



**JAEA**

# **Measurements of Thermal-Neutron Capture Cross-Sections for Radioactive Nuclides**

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*ex. 2*  $^{241}\text{Am}(n, \gamma) ^{242g}\text{Am}$  reaction

*ex.3* other

Prompt  $\gamma$ -ray spectroscopic analysis

$^{107}\text{Pd}(n_{\text{th}}, \gamma) ^{108}\text{Pd}$  cross-section

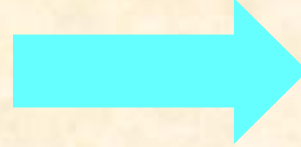
$^{93}\text{Zr}(n_{\text{th}}, \gamma) ^{94}\text{Zr}$  cross-section

## 4. Highlight Data

FP and MA cross-section data by JAEA

# 1. Motivation

**Nuclear Cross-Section Data**



**Evaluated Data File**

*ex. JENDL*

.Transmutation study, Fuel Burn-up, Fission Products, *etc.*

.Fast Breeder Reactor, ADS Reactor, Innovative Reactor, *etc.*

$^{90}\text{Sr}$   $^{137}\text{Cs}$   $^{99}\text{Tc}$   $^{129}\text{I}$

$^{134,135}\text{Cs}$   $^{166\text{m}}\text{Ho}$

$^{237}\text{Np}$   $^{238}\text{Np}$

$^{241}\text{Am}$   $^{243}\text{Am}$  ...

$^{244}\text{Cm}$   $^{242}\text{Cm}$  ...

$^{107}\text{Pd}$   $^{93}\text{Zr}$  ...

@JRR-3M.KUR *etc.*

$^{237}\text{Np}$  .KUR LINAC

$^{99}\text{Tc}$ ,  $^{237}\text{Np}$ ,  $^{129}\text{I}$

Fast Neutron Capture  
Cross-Sections

.TokyoUniv. "YAYOI"

Thermal  
Cross-Section  $s_0$

Thermal

R.I.  $s(E)$

Epi-Thermal

Fast Neutron

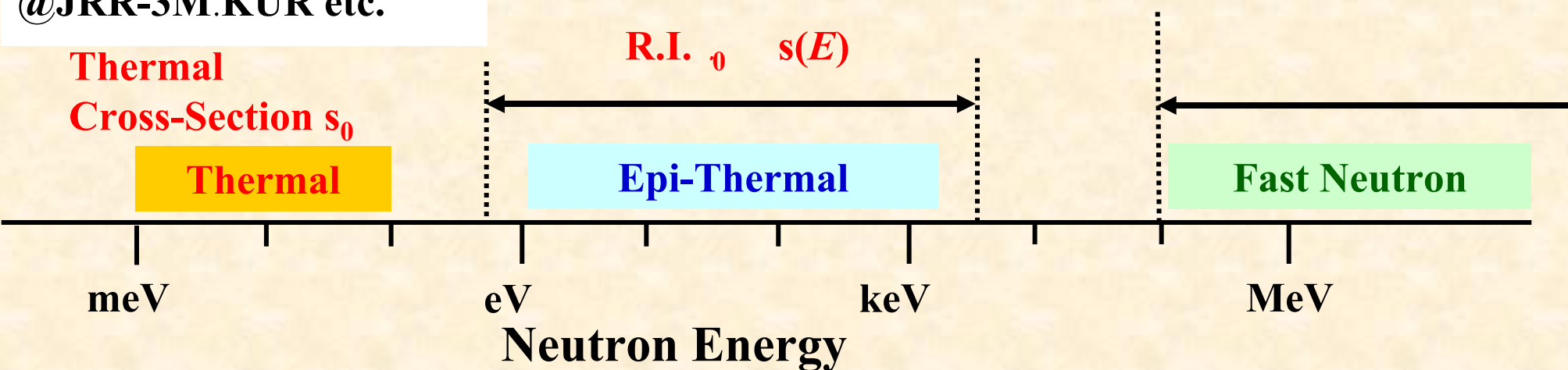
meV

eV

keV

MeV

Neutron Energy



# Problems in Cross-Section Data for FP & MA

## A very few reliable data

### 1. Discrepancies among reported data

$^{90}\text{Sr}$ . 0.8 to 0.015 (b)

$^{137}\text{Cs}$ . 0.11 to 0.25 (b)

### 2. Relatively large errors

$^{241}\text{Am}$ .  $780 \pm 50$ ,  $636 \pm 46$ , 602 (b)

### 3. A few of exp. Data $^{135}\text{Cs}$ , $^{107}\text{Pd}$ , $^{93}\text{Zr}$ etc.

### 4. No Data $^{238}\text{Np}$

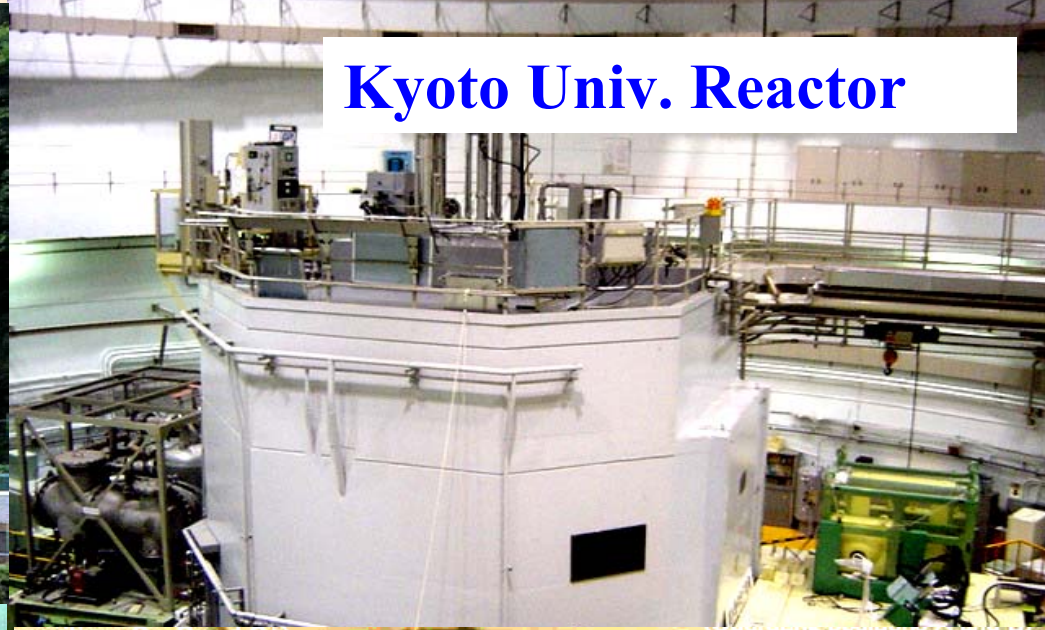


Aimed to re-measurement the cross-sections  
for important FP's and MA's

## 2.Methods -Neutron Activation Analysis-

Rikkyo Univ. Reactor

Kyoto Univ. Reactor

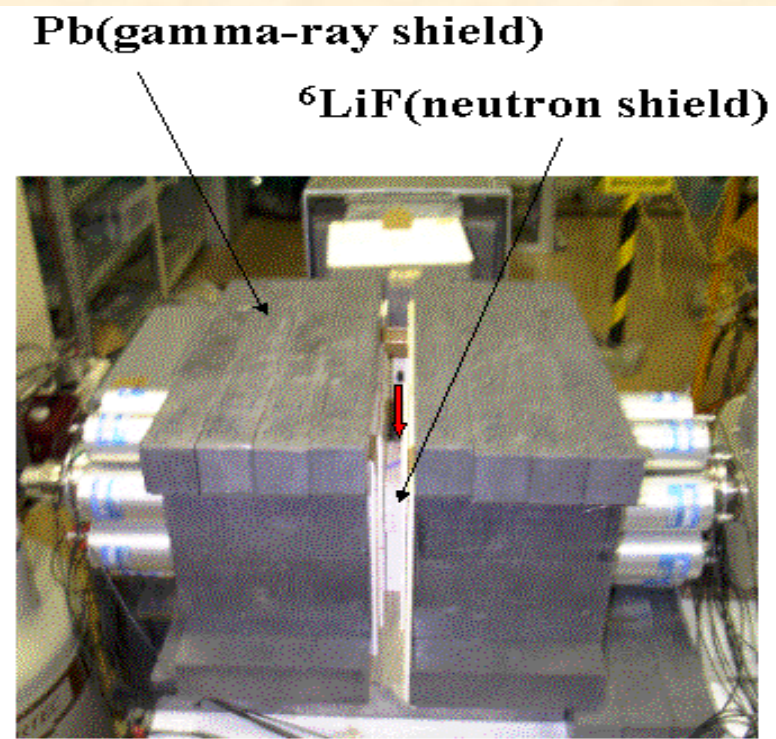
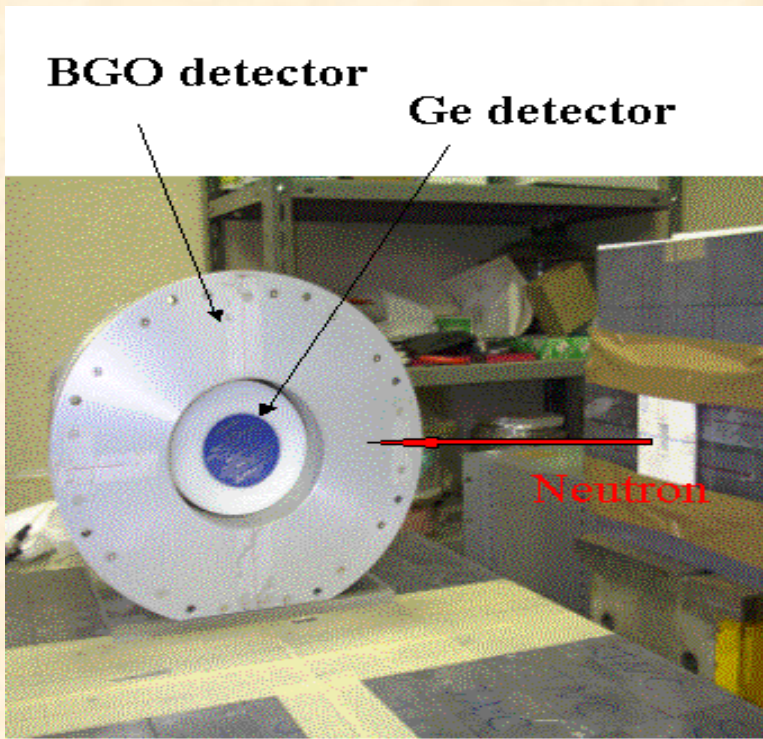
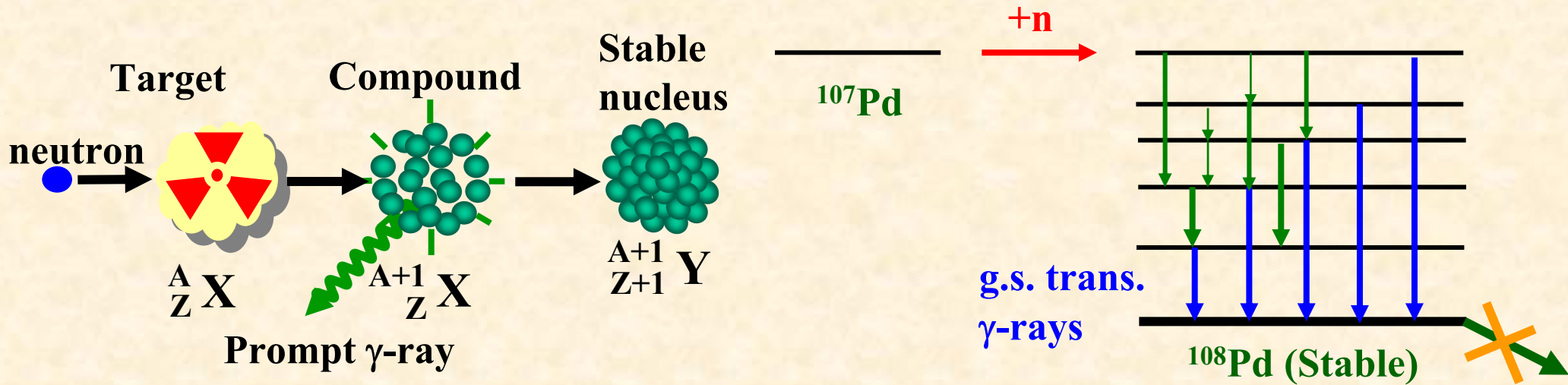


JRR-3M

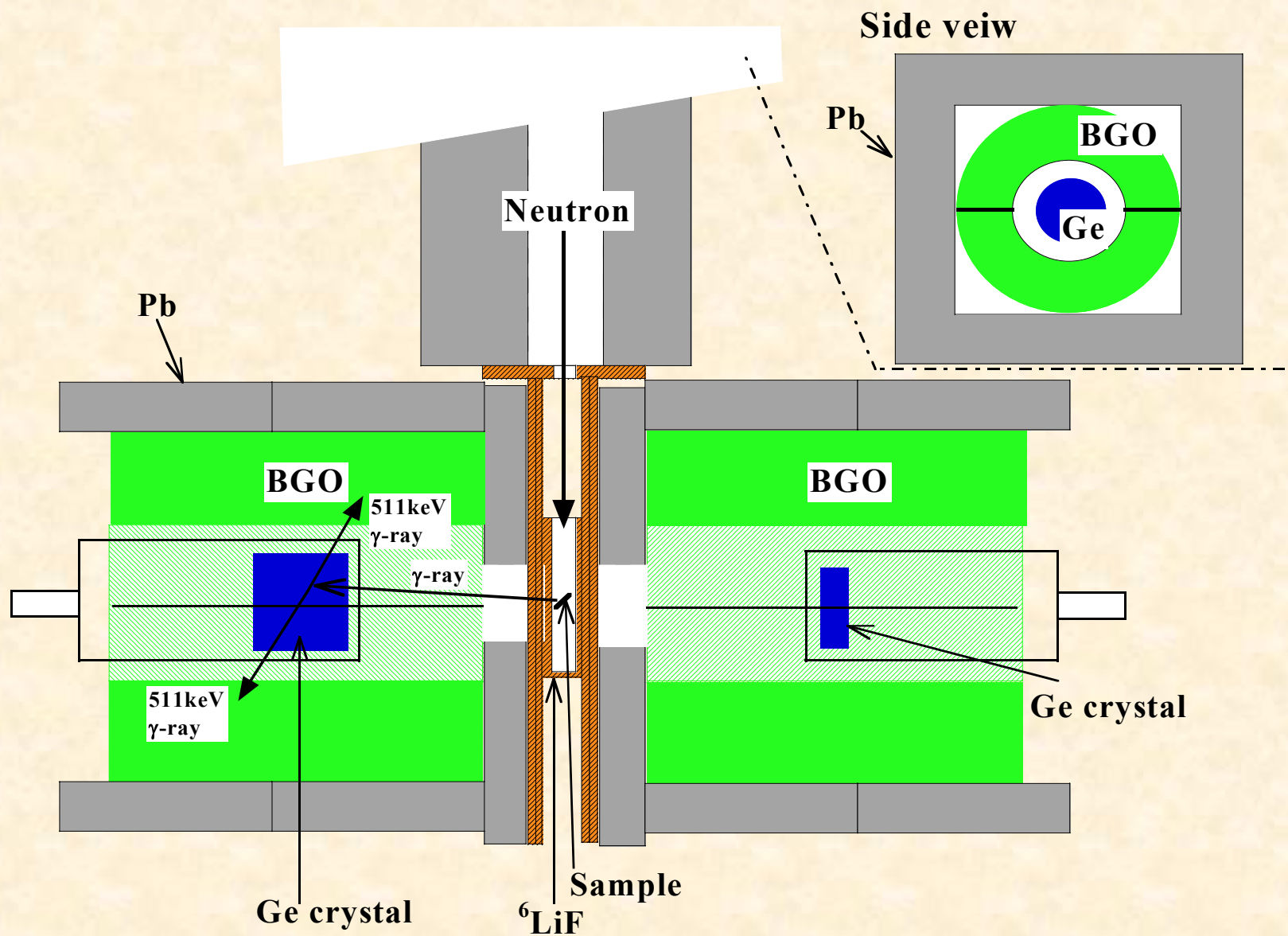


YAYOI

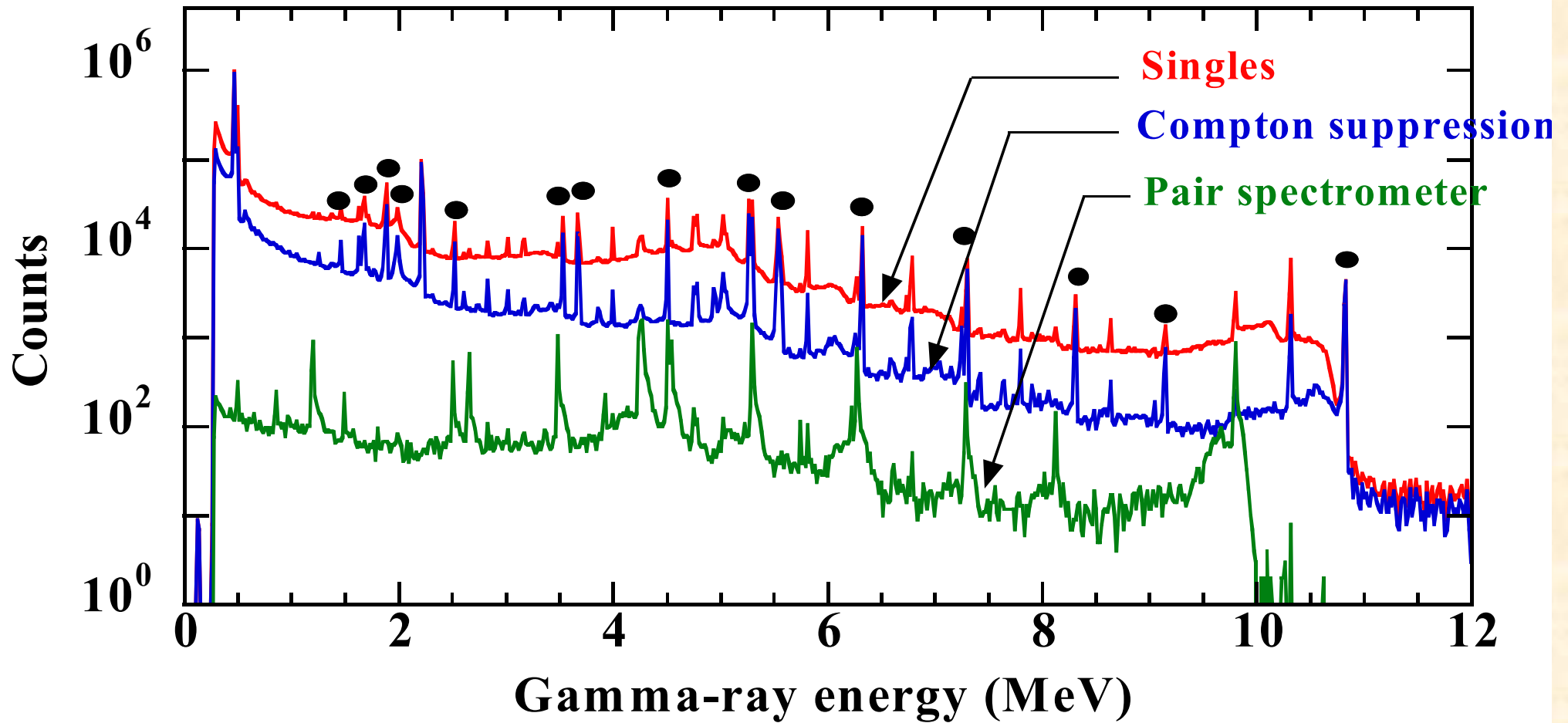
# -Prompt $\gamma$ -ray Spectroscopic Analysis-



