Greenwood *et al*, 1997).

Level energy (keV)	P_β(%)	
	NDS	TAGS
0.0	0.0	0.0
1435.9	4.3	3.76
1898.7	13.7	13.94
2217.9	13.0	13.23
2307.6	7.3	6.10
2415.5	0.63	0.64
2445.7	44.0	44.76
2583.1	1.67	1.42
2639.5	8.80	7.32
2779.5	1.59	0.81
2851.6	0.20	0.20
2880.9	0.54	0.55
2931.5	0.21	0.21
2991.2	0.64	0.61
3049.9	0.17	0.17
3163.6	0.34	0.35
3242.6	0.27	0.54
3257.7	0.06	0.33
3339.0	0.17	0.33
3352.6	0.035	0.036
3367.0	0.23	0.23
3437.4	0.011	0.011
3442.3	0.011	0.011
3510 P	-	0.25
3647.0	0.43	0.66
3652.6	0.005	0.005
3694.0	0.30	0.46
3825 P	-	0.15
3922.6	0.21	0.25
3935.2	0.47	0.61
4012.3	0.08	0.08
4080.1	0.18	0.18
4242.5	0.10	0.28
4370 P	-	0.10
4508.1	0.16	0.41
4629.8	0.26	0.72
4850 P	-	0.20
5080 P	-	0.046

NDS: Nuclear Data Sheets, **69**(1993)69.
P: placement of pseudolevel.

FIXES

(1) Fission yields: systematic fitting procedures

(2) Decay data: gross energy measurements by TAGS

FISPIN10 Decay Heat Calculations

Robert Mills: NNL, UK

- JEFF-3.1 decay data files, and JEFF-3.1 fission yield files

- FISPIN10 – used to calculate FP number densities following a single fission
- decaying FPs from 0.1 to 10^{+9} secs after fuel irradiation
- FPs > 0.5% of the resulting decay heat as a function of cooling time after irradiation for $^{235}U_{th}$, $^{239}Pu_{th}$, $^{238}U_{f}$, $^{232}Th_{f}$
- ordered in terms of those that contribute the most uncertainty to the overall uncertainty of the calculated decay heat

$^{239}Pu_{th}$: 1.0 s cooling time

Nuclide	Energy release rate eV per second per fission			
	Ebeta	Egamma	Ealpha	
41-Nb-103	2.3526E+04	2.3526E+04	0.0000E+00	6%
41-Nb-104M	1.2554E+04	1.2554E+04	4.4028E-01	3%
39-Y-98M	1.4565E+04	1.7209E+04	8.2312E+01	4%
41-Nb-102	1.6423E+04	1.6423E+04	0.0000E+00	**4%**
9-Y-99	2.0826E+04	6.4749E+03	5.1960E+01	3.2%
38-Sr-97	1.1495E+04	1.1495E+04	5.3695E-01	**2.7%**
43-Tc-109	8.4527E+03	8.4527E+03	8.5152E-01	2% ....etc.

$^{239}Pu_{th}$: 1 min cooling time

Nuclide	Energy release rate eV per second per fission			
	Ebeta	Egamma	Ealpha	
53-I-137	4.3704E+02	2.8630E+02	9.2438E+00	**3.2%**
53-I-136	3.5992E+02	4.2571E+02	0.0000E+00	
41-Nb-98	6.2536E+02	1.0327E+02	0.0000E+00	
52-Te-135	4.4471E+02	7.0002E+01	0.0000E+00	
53-I-136m	3.6907E+02	3.6907E+02	0.0000E+00	
52-Te-136	1.1528E+02	1.7469E+02	0.0000E+00	
51-Sb-133	7.6329E+01	2.5365E+02	0.0000E+00	
44-Ru-109	1.7440E+02	2.7795E+02	0.0000E+00	
43-Tc-107	5.0802E+02	1.2722E+02	0.0000E+00	
42-Mo-105	6.8414E+02	1.9630E+02	0.0000E+00	
43-Tc-106	8.3182E+02	9.3787E+02	0.0000E+00	..............etc.

