Measurement of neutron induced reaction cross-sections of structural materials and compilation of Indian experimental nuclear data in IAEA-EXFOR database

Paresh M. Prajapati
Junior Research Fellow (DAE-BRNS)
Physics Department, Faculty of Science
The M.S. University of Baroda, Vadodara – 390002

Working Place : Reactor Physics Design Division/
Radiochemistry Division,
Bhabha Atomic Research Centre,
Trombay, Mumbai – 400085

E-mail ID: paresh_21soft@yahoo.co.in

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1. Motivation of Experiments

Zr, Ni and Fe are an important component of the structural materials used in traditional and advanced nuclear reactors. Because of its very low absorption cross-sections of thermal neutrons and resistance to corrosion, these materials are frequently used as cladding of fuel rods, calandria vessel and pipe lines of secondary coolant circuit in nuclear reactors in. But its cross-sections database especially for neutron threshold reactions are rather sparse. Besides applications, excitation functions of neutron threshold reactions are of considerable interest for testing nuclear models.

An examination of International Atomic Energy Agency-Exchange Format (IAEA-EXFOR) database shows that a significant discrepancy as well as gaps exits in the measured experimental data for many neutron threshold reactions in MeV region and at thermal energies. It also indicates that there has been no neutron capture or (n,γ) cross-section measurements available beyond 2 MeV region of neutron energy. Furthermore, literature survey shows that most of the thermal neutron activations cross-sections for Zirconium isotopes were made in reactors with neutron spectra and therefore were not precise thermal cross-section measurements.

Motivated by these requirements, the neutron induced reaction cross-sections for $^{94}Zr(n,\gamma)^{95}Zr$, $^{96}Zr(n,\gamma)^{97}Zr$, $^{90}Zr(n,p)^{90}Y^{m}$, ^{58}Fe (n, γ)^{59}Fe, ^{56}Fe (n, p)^{56}Mn, ^{64}Ni (n, γ) ^{65}Ni and ^{58}Ni (n, p) ^{58}Co were determined at neutron energies of thermal (0.0253 eV), 2.45 MeV, 3.7 MeV and 7. 3 MeV using activation and off-line γ-ray spectrometric technique. $^{2}H(D,n)^{3}He$ reaction was used to produce quasi-monoenergetic neutrons of energy (En) = 2.45 MeV by Purnima neutron generator while average neutrons of energy (En) = 3.7 MeV and 7.3 MeV were produced by $^{7}Li(p,n)^{7}Be^{*}$ reaction using BARC-TIFR Pelletron facility. For determination of “thermal neutron” activation cross-sections, thermal column of swimming pool type APSARA reactor was utilized. The present measurement at thermal neutron energy is compared with experimental data from EXFOR and is used to validate methodology applied here. Theoretical calculations were performed using TALYS 1.2 code
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2. Experimental Method, calculations and results

A: Thermal neutron activation with APSARA reactor

1. A known amount of natural Zr , Ni, and Fe metal foils of thickness about 1 mm and neutron flux monitor Au metal foil (0.0215 gm) were wrapped separately with 0.025 mm thick super pure Aluminum and doubly sealed with Alkathene bags.