# Ge & BGO Detector Geometry

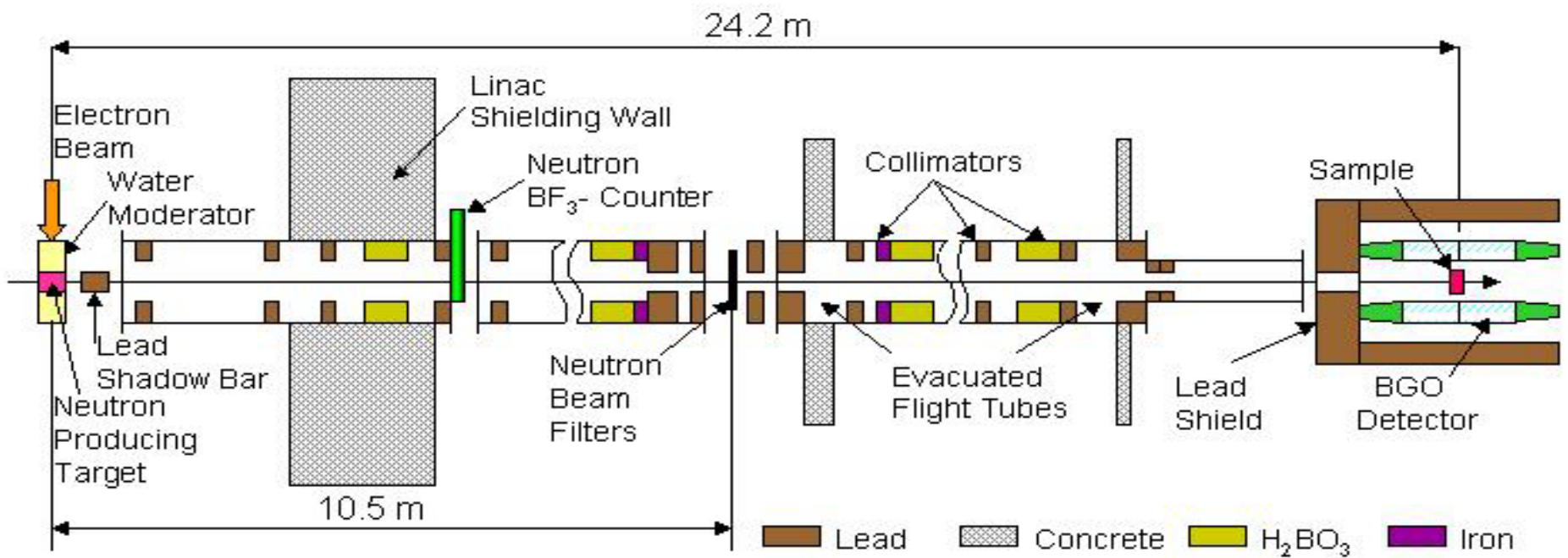


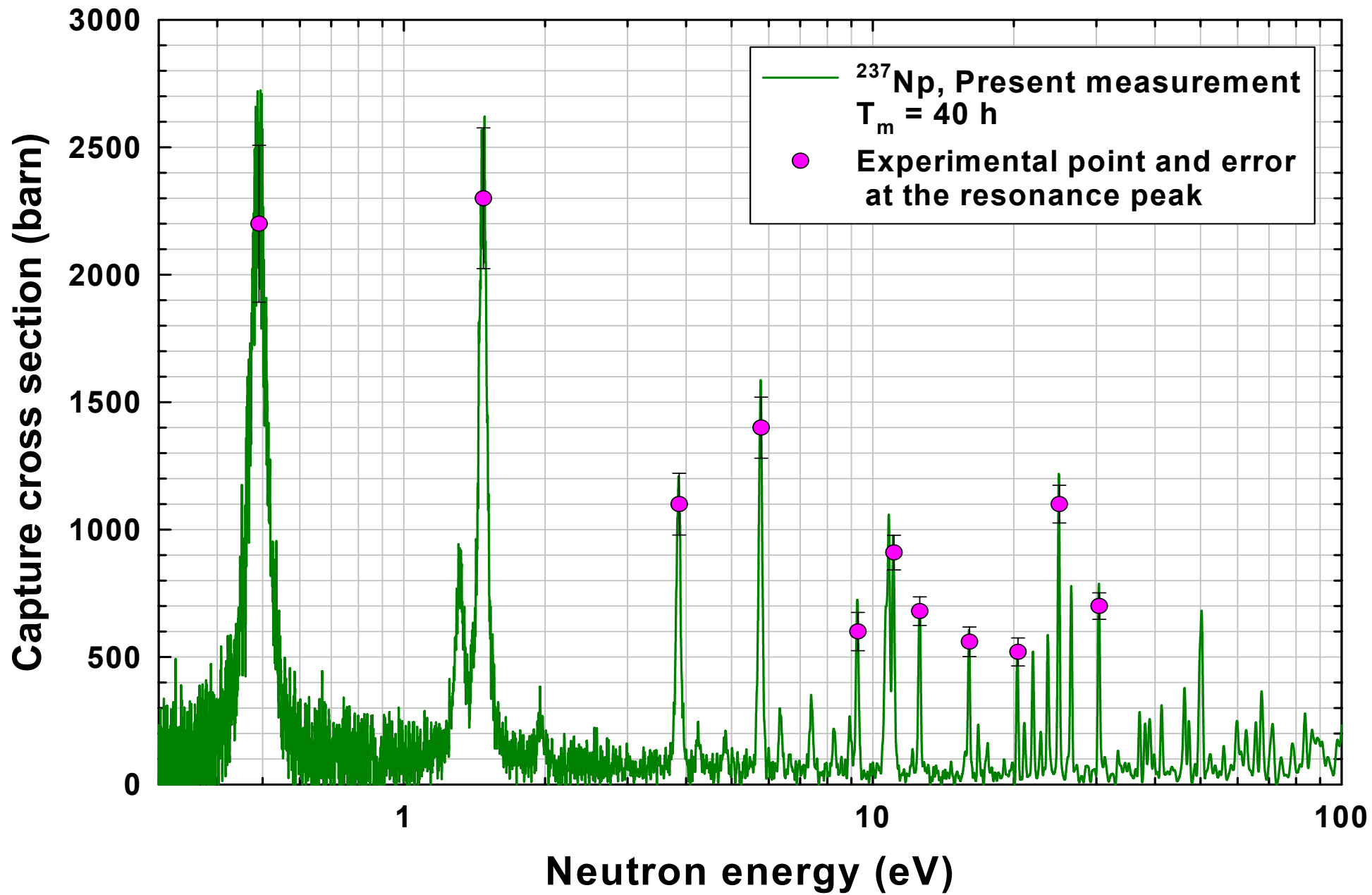
# An Example of $\gamma$ -ray spectrum





# -Time-Of-Flight Analysis-





### 3. Experiment Ex.1 $^{90}\text{Sr}(n,\gamma)^{91}\text{Sr}$ reaction

Only upper limits of  $I_0$

$$\sigma_0 = 15.3^{+1.3}_{-4.2} \text{ (mb)}$$
$$I_0 < 160 \text{ (mb)}$$

(1994 JRR-4)

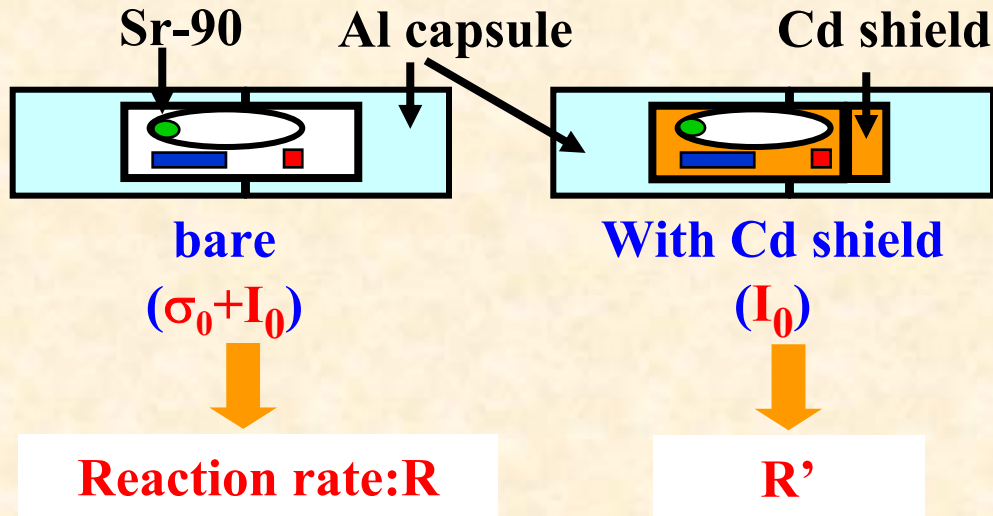
Al capsules for confinement of samples

Al capsule also play the role as a **heat sink**

**Modified Cadmium-ratio Method**

**Multi-flux monitors**

Extract  $\sigma_0$  from the reaction rate R



Kyoto Univ. Reactor : KUR @ Hydraulic Facility

Neutron Flux :  $1 \times 10^{14}$  n/cm<sup>2</sup>sec

Irradiation: 10 hours



# Ex.1 $^{90}\text{Sr}(n,\gamma)^{91}\text{Sr}$ reaction -Results-

Author	Thermal Cross Section, $\sigma_0$ (mb)	Resonance Integral, $I_0$ (mb)
<b>Present Work</b>	<b><math>10.1 \pm 1.3</math></b>	<b><math>104 \pm 16</math></b>
Harada <i>et al.</i> [1994]	$15.3^{+1.3}_{-4.2}$	<160

S.Nakamura *et al.* : *J. Nucl. Sci. Technol.*, 38, No.12, 1029(2001).

Measured  $\sigma_0$  and  $I_0$  separately

First data for  $I_0$

Error of  $\sigma_0$  was improved to 1/2

Adopted as a part of JENDL-4

# Ex.2 $^{241}\text{Am}(n,\gamma)^{242\text{g}}\text{Am}$ reaction

$^{241}\text{Am}(n,\gamma)^{242\text{g}}\text{Am}$  reaction cross-section

Differences between the reported data: 600 to 800 (b)

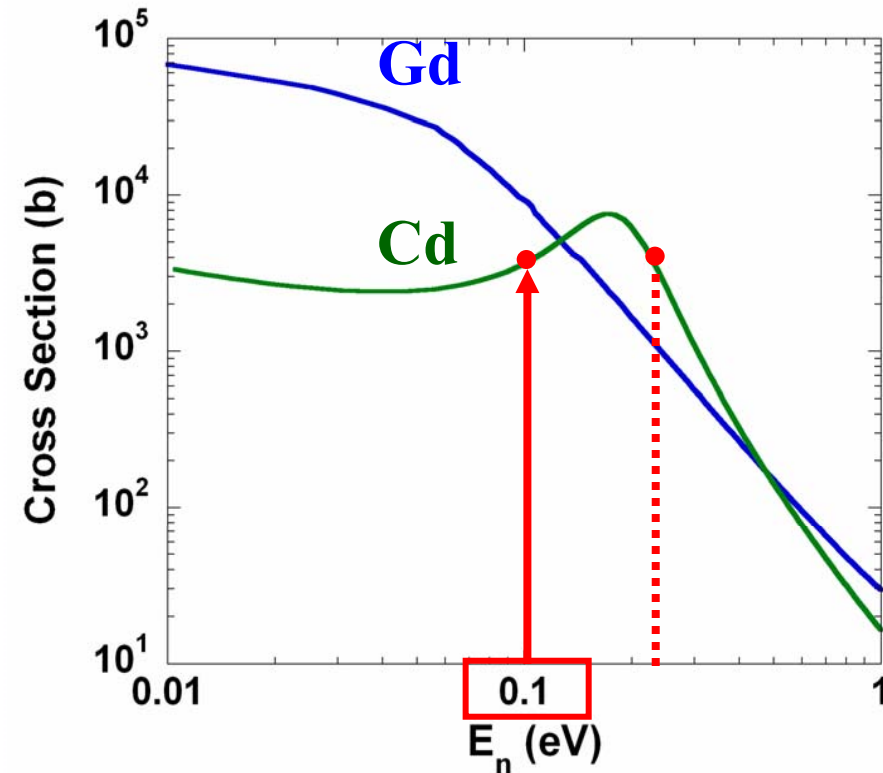
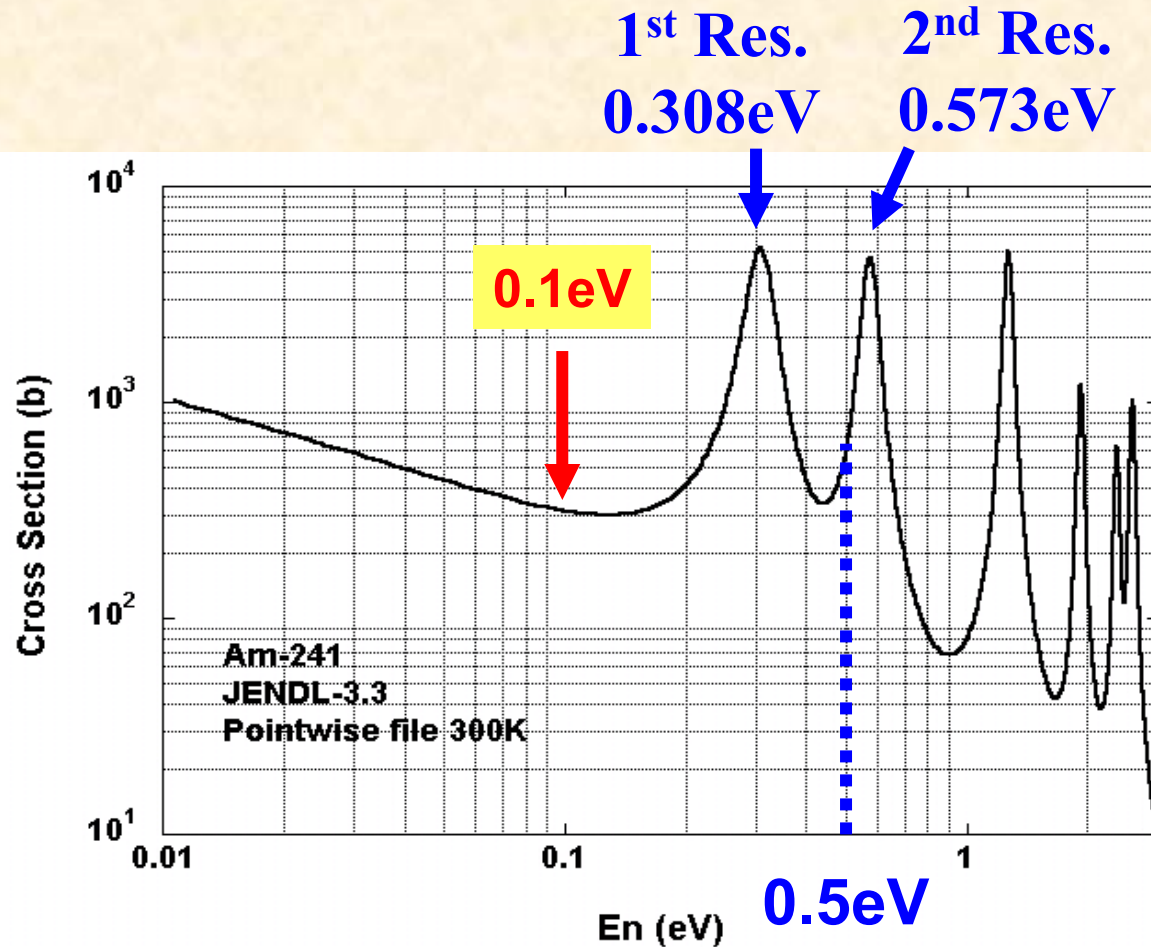
**Cd shield**

Difficult to set the cut-off into the low energy



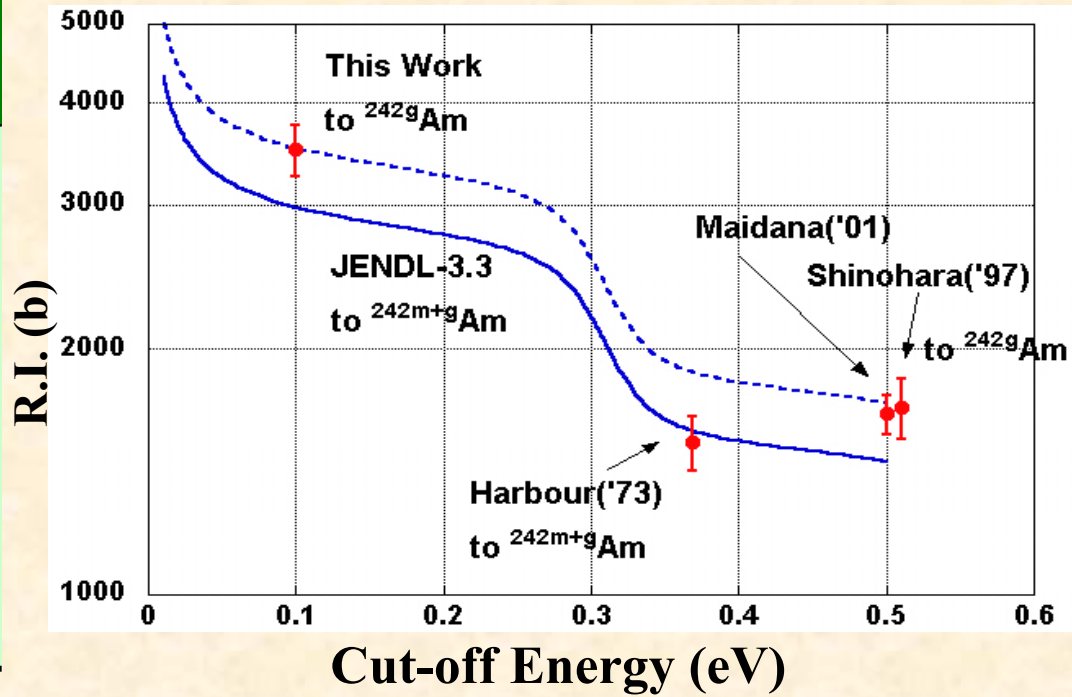
**Gd shield**

Set the cut-off to **0.1eV**



# Ex.2 $^{241}\text{Am}(n,\gamma)^{242g}\text{Am}$ reaction -Results-

Authors	$\sigma_{0g}$ (b)	$I_{0g}$ (b)	Cut-off (eV)
This Work	$628 \pm 22$	$3.5 \pm 0.3$ k	0.107
Fioni	$636 \pm 46$	---	---
Shinohara.	$768 \pm 58$	$1694 \pm 146$	0.5
Gavrulov	$780 \pm 50$	---	---
Harbour	$748 \pm 20$	$1330 \pm 117$	0.369
Bak	$670 \pm 60$	2100	---
Deal	770	---	---

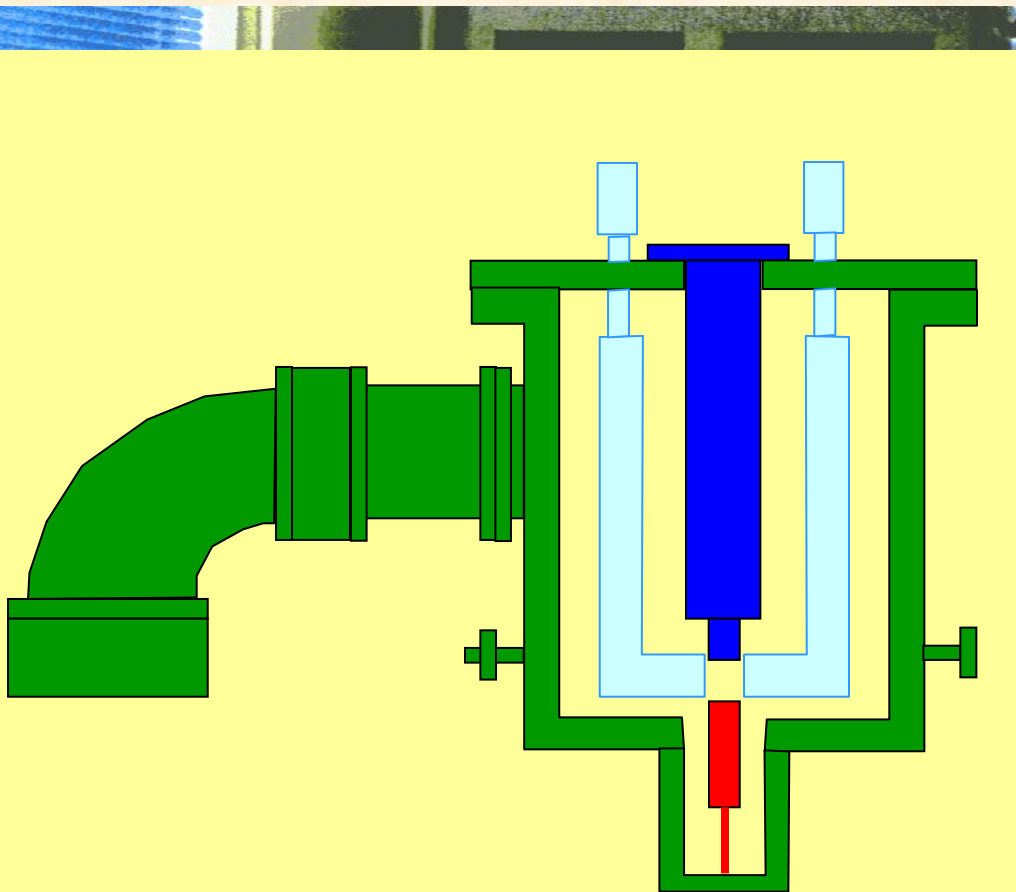


The differences among past data was caused by the treatment of the first resonance peak (0.308eV).

## Another nuclides

**Difficult and/or impossible to supply samples:** ex.  $^{135}\text{Cs}$  ( $2.3 \times 10^6$ yr)

Use  $^{135}\text{Cs}$  contained in  $^{137}\text{Cs}$  std. sol. as an impurity



**Mass Analyzer for *NAA***

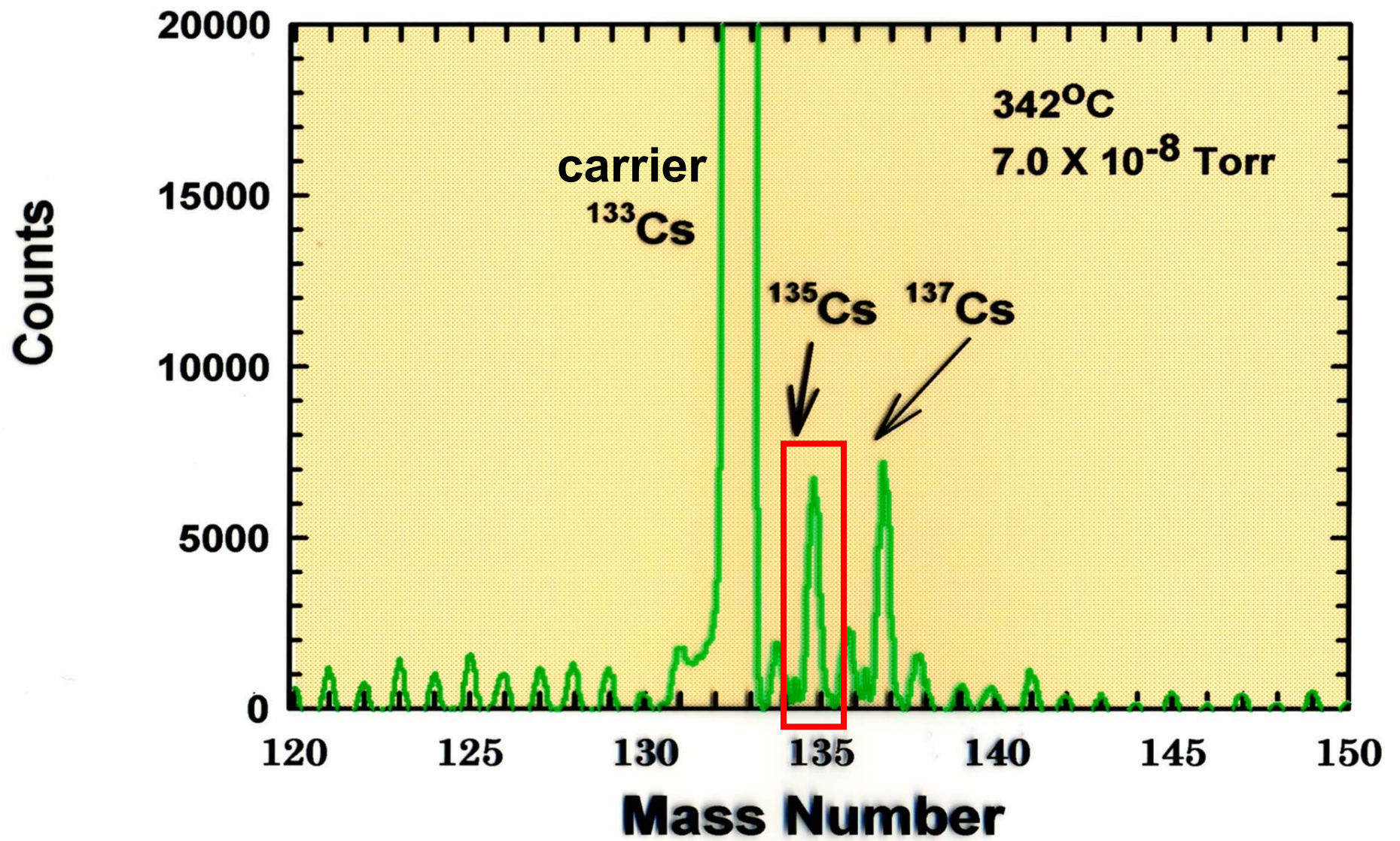
**Quadrupole Mass Filter**

**Liq. N Cooling Trap**

**High Vacuum :  $10^{-10}$  Torr**

**Detection limit : 300 ng**

# Isotope Ratio $^{135}\text{Cs}/^{137}\text{Cs}$





# 4. Highlight Data

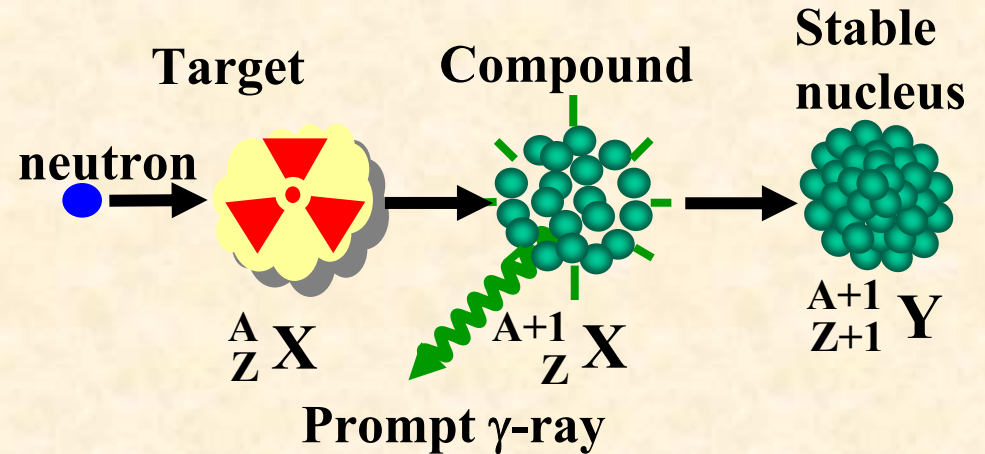
## FP's & MA's cross-section data by JAEA