$^{239}Pu_{th}$: 1.1 h cooling time

Nuclide	Energy release rate eV per second per fission			
	Ebeta	Egamma	Ealpha	
53-I-134	4.2397E+00	1.7166E+01	0.0000E+00	12%
51-Sb-130	8.3440E-01	3.6537E+00	0.0000E+00	2.5%
57-La-143	1.7144E+00	3.4970E-01	0.0000E+00	**1.1%**
55-Cs-138	1.0023E+01	1.8952E+01	0.0000E+00	
43-Tc-104	5.7022E+00	6.7564E+00	0.0000E+00	
52-Te-134	1.3600E+00	5.3068E+00	0.0000E+00	
52-Te-133M	9.4822E-01	4.5968E+00	0.0000E+00	
51-Sb-131	1.1401E+00	3.4346E+00	0.0000E+00	
43-Tc-101	2.9988E+00	2.1049E+00	0.0000E+00	..............etc.

$^{239}Pu_{th}$: 1.04 d cooling time

Nuclide	Energy release rate eV per second per fission			
	Ebeta	Egamma	Ealpha	
53-I-135	5.2571E-02	2.2648E-01	0.0000E+00	10%
54-Xe-135	1.2904E-01	1.0169E-01	0.0000E+00	8%
53-I-133	1.1171E-01	1.6708E-01	0.0000E+00	
51-Sb-128	1.3006E-02	8.1520E-02	0.0000E+00	
46-Pd-109	4.2663E-02	7.6214E-05	0.0000E+00	
53-I-132	4.6180E-02	2.0961E-01	0.0000E+00	
41-Nb-97M	2.6280E-03	1.3283E-01	0.0000E+00	
39-Y-93	1.2054E-01	9.7878E-03	0.0000E+00	
41-Nb-97	9.6867E-02	1.3737E-01	0.0000E+00	............etc.

$^{235}U_{th}$: Dominant Uncertainties

- Short cooling times – uncertainties in FYs
- Half-lives:

  $^{98}Y^{m}$ (± 10%), ^{100}Nb (± 13%), ^{102}Nb (± 15%)
- Energy release:

  ^{87}Br (± 17%), ^{89}Sr (± 40%), ^{97}Sr (no uncertainty), ^{101}Nb (± 15%), ^{102}Nb (no uncertainty), ^{143}La (± 53%)

Decay Heat and Problem Fission Products, 2007

- WPEC: Working Party on international nuclear data Evaluation Cooperation – Subgroup 25 formed in April 2005
- December 2005: first meeting at the IAEA
- May 2006: second meeting at OECD/NEA
- NEA/WPEC-25 report published in 2007

Decay Heat and Problem Fission Products, 2007

Decay heat calculations:

- avoidance of nuclear theory (minimal usage)
- experimental TAGS data
- experimental decay scheme data: β - and γ -feeding

Aims of WPEC Subgroup 25:

- identify nuclides that contribute significantly between 10 and 5000 secs
- possible inadequate decay data (pandemonium effect)
- new β -feeding measurements by means of TAGS
- determine β -decay directly to daughter ground state

Decay Heat and Problem Fission Products, 2007

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C.J. Dean	Serco Assurance, UK
W. Gelletly	University of Surrey, UK
R.W. Mills	Nexia Solutions Ltd., UK
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P. Möller	LANL, USA
A. Sonzogni	BNL, USA