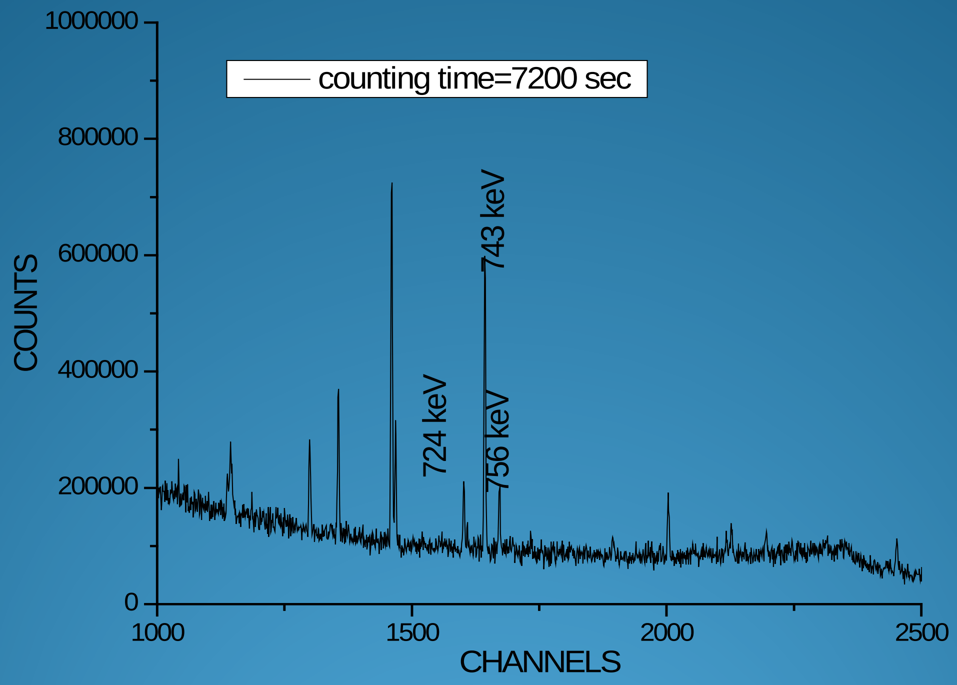
2. These samples were kept inside an irradiation capsule made of polypropylene. The capsule containing samples were doubly resealed with Alkathene.

3. These samples were irradiated in the thermal column of swimming pool type APSARA reactor for the period of 6 hours and 30 minutes.

4. After sufficient cooling, the irradiated samples along with Al wrapper were mounted on different Perspex plates and taken for gamma-ray spectrometry. Radioactivity in the irradiated samples were measured using energy and efficiency calibrated 80 cm^3 high-purity germanium (HPGe) detector coupled to a PC based 4K channel analyzer in live time mode. The efficiency of the detector was 20 % with energy resolution of 1.8 keV FWHM at 1332.0 keV peak of ^{60}Co.

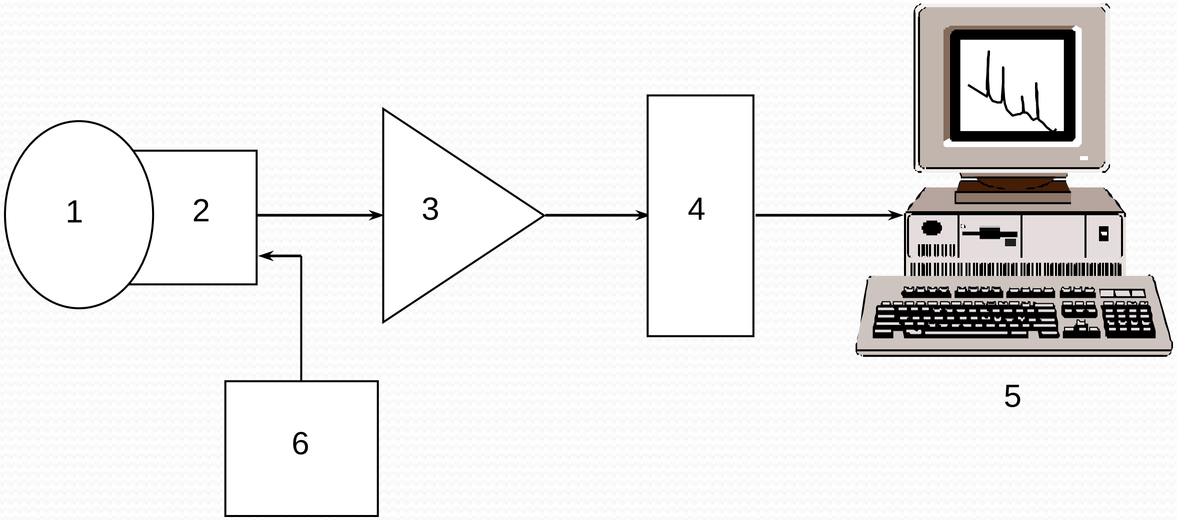
5. The standard ^{152}Eu source (having gamma-rays in the energy of 121.8-1408 keV was used for energy and efficiency calibration. The dead time of the detector system during counting was always kept less than 10 % by placing the sample at a suitable distance to avoid pile up effects.

Fig.1 Gamma-ray spectrum of irradiated natural Zr showing the γ-ray energy of ^{95}Zr and ^{97}Zr



The gamma-ray spectra, as shown in Fig. 1, were analyzed with peak fitting program PHAST which can search for up to 500 peaks and fit model peak shape.

