

# Measurement and EXFOR Compilation of Neutron-Induced Activation Cross Sections in the Energy Range up to 20 MeV

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# Outline

- Activation data: needs and applications
- Activation method
- Main accelerator-based neutron source reactions
- Sample composition
- Neutron spectrum characterisation
- Coincidence summing effects
- Detector efficiency determination
- Nuclide identification by  $E_g$  and  $T_{1/2}$
- Uncertainty analysis
- Constructing a covariance matrix from EXFOR uncertainties
- Isomeric ratio measurements
- ${}^9\text{Be}(d,n)$  thick target neutron spectrum
- Spectrum average cross section

### **Institute for Nuclear Research and Nuclear Energy, Sofia, Bulgaria**

- Dates May 1993 - present
- Occupation or position held Physicist 1993-1996; Research fellow 1996-2003; Assistant Professor 2003 - 2010, Associate Professor 2010-.
- Main activities and responsibilities
- Nuclear research applying different experimental techniques, including:
    - (n,2n) cross section measurements based on neutron detection time distribution.
    - Reaction cross section measurements by activation method

### **International Atomic Energy Agency, Nuclear Data Section**

- Dates October 2010 – October 2017
- Occupation or position held Nuclear Physicist
- Main activities and responsibilities
- Survey of the scientific literature and compilation of the publications reporting experimental nuclear reaction data for the EXFOR database.
  - Organizing consultants' meetings and workshops.

### **European Commission, Institute for Reference Material and Measurements, Geel, BELGIUM**

- Dates March 2005 – February 2009
- Occupation or position held National Seconded Expert
- Main activities and responsibilities
- Neutron-induced activation cross section measurements on isotopes of Zr, Ta, W, Hf, Cr etc.
  - Development of a Monte-Carlo model of the HPGe detector.
  - $^{241}\text{Am}(n,2n)$  reaction study as part of broad collaboration between IRMM, ITU, and CEA (France).

### **Physikalisch-Technische Bundesanstalt (PTB)**

Dates April 2010 – September 2010

Occupation or position held Visiting Scientist

- Main activities and responsibilities
- Experimental study of neutron-induced reaction cross sections on Mo isotopes.
  - Re-establish the procedure for activation measurements after discontinuation of several years.

### **Institute of Experimental Physics, Debrecen, HUNGARY**

Dates January 1995 – January 1996

- Main activities and responsibilities
- Study of reaction cross sections induced by quasi-monoenergetic neutrons and neutrons with spectral energy distribution from  ${}^9\text{Be}(d,n)$  reaction by activation methods.
  - ${}^2\text{H}(d,n){}^3\text{He}$  and  ${}^9\text{Be}(d,n){}^{10}\text{B}$  neutron flux determination using activation method and PHRS
  - $(n,p)$  and  $(n,\alpha)$  reaction cross section systematics at 14 MeV.

### **European Commission, Institute for Reference Material and Measurements, Geel, BELGIUM**

Dates February 2001 – January 2003

- Principal subjects
- Neutron-induced activation cross sections measurements on different isotopes of Co, Ni, Cu, Zr, and Pb from the threshold up to 20 MeV.
- ${}^{58}\text{Ni}(n,p){}^{58\text{m,g}}\text{Co}$ , the  ${}^{60}\text{Ni}(n,p){}^{60\text{m,g}}\text{Co}$  and  ${}^{59}\text{Co}(n,2n){}^{58\text{m,g}}\text{Co}$  isomeric ratio measurements.

# Activation data: needs and applications

- Activation of materials : isotopes, activity
- Dose rates : handling limits, storage
- Heating : cooling , storage conditions
- Charge particle production : gas production, radiation damage
- Scattering cross sections  $(n, xn)$   $x=1,2 \dots$  : neutron transport
- Data for nuclear models development

# Activation data in EXFOR

## METHOD = ACTIV

Request #201

Access-Level=2 /pdf/

**Attention: Huge data request**

Found: Entries:3425; Subentries:30810; Datasets:28189 (Limit exceeded by factor of 5.7)

Go back, check and narrow your request

BACK

Continue your request with limited output:  
produce list of Entries

LIST

### EXFOR General Statistics

Information updated: 18-Oct-2016, 11:13:14

Database as of: 2016-10-18

Number of ENTRY	21424	experimental works
Number of SUBENT	149753	data tables (can contain data of more than one reaction)
Number of Datasets	166549	data tables of reactions
Number of Datapoints	13981410	total number of data points

Percent: [Counts]/[Number of ENTRY], i.e. = [Counts]/21424

Note.  $\Sigma$ [Percent] of a table below can be > 100% because one experimental work can contain many data tables with data of many types

### EXFOR Quantity

#	Code	Quantity	Counts	Percent
1	CS	Cross section data	10942	51
2	DAP	Partial differential data with respect to angle	4175	19.4
3	DA	Differential data with respect to angle	4152	19.3
4	RP	Resonance parameters	1924	8.98

ENTRY : 16%  
SUBENT : 21 %  
Datasets : 17 %

# Activation method

$$\sigma_x = \sigma_{st} \frac{A_x [I\epsilon Fn]_{st}}{A_{st} [I\epsilon Fn]_x} \frac{C_{flux,x}}{C_{flux,st}} \frac{C_{low,x}}{C_{low,st}} \frac{C_{coins,x}}{C_{coins,st}}$$

$$F = \frac{\lambda}{\exp(-\lambda t_c)(1 - \exp(-\lambda t_e))(1 - \exp(-\lambda t_m))}$$