Decay Heat and Problem Fission Products, 2007

Decay Heat and Problem Fission Products, 2007

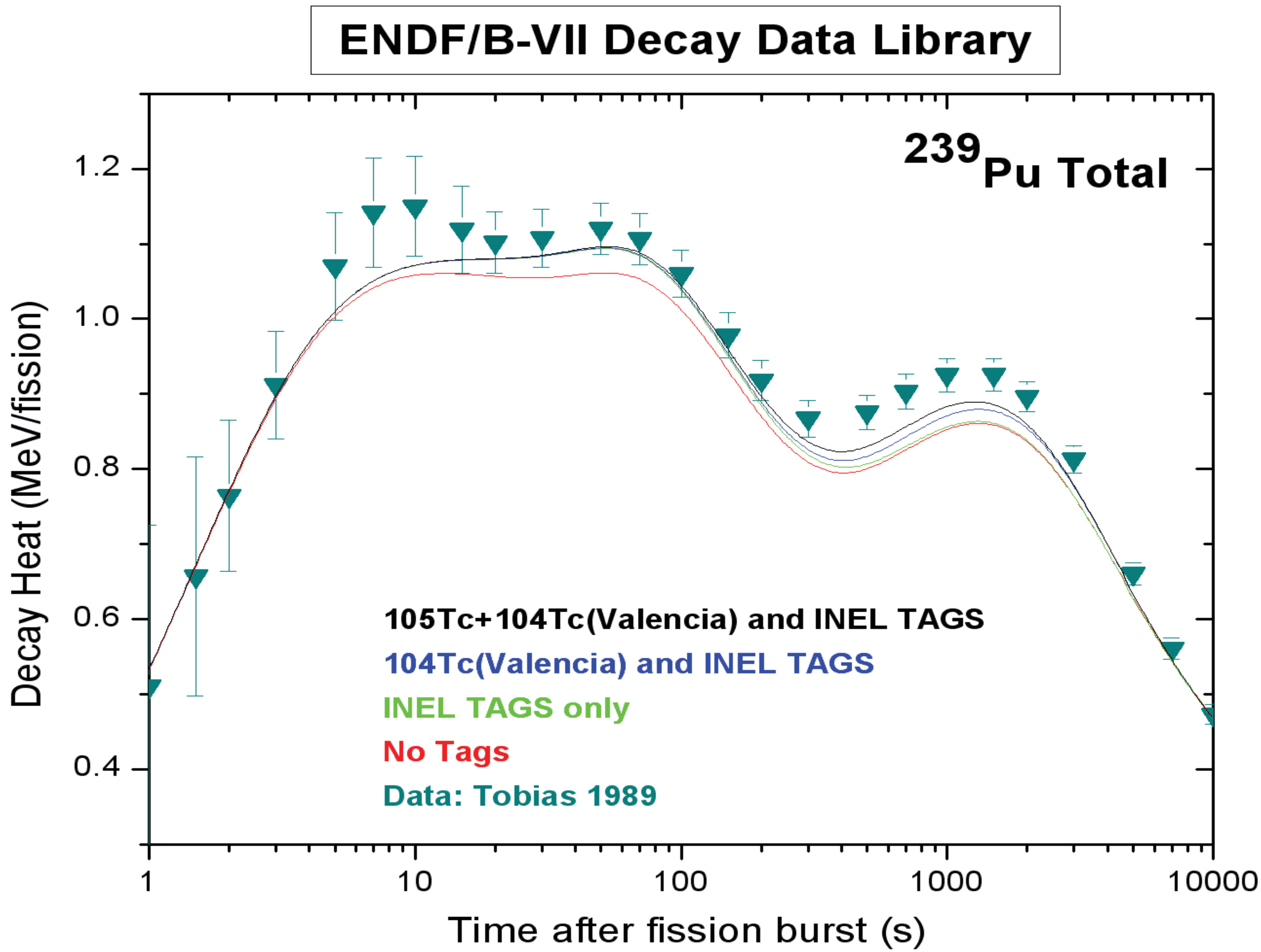
Priority 1:

Radionuclide	Half-life	Radionuclide	Half-life
35-Br-86	55.1 s	43-Tc-103	54.2 s
35-Br-87	55.65 s (β^-,n)	43-Tc-104	18.3 min
35-Br-88	16.36 s (β^-,n)	43-Tc-105	7.6 min
36-Kr-89	3.15 min	43-Tc-106	35.6 s
36-Kr-90	32.32 s	51-Sb-132	2.79 min
41-Nb-98	2.86 s	53-I-136	83.4 s
41-Nb-99	15.0 s	53-I-136m	46.9 s
41-Nb-100	1.5 s	53-I-137	24.13 s (β^-,n)
41-Nb-101	7.1 s	54-Xe-137	3.82 min
42-Mo-103	67.5 s	54-Xe-139	39.68 s
42-Mo-105	35.6 s	54-Xe-140	13.6 s
43-Tc-102	5.28 s		

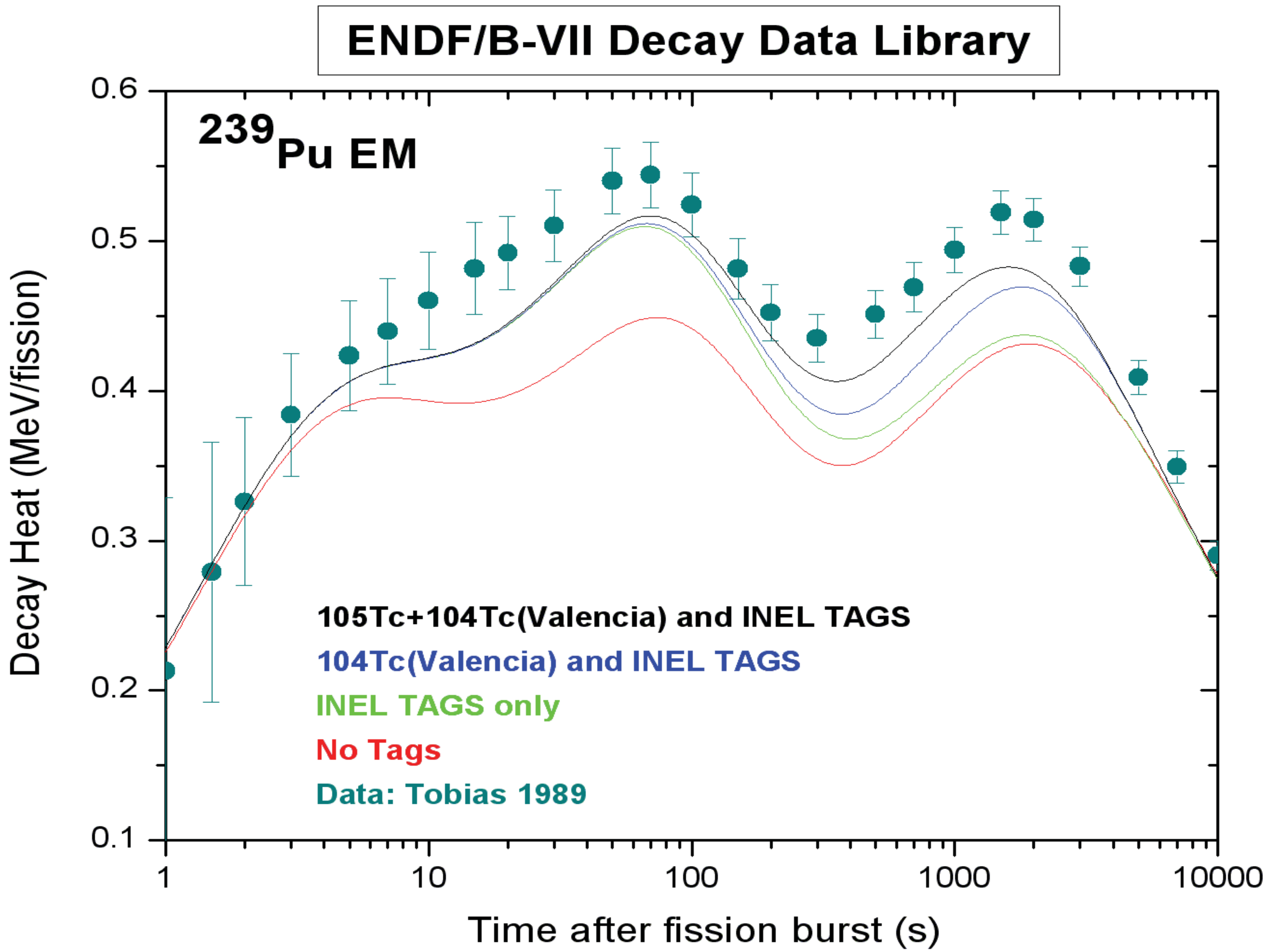
Decay Heat and Problem Fission Products, 2007