Nuclide	Past Data □Author, Year□	JAEA Data	Nuclide	Past Data □Author, Year□	JAEA Data
<sup>90</sup> Sr	$\sigma_{\text{eff}} \square 0.8 \pm 0.5 \square$ (Zeisel 1966)	$\sigma_0 \square 10.1 \pm 1.3 \text{m} \square$ $I_0 \square 104 \pm 16 \text{m} \square$ (2001)	<sup>166m</sup> Ho	$\sigma_0 \square 9140 \pm 650 \square$ $I_0 \square 1140 \pm 90 \square$ □Masyanov 1993□	$\sigma_{\text{eff}} \square 3 \pm 1 \text{k} \square \square 2000 \square$ $\sigma_0 \square 3.11 \pm 0.82 \text{k} \square$ $I_0 \square 10.0 \pm 2.7 \text{k} \square$ (2002)
<sup>99</sup> Tc	$\sigma_0 \square 20 \pm 2 \square$ $I_0 \square 186 \pm 16 \square$ □Lucas 1977□	$\sigma_0 \square 22.9 \pm 1.3 \square$ $I_0 \square 398 \pm 38 \square$ □1995□			
<sup>129</sup> I	$\sigma_0 \square 27 \pm 2 \square$ $I_0 \square 36 \pm 4 \square$ □Eastwood 1958□	$\sigma_0 \square 30.3 \pm 1.2 \square$ $I_0 \square 33.8 \pm 1.4 \square$ □1996□	<sup>237</sup> Np	$\sigma_0 \square 158 \pm 3 \square$ $I_0 \square 652 \pm 24 \square$ □Kobayashi 1994□	$\sigma_0 \square 141.7 \pm 5.4 \square$ $I_0 \square 862 \pm 51 \square$ (2003□ $\sigma_0 \square 169 \pm 6 \square$ (2006)
<sup>127</sup> I	$\sigma_0 \square 4.7 \pm 0.2 \square$ $I_0 \square 109 \pm 5 \square$ □Friedmann 1983	$\sigma_0 \square 6.40 \pm 0.29 \square$ $I_0 \square 162 \pm 8 \square$ □1999□	<sup>238</sup> Np	<b>No Data</b>	$\sigma_{\text{eff}} \square 479 \pm 24 \square$ (2004)
<sup>135</sup> Cs	$\sigma_0 \square 8.7 \pm 0.5 \square$ $I_0 \square 61.7 \pm 2.3 \square$ □Baerg 1958□	$\sigma_0 \square 8.3 \pm 0.3 \square$ $I_0 \square 38.1 \pm 2.6 \square$ □1997□	<sup>241</sup> Am	$\sigma_{0g} \square 768 \pm 58 \square$ $I_0 \square 1694 \pm 146 \square$ □Shinohara 1997	$\sigma_{0g} \square 628 \pm 22 \square$ $I_0 \square 3.5 \pm 0.3 \text{k} \square$ (2007)
<sup>134</sup> Cs	$\sigma_{\text{eff}} \square 134 \pm 12 \square$ (Bayly 1958)	$\sigma_{\text{eff}} \square 141 \pm 9 \square$ (1999)	<sup>243</sup> Am	$\sigma_{0m} \square 80 \square,$ $\sigma_{0g} \square 4.3 \square$ $\sigma_{0m+g} \square 84.3 \square$ □Ice 1966□	$\sigma_{\text{eff}} \square 174.0 \pm 5.3 \square$ (2006)
<sup>133</sup> Cs	$\sigma_0 \square 30.4 \pm 0.8 \square$ $I_0 \square 461 \pm 25 \square$ □Baerg 1960□	$\sigma_0 \square 29.0 \pm 1.0 \square$ $I_0 \square 298 \pm 16 \square$ □1999□			

# Limit of application by NAA

Impossible to measure the cross-sections by NAA !!

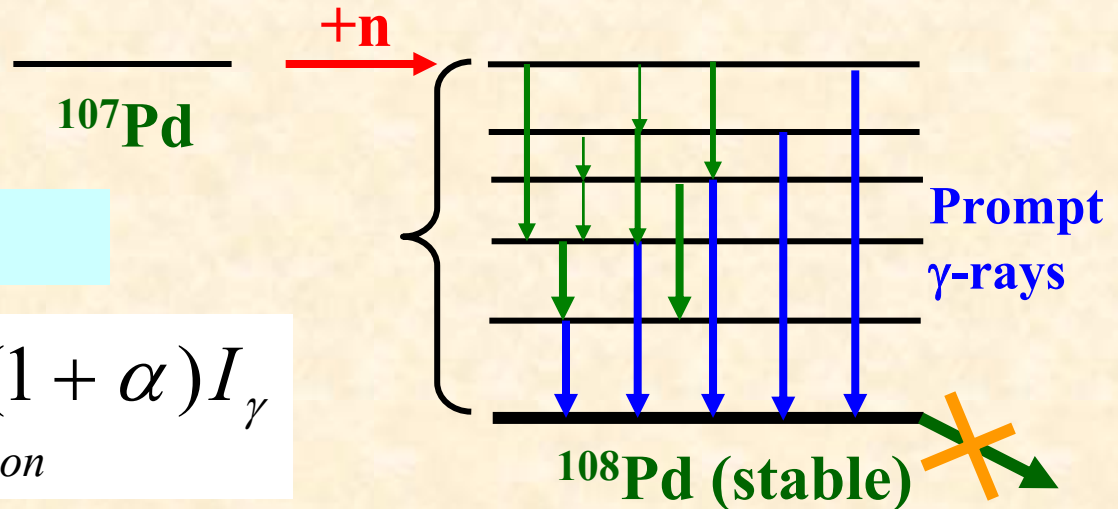
ex.  $^{107}\text{Pd}$  ( $6.5 \times 10^6 \text{y}$ )  
 $^{93}\text{Zr}$  ( $1.53 \times 10^6 \text{y}$ )



Prompt  $\gamma$ -ray spectroscopy

**g.s. Trans.  
Method**

$$\sigma = \sum_{g.s. transition} (1 + \alpha) I_{\gamma}$$



$\alpha$  : Internal Conversion Coefficient

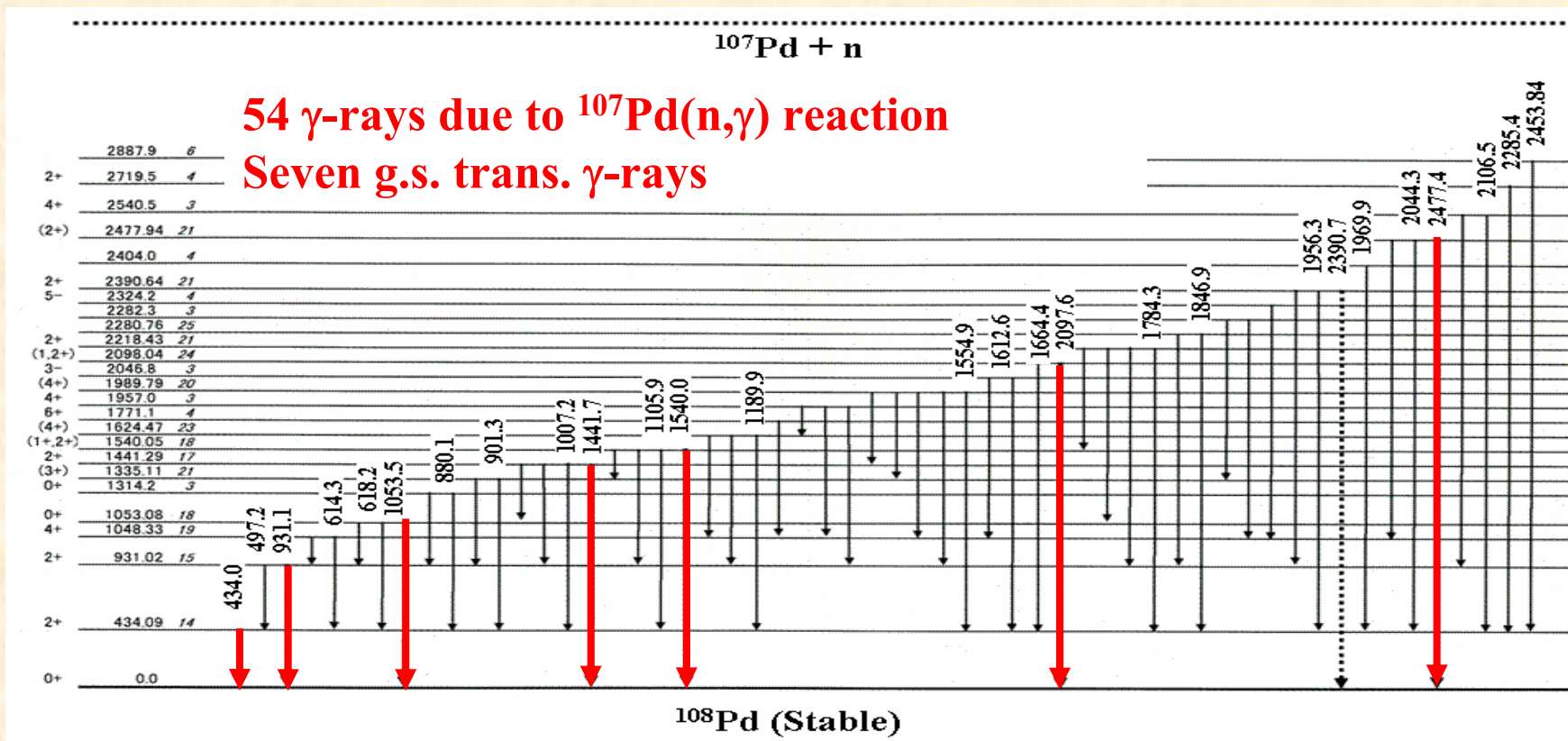
$I_{\gamma}$  :  $\gamma$ -ray intensity feeding to g.s. (unit.barn)

Stand. Cross-section 332 (mb) of  $\text{H}(n,\gamma)$

**8-MW Los Alamos  
Omega West Reactor (OWR)  
(Shutdown in 1994)**

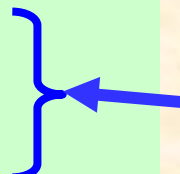


# Application to cross-section measurement for $^{107}\text{Pd}$



Author	Cross Section $\sigma_0$ (b)
Present Work	$9.16 \pm 0.27$
JENDL-3.3	2.0071
T.O.I. 8ed	$1.8 \pm 0.2$
Mughabghab	$1.8 \pm 0.2$

**Obtained  $^{107}\text{Pd}$  cross-section experimentally !  
 About 5 times larger than eval. data! !!**



**Calc. values by res. parameters**

# Result of thermal-neutron capture cross-section for $^{93}\text{Zr}(n_{\text{th}},\gamma)$ and $^{91}\text{Zr}(n_{\text{th}},\gamma)$ reactions

References		$\sigma_0$ for $^{91}\text{Zr}$ (b)	$\sigma_0$ for $^{93}\text{Zr}$ (b)
H.Pomerance <sup>a)</sup>	1952	$1.52 \pm 0.12$	$1.3 < \sigma_0 < 4$
Garrison <i>et al.</i> <sup>b)</sup>	1962	$1.2 \pm 0.32$	$1.1 \pm 0.4$
Clayton <sup>c)</sup>	1972	1.579	1.996
Mughabghab <i>et al.</i>	1981	$1.24 \pm 0.25$	$1.3 < \sigma_0 < 4$
Table of Isotopes <i>8ed</i>	1998	$1.24 \pm 0.25$	$2.7 \pm 1.4$
JENDL-3.3	2002	1.247	2.239
<b>This Work</b>		<b><math>1.30 \pm 0.04</math></b>	<b><math>0.63 \pm 0.02</math></b>

a) Measurements with ORNL pile oscillator

b) Statistical model estimates

c) Calculation by the resonance parameters from BNL-325

*Thank you  
for your kind attention*

## Supplement A:

Analysis of Prompt  $\gamma$  rays  
emitted from  $^{107}\text{Pd}(n_{\text{th}},\gamma)$  Reaction

# Analysis of Prompt $\gamma$ ray from $^{107}\text{Pd}(n_{\text{th}},\gamma)$ Reaction

Reported Data

$^{107}\text{Pd}.T_{1/2}=6.5\times 10^6$  yr

Refs.	$\sigma_0$ (b)	$I_0$ (b)
JENDL-3.3	2.0071	112.2
Macklin ('85)		$108.1 \pm 4.3^{*1}$
Holden ('81) ENSDF of U.S.NNDC	$1.8 \pm 0.2$ (b)	86.6
Singh ('78)	$1.8^{*2}$	$87^{*2}$

**\*1** Cal. val. with 130 resonances up to 3.5keV

**\*2** Cal. Val. with 34  $^{107}\text{Pd}$  resonances below 700eV



# 8-MW Los Alamos Omega West Reactor (OWR)

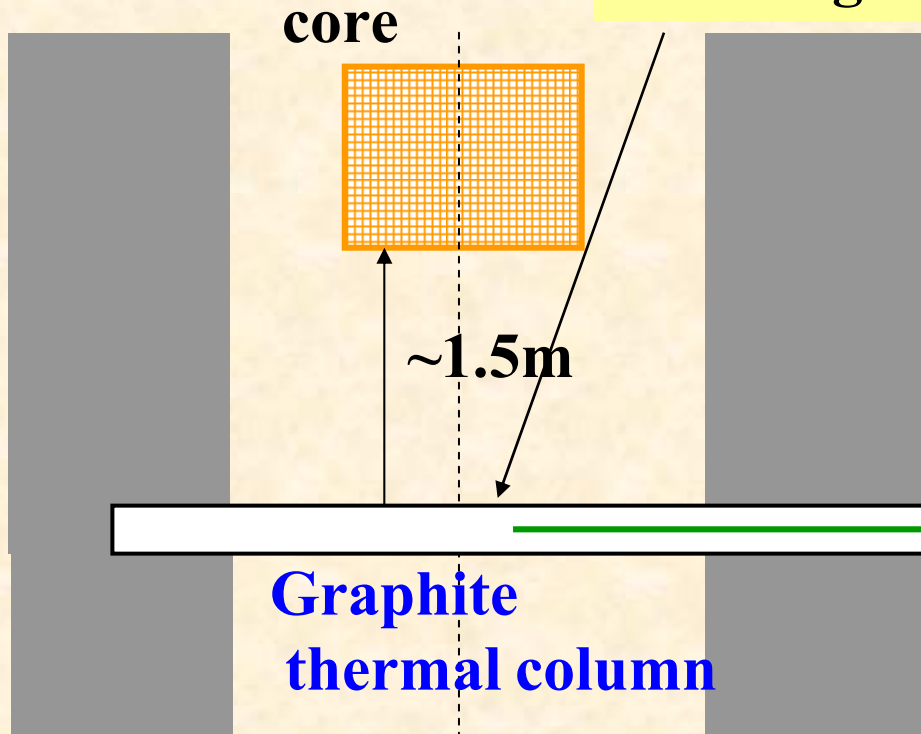


# 8-MW Los Alamos

# Exp. in 1988

## Omega West Reactor (OWR)

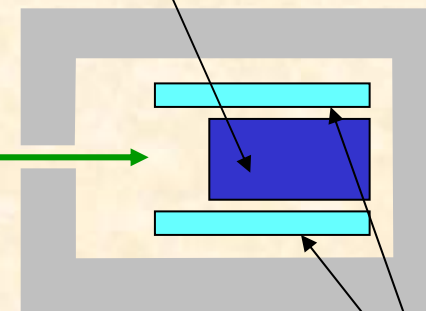
**Target**  
201.4-mg  $^{107}\text{Pd}$   
100.0-mg  $\text{CH}_2$



**Flux  $\sim 6 \times 10^{11}$  n/cm<sup>2</sup>s**  
**Cd(In) ratio  $\sim 2000$**   
**Maxwell distribution**

Mass	Abundance(%)
104	1.61 2
105	48.50 5
106	22.90 5
<b>107</b>	<b>15.54 5</b>
108	8.77 2
110	2.68 2