*A - activity*

*I - gamma-ray emission probability*

*ε - detector efficiency*

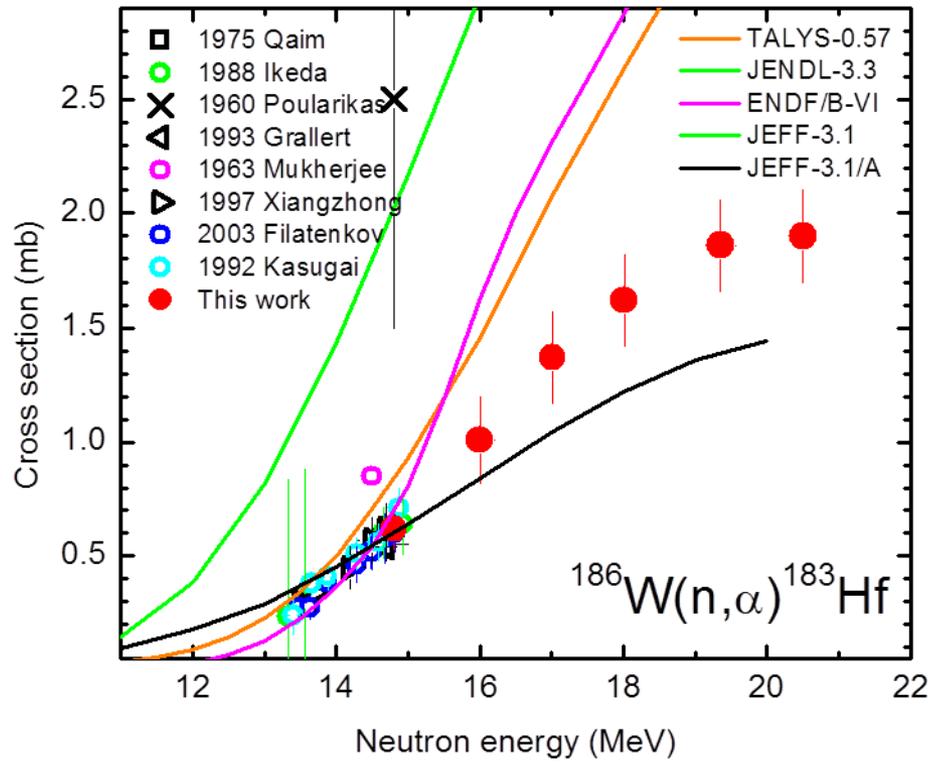
*n - number of atoms in the target*

*C - correction factors*

*σ<sub>st</sub> - <sup>27</sup>Al(n, α)<sup>24</sup>Na standard reaction cross*

# Employed mono- and quasymonoenergetic neutron source reactions

- Neutron generator (D-D: 2-3 MeV, D-T: 13.5-14.8 MeV): high intensity; low background; neutron emission in  $4\pi$ ; narrow energy range.
- Cyclotron (D-D: 4-13 MeV): neutron emission predominantly in forward direction; quasi-monoenergetic neutrons background of low-energy neutrons.
- Van de Graaff accelerator (D-D (gas target), D-T 13.8-20.5 MeV): wider energy range; neutron emission in  $4\pi$ ; quasi-monoenergetic neutrons background of low-energy neutrons; relatively low intensity.



# Sample composition

Interferences in production of the same radionuclide from different reactions on different isotopes in the sample.

42	<sup>92</sup> Mo STABLE 14.5% (n,γ)	<sup>93</sup> Mo 4.0E+3 Y ε: 100.00%	<sup>94</sup> Mo STABLE (n,3n)	<sup>95</sup> Mo STABLE 15.84% (n,p)	<sup>96</sup> Mo STABLE 16.67% (n,n+p)	<sup>97</sup> Mo STABLE 9.05%	<sup>98</sup> Mo STABLE 24.39%	<sup>99</sup> Mo 65,976 H β-: 100.00%	<sup>100</sup> Mo 7.3E+18 Y 9.82% 2β-: 100.00%
41	<sup>91</sup> Nb 6.8E+2 Y ε: 100.00%	<sup>92</sup> Nb 3.47E+7 Y ε: 100.00% β-: < 0.05%	<sup>93</sup> Nb STABLE 100% (n,2n)	<sup>94</sup> Nb 1.03E+4 Y β-: 100.00%	<sup>95</sup> Nb 34.5 D β-: 100.00%	<sup>96</sup> Nb 23,35 H β-: 100.00%	<sup>97</sup> Nb 72.1 M β-: 100.00%	<sup>98</sup> Nb 2.86 S β-: 100.00%	<sup>99</sup> Nb 15.0 S β-: 100.00%

samples with different isotopic composition

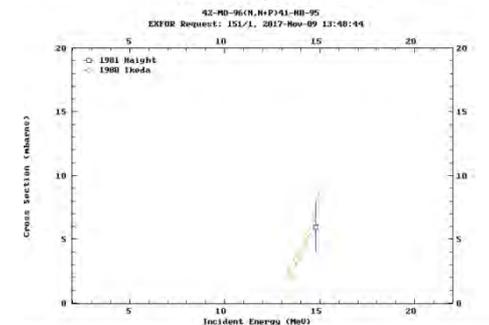
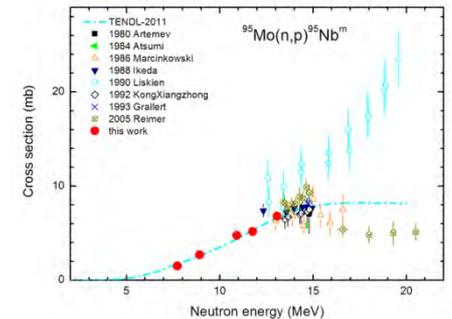
$$\sigma_1 = \frac{a_{y2}\sigma_x - a_{x2}\sigma_y}{a_{x1}a_{y2} - a_{x2}a_{y1}}$$

$$\sigma_2 = \frac{a_{x1}\sigma_y - a_{y1}\sigma_x}{a_{x1}a_{y2} - a_{x2}a_{y1}}$$

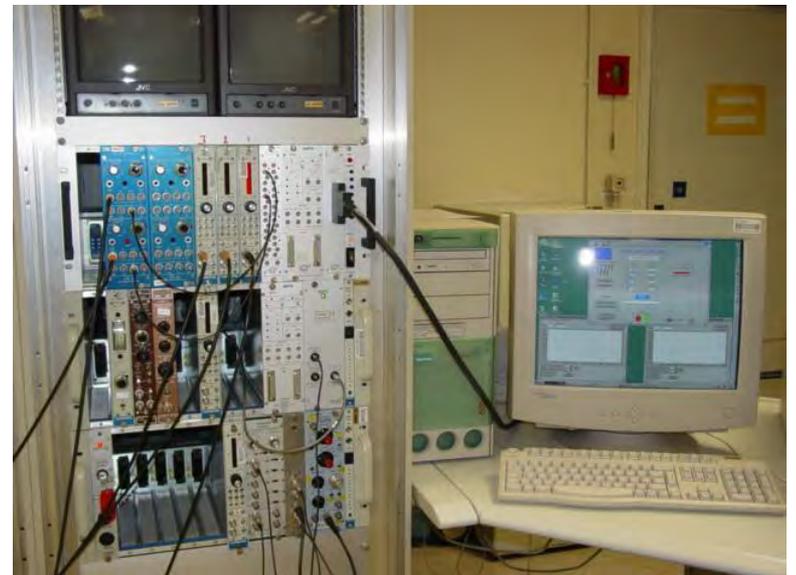
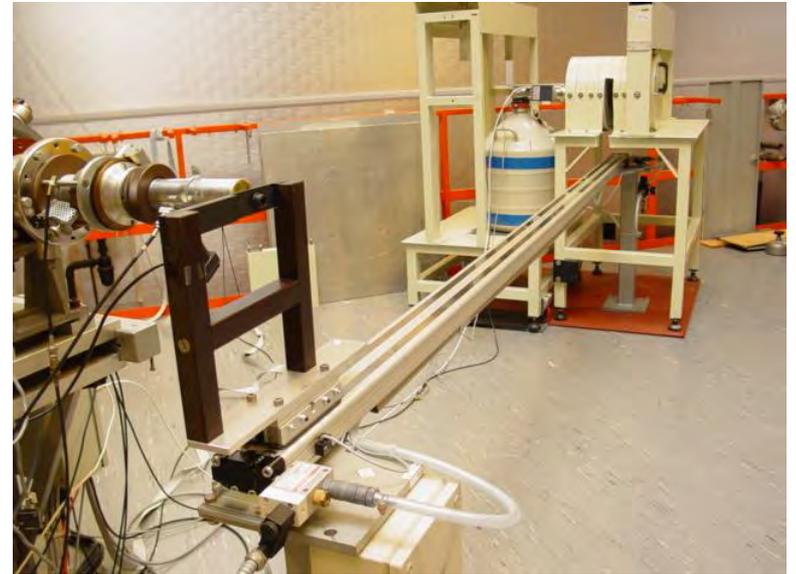
$\sigma_i$  -  $i=1,2$  reaction cross sections