Schematic of HPGe gamma ray spectrometry setup



1: High-Purity Coaxial Germanium detector (HPGe)
2: Preamplifier
3: Amplifier
4: 4 K Multichannel analyzer
5: PC
6: Bias supply (High Voltage: +2500 v)

B: Neutron capture cross-section measurement of $^{94}Zr(n,\gamma)^{95}Zr$ at neutron energy of 2.45 MeV using Purnima Neutron Generator.

1. Purnima Neutron Generator is 300 KV DC electrostatic accelerator (based on Cockcroft and Walten type multiplier) in which D^+ ion beam is accelerated and bombarded on deuterium target.
2. It produces quasi-monoenergetic neutrons of energy 2.45 MeV based on the $D(D,n)^3He$ fusion reaction. The operating parameter of the neutron generator for the experiment were 115 µA D^+ ion beam current, 100 kV acceleration voltage and vacuum inside the system was maintained at pressure of 3×10^{-6} mbar.
3. Natural Zirconium metal foil of amount 0.0952 gm with thickness of 1 mm and Indium metal foil (neutron flux monitor) of amount 0.057 gm with same thickness as Zr metal foil were wrapped separately with 0.025 mm thick super pure Aluminum foil. These samples were placed at the neutron source (deuterium target) respect to zero degree with incident beam direction and irradiated for the period of 2 hours and 30 minutes. After sufficient cooling, high resolution gamma-ray spectrometry of these activated samples was performed using energy and efficiency calibrated HPGe detector as mentioned in the section A.

3. Neutron cross-section measurements at average neutron energies of 3.7 MeV and 7.3 MeV using BARC- TIFR Pelletron.

- The experiments were carried out BARC-TIFR Pelletron facility at TIFR, Mumbai.
- Proton beam of energies 5.6 MeV and 12 MeV bombarded on Li target to produces average neutrons of energy 3.7 MeV and 7. 3 MeV respectively.
- Li foil was wrapped with tantalum foil to stop the proton beam.
- Neutron Production :

$$P + {}^{7}Li \rightarrow n + {}^{7}Be - 1.64\ MeV \quad (E_{th}=1.880\ MeV)$$

- Both the samples were placed at 0^0 angle with respect to incident neutron beam.