**Ge(Li) detector**

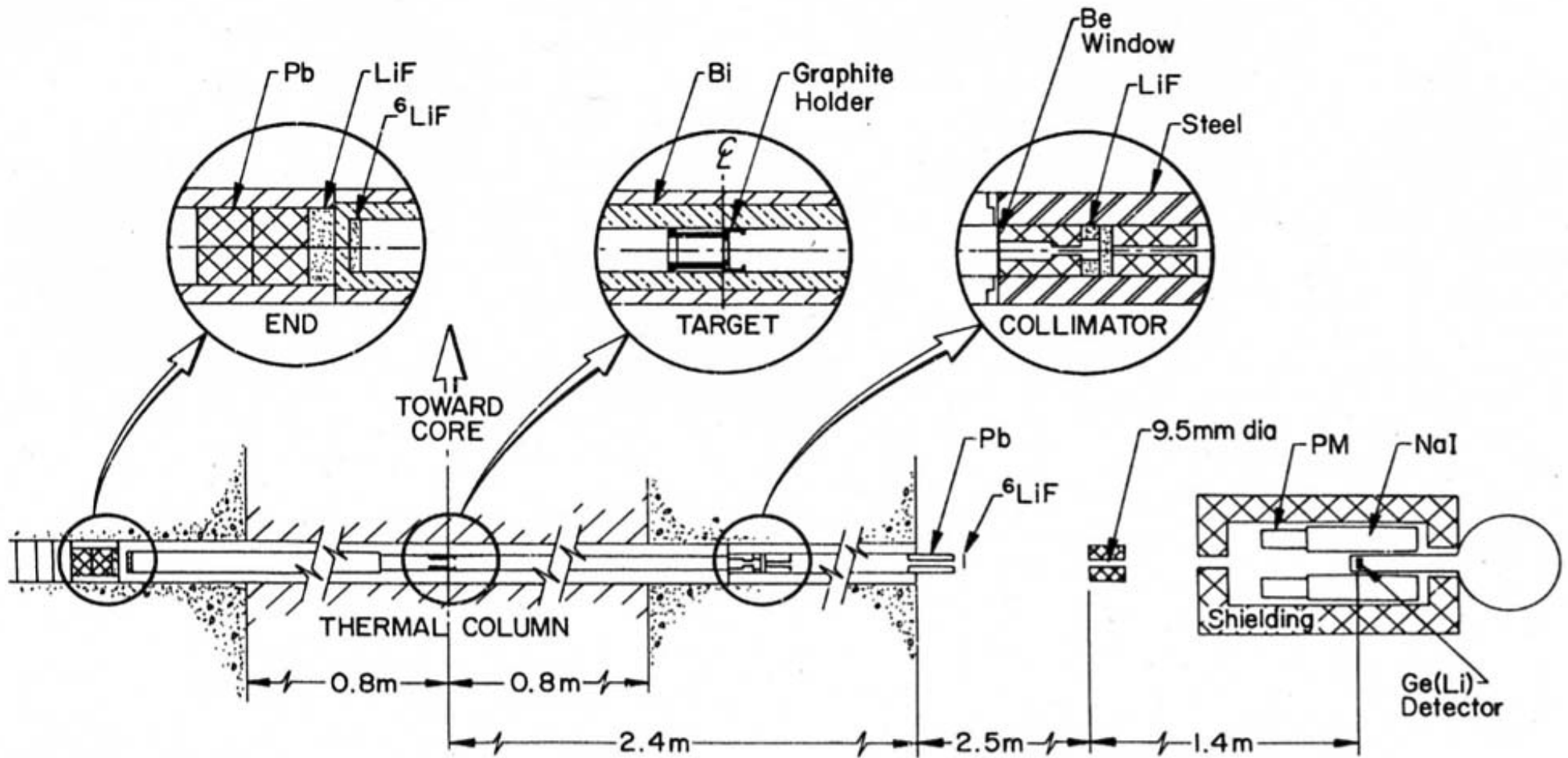


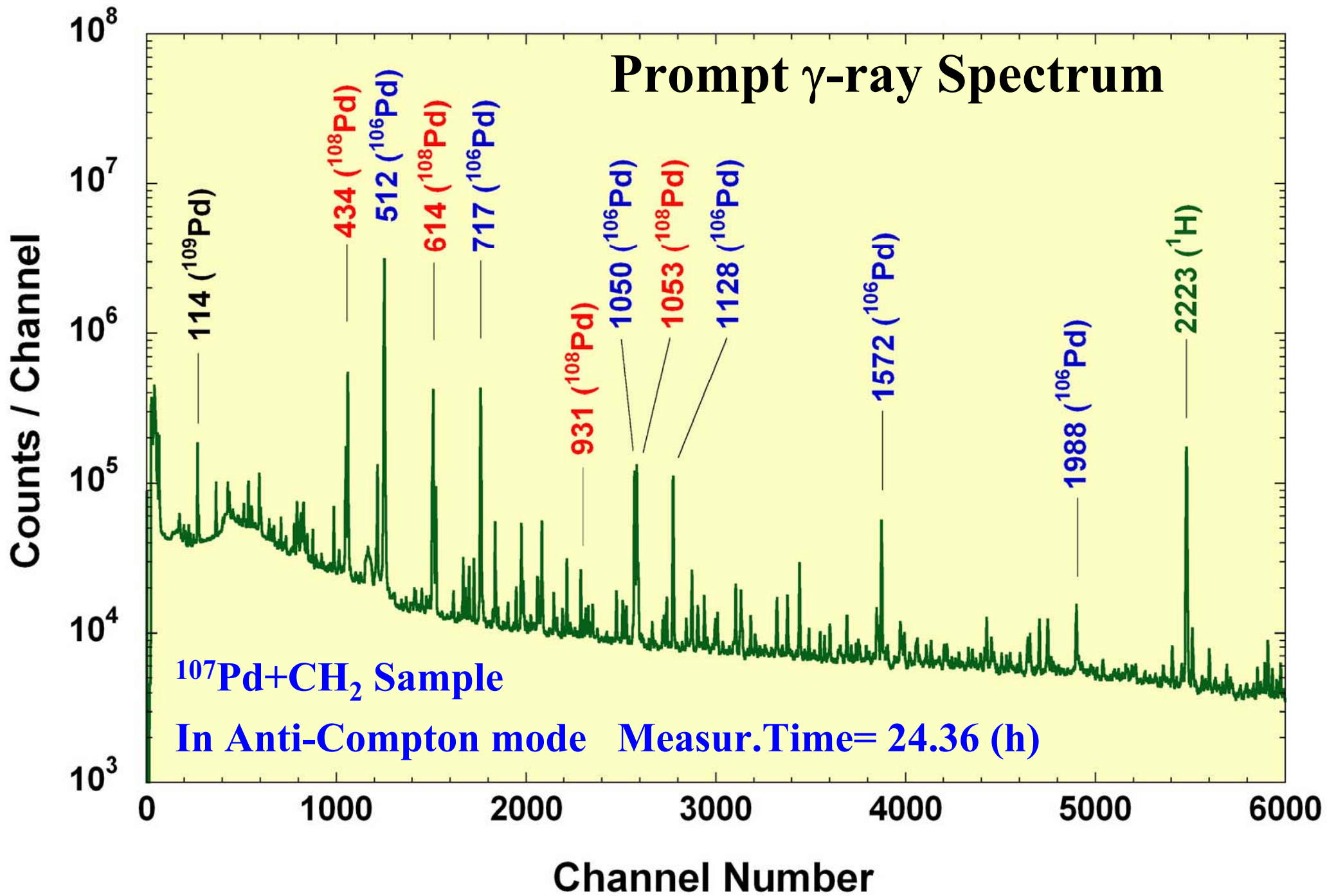
**Pb shield**

**NaI(Tl) detector**

# Experimental arrangement of the target, collimator, and detector at the Los Alamos Omega West Reactor

Phys.Rev.C, 32,18,1985. S.Raman *et al.*





# Analysis Process

$^{107}\text{Pd}+100\text{mg CH}_2$   
Anti-Compton Spectrometer Data  
Low Energy region: 0.3.3 MeV  
663 peaks

Select prompt  $\gamma$ -rays  
with known levels

Test of selected  $\gamma$ -rays  
with Gtol Code

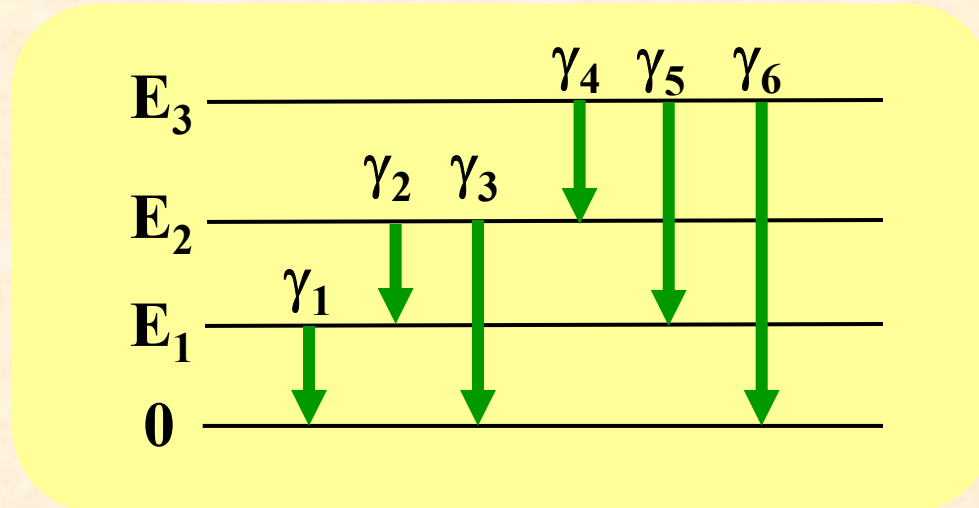
Efficiency correction  
calibration by  $^1\text{H}+n$  cross section  
 $332.6 \pm 0.7(\text{mb})$

Emission Intensity ( $I_\gamma$ )  
for  $\gamma$ -rays from  $^{107}\text{Pd}(n,\gamma)$  reaction

Lower limit of cross section  $\sum_{\text{g.s.}} I_\gamma$

# Authorization by Gtol Code

Program that calculates optimized energy levels by least-squares method with the information on  $\gamma$ -ray energies



U.S. National Nuclear Data Center

ENSDF Analysis and Utility Programs

Version 6.4b [ Dec.3,2003]

ENSDF data set

# Emission Intensity: $I_\gamma$

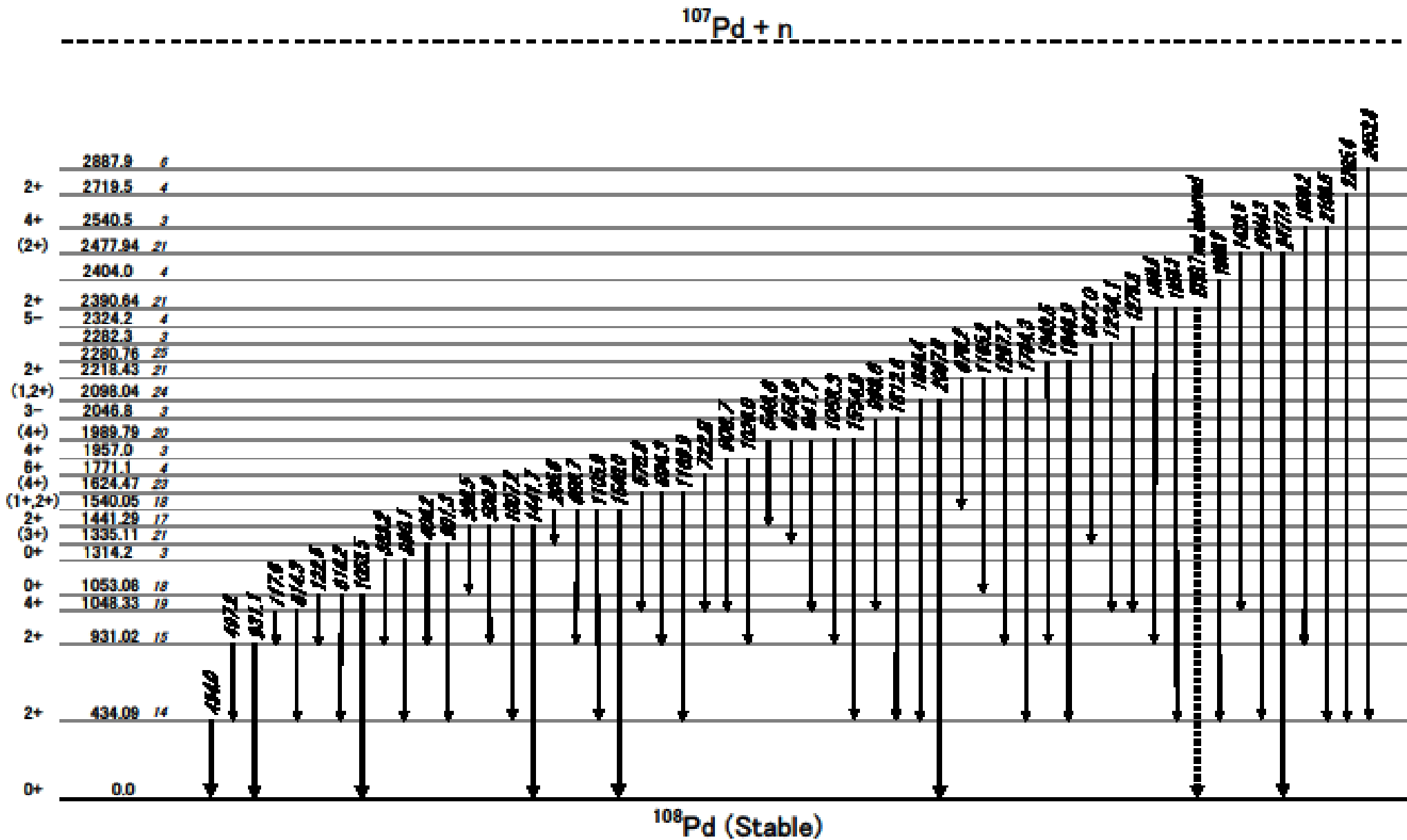
Intensity  $I_\gamma$  of prompt  $\gamma$  ray from  $^{107}\text{Pd}(n,\gamma)$  reaction are given by following equation:

$$I_\gamma = \frac{\varepsilon_H n_H \sigma_H}{n_{pd} Y_H} \cdot \frac{Y_{Pd}}{\varepsilon_{pd}}$$

where

- $n_H \cdot n_{Pd}$  : Target amounts of H and  $^{107}\text{Pd}$
- $\varepsilon_H$  : Detection efficiency fo 2.2-MeV  $\gamma$  ray
- $\varepsilon_{Pd}$  : Detection efficiency fo 2.2-MeV  $\gamma$  ray  $^{108}\text{Pd}$
- $\sigma_H$  :  $^1\text{H}$  cross-section  $332.6 \pm 0.7(\text{mb})$
- $Y_H$  : Yield of 2.2-MeV  $\gamma$  ray
- $Y_{Pd}$  : Yield of prompt  $\gamma$  ray from  $^{108}\text{Pd}$

# Extraction of prompt $\gamma$ rays





# Results of authorization

Level (keV)	$J_{\pi}$	Deexciting $\gamma$ rays	Level (keV)	$J_{\pi}$	Deexciting $\gamma$ rays
0.0	$0^+$		2046.2 3	$3^-$	998.0, 1612.3
434.01 15	$2^+$	433.1	2097.87 23	$(1,2^+)$	1664.2, 2097.5
930.54 17	$2^+$	496.4, 930.5	2217.74 21	$2^+$	677.5, 1164.6, 1287.3, 1784.1
1047.87 20	$4^+$	613.5	2281.1 3		1351.0, 1846.7
1053.11 21	$0^+$	619.1, 1053.1	2282.49 12		946.4, 1233.6
1313.4 3	$0^+$	382.3, 879.5	2325.3 7	$5^-$	1277.4
1334.73 25	$(3^+)$	900.6	2390.36 21	$2^+$	1459.4, 1956.2, 2390.7
1440.90 21	$2^+$	1006.6, 1441.3	2403.8 4		1969.8
1539.75 20	$(1^+,2^+)$	1105.3, 1539.7	2477.65 20	$(2^+)$	1429.2, 2044.2, 2477.6
1623.7 3	$(4^+)$	693.5, 1189.4	2540.1 3	$4^+$	1608.9, 2106.4
1956.0 3	$4^+$	908.1, 1025.4	2719.4 4	$2^+$	2285.4
1988.79 21	$(4^+)$	548.0, 654.1, 941.1, 1057.8, 1554.6			

# Lower limit of thermal-neutron capture cross-section

Emission Intensities:  $I_g$  of  $\gamma$ -rays feeding to the ground state of  $^{108}\text{Pd}$

Observed $\gamma$ rays (keV)	Intensity $I_\gamma$ (mb)
434.0	$7588 \pm 255$
931.1	$606 \pm 19$
1053.5	$488 \pm 73$
1441.7	$214 \pm 8$
1540.0	$104 \pm 6$
2097.6	$83 \pm 6$
2477.4	$72 \pm 7$

$$\sum_{g.s.} I_\gamma \cdot (1 + \alpha_T) = 9.16 \pm 0.27 \quad (b)$$

# Gamma-ray intensity balance for the 434keV level

	$E_\gamma$ (keV)	$I_\gamma$ (mb)	$E_\gamma$ (keV)	$I_\gamma$ (mb)
<b>OUT</b>	<b>434.0</b>	<b>7588<math>\pm</math>255</b>		
<b>IN</b>	<b>497.2</b>	<b>1940<math>\pm</math>63</b>	<b>1664.4</b>	<b>34<math>\pm</math>4</b>
	<b>614.3</b>	<b>2168<math>\pm</math>81</b>	<b>1784.3</b>	<b>117<math>\pm</math>7</b>
	<b>618.2</b>	<b>454<math>\pm</math>30</b>	<b>1946.9</b>	<b>133<math>\pm</math>6</b>
	<b>880.1</b>	<b>93<math>\pm</math>4</b>	<b>1956.3</b>	<b>45<math>\pm</math>5</b>
	<b>901.3</b>	<b>713<math>\pm</math>23</b>	<b>1969.9</b>	<b>33<math>\pm</math>4</b>
	<b>1007.2</b>	<b>399<math>\pm</math>13</b>	<b>2044.3</b>	<b>133<math>\pm</math>8</b>
	<b>1105.9</b>	<b>169<math>\pm</math>9</b>	<b>2106.5</b>	<b>103<math>\pm</math>7</b>
	<b>1189.9</b>	<b>39<math>\pm</math>5</b>	<b>2285.4</b>	<b>82<math>\pm</math>8</b>
	<b>1554.9</b>	<b>42<math>\pm</math>8</b>	<b>2453.8</b>	<b>27<math>\pm</math>6</b>
	<b>1612.6</b>	<b>374<math>\pm</math>18</b>		

**IN : 7.10 $\pm$ 0.11 (b)**

**OUT : 7.59 $\pm$ 0.26 (b)**

# Result of thermal-neutron capture cross-section for $^{107}\text{Pd}(n_{\text{th}},\gamma)^{108}\text{Pd}$ reaction

Refs.	$\sigma_0$ (b)	$I_0$ (b)
<b>This Work</b>	<b><math>9.16 \pm 0.27</math></b>	
<b>JENDL-3.3</b>	<b>2.0071</b>	<b>112.2</b>
<b>Macklin ('85)</b>		<b><math>108.1 \pm 4.3^{*1}</math></b>
<b>Holden ('81)</b> ENSDF of U.S.NNDC	<b><math>1.8 \pm 0.2</math>(b)</b>	<b>86.6</b>
<b>Singh ('78)</b>	<b><math>1.8^{*2}</math></b>	<b><math>87^{*2}</math></b>

**\*1** Cal. val. with 130 resonances up to 3.5keV

**\*2** Cal. Val. with 34  $^{107}\text{Pd}$  resonances below 700eV

# Result of thermal-neutron capture cross-section for $^{105}\text{Pd}(n_{\text{th}},\gamma)^{106}\text{Pd}$ reaction

References		$\sigma_0$ (b)
Table of Isotopes 8th	1998	$20.0 \pm 3.0$
JENDL-3.3	2002	20.25
Mughabghab <i>et al.</i>	2003	$21.0 \pm 1.5$
Firestone <i>et al.</i>	2005	$21.1 \pm 1.5^*$
<b>This Work</b>		<b><math>19.1 \pm 0.5^*</math></b>

\* Prompt g-ray analysis

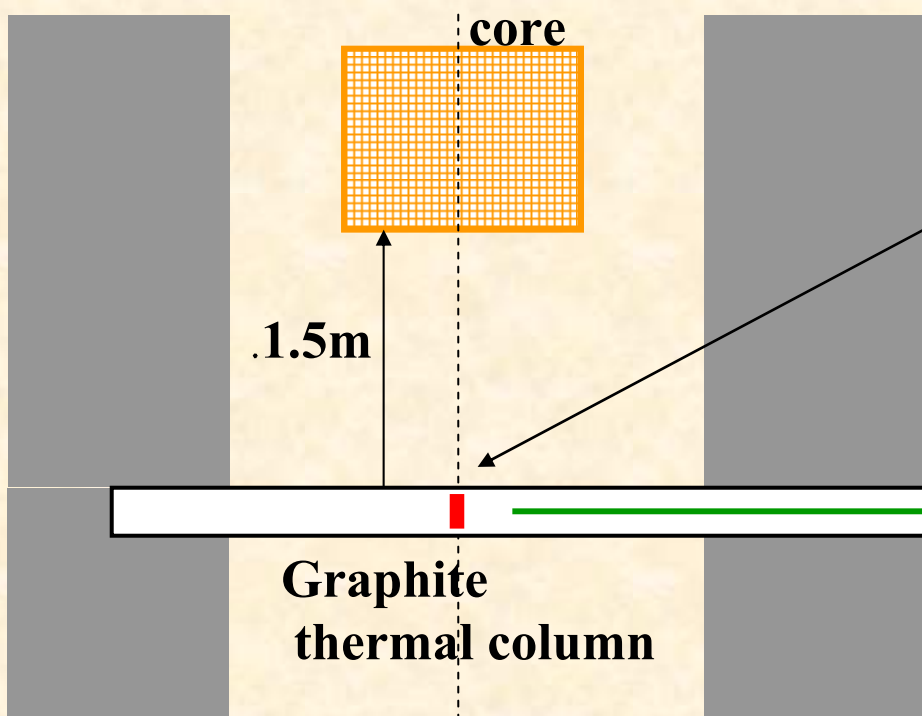
## Supplement B:

Analysis of Prompt  $\gamma$  rays  
emitted from  $^{93}\text{Zr}(n_{\text{th}},\gamma)$  Reaction

# Experiment

8MW Los Alamos

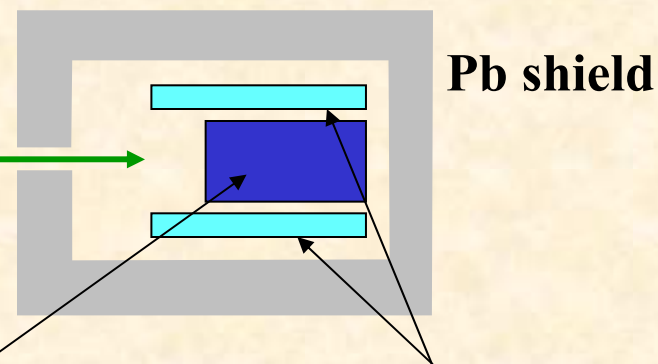
Omega West Reactor



Flux  $.6 \times 10^{11}$  n/cm<sup>2</sup>s  
Cd(In) ratio . 2000  
Maxwell distribution

Mass	Abundance(%)
90	2.29 ± 0.05
91	18.61 ± 0.10
92	18.95 ± 0.10
93	19.98 ± 0.10
94	20.50 ± 0.10
96	19.67 ± 0.10