$\sigma_s$ , effective cross sections for the sample  $s = x,y$  isotopic composition

$a_{si}$  - atom fraction of the target isotope in the sample



# Irradiation geometry



# Neutron spectrum characterisation

- **Mean neutron energy** and energy distribution for the irradiation geometry : the reaction **kinematics**
- **Spectrum unfolding** : **time-of-flight** measurements in combination with **threshold activation** measurements

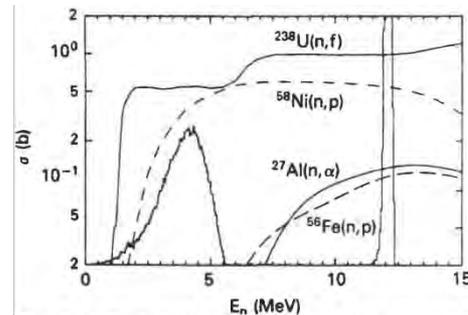
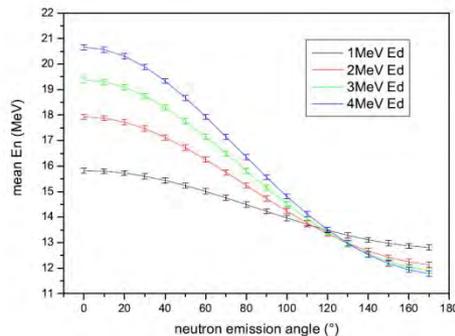
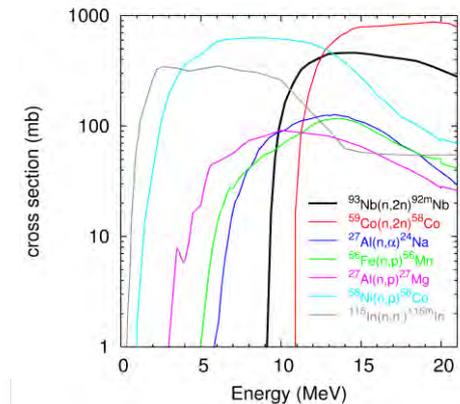
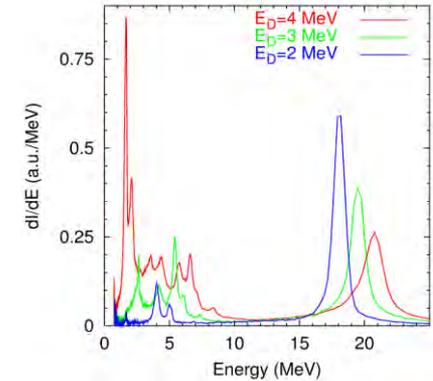


Fig. 1. Evaluated cross sections and the 0-deg neutron spectrum of the  $D(d,n)^3He$  reaction at 9.02 MeV. Cabral et al., Nucl. Sci. Eng. 106, 308-317 (1990).

# Coincidence summing effects I

3 decays found: **01** 100.0%  $\beta^-$  14.997 h  $^{24}_{11}\text{Na}_{13} \Rightarrow ^{24}_{12}\text{Mg}_{12}$  **02** 99.95% IT 20.18 ms  $^{24m}_{11}\text{Na}_{13} \Rightarrow ^{24}_{11}\text{Na}_{13}$  **03** 0.05%  $\beta^-$  20.18 ms  $^{24m}_{11}\text{Na}_{13} \Rightarrow ^{24}_{12}\text{Mg}_{12}$

**01 Evaluation:** R.B. FIRESTONE Publication cut-off: 3-Oct-2006 ENSDF insertion: 2007-10 Publication: Nuclear Data Sheets

Parent	T <sub>1/2</sub>	E <sub>x</sub> [keV]	J <sup>π</sup> order	Decay	Q decay note on Q value	Daughter	Comments
$^{24}_{11}\text{Na}_{13}$	14.997 h 12	0	4+	$\beta^-$ 100 %	5515.611 39	$^{24}_{12}\text{Mg}_{12}$	

see the ENSDF source

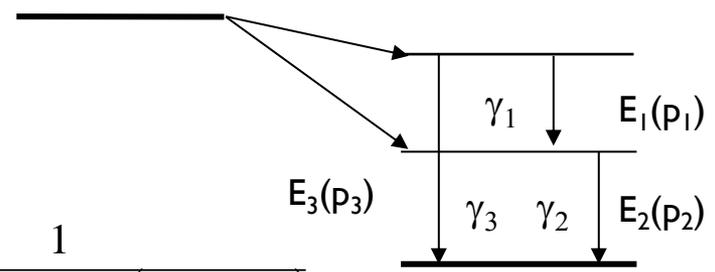
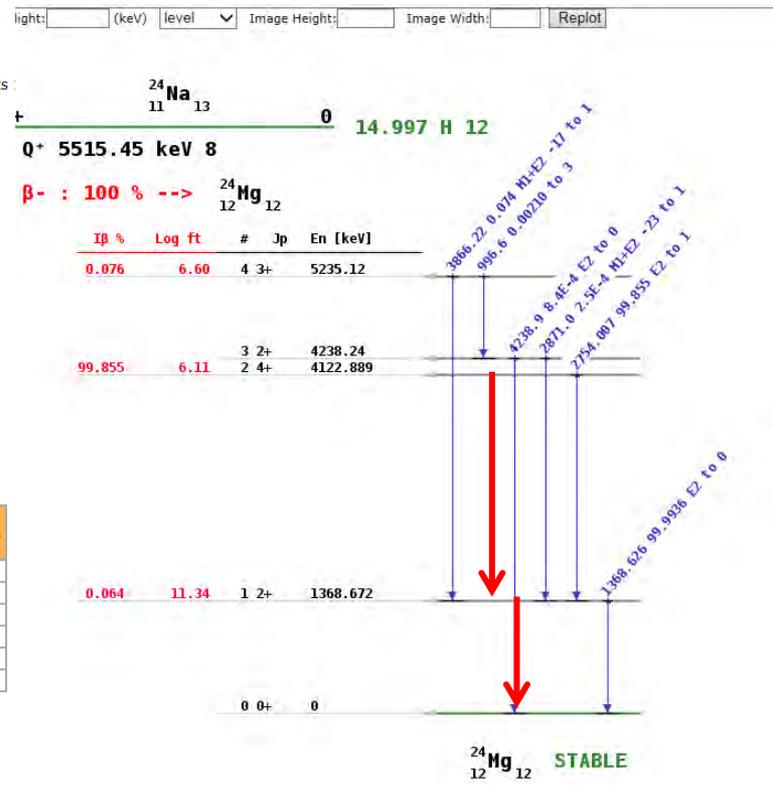
Note: Q-value used in ENSDF to determine displayed decay data is: 5515.61 4 keV - see note on Q value