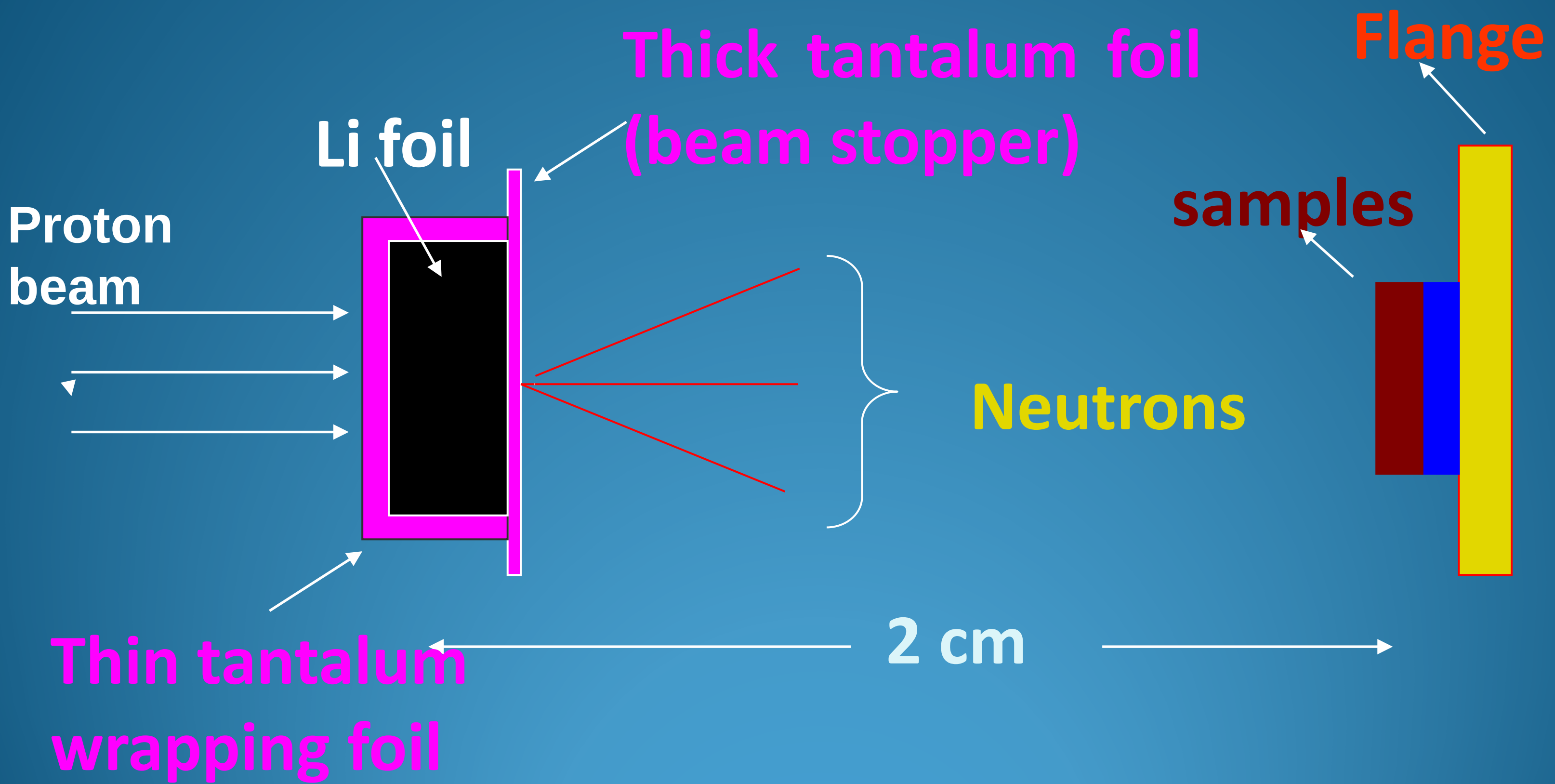
♦ The uranium metal foil was used to measure the neutron flux.

♦ The samples natural Zr, Fe and Ni were irradiated for 4 to 6 hours with average neutron energy of 3.7 MeV and 7.3 MeV from ^{7}Li (p,n) ^{7}Be reaction of 5.6 MeV and 12 MeV proton beam respectively at the 6 meter height main line of Pelletron facility.

♦ After sufficient cooling, high resolution gamma-ray spectrometry of these activated samples was performed using energy and efficiency calibrated HPGe detector as mentioned in the section A.

♦ `

The neutrons produced in $^{7}Li(p,n)$ reaction with 5.6 MeV and 12 MeV proton beam were not mono-energetic. Thus the neutron spectrum, obtained by using EMPIRE-2.1 as shown in fig. 2, had a peak at 3.7 and 8.8 MeV respectively for 5.5 MeV and 12 MeV with tailing towards lower energy. The flux-weighted average neutron energy were calculated to be 3.7 MeV and 7.3 MeV.