Irrad. Target  
114.0-mg <sup>93</sup>Zr  
100.0-mg (CH<sub>2</sub>)<sub>n</sub>

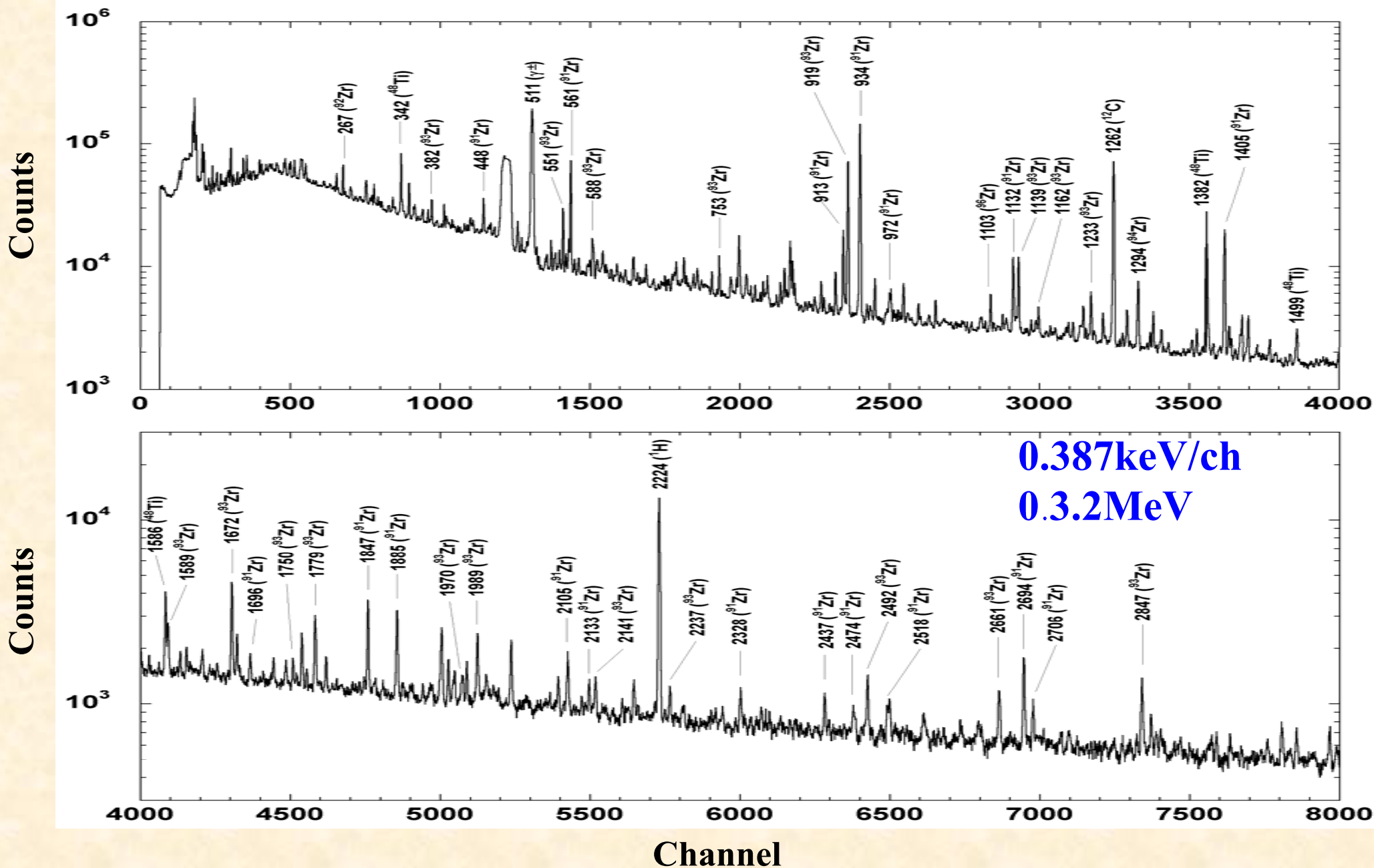


Ge(Li) detector

NaI(Tl) detector

Anti-Compton mode  
Pair-spectrometer mode

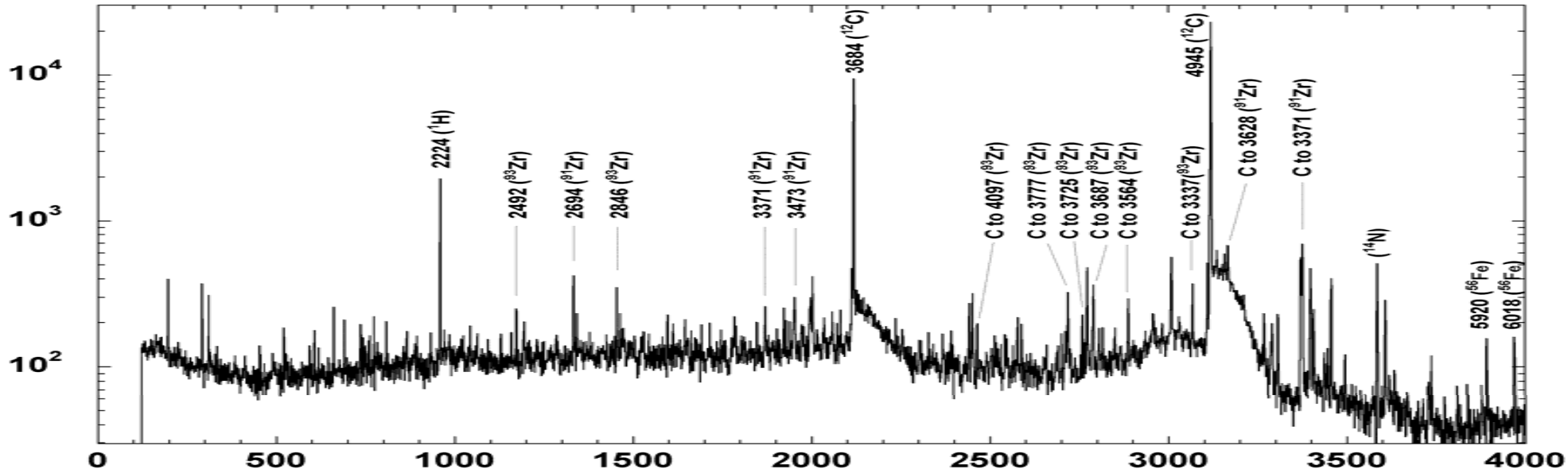
# Measured Spectra (Anti-Compton mode)



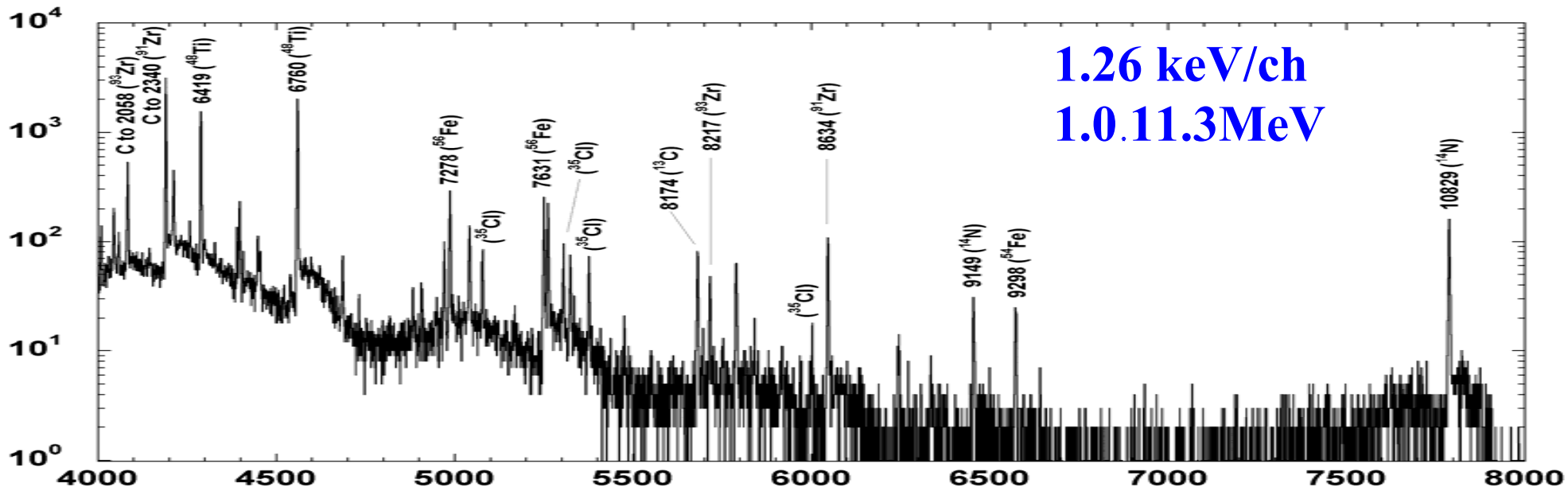


# Measured Spectra (Pair-spectrometer mode)

Counts



Counts



Channel

# Analysis Process

.Zr + 100 mg (CH<sub>2</sub>)<sub>n</sub>  
.Anti-Compton mode  
.3.176 MeV  
. 219 peaks

Efficiency correction  
calibration by <sup>1</sup>H+n  
 $\sigma = 332.6 \pm 0.7(\text{mb})$

$$I_{\gamma} = \frac{\varepsilon_H n_H \sigma_H}{n_{Zr} Y_H} \cdot \frac{Y_{Zr}}{\varepsilon_{Zr}}$$

Intensity for 918 keV  $\gamma$ -ray

Zr  
Anti-Compton mode  
.3.176 MeV  
530 peaks

Zr  
Pair Spectrometer mode  
1.0. 11.3 MeV  
162 peaks

Gtol Code

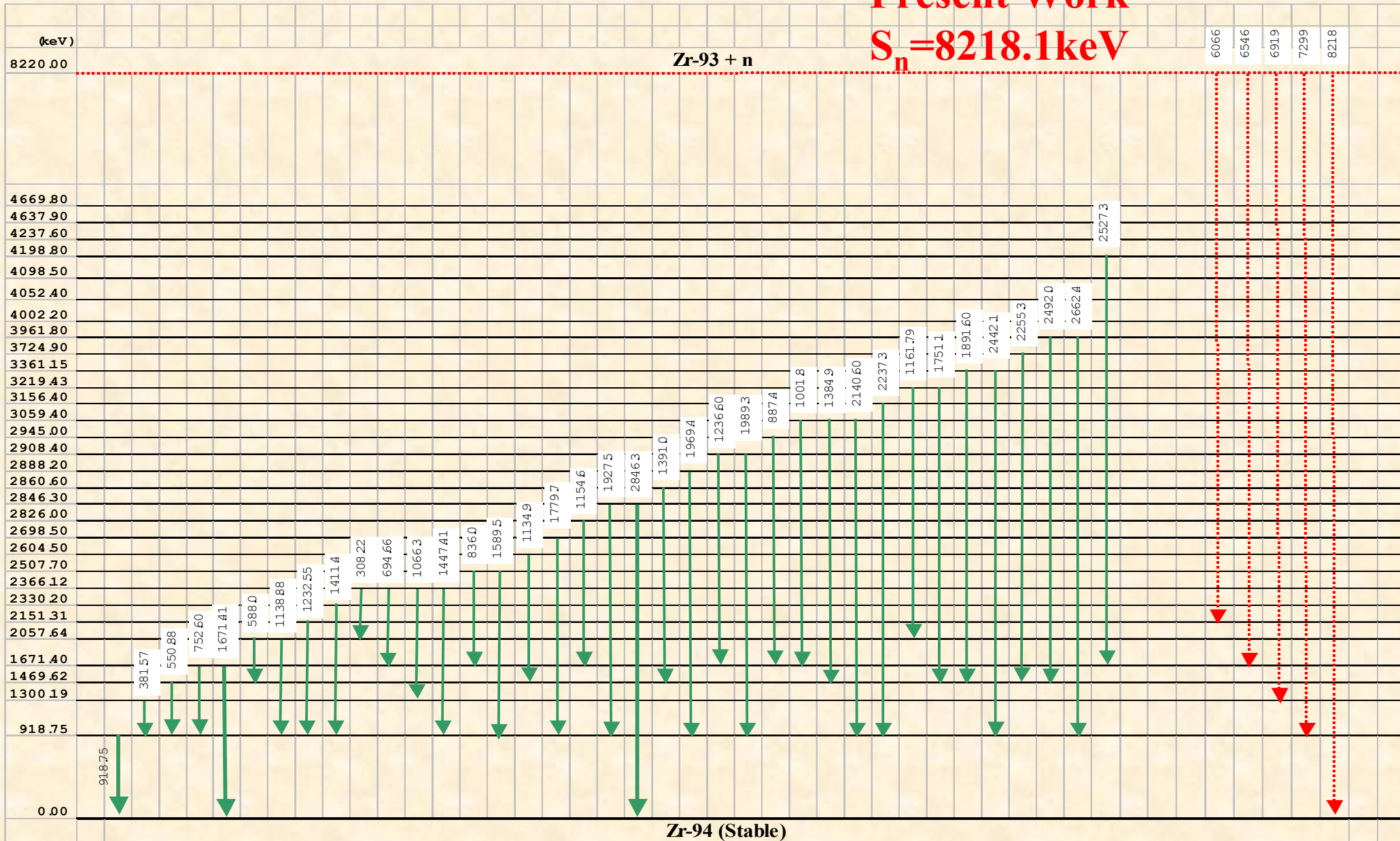
Intensities  $I_{\gamma}$  (mb)

lower limit of cross sections  
 $\Sigma I_{\gamma}$  g.s.

# Prompt $\gamma$ rays due to $^{93}\text{Zr}(n_{\text{th}},\gamma)$ reaction

Present Work

$S_n = 8218.1 \text{ keV}$



# Lower limit of thermal-neutron capture cross-section

Emission Intensities:  $I_\gamma$  of  $\gamma$ -rays feeding to the ground state of  $^{94}\text{Zr}$

Observed $\gamma$ rays (keV)	Intensity $I_\gamma$ (mb)
918.8	$543.5 \pm 3.7$
1671.5	$53.1 \pm 1.1$
2846.5	$27.1 \pm 0.7$
8217.7	$4.6 \pm 0.4$

$$\sum_{g.s.} I_\gamma = 0.63 \pm 0.02 \quad (b)$$

## Result of thermal-neutron capture cross-section for $^{93}\text{Zr}(n_{\text{th}},\gamma)$ and $^{91}\text{Zr}(n_{\text{th}},\gamma)$ reactions

References		$\sigma_0$ for $^{91}\text{Zr}$ (b)	$\sigma_0$ for $^{93}\text{Zr}$ (b)
H.Pomerance <sup>a)</sup>	1952	$1.52 \pm 0.12$	$1.3 < \sigma_0 < 4$
Garrison <i>et al.</i> <sup>b)</sup>	1962	$1.2 \pm 0.32$	$1.1 \pm 0.4$
Clayton <sup>c)</sup>	1972	1.579	1.996
Mughabghab <i>et al.</i>	1981	$1.24 \pm 0.25$	$1.3 < \sigma_0 < 4$
Table of Isotopes <i>8ed</i>	1998	$1.24 \pm 0.25$	$2.7 \pm 1.4$
JENDL-3.3	2002	1.247	2.239
<b>This Work (lower limit)</b>		<b><math>1.30 \pm 0.04</math></b>	<b><math>0.63 \pm 0.02</math></b>

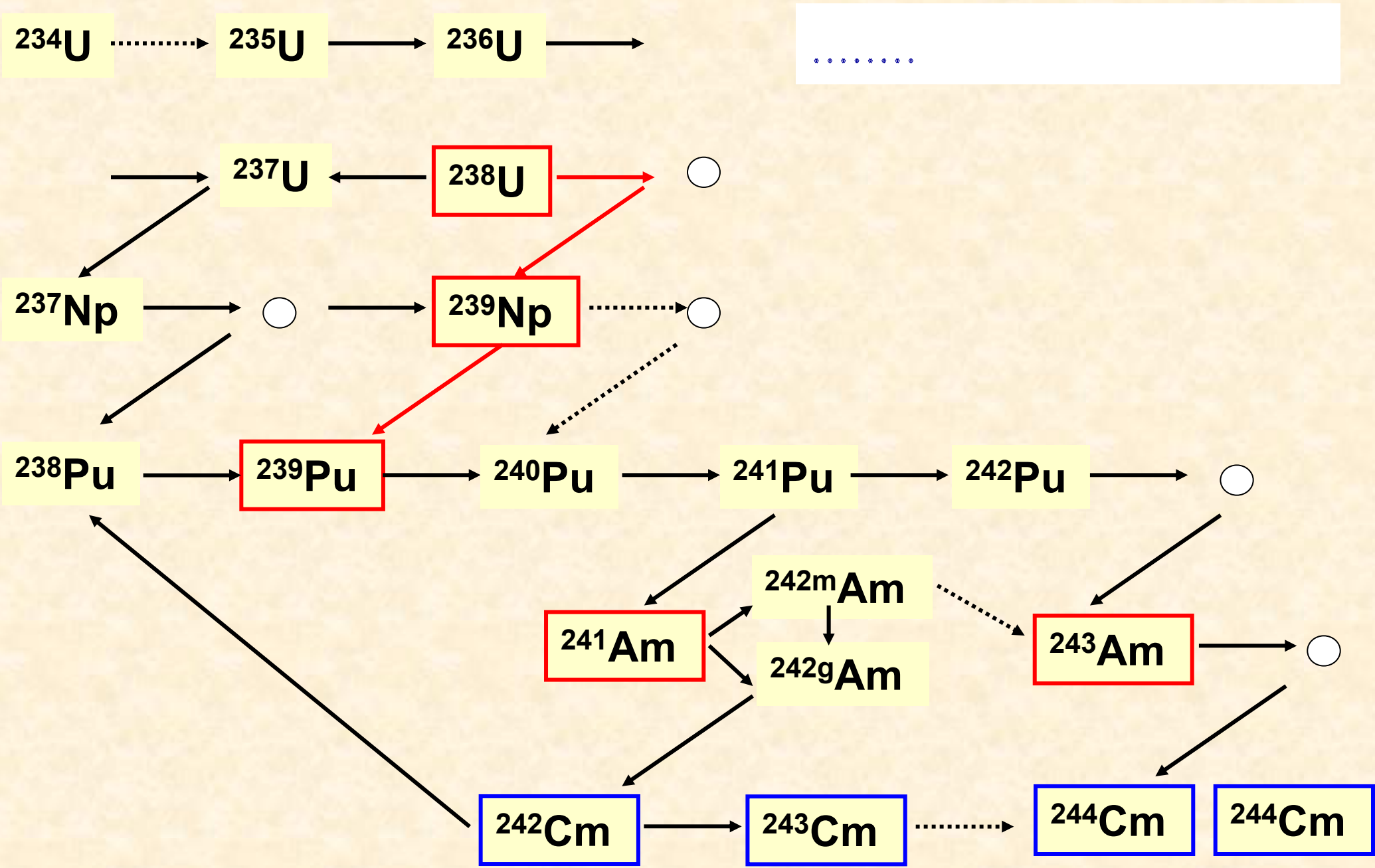
a) Measurements with ORNL pile oscillator

b) Statistical model estimates

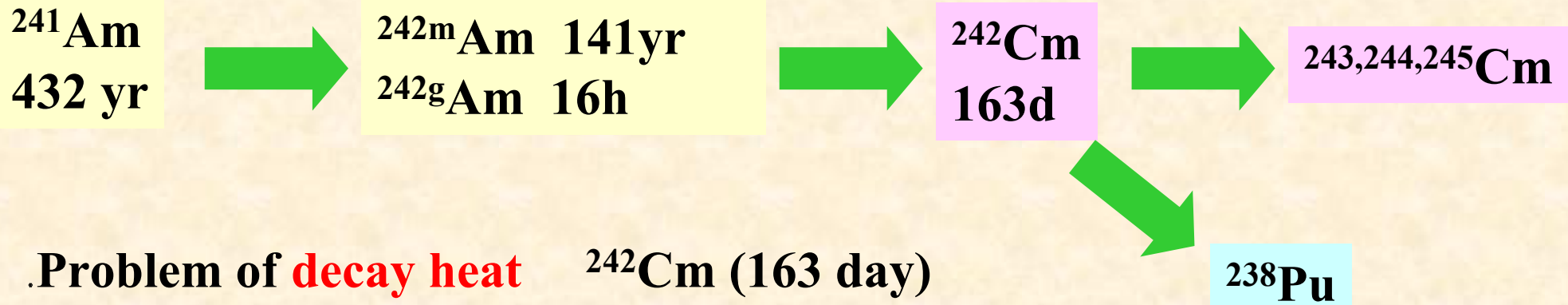
c) Calculation by the resonance parameters from BNL-325

## Supplement C:

# Thermal-Neutron Capture Cross-Section And Resonance Integral of $^{241}\text{Am}(n,\gamma)^{242\text{g}}\text{Am}$ Reaction



# Motivation



.Problem of **decay heat**  $^{242}\text{Cm}$  (163 day)

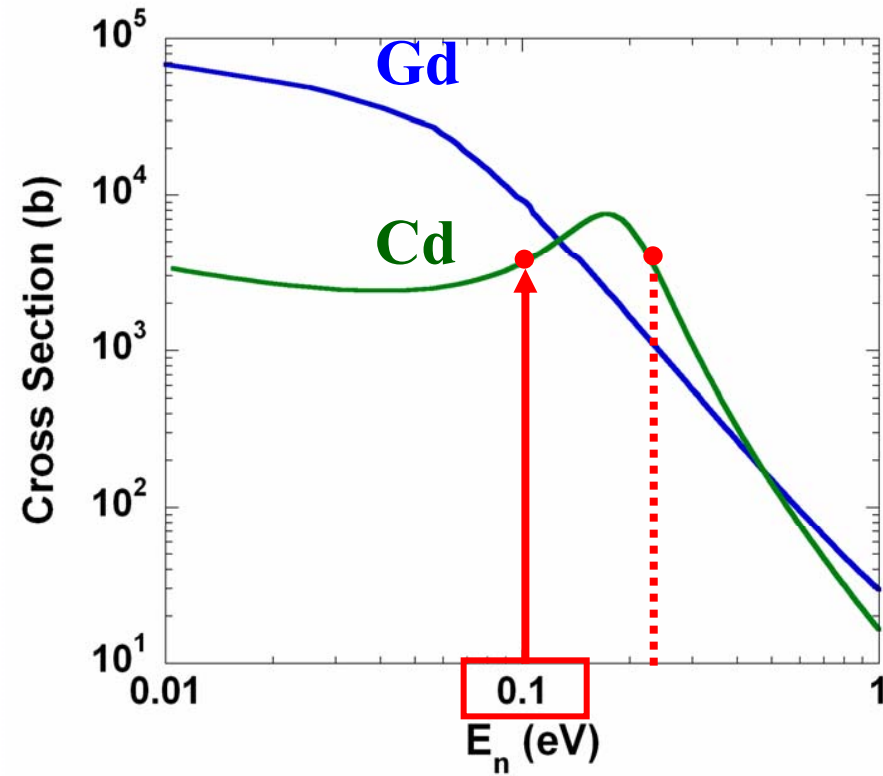
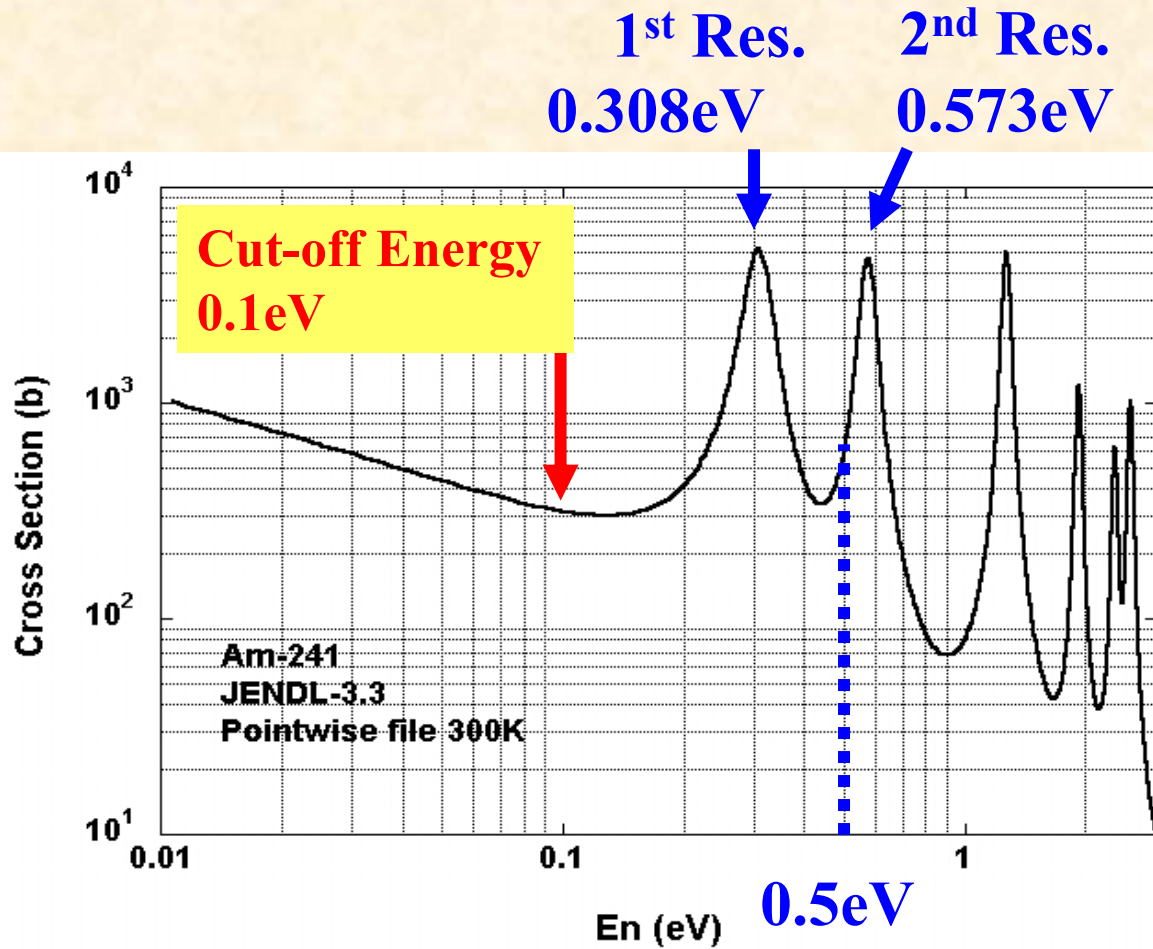
.Production of **long-lived Cm isotopes**