## Beta -

<E <sub>β</sub> > [keV]	I <sub>β</sub> (abs) [%]	Daughter level [keV]	J <sup>n</sup>	E <sub>β</sub> , max [keV]	logft	Transition type	Comments
89.24 22	0.076 3	5235.12 4	3+	280.33 9	6.60 2		
554.1 3	<b>99.855 5</b>	4122.889 12	4+	1392.56 8	6.11 1		
1865.5 3	0.064 6	1368.672 5	2+	4146.78 8	11.34 4		

## Gamma

E <sub>γ</sub> [keV]	I <sub>γ</sub> (abs) [%]	Initial level [keV]	J <sup>n</sup>	Final level [keV]	J <sup>n</sup>	Mult.	δ	α <sub>T</sub>	Comments
996.6 10	0.00210 20	5235.12 4	3+	4238.24 3	2+				
1368.626 5	<b>99.9936 15</b>	1368.672 5	2+	0	0+	E2		1.3E-5	
2754.007 11	99.855 5	4122.889 12	4+	1368.672 5	2+	E2			
2871.0 10	2.5E-4 4	4238.24 3	2+	1368.672 5	2+	M1+E2	-23 9		
3866.22 15	0.074 3	5235.12 4	3+	1368.672 5	2+	M1+E2	-17 4		
4238.9 10	8.4E-4 10	4238.24 3	2+	0	0+	E2			



$$C_{\text{coin}3} = \frac{1}{1 + p_{\gamma 1} \cdot \epsilon_{\text{peff}1} \cdot \epsilon_{\text{peff}2} / (p_{\gamma 3} \cdot \epsilon_{\text{peff}3})}$$

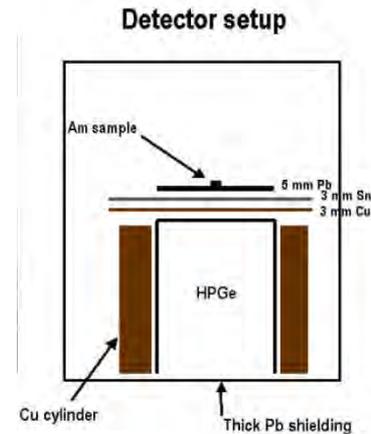
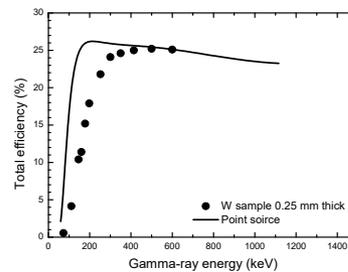
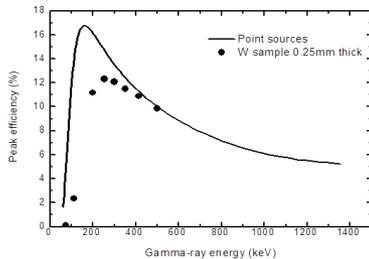
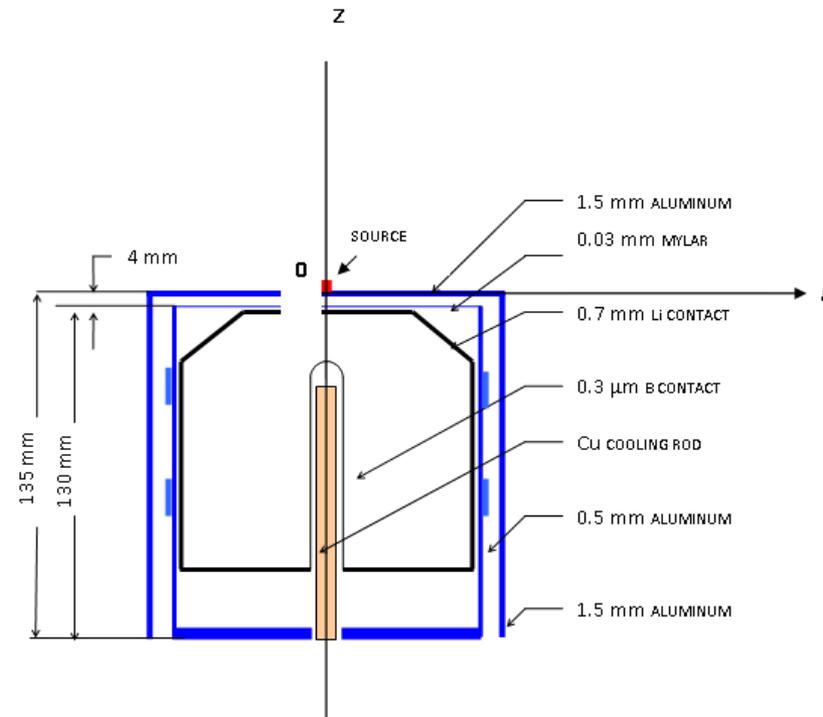
$$C_{\text{coin}1} = \frac{1}{1 - \epsilon_{t2}}$$

$$C_{\text{coin}2} = \frac{1}{1 - (p_{\gamma 1} / p_{\gamma 2}) \epsilon_{t1}}$$



# Detector efficiency

The **Monte Carlo simulation** of the detector response allows taking into account the detailed characteristics of the detector and samples (complex shape, sample matrix,  $\gamma$ -ray self-attenuation, volume activity distribution, coincidence summing effects, etc.