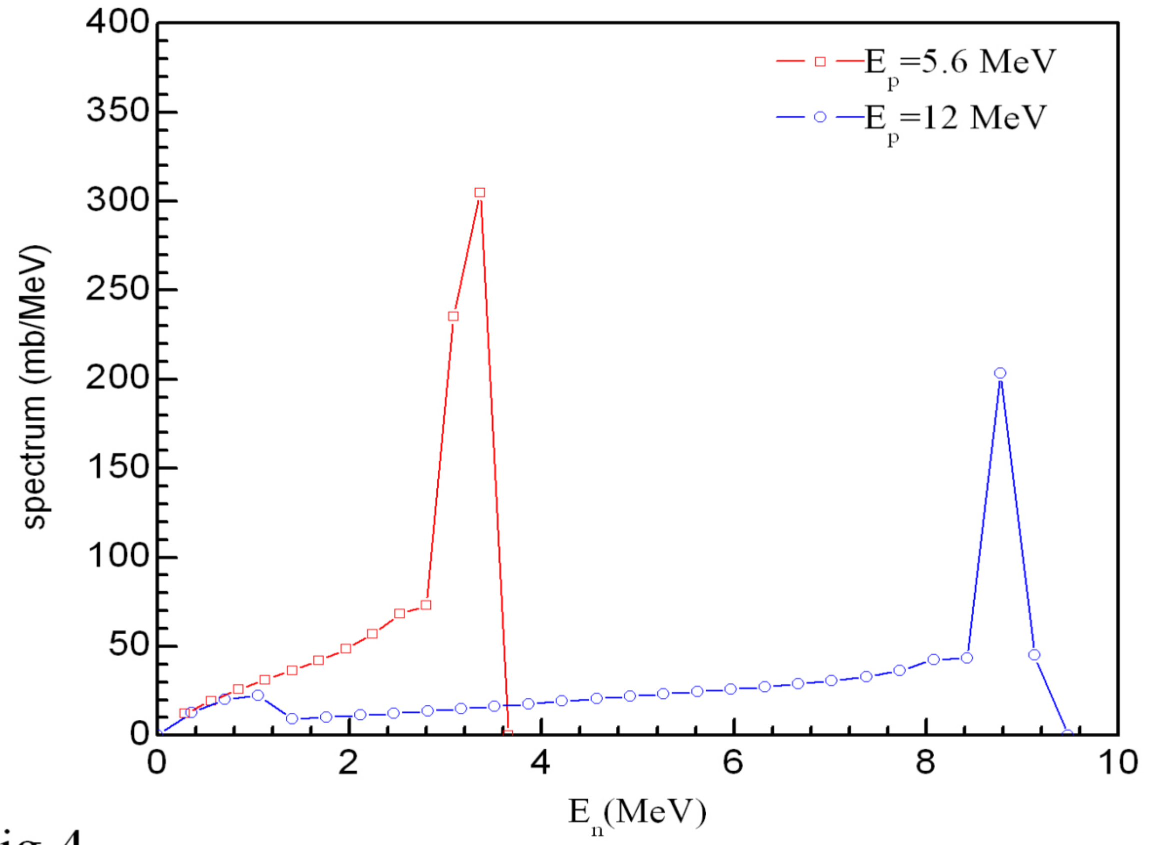


Fig.4

Fig. Neutron spectrum from $^{7}Li(p,n)$ reaction irradiated with proton energy of 5.6 MeV and 12 MeV calculated using EMPIRE-2.1 computer code.

Table 1. Nuclear Spectroscopic Data used for the neutron flux and cross-section section calculation

Nuclide	Half life	γ-ray energy (keV)	γ-ray abundance (%)
^{198}Au	2.695 d	411.80	96.0
$^{115}In^{m}$	4.486 h	336.24	45.90
^{95}Zr	64.02 d	724.90 756.72	44.17 54.17
^{97}Zr	16.91 h	743.36	93.0
$^{90}Y^{m}$	3.19 h	202.51 479.17	97.3 90.72
^{59}Fe	44.503 d	1099.251 1291.59	56.5 43.2
^{56}Mn	2.58 h	846.7	98.9
^{58}Co	70.78 d	810.77	99

Neutron Flux & Reaction Cross-section Calculation

★From observed activity (A_{obs}) of a particular reaction product, neutron flux (φ) and reaction cross-sections (σ) were obtained using following decay equation ,

$$A_{obs} = N\,\sigma\,\varphi a\,\varepsilon\,(1-e^{-\lambda t})\,e^{-\lambda T}(1 - e^{-\lambda \Delta T}) \;/\; \lambda$$

where N = Number of atoms of the isotope of the element

σ = cross – section

a = γ-ray abundance

φ = Neutron Flux

ε = Detector efficiency

λ = Decay constant

t = Irradiation time

T = Cooling time

ΔT = Counting time

Table.2. Experimentally measured reaction cross-sections at different neutron energy