.Discrepancies among the reported data:  $\sigma_0$  about **20%**

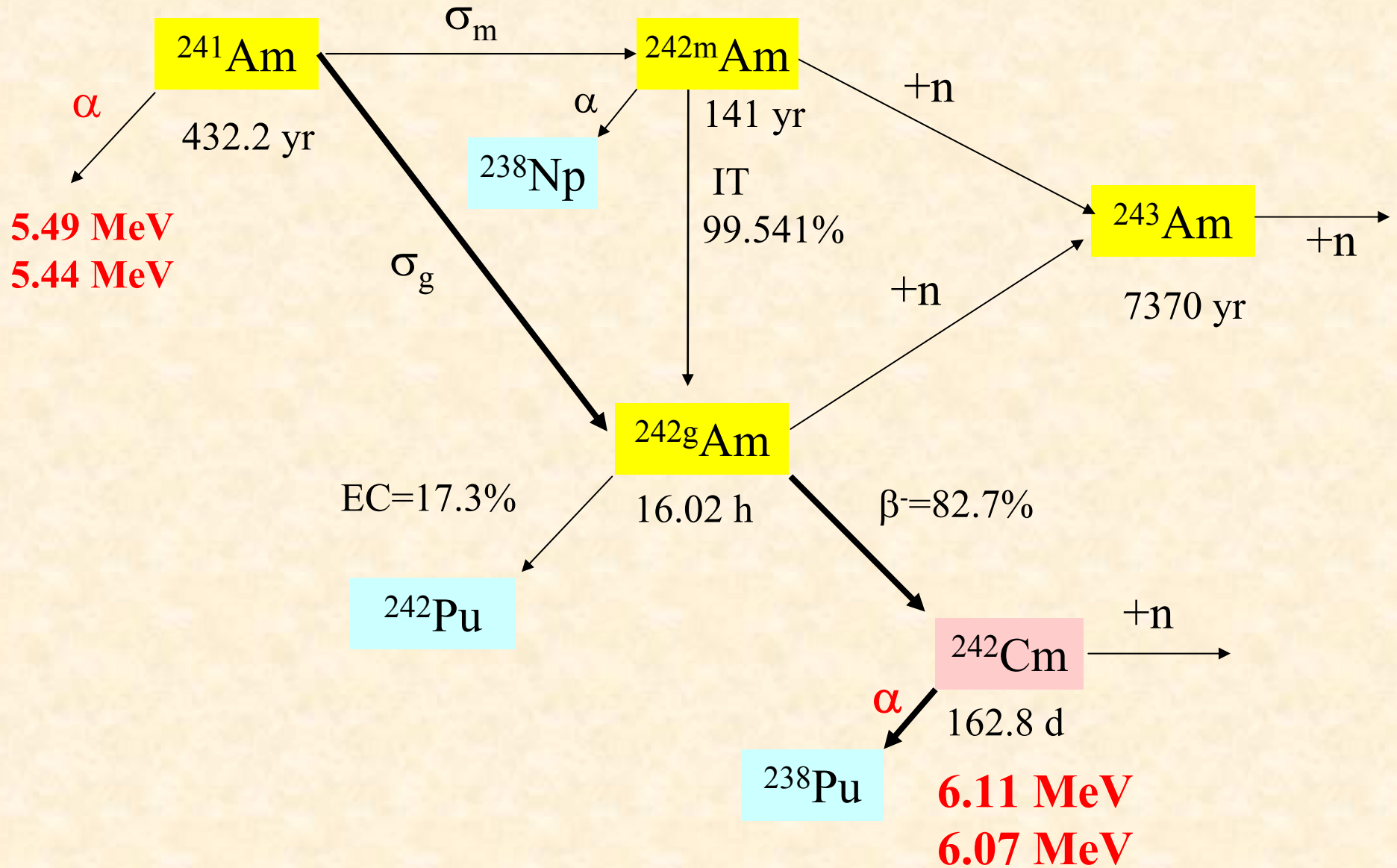
Authors	Year	$\sigma_{0,g}$ (b)	$I_{0,g}$ (b)
Maidana et al.	(2001)	<b>602±9</b>	1665±91
Fioni et al.	(2001)	<b>636±46</b>	-----
Shinohara et al.	(1997)	<b>768±58</b>	1694±146
Gavrulov et al.	(1977)	<b>780±50</b>	-----
Harbour et al.	(1973)	<b>748±20</b>	1330±117
Bak et al.	(1967)	<b>670±60</b>	2100
Deal et al.	(1964)	<b>770</b>	-----



# Cross Section Curve of $^{241}\text{Am}(n,\gamma)$ reaction



# Partial Decay Scheme



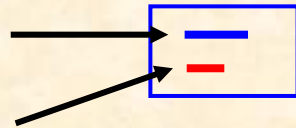
# Irradiation of samples

**Am-241**

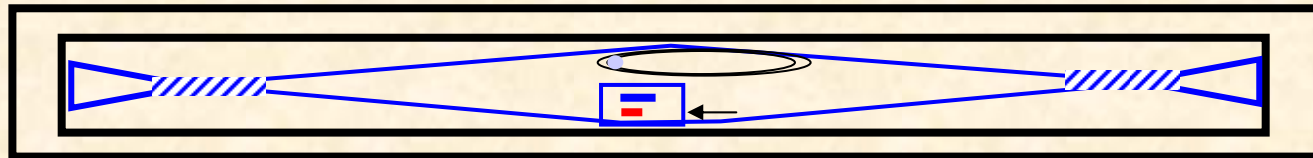
**100Bq**

Co/Al 1 mm

Au/Al 0.5 mm



High purity quartz tube  
8mmf, 50mm in length



**Purity  $^{241}\text{Am}$  : 99.93 %**

**$^{243}\text{Am}$  : 0.0647%**

**Other : 0.0002%**

**Am-241**

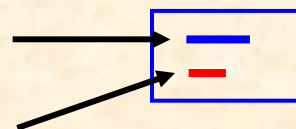
**500Bq**

Co/Al 1 mm

Au/Al 0.5 mm



20mm



**KUR @Long-Irradiation Plug  
Irradiation for 68Hours**

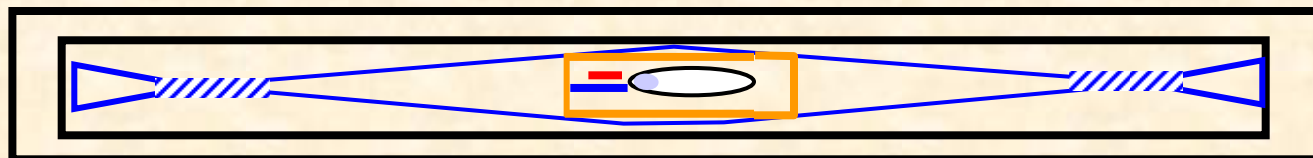
**Gd foil 25mm<sup>t</sup>**

**Cut-off Energy  
0.107eV**

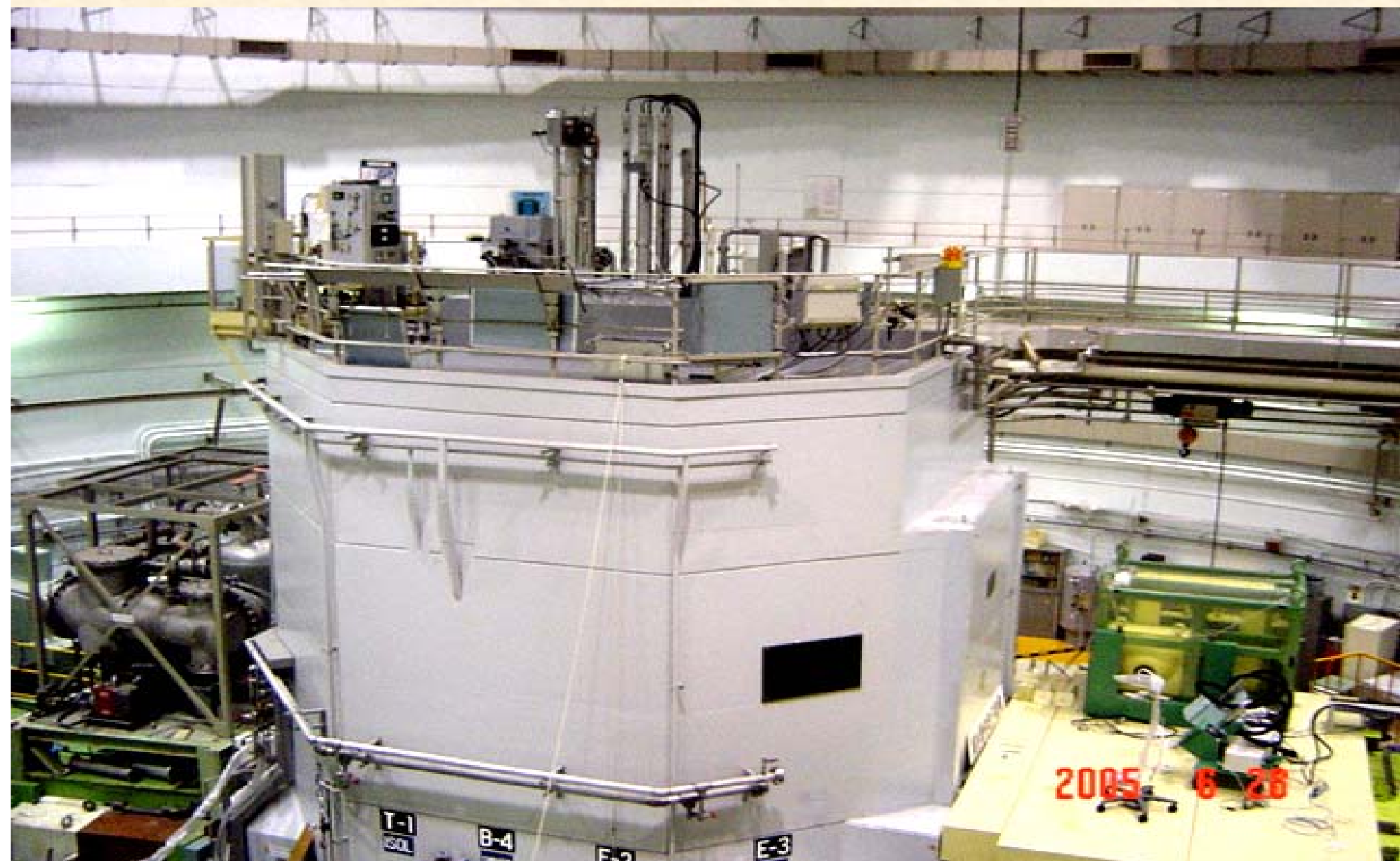


10mmf,30mm

**Gd covered capsule**

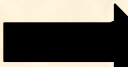


# Kyoto University Reactor: KUR @KURRI

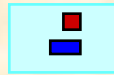


# Measurements of Samples

## 20 Days Cooling after Irradiation

$^{242}\text{gAm}$    $^{242}\text{Cm}$

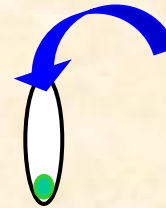
flux monitors



$\gamma$  ray measurement  
with a Ge detector  
flux information

Irradiated

$^{241}\text{Am}$  sample



3M  $\text{HNO}_3$   
Add 30ml solution

$^{241}\text{Am}$

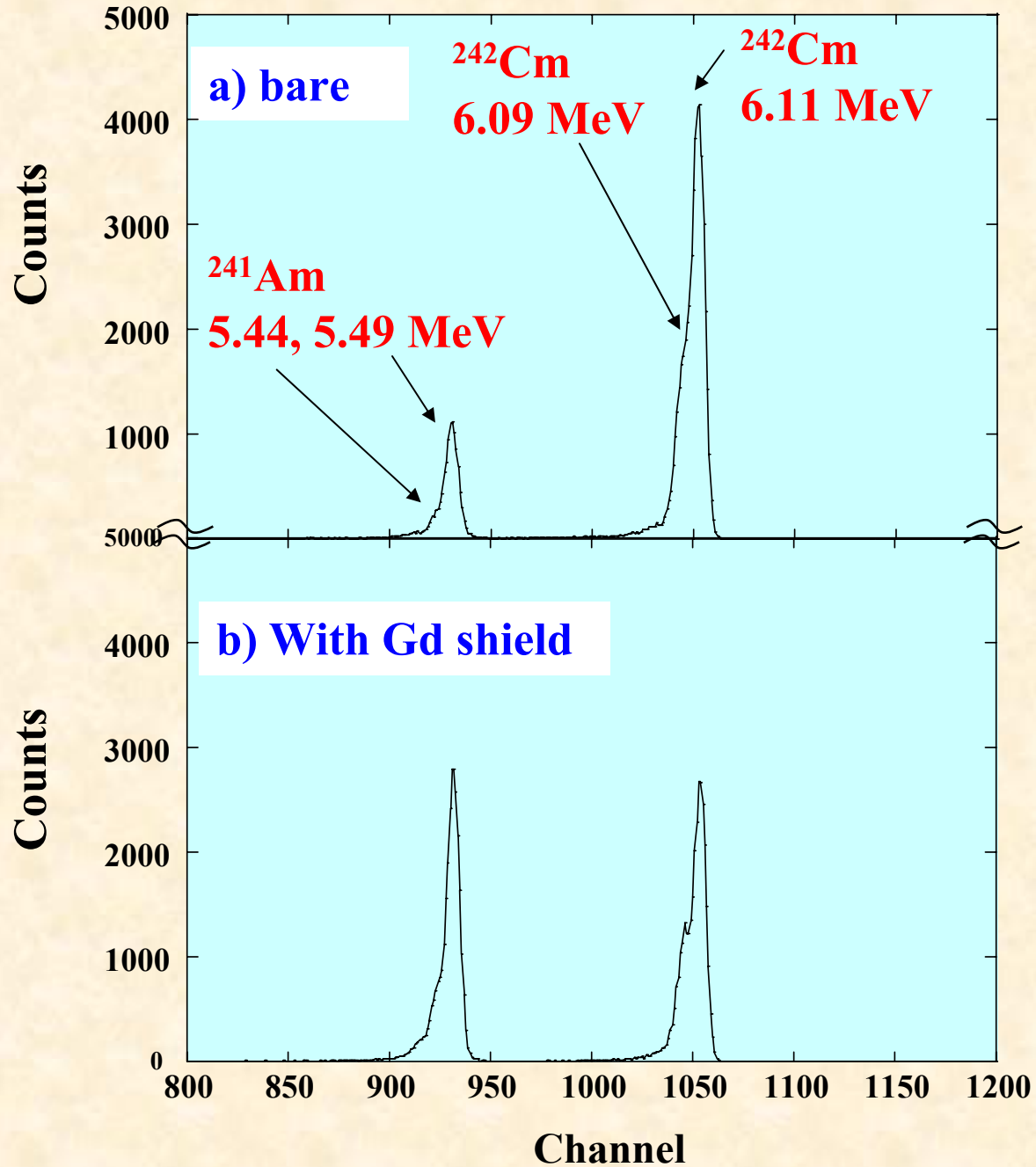
$^{242}\text{Cm}$



EG&G ORTEC  
SOLOIST

$\alpha$ -ray measurements

# $\alpha$ -ray spectra of irradiated $^{241}\text{Am}$ sample



Energy Resolution 45keV

$^{241}\text{Am}$  :  $11421 \pm 107$  Cnts.

$^{242}\text{Cm}$  :  $47259 \pm 217$  Cnts.

$^{241}\text{Am}$  :  $28510 \pm 169$  Cnts.

$^{242}\text{Cm}$  :  $30736 \pm 175$  Cnts.

# Analysis - the Westcott's convention-

$$\frac{R}{\sigma_0} = g G_{th} \phi_1 + \phi_2 \cdot s_0 G_{epi}$$

for irradiation without a Gd shield,

$$\frac{R'}{\sigma_0} = g G_{th} \phi'_1 + \phi'_2 \cdot s_0 G_{epi}$$

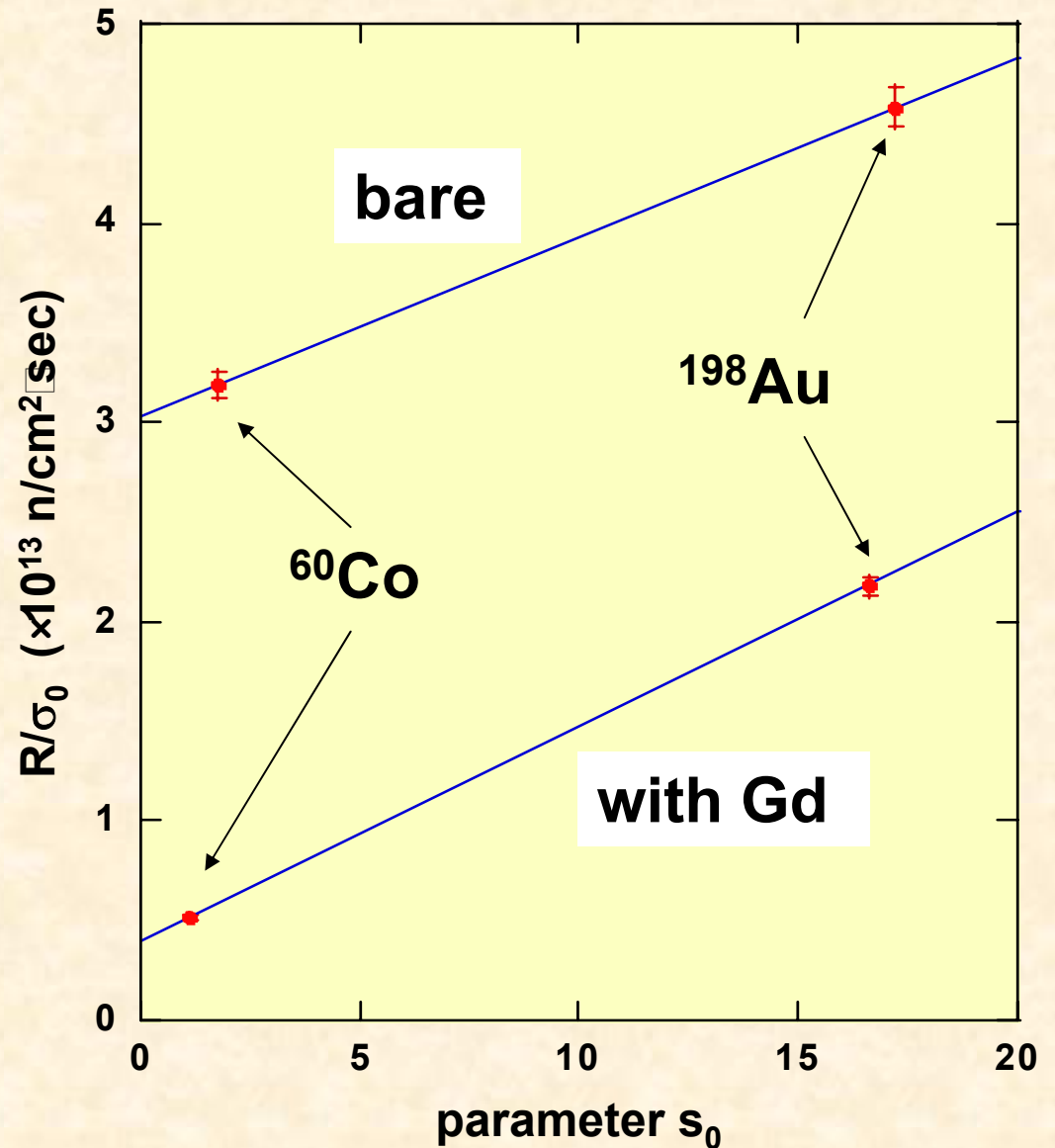
for irradiation with a Gd shield.  
where

$$s_0 = \sqrt{\frac{4}{\pi}} \cdot \frac{I'_0}{\sigma_0}$$

$I'_0$  is the resonance integral  
after subtracting the  $1/v$  component  
 $g : 1.051$  for  $^{241}\text{Am}$