Priority 2:

Radionuclide	Half-life	Radionuclide	Half-life
37-Rb-90m	**258 s** repeat INEL TAGS	**41-Nb-102**	**1.3 s**
37-Rb-92	**4.49 s** **small (β^-,n)**	**43-Tc-107**	**21.2 s**
38-Sr-89	**50.53 d**	**52-Te-135**	**19.0 s**
38-Sr-97	**0.429 s** **possible (β^-,n)**	**56-Ba-145**	**4.31 s** repeat INEL TAGS
39-Y-96	**5.34 s**	**57-La-143**	**14.2 min** repeat INEL TAGS
40-Zr-100	**7.1 s**	**57-La-145**	**24.8 s** repeat INEL TAGS



ENDF/B-VII Decay Data Library
239Pu Total
105Tc+104Tc(Valencia) and INEL TAGS
104Tc(Valencia) and INEL TAGS
INEL TAGS only
No Tags
Data: Tobias 1989
Decay Heat (MeV/fission)
1.2
1.0
0.8
0.6
0.4
1
10
100
1000
10000
Time after fission burst (s)



ENDF/B-VII Decay Data Library
239Pu EM
Decay Heat (MeV/fission)
Time after fission burst (s)
0.6
0.5
0.4
0.3
0.2
0.1
1
10
100
1000
10000
105Tc+104Tc(Valencia) and INEL TAGS
104Tc(Valencia) and INEL TAGS
INEL TAGS only
No Tags
Data: Tobias 1989

Decay Heat and Problem Fission Products, 2009/10
Th-U fuel

India is especially interested in the Th-U fuel cycle

(1). No substantial experimental benchmarks

(2). Collaboration (Manipal University (Mohini Gupta + Alan Nichols)) to assess

- decay heat profile
- possible candidates for TAGS

Decay Heat and Problem Fission Products, 2009/10
Th-U fuel

Calculations used DECROI code - Olivier Bersillon (CEA Bruyères-le-Châtel, France)

- neutron burst
- 10, 100, 1000, 5000 and 10000 s cooling times

Comparable aims to FISPIN calculations of Robert Mills for U-Pu fuels

Decay Heat and Problem Fission Products, 2009/10

Th-U fuel - radionuclides in common with Greenwood studies

10 secs: **^{91}Rb, ^{93}Rb, ^{94}Sr, ^{95}Sr, ^{140}Cs, ^{141}Cs, ^{143}Ba, ^{144}Ba, ^{145}Ba, ^{144}La, ^{145}La**

100 secs: **^{89}Rb, ^{90}Rb, ^{90}Rb^{m}, ^{91}Rb, ^{93}Sr, ^{94}Sr, ^{95}Sr, ^{94}Y, ^{95}Y, ^{139}Cs, ^{140}Cs, ^{141}Cs, ^{141}Ba, ^{142}Ba, ^{143}La, ^{144}La, ^{145}La, ^{145}Ce, ^{147}Ce, ^{148}Ce, ^{148}Pr**