Energy (MeV)	Reaction	Cross-section (barn)	Talys 1.0 (barn)	EXFOR (barn)	JENDL 4.0 (barn)	ENDF/B-VII (barn)
Thermal Neutron	$^{94}Zr(n,\gamma)^{95}Zr$	0.051±0.0037	---------	0.047 to 0.075	0.050	0.049
Thermal Neutron	$^{96}Zr(n,\gamma)^{97}Zr$	0.024±0.0028	---------	0.02 to 0.1	0.020	0.022
Thermal Neutron	$^{58}Fe(n, \gamma)^{59}Fe$	1.04±0.042	---------	0.106-1.31	1.30049	1.14965
Thermal Neutron	$^{64}Ni(n, \gamma)^{64}Ni$	1.3706±0.042	----------	1.35-1.96	1.48036	1.5184
2.45	$^{94}Zr(n,\gamma)^{95}Zr$	0.0054±0.0009	0.0030	---------	0.0032- 0.0019	0.0079- 0.0050
3.7	$^{58}Ni(n,p)^{58}Co$	0.317±0.036	0.297	0.298-0.333	0.27563-0.35607	0.212148-0.359928
7.3	$^{90}Zr(n,p)^{90}Y^{m}$	0.0021±0.0004	0.0025 (0.0021)*	0.0010-0.0049	---------	---------
7.3	$^{56}Fe(n,p)^{56}Mn$	0.033±0.0004	0.041(0.036)*	0.031-0.044	0.027281-0.04183	`0.027- 0.0029

*Flux weighted average

Fig. Plot of experimental and theoretical $^{94}Zr(n,\gamma)^{95}Zr$ reaction cross-section as a function of neutron energy from 1 keV to 20 MeV from present work and from EXFOR in open circle.