Self-attenuation correction:  
 standard formula 0.85  
 MCNP 0.65

# Nuclide identification by $E_\gamma$ and $T_{1/2}$

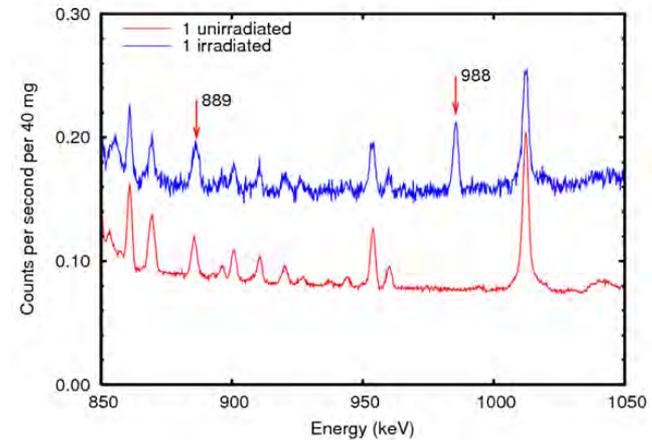
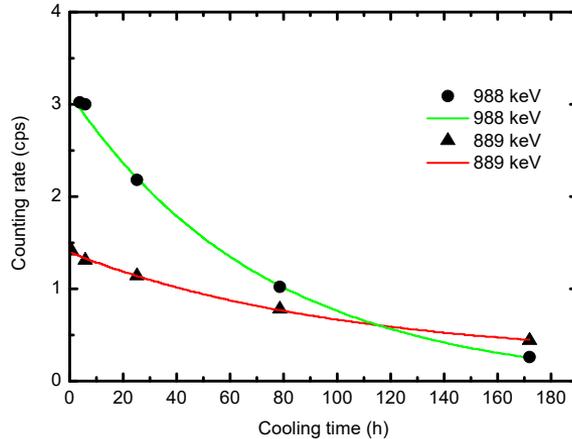
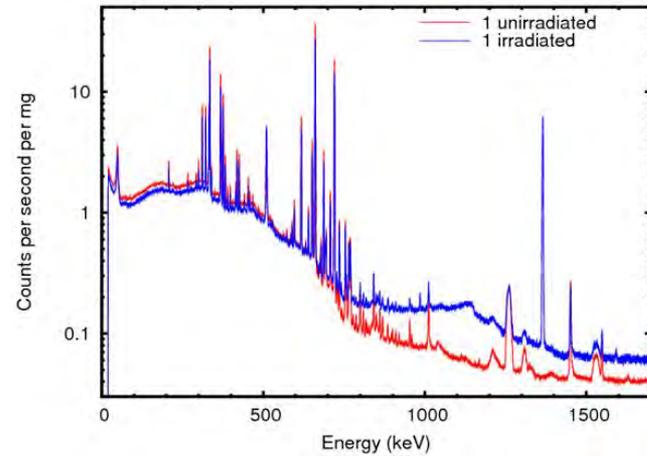
$^{240}\text{Am}$  decay data

$T_{1/2}$  50.8(3) h

E (keV)  $I_\gamma$  (%)

988 73(4)

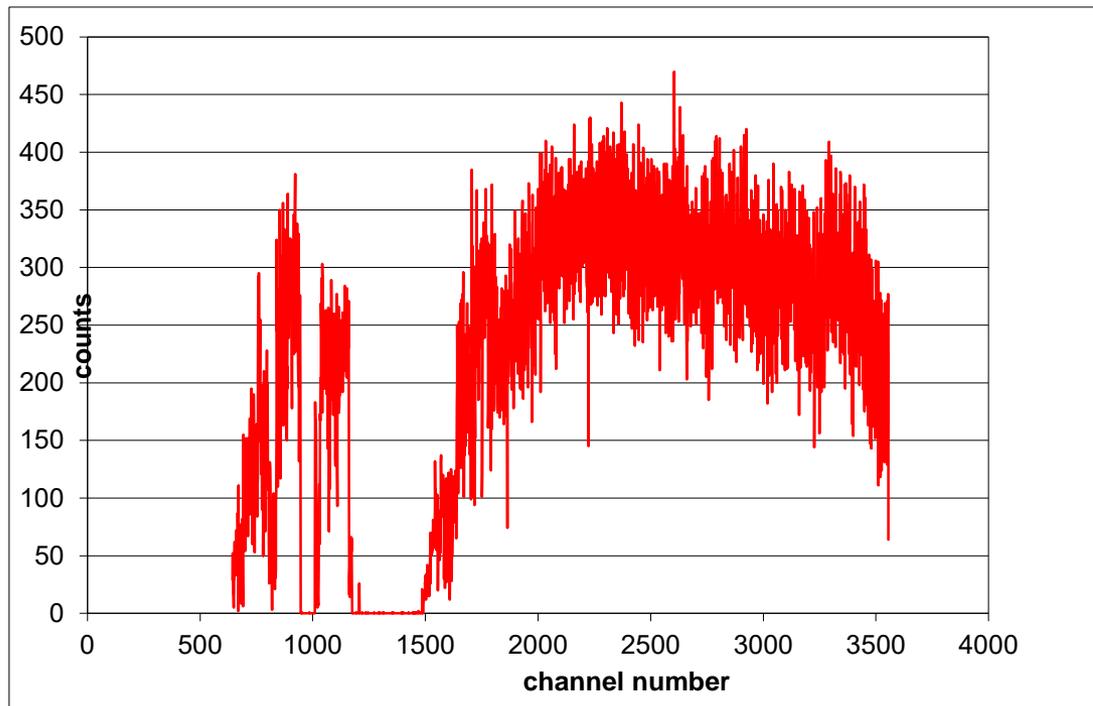
889 25.1(1.3)



fitted  $T_{1/2}(988 \text{ keV}) = 50.88 \text{ h}$

# Neutron flux variation during irradiation

$$C_{\text{flux}} = \frac{\bar{\Phi}(1 - e^{-\lambda t_r})}{\sum_{i=1}^m \Phi_i(1 - e^{-\lambda \Delta t})e^{-\lambda(m-i)\Delta t}}$$