1000 secs: **^{89}Rb, ^{90}Rb, ^{90}Rb^{m}, ^{93}Sr, ^{94}Y, ^{95}Y, ^{138}Cs, ^{139}Cs, ^{141}Ba, ^{142}Ba, ^{142}La, ^{143}La, ^{145}Ce, ^{146}Ce, ^{146}Pr, ^{147}Pr**

Decay Heat and Problem Fission Products, 2009/10

Th-U fuel - radionuclides in common with Greenwood studies

5000 secs: **^{89}Rb, ^{94}Y, ^{95}Y, ^{138}Cs, ^{141}Ba, ^{142}La, ^{143}La, ^{146}Pr, ^{149}Nd**

10000 secs: **^{94}Y, ^{138}Cs, ^{142}La, ^{146}Pr, ^{149}Nd**

Decay Heat and Problem Fission Products, 2009/10
Th-U fuel - requirements in common with decay-heat needs of U-Pu fuel

10 secs: **^{86}Br, $^{87}Br(\beta^-,n)$, $^{88}Br(\beta^-,n)$, ^{90}Kr, ^{92}Rb, ^{96}Y, ^{99}Zr, ^{100}Zr, ^{98}Nb, ^{99}Nb, ^{100}Nb, ^{101}Nb, ^{102}Nb, ^{135}Te, $^{136}I^m$, $^{137}I(\beta^-,n)$, ^{139}Xe, ^{140}Xe**

100 secs: **^{86}Br, $^{87}Br(\beta^-,n)$, ^{89}Kr, ^{90}Kr, ^{98}Nb, ^{103}Mo, ^{103}Tc, ^{132}Sb, ^{136}I, $^{136}I^m$, $^{137}I(\beta^-,n)$, ^{137}Xe, ^{139}Xe**

1000 secs: **^{89}Kr, ^{102}Tc, ^{104}Tc, ^{137}Xe**

5000 secs: **^{104}Tc**

Decay Heat Calculations: Assessment of Fission Product Decay Data Requirements for Th-U fuel

IAEA report INDC(NDS)-0577, May 2010

www-nds.iaea.org/reports-new/indc-reports/indc-nds/indc-nds-0577.pdf

Identified 40 top contributors to decay heat at specific cooling times

% decay heat contribution
Q-value and level energies
assessment of known decay scheme

- 10 s: 11 fission products important to ^{233}U c.f. ^{235}U
  9 possible 'new' TAGS candidates
- 100 s: 6 fission products important to ^{233}U c.f. ^{235}U
  4 possible 'new' TAGS candidates
- 1000 s: 19 fission products important to ^{233}U c.f. ^{235}U
  7 possible 'new' TAGS candidates

etc.

Decay Heat and Problem Fission Products, 2009/10 - required for Th-U fuel

10 secs: ^{86}Se, ^{89}Br, ^{91}Kr, ^{94}Rb, $^{96}Y^{m}$, ^{97}Y, $^{100}Nb^{m}$, $^{102}Nb^{m}$, $^{146}La^{m}$

100 secs: ^{85}Se, ^{98}Zr(?), $^{99}Nb^{m}$, ^{133}Sb

1000 secs: ^{84}Br, ^{87}Kr, ^{92}Sr, ^{101}Mo, $^{129}Sb^{m}$, $^{130}Sb^{m}$, ^{138}Xe

5000 secs: ^{87}Kr, ^{88}Rb, ^{92}Sr, $^{128}Sb^{m}$, $^{129}Sb^{m}$, ^{139}Ba, ^{141}La

QUESTION

Are we striving for perfection when good is good enough ?

Significant gaps remain in measured fission yields and decay data, implying we are still some way from such an enviable position ………
and then there are concerns at very long cooling times for high burn-up fuels (?)

Decay Heat: thanks for all the advice and help

Alan Tobias (CEGB, UK), at the beginning …….

Francois Storrer (CEA, France), in those middle Ages …….

Olivier Bersillon (CEA, France), Robert Mills (NNL, UK) and Mohini Gupta (Manipal University, India), over the recent past