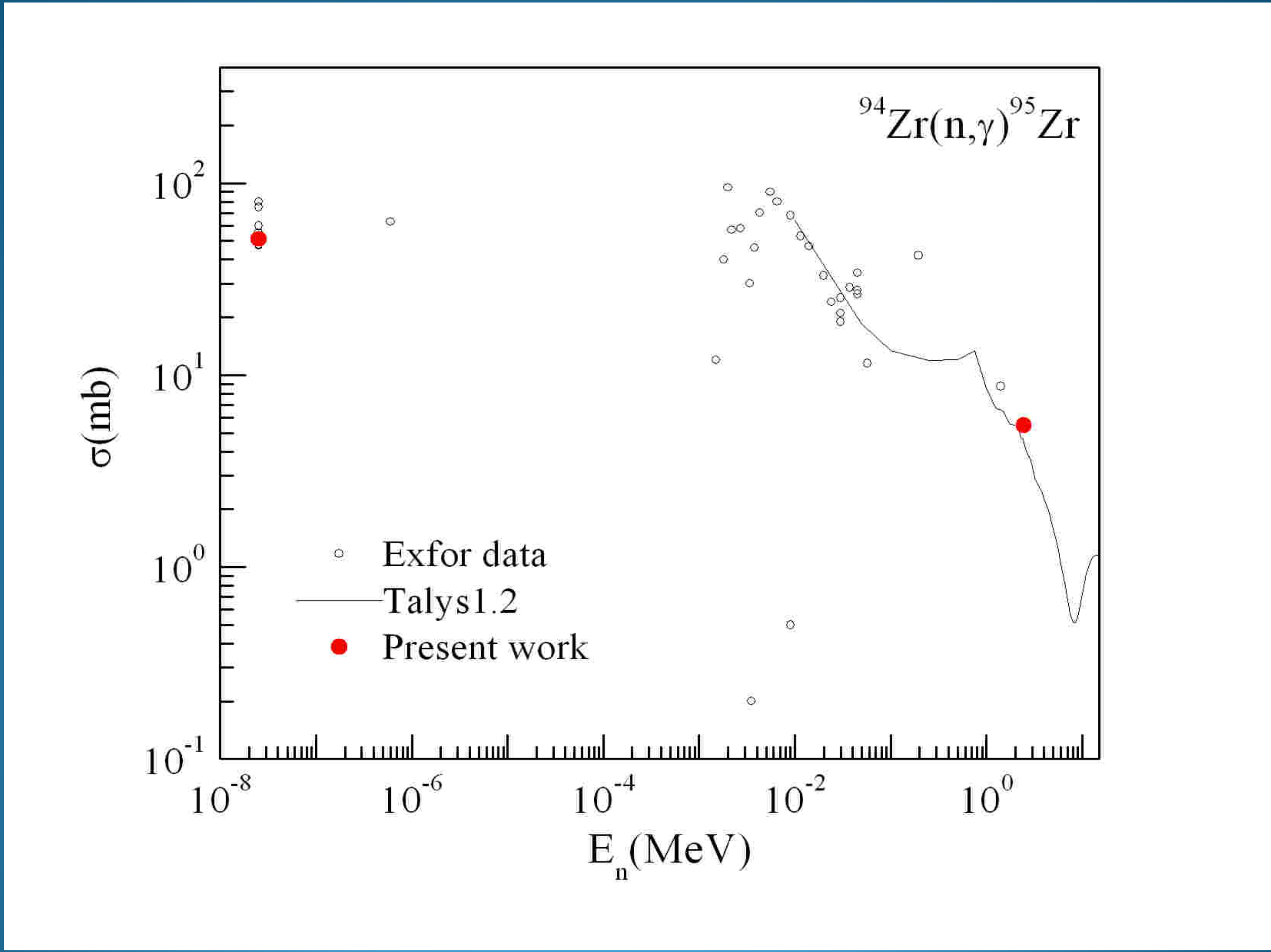
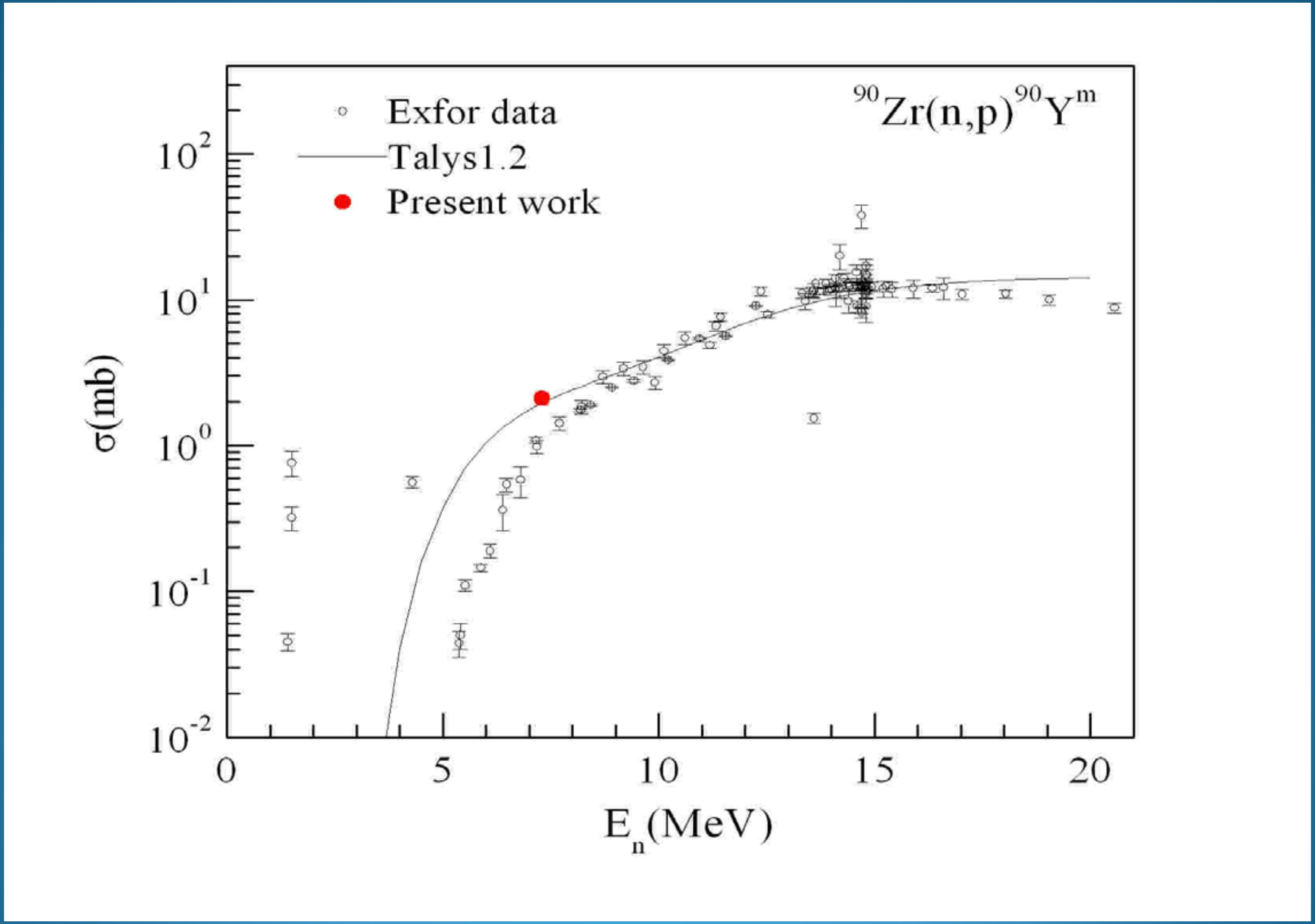