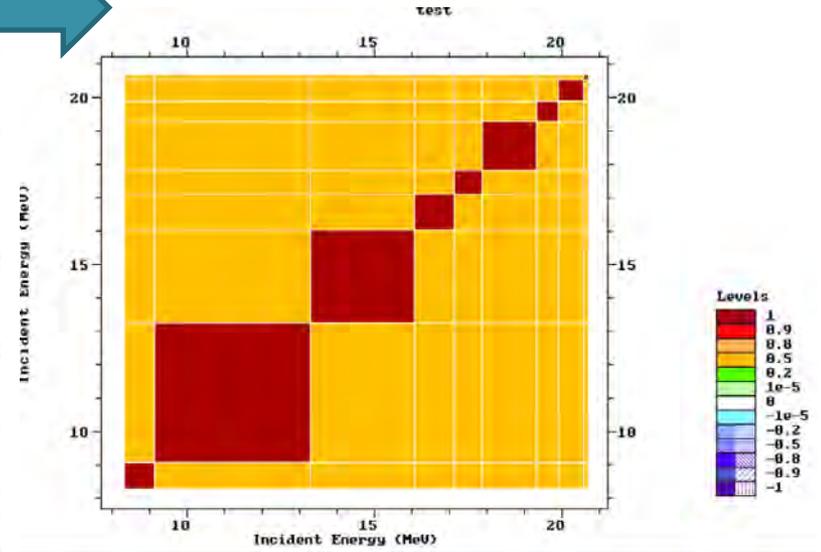
# Constructing a covariance matrix from EXFOR uncertainties

```

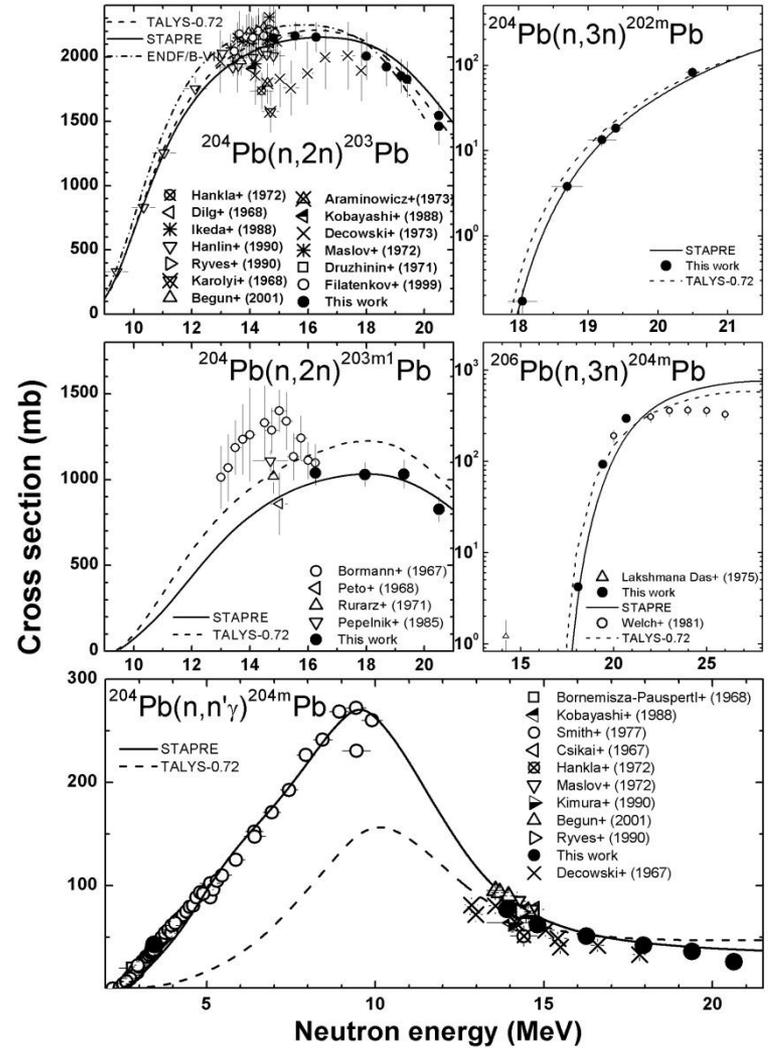
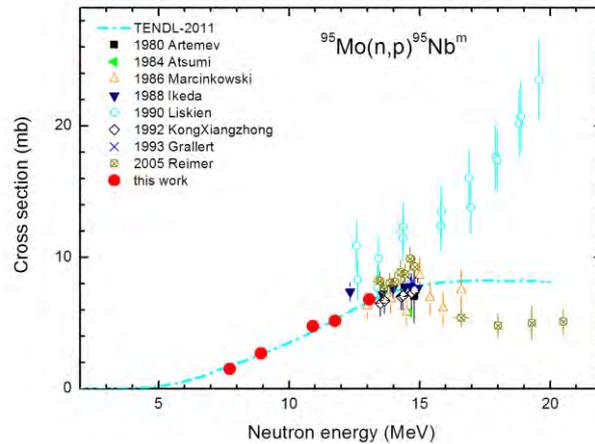
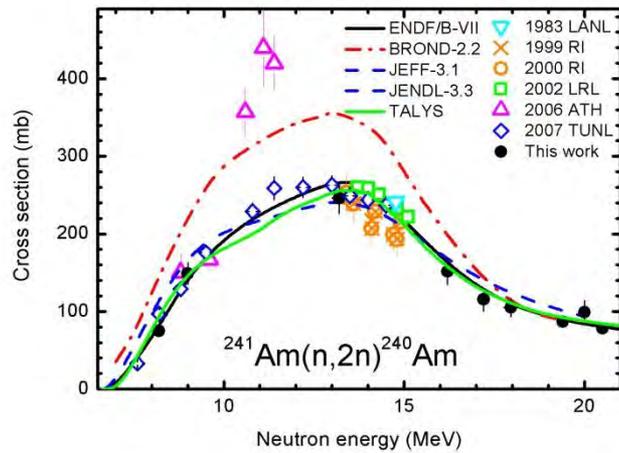
ERR-ANALYS (ERR-T,,,P) Total uncertainty
(MONIT-ERR,,,P) 27Al(n,a) standard x-section (1.6-5.4%)
(ERR-1,,,U) Counting of 240Am activity (1.4-6.3%)
(ERR-2,,,U) Counting of 24Na activity (0.7-2.0%)
(ERR-3,,,F) Intensity of 240Am gamma line (1.2%)
(ERR-4,,,U) Number of 27Al in sample (0.1%)
(ERR-5,,,P) Number of 241Am in sample (0.3%)
(ERR-6,,,F) 24Na/240Am efficiency ratio (3.0%)
(ERR-7,,,F) Correction for decay of 240Am (0.4-0.9%)
(ERR-8,,,U) Correction for secondary neutron (<1.4%)

# /SUBENT 23114002
    
```

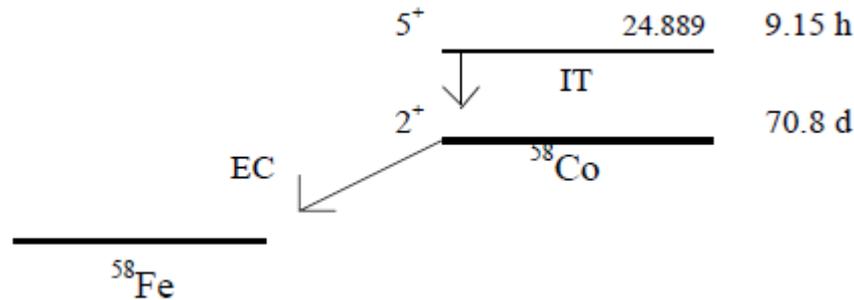
No.	1	2	3	4	5	6	7	8	9
Energy (eV) *0.001	8340.	9150.	13330.	16100.	17160.	17900.	19360.	19950.	20610.
Data (B) *1000.	96.8	162.9	241.8	152.4	116.1	105.7	89.5	102.1	77.9
<b>Uncertainties defined in C5 (C4++)</b>									
Total (%)	6.5	5.7	4.6	4.6	4.4	4.4	8.2	5.8	8.8
Statistical (%) <i>empty</i>	-	-	-	-	-	-	-	-	-
Systematic (%)	6.4	5.6	4.5	4.5	4.3	4.3	8.1	5.7	8.8
<b>Uncertainties given in EXFOR</b>									
ERR-T (%)	6.5	5.7	4.6	4.6	4.4	4.4	8.2	5.8	8.8
ERR-1 (%)	5.0	4.0	2.5	2.1	1.5	1.3	6.3	1.4	5.7
ERR-2 (%)	1.0	1.0	1.0	1.0	1.0	0.7	2.0	1.0	1.6
ERR-3 (%) <i>const</i>	1.2	=	=	=	=	=	=	=	=
ERR-4 (%) <i>const</i>	0.1	=	=	=	=	=	=	=	=
ERR-5 (%) <i>const</i>	0.3	=	=	=	=	=	=	=	=
ERR-6 (%) <i>const</i>	3.0	=	=	=	=	=	=	=	=
ERR-7 (%)	0.9	0.6	0.4	0.6	0.6	0.7	0.6	0.6	0.6
ERR-8 (%)	-	-	0.3	0.3	0.3	0.3	1.3	1.4	1.4
MONIT-ERR (%)	1.9	1.9	1.6	2.0	2.0	2.2	3.1	4.1	5.4