Resonance Integral  $I_0$

$I_0 = I'_0 + 0.9725 \sigma_0$   
for **cut-off energy of 0.107 eV**



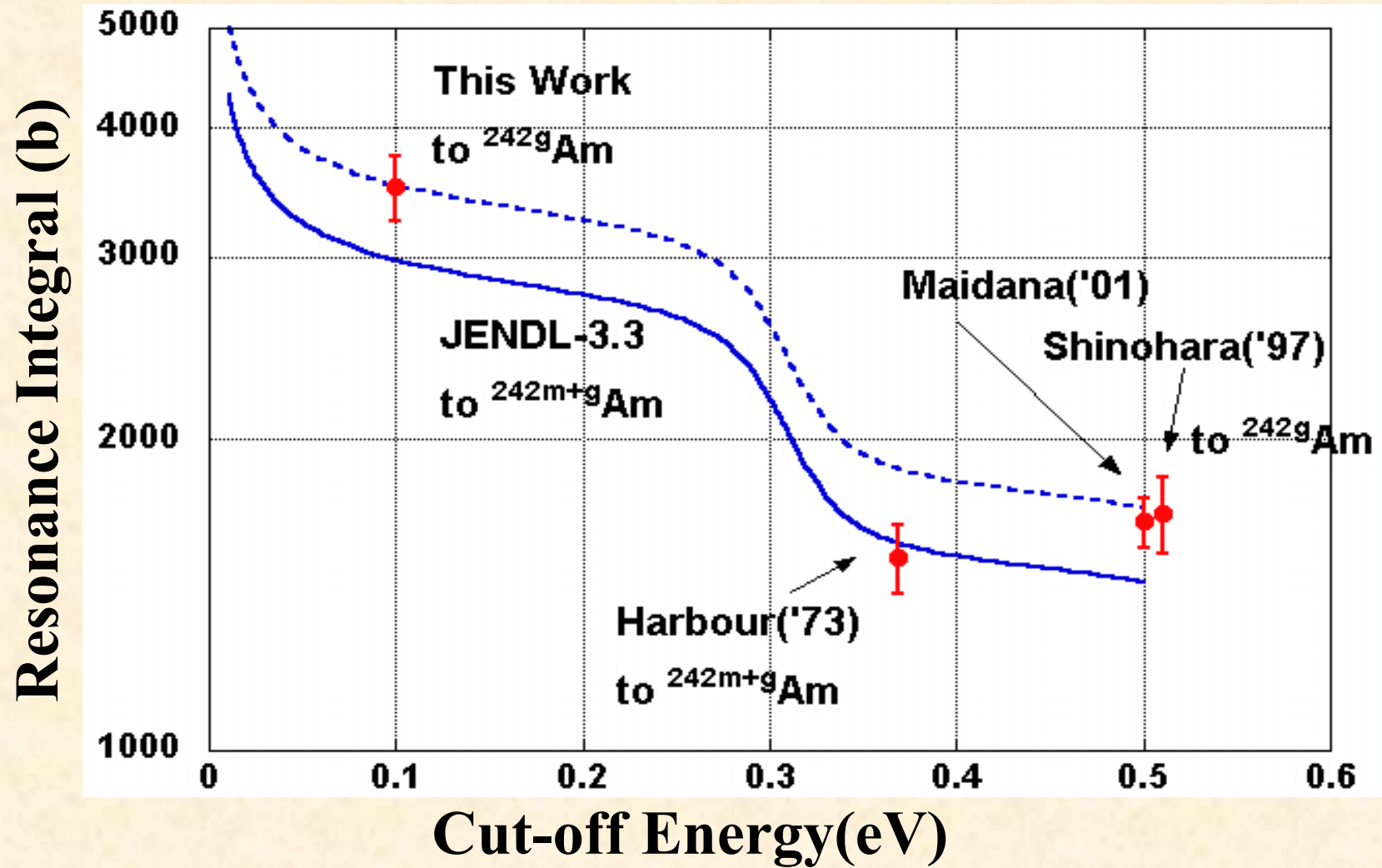
# Results of $\sigma_0$ and $I_0$ for the $^{241}\text{Am}(n,\gamma)^{242g}\text{Am}$ reaction

Authors & Year	$\sigma_{0g}$ (b)	$I_g$ (b)	Cut-off Energy(eV)
<b>This Work</b>	<b>628±17</b>	<b>3.5±0.3 k</b>	<b>0.107</b>
JENDL-3.3 (2002)	639.4*	1456 *	
Maidana et al. (2001)	602±9	1665±91	0.5
Fioni et al. (2001)	636±46	----	----
Shinohara et al. (1997)	768±58	1694±146	0.5
Gavrulov et al. (1977)	780±50	---	---
Harbour et al. (1973)	748±20	1330±117	0.369
Bak et al. (1967)	670±60	2100	
Deal et al. (1964)	770	----	----

$^{241}\text{Am}$  to  $^{242m+g}\text{Am}$



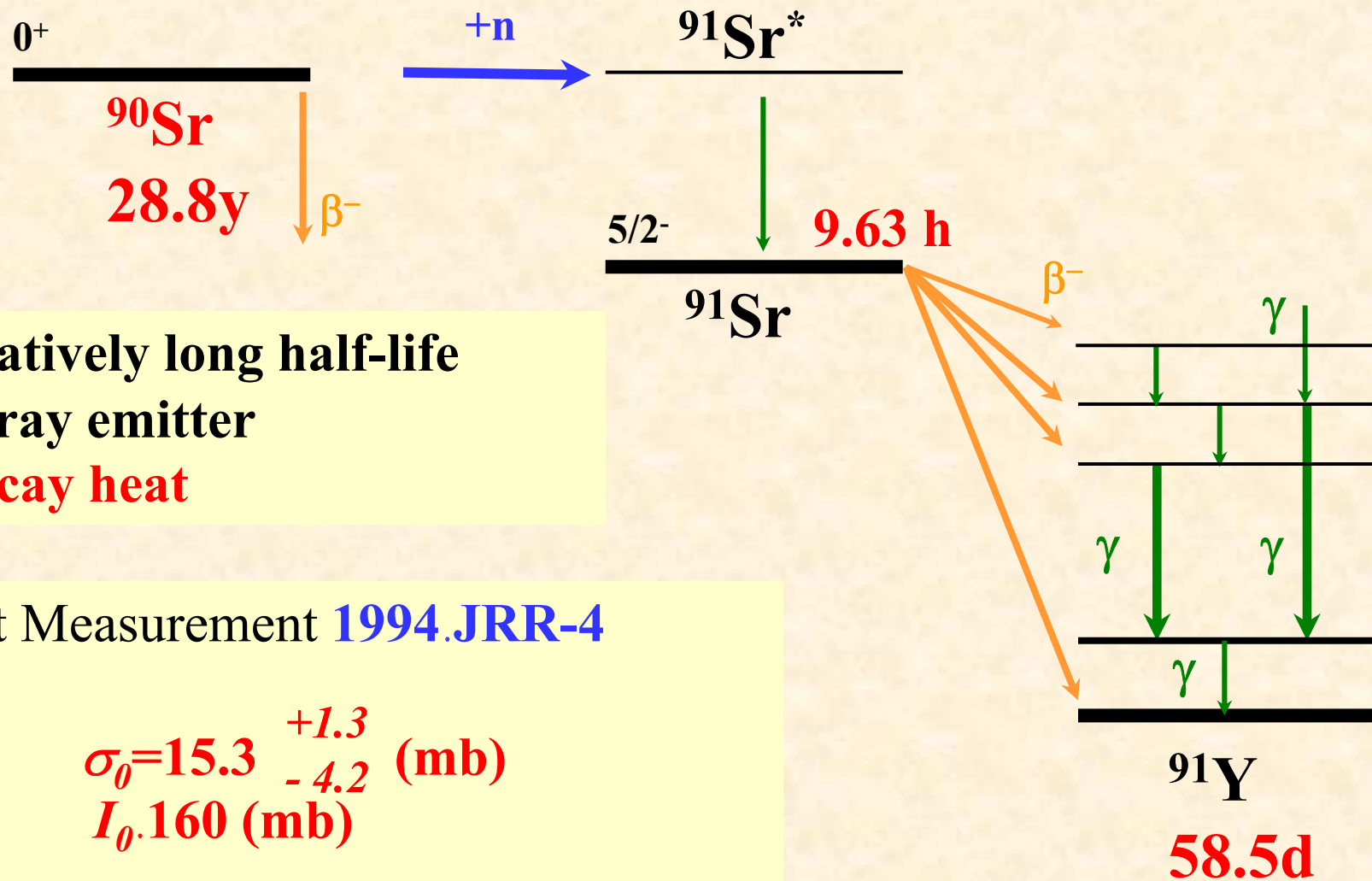
# Result of $I_0$ for the $^{241}\text{Am}(n,\gamma)^{242g}\text{Am}$ reaction



## Supplement D:

# Thermal-Neutron Capture Cross-Section and Resonance Integral of $^{90}\text{Sr}(n,\gamma)^{91}\text{Sr}$ Reaction

# $^{90}\text{Sr}(n,\gamma)^{91}\text{Sr}$ reaction Cross-Section



- .relatively long half-life
- .  $\beta$ -ray emitter
- . Decay heat

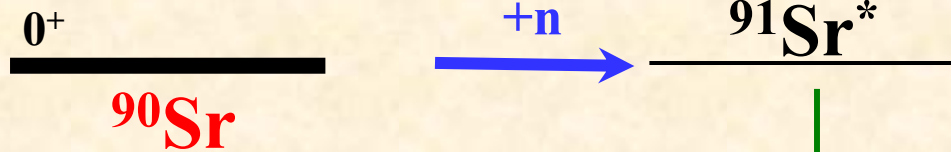
Past Measurement **1994.JRR-4**

$$\sigma_0 = 15.3^{+1.3}_{-4.2} \text{ (mb)}$$

$$I_0 = 0.160 \text{ (mb)}$$

.upper limit just for  $I_0$

# Experiment Procedure



.Irradiation Sample  
 $^{90}\text{Sr}$  Stand.Sol.  $^{85}\text{Sr}$  sol. (as tracer)

.Au, Co neutron flux monitors

.Irradiation .KUR Hyd.  
 Irrad. With Cd shield capsule

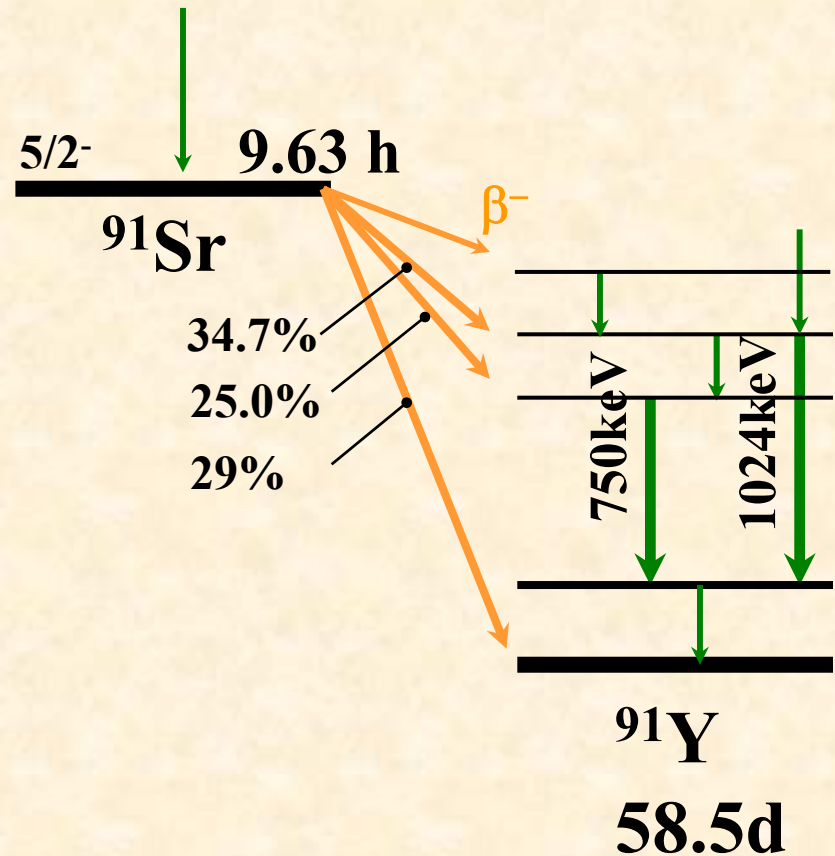
.Chemical Treatment  
 huge  $\gamma$ -ray B.G. due to  $^{24}\text{Na}(15\text{h})$   
 1369keV.2754keV

.Singles Measurement with Ge detector

Yield of  $^{91}\text{Sr}$  ➔ Reaction rate

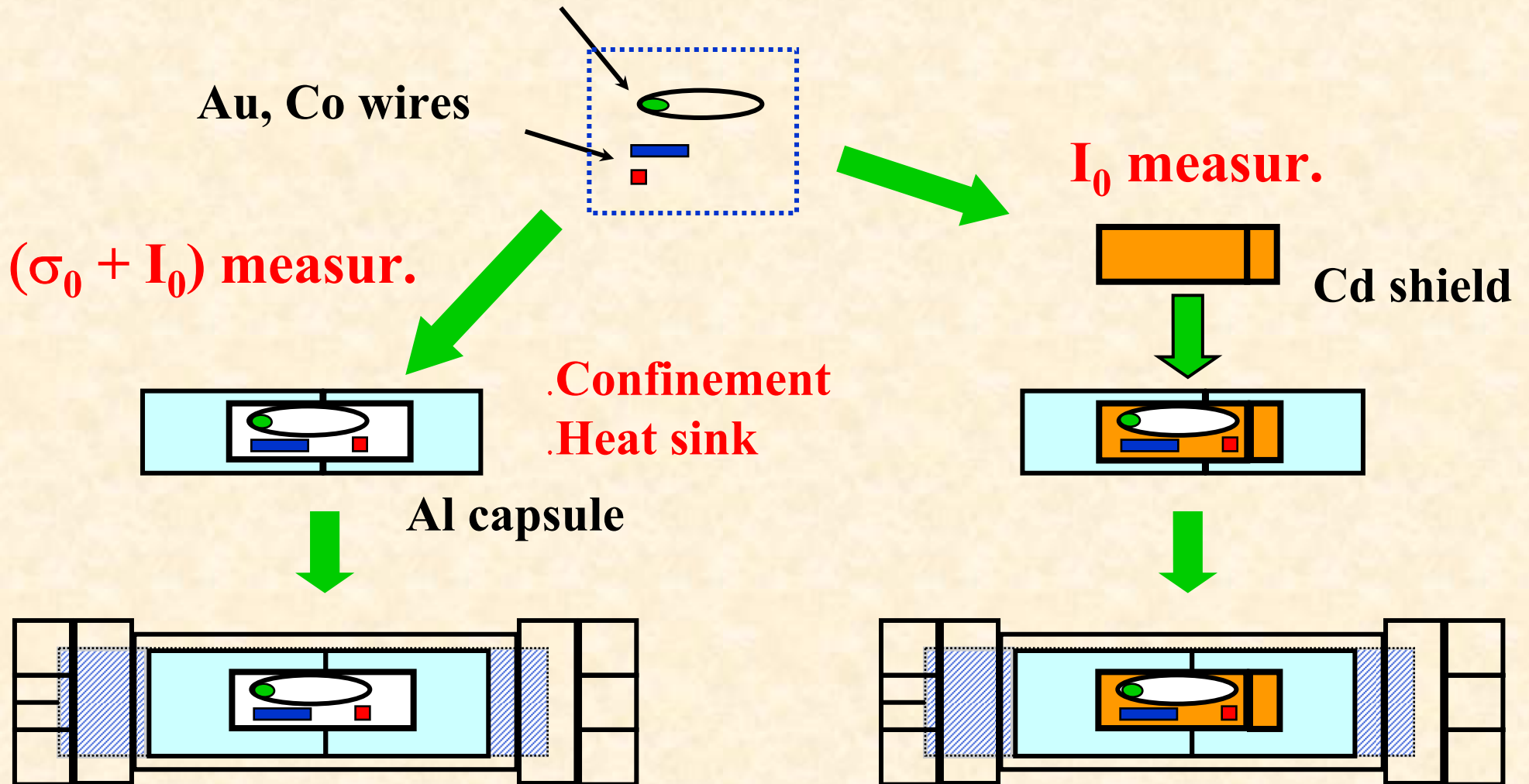
Induce Activities of Monitors ➔ Neutron Fluxes

.Derivation of  $\sigma_0$  and  $I_0$



# Irradiation Targets

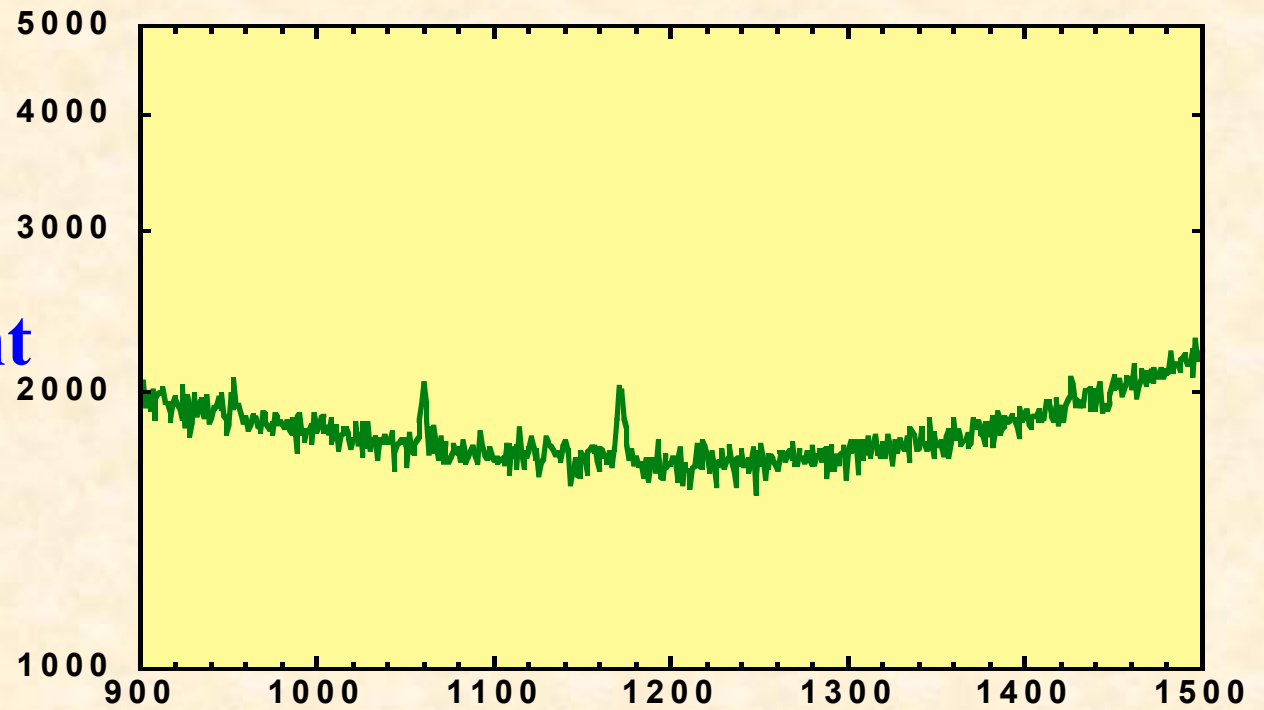
$^{90}\text{Sr}$  sol.  $^{85}\text{Sr}$  sol. (.as Tracer.



KUR .Hyd  $1 \times 10^{14}$  n/cm<sup>2</sup>sec

# $\gamma$ -ray Spectra

Before  
Chemical Treatment

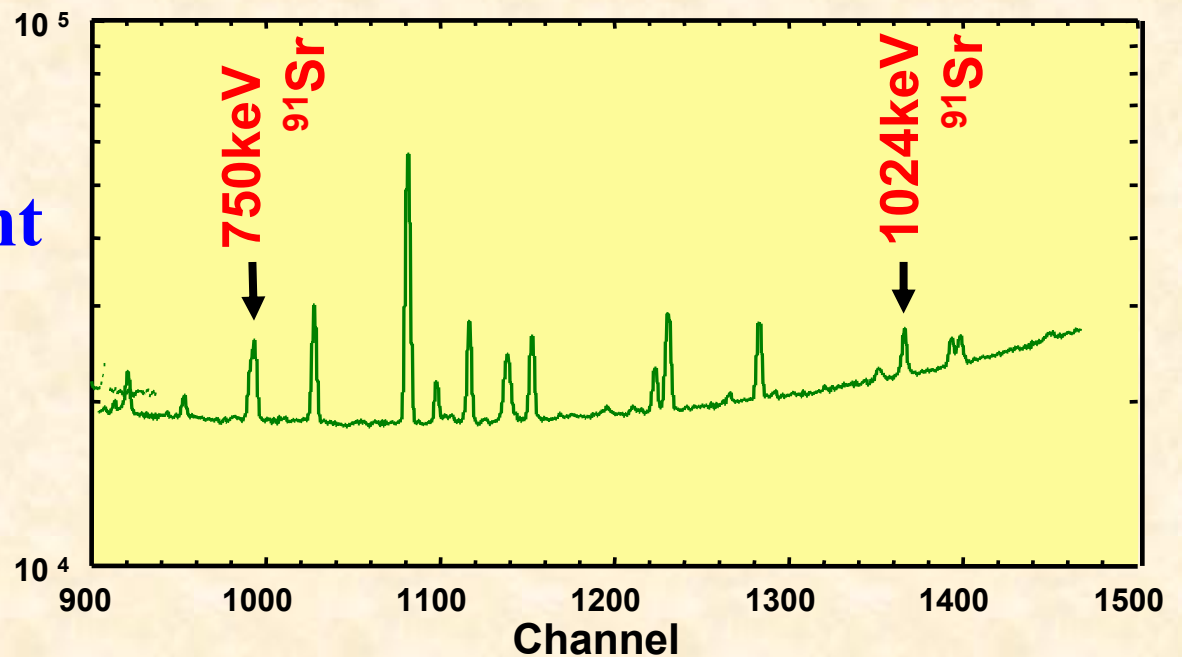


Technique of  
solvent extract

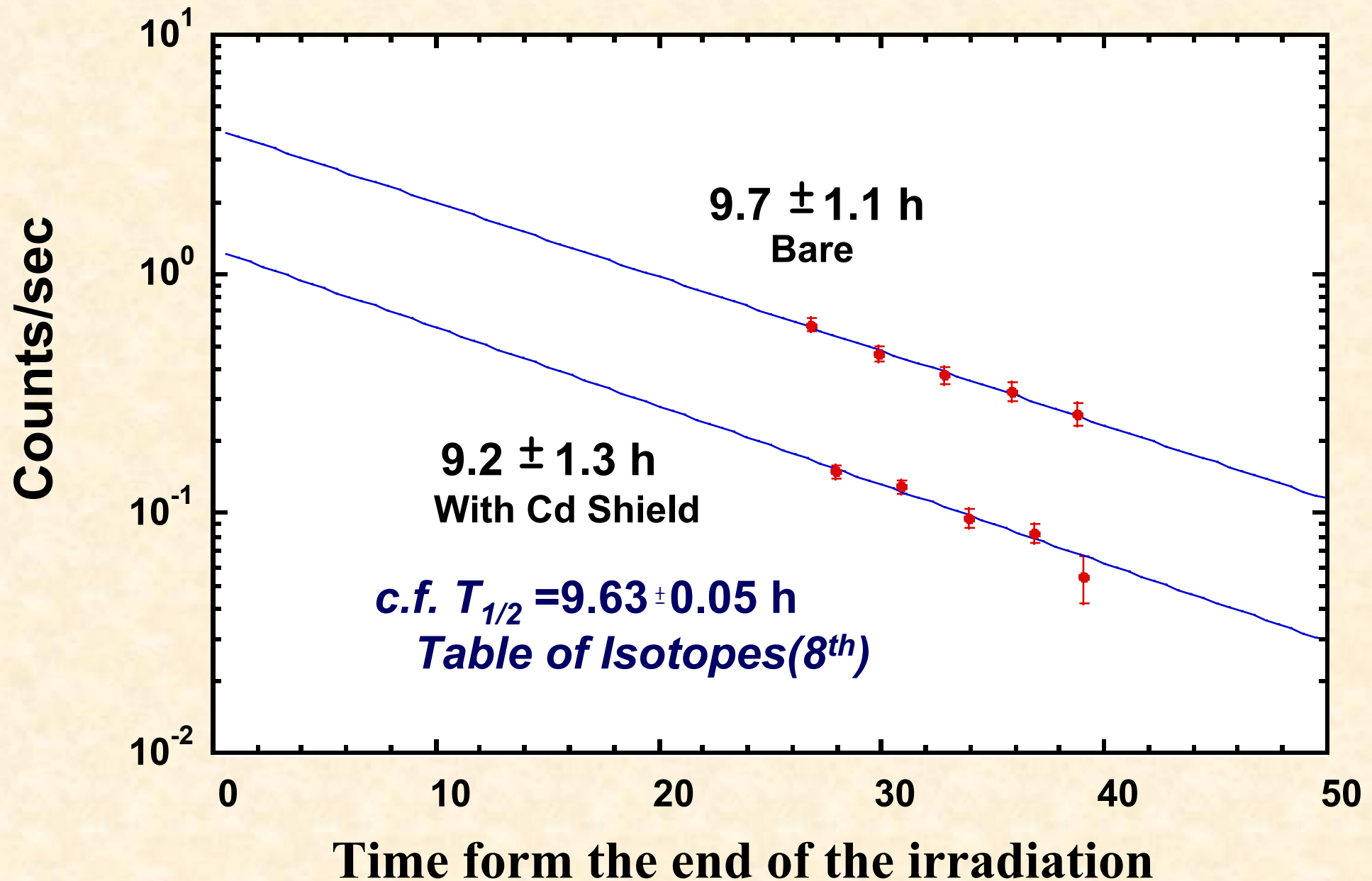


Reduction of  $^{24}\text{Na}$ !