# Some results



# Isomeric ratio measurements



$$N_g(t_c) = n\Phi \left\{ \left( \frac{p\sigma_m + \sigma_g}{\lambda_g} + \frac{p\sigma_m}{\lambda_m - \lambda_g} \right) \left( 1 - e^{-\lambda_g T} \right) e^{-\lambda_g t_c} - \frac{p\sigma_m}{\lambda_m - \lambda_g} \left( 1 - e^{-\lambda_m T} \right) e^{-\lambda_m t_c} \right\}$$

if we substitute

$$A = \left\{ \left( \frac{p\sigma_m + \sigma_g}{\lambda_g} + \frac{p\sigma_m}{\lambda_m - \lambda_g} \right) \left( 1 - e^{-\lambda_g T} \right) \right\}$$

$$B = \frac{p\sigma_m}{\lambda_m - \lambda_g} \left( 1 - e^{-\lambda_m T} \right)$$

then

$$N_g(t_c) = n\Phi \left( A e^{-\lambda_g t_c} + B e^{-\lambda_m t_c} \right)$$

The coefficients A and B can be determined by fitting the experimental data, and the isomeric cross section ratio is then given by:

$$\frac{\sigma_m}{\sigma_m + \sigma_g} = \frac{(\lambda_m - \lambda_g)}{\lambda_g} \left( \frac{B}{\left( \frac{1 - e^{-\lambda_m T}}{1 - e^{-\lambda_g T}} \right) A - B} \right)$$

# ${}^9\text{Be}(d,n)$ thick target neutron spectrum

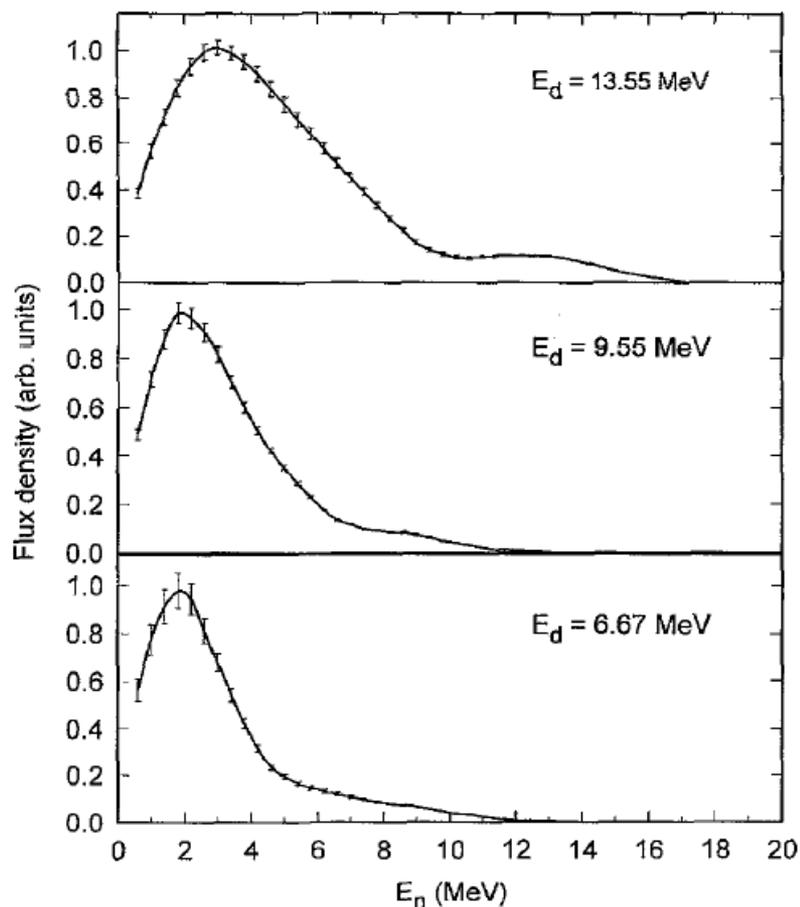


Fig. 7. Multiple foil activation unfolding of neutron spectra from a thick Be target at  $E_d = 13.55, 9.55$  and  $6.67$  MeV.

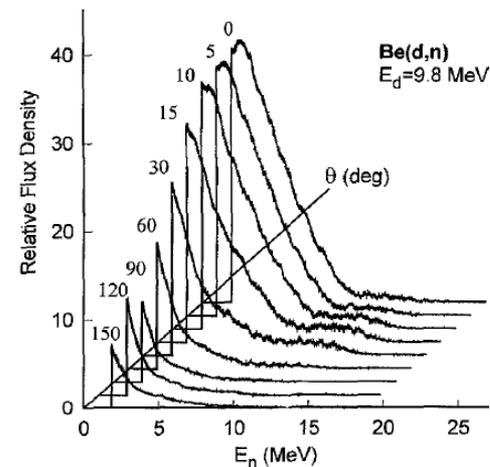
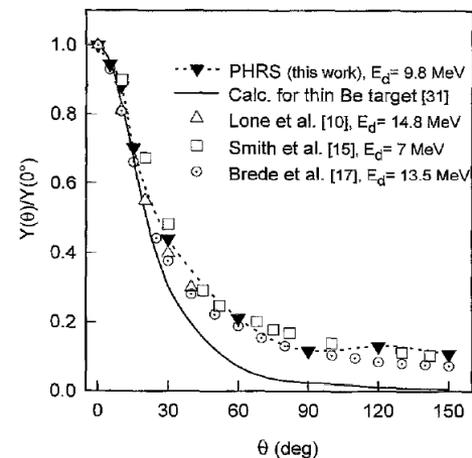


Fig. 9. Relative flux density of  ${}^9\text{Be}(d,n)$  neutrons vs. emission angle measured by PHRS.



# Spectrum average cross sections

$$\bar{\sigma} = \frac{\int_{E_1}^{E_2} \sigma(E)\Phi(E)dE}{\int_{E_1}^{E_2} \Phi(E)dE} = \int_{E_1}^{E_2} \sigma(E)\chi(E)$$

TABLE V

Measured and Calculated Be(d,n) Spectrum-Averaged Data for (n,p) Reactions

Reaction	Measured (mb)	ENDF-B/VI	IRDF90	JEF-2	JENDL3	CENDL2	SINCROS II	ADL-3	BROND
$^{58}\text{Ni}(n,p)^{58m+p}\text{Co}$	$325.65 \pm 30$	316.41	320.52	316.41	312.74			201.46	324.02 <sup>a</sup>
$^{60}\text{Ni}(n,p)^{60m+p}\text{Co}$	$18.54 \pm 1.9$	19.43 <sup>a</sup>	19.81	19.43 <sup>a</sup>	25.56			10.91	25.84
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	$111.34 \pm 10$		119.00 <sup>a</sup>	135.86			126.81	133.01	
$^{59}\text{Co}(n,p)^{59}\text{Fe}$	$9.01 \pm 0.9$	9.01 <sup>a</sup>		9.01 <sup>a</sup>	9.02	8.77		13.55	
$^{90}\text{Zr}(n,p)^{90m}\text{Y}$	$0.56 \pm 0.05$						0.48 <sup>a</sup>	5.49	
$^{91}\text{Zr}(n,p)^{91m}\text{Y}$	$0.86 \pm 0.09$						0.81 <sup>a</sup>	2.99	
$^{92}\text{Zr}(n,p)^{92}\text{Y}$	$0.59 \pm 0.06$	0.87		0.77	0.58		0.83	0.41	0.52
$^{94}\text{Zr}(n,p)^{94}\text{Y}$	$0.12 \pm 0.02$	0.30		0.29	0.14		0.11	0.12 <sup>a</sup>	0.17
$^{91}\text{Mo}(n,p)^{91}\text{Nb}$	$34.42 \pm 4.0$						30.28 <sup>a</sup>	40.21	
$^{96}\text{Mo}(n,p)^{96}\text{Nb}$	$0.62 \pm 0.07$			0.84	0.64		0.66	0.62 <sup>a</sup>	

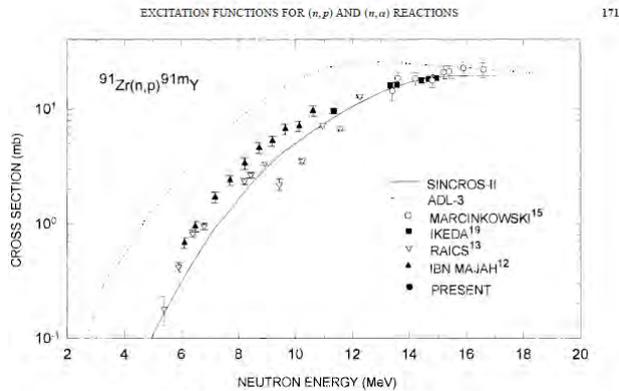


Fig. 8. Hydrogen production via the  $^{91}\text{Zr}(n,p)^{91m}\text{Y}$  reaction.

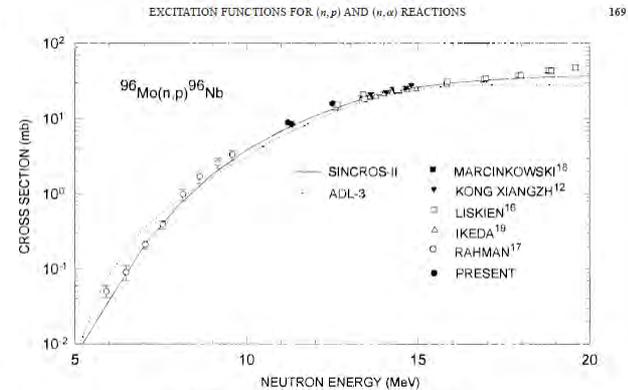
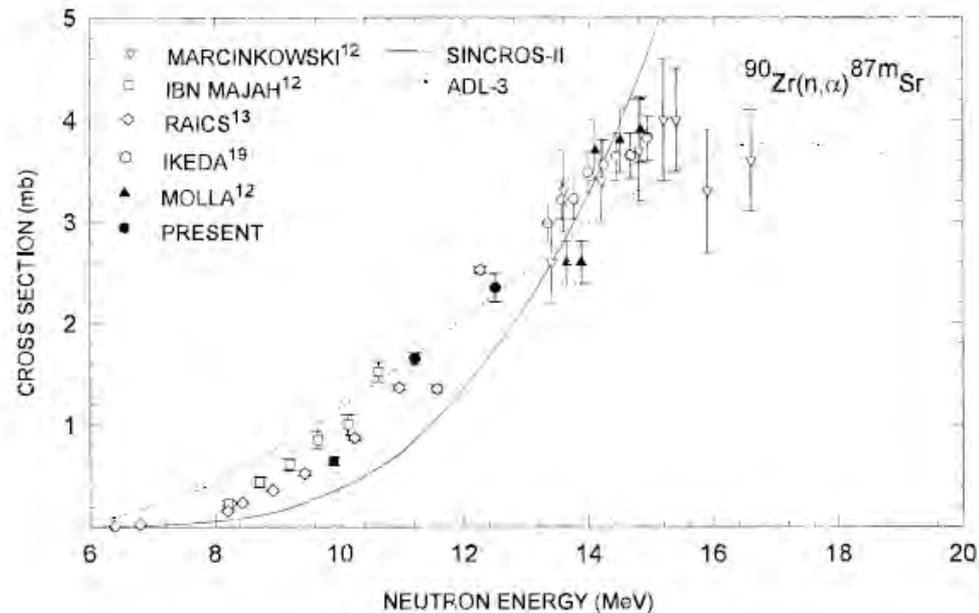


Fig. 4. Hydrogen production via the  $^{96}\text{Mo}(n,p)^{96}\text{Nb}$  reaction.

TABLE IV

Measured and Calculated Be(*d, n*) Spectrum-Averaged Data for (*n, α*) Reactions

Reaction	Measured (mb)	ENDF-B/VI	IRDF90	JEF-2	JENDL3	CENDL2	SINCROS II	ADL-3	BROND
$^{54}\text{Fe}(n, \alpha)^{51}\text{Cr}$	$7.80 \pm 0.80$	9.96		10.11	9.62			11.24	7.25 <sup>a</sup>
$^{51}\text{V}(n, \alpha)^{48}\text{Sc}$	$0.61 \pm 0.06$		0.62 <sup>a</sup>		0.67		0.52	0.47	
$^{59}\text{Co}(n, \alpha)^{56}\text{Mn}$	$2.10 \pm 0.18$	2.35	2.40	2.35	2.45	2.40		2.13 <sup>a</sup>	
$^{68}\text{Zn}(n, \alpha)^{65}\text{Ni}$	$0.67 \pm 0.07$						0.86 <sup>a</sup>	0.42	
$^{90}\text{Zr}(n, \alpha)^{87\text{m}}\text{Sr}$	$0.12 \pm 0.02$						0.08 <sup>a</sup>	0.18	

Fig. 7. Helium production via the  $^{90}\text{Zr}(n, \alpha)^{87\text{m}}\text{Sr}$  reaction.