After treatment



# Decay curves of 1024-keV $\gamma$ ray



# Analysis

## Westcott's convention

$$\frac{R}{\sigma_0} = g G_{th} \phi_1 + \phi_2 \cdot s_0 G_{epi}$$

for irradiation without a Gd shield,

$$\frac{R'}{\sigma_0} = g G_{th} \phi'_1 + \phi'_2 \cdot s_0 G_{epi}$$

for irradiation with a Gd shield.

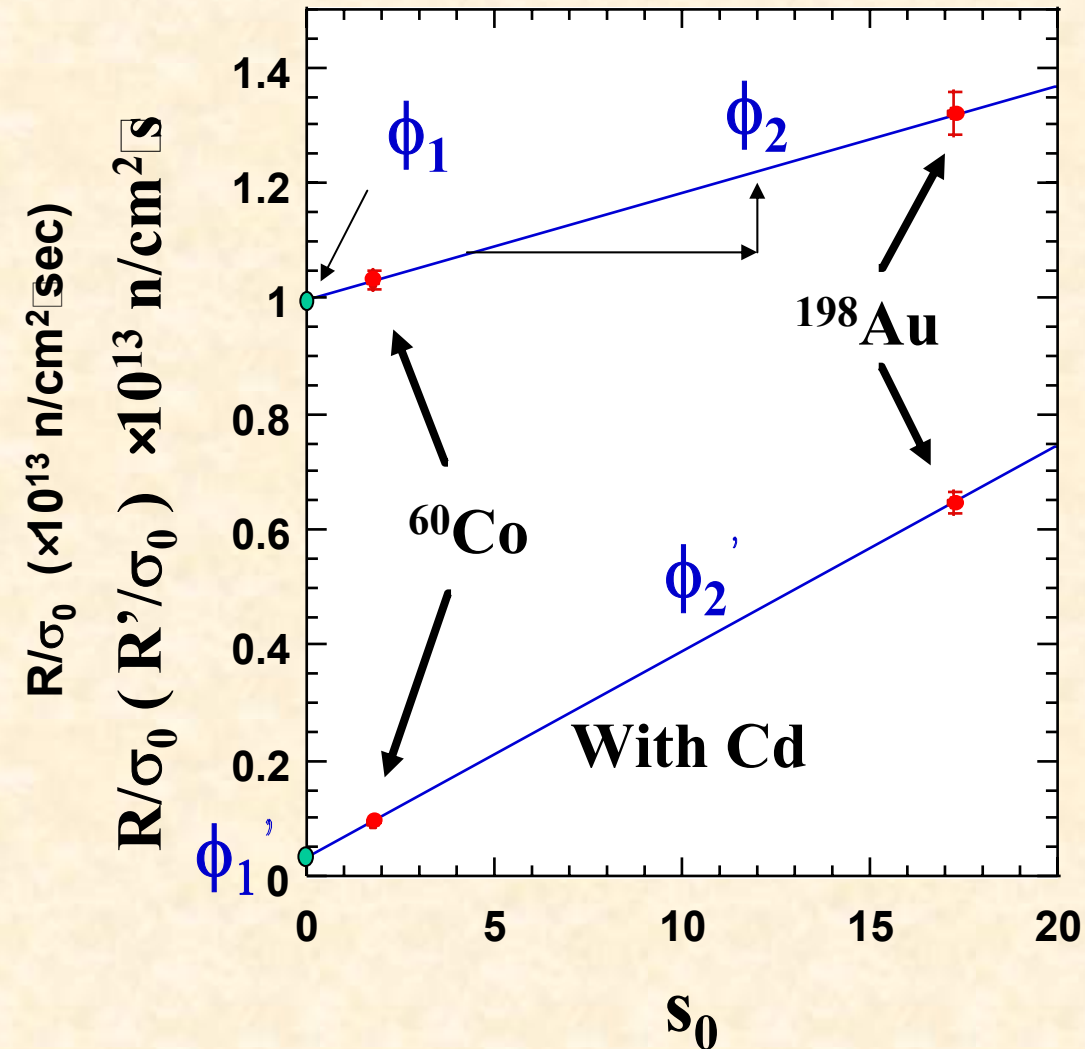
where

$$s_0 = \sqrt{\frac{4}{\pi} \cdot \frac{I'_0}{\sigma_0}}$$

$I'_0$  is the resonance integral  
after subtracting the  $1/v$  component  
 $g$  : deviation from  $1/v$  low

Resonance Integral  $I_0$

$I_0 = I'_0 + 0.45 \sigma_0$   
for cut-off energy of 0.5 eV



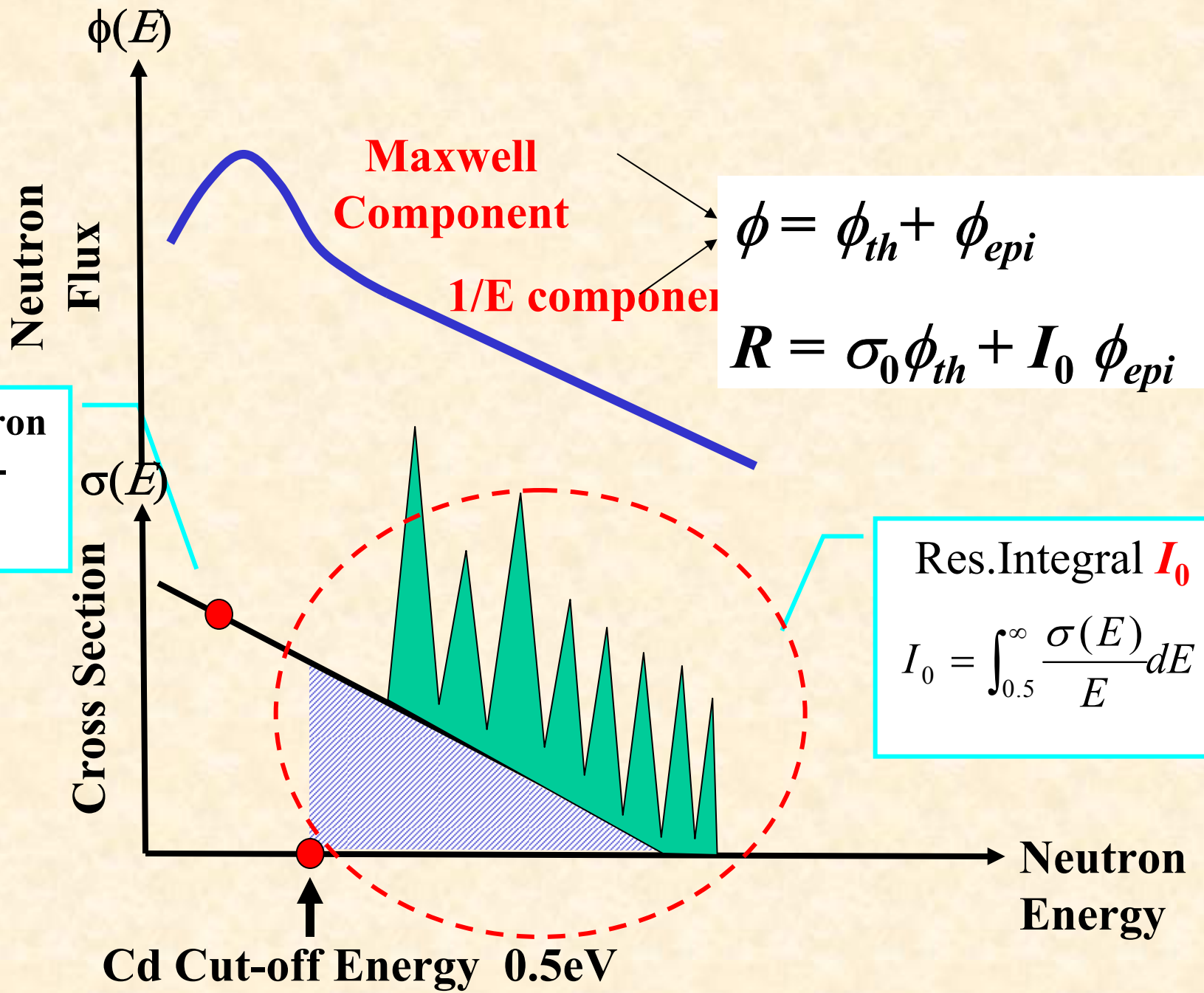


# Results

Author	Thermal Cross Section, $\sigma_0$ (mb)	Resonance Integral, $I_0$ (mb)
Zeisel [66]	$800 \pm 500$	-----
McVey <i>et al.</i> [83]	$14.0 \pm 2.4$	-----
Lone <i>et al.</i> [93]	$9.7 \pm 0.7$	-----
Harada <i>et al.</i> [94]	$15.3^{+1.3}_{-4.2}$	.160
<b>Present Work</b>	<b><math>10.1 \pm 1.3</math></b>	<b><math>104 \pm 16</math></b>

## **Supplement E:**

# **Brief Outline of Analysis In the Basis of Westcott's Convention**



Thermal-Neutron  
Capture Cross-  
Section  $\sigma_0$

Res. Integral  $I_0$

$$I_0 = \int_{0.5}^{\infty} \frac{\sigma(E)}{E} dE$$

$$R = \sigma_0 \cdot \phi_{th} + RI \cdot \phi_{epi}$$

$$R' = \quad \quad \quad RI \cdot \phi_{epi}$$

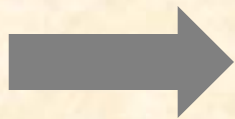
## Cadmium Ratio Method

Cadmium ratio  $R_{Cd}$

$$R_{Cd} = \frac{\sigma_0 \cdot \phi_{th} + RI \cdot \phi_{epi}}{RI \cdot \phi_{epi}} = \frac{\phi_{th}}{\phi_{epi}} \cdot \frac{\sigma_0}{RI} + 1$$


$$R_{Cd,x} = \frac{\phi_{th}}{\phi_{epi}} \cdot \frac{\sigma_{0,x}}{RI_x} + 1$$

$$R_{Cd,Co} = \frac{\phi_{th}}{\phi_{epi}} \cdot \frac{\sigma_{0,Co}}{RI_{Co}} + 1$$



$$RI_x = RI_{Co} \frac{(R_{Cd,Co} - 1) \cdot \sigma_{0,x}}{(R_{Cd,x} - 1) \cdot \sigma_{0,Co}}$$

# Modified Cadmium Ratio Method


$$R = n \nu_0 \hat{\sigma} = n \nu_0 \sigma_0 (g G_{th} + r (T / T_0)^{1/2} s_0 G_{epi})$$

*Westcott's  
Convention*



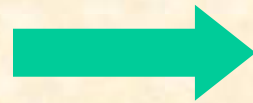
Siple Neutron-Flux notations

Bare:  $R / \sigma_0 = \phi_1 \cdot G_{th} + \phi_2 \cdot s_0 \cdot G_{epi}$

with Cd:  $R' / \sigma_0 = \phi_1' \cdot G_{th} + \phi_2' \cdot s_0 \cdot G_{epi}$

$$\text{Nuclide: } x \left\{ \begin{array}{l} R_x / \sigma_{0,x} = \phi_1 \cdot G_{th,x} + \phi_2 \cdot s_{0,x} \cdot G_{epi,x} \\ R'_x / \sigma_{0,x} = \phi'_1 \cdot G_{th,x} + \phi'_2 \cdot s_{0,x} \cdot G_{epi,x} \end{array} \right.$$

Neutron flux components  $\phi_{1,2}, \phi'_{1,2}$



**Multi-flux Monitor Method**

$$R_x / \sigma_{0,Au} = \phi_1 \cdot G_{th,Au} + \phi_2 \cdot s_{0,Au} \cdot G_{epi,Au}$$

$$R'_{Au} / \sigma_{0,Au} = \phi'_1 \cdot G_{th,Au} + \phi'_2 \cdot s_{0,Au} \cdot G_{epi,Au}$$

$$R_{Co} / \sigma_{0,Co} = \phi_1 \cdot G_{th,Co} + \phi_2 \cdot s_{0,Co} \cdot G_{epi,Co}$$

$$R'_{Co} / \sigma_{0,Co} = \phi'_1 \cdot G_{th,Co} + \phi'_2 \cdot s_{0,Co} \cdot G_{epi,Co}$$

# Analysis on the basis of the Westcott's convention

$$\frac{R}{\sigma_0} = g G_{th} \phi_1 + \phi_2 \cdot s_0 G_{epi}$$

for irradiation without a Gd shield,

$$\frac{R'}{\sigma_0} = g G_{th} \phi'_1 + \phi'_2 \cdot s_0 G_{epi}$$

for irradiation with a Gd shield.

where

$$s_0 = \sqrt{\frac{4}{\pi} \cdot \frac{I'_0}{\sigma_0}}$$

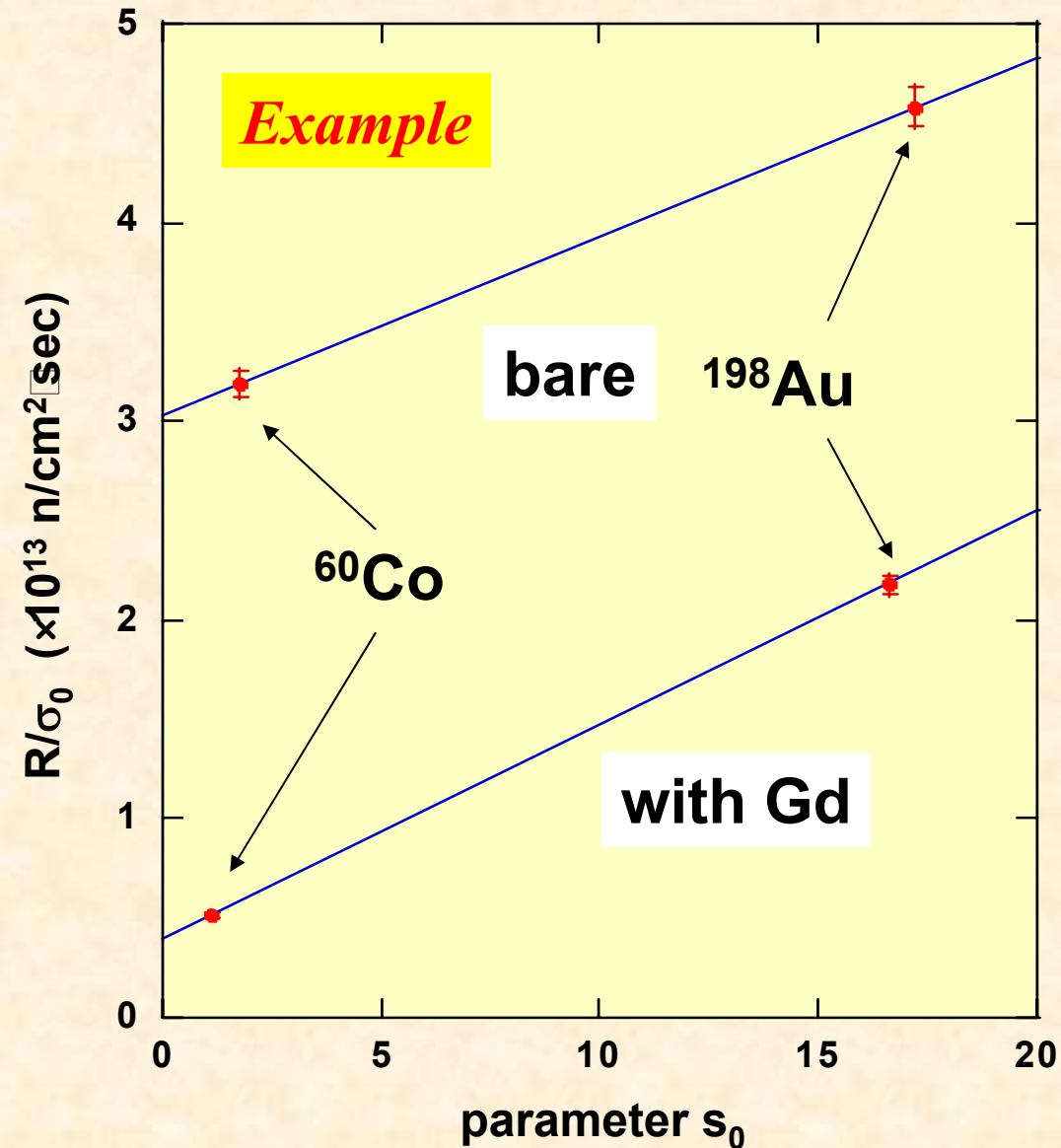
$I'_0$  is the resonance integral  
after subtracting the  $1/v$  component

$g$  : deviation from  $1/v$  low

Resonance Integral  $I_0$

$$I_0 = I'_0 + 0.45 \sigma_0$$

for cut-off energy of 0.5 eV



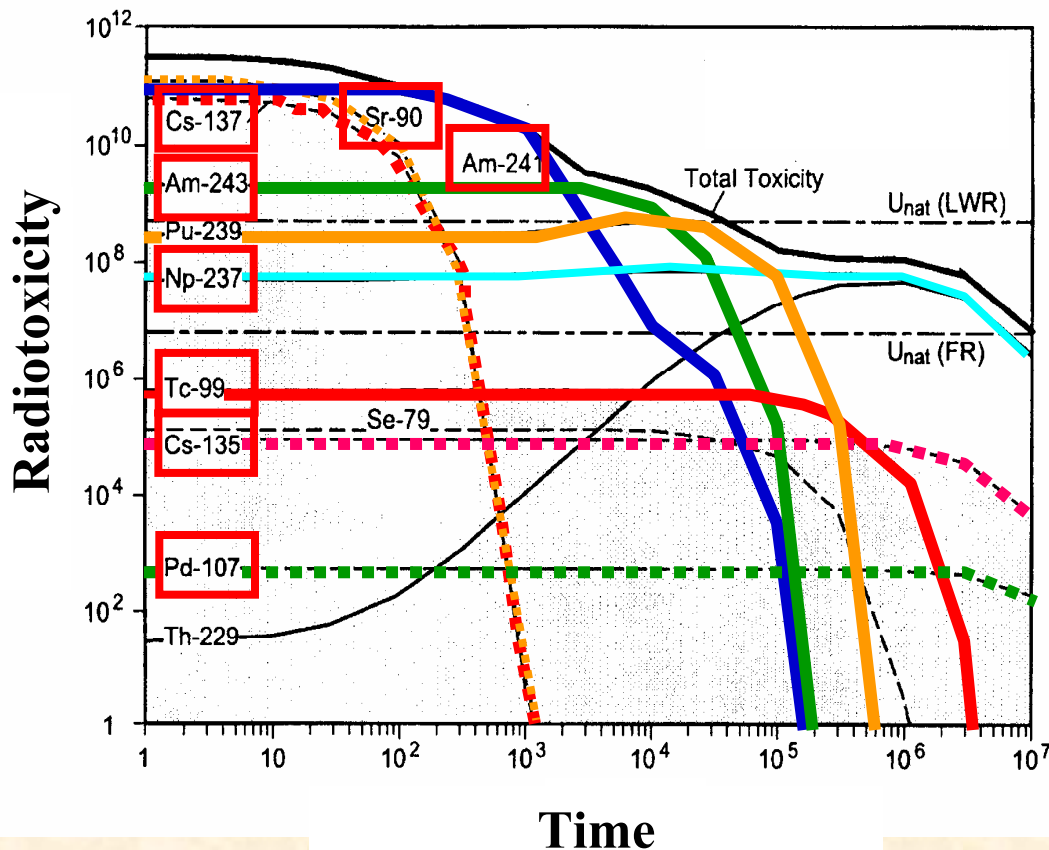
**Supplement F:**

**Others**



# Motivation

High Level Nuclear Wastes  
Log-Lived Fission Products (FP's)  
Minor Actinides (MA's)



Reduction of  
Disposal Managements  
Load to Environment

Nuclear Transmutation

Cross-Section Data  
to Evaluate Reaction Rates  
*WRENDA: World Request  
List for Nuclear Data*

Important FP's.Me's  
Selection of Nuclides  
*High Priority Request List*

## Transmutability of long-lived fission products

Fission product	Decay $T_{1/2}$ (y)	Trans $T_{1/2}$ (y)	Isotopic separation	Transmutable (yes/no)
$^{129}\text{I}$	$1.6 \times 10^7$	51	no	yes
$^{135}\text{Cs}$	$2.3 \times 10^6$	170	yes	no
$^{99}\text{Tc}$	$2.1 \times 10^5$	51	no	yes
$^{126}\text{Sn}$	$1.0 \times 10^5$	$4.4 \times 10^3$	yes	no
$^{79}\text{Se}$	$6.5 \times 10^4$	$2.2 \cdot 10^3$	yes	no

Reference. NEA2002  
Accelerator-driven Systems (ADS)  
and Fast Reactors (FR) in  
Advanced Nuclear Fuel Cycles

### Example of LSD for the reaction rate of $^{99}\text{Tc}$