Fig. Plot of experimental and theoretical $^{90}Zr(n,\gamma)^{90}Y^{m}$ reaction cross-section as a function of neutron energy from 1 keV to 20 MeV from present work and from EXFOR in open circle.



0	Preliminary	For internal center use
1	NNDC (Brookhaven)	Neutron nuclear data
2	NEA-DB (Paris)	Neutron nuclear data
3	NDS (Vienna)	Neutron nuclear data
4	CJD (Obnisk)	Neutron nuclear data
9	NDS (Vienna)	Dictionary transmission
A	CAJaD (Moscow)	Charged-particle nuclear data
B	KaChaPag (Karlsruhe)	Charged-particle nuclear data
C	NNDC (Brookhaven)	Charged-particle nuclear data
D	NDS (Vienna)	Charged-particle nuclear data
E	JCPRG (Sapporo)	Charged-particle nuclear data
F	NPDC (Sarov)	Charged-particle nuclear data
G	NDS (Vienna)	Photonuclear data
H	NNDC (Brookhaven)	Special internal use for relativistic particle reaction data
L	NNDC (Brookhaven)	Photonuclear data
M	CDFE (Moscow)	Photonuclear data
N	NEA-DB (Paris)	Special use for memos only
O	NEA-DB (Paris)	Charged-particle nuclear data
P	NNDC (Brookhaven)	Charged-particle nuclear data from MacGowen file
Q	CJD (Obnisk)	Photonuclear data
R	RIKEN	Charged-particle nuclear data
S	CNDC	Charged-particle nuclear data
T	VNIIEF?NNDC	Charged-particle nuclear data
V	NDS (Vienna)	Special use for selected evaluated neutron data “VIEN” file.

3. List of successful Indian exfor entries into IAEA-EXFOR database

S. No	Entry No.	Title of the Paper	Authors	Reference
1	33020(n,f)	Fragment angular momenta in low and medium energy fission of 242-Pu	B.S.Tomar, A.Goswami, S.K.Das, T.Datta, B.K.Srivastava, A.G.C.Nair, Satya Prakash, M.V.Ramaniah	J,ZP/A,327,225,1987
2	33021	Effect of shell closure proximity on fragment angular momenta in 241-Pu(n,f)	S.P.Dange, H.Naik, T.Datta, R.Guin, Satya Prakash, M.V.Ramaniah	J,JRN,108,269,1987
3	33022	Fission fragment angular momentum in the spontaneous fission of 244-Cm	H.Naik, R.J.Singh and R.H.Iyer	J,RCA,92,1,2004
4	D6044(a,f)	Fragment angular momenta in low and medium energy fission of 242-Pu	B.S.Tomar, A.Goswami, S.K.Das, T.Datta, B.K.Srivastava, A.G.C.Nair, Satya Prakash, M.V.Ramaniah	J,ZP/A,327,225,1987
5	D6067	Large pre-equilibrium contribution in A + nat.Ni interactions at ~ 8-40 MeV	Abhishek Yadav, Pushpendra P.Singh, Manoj K.Sharma, Devendra P. Singh, Unnati, B.P.Singh, R.Prasad , M.M.Musthafa	J,PR/C,78,044606,2008
6	33023	Determination of the 233-Pa (n,f) reaction cross-section from 11.5 to 16.5 MeV neutron energy by the hybrid surrogate ratio approach	B.K.Nayak, A. Saxena, D.C.Biswas, E.T.Mirgule,B. V.John, S.Santra, R.P.Vind, R.K.Choudhrury, S.Ganesan	J,PR/C,78,061602,2008
7	D6075	Determination of the 233-Pa (n,f) reaction cross-section from 11.5 to 16.5 MeV neutron energy by the hybrid surrogate ratio approach	B.K.Nayak, A. Saxena, D.C.Biswas, E.T.Mirgule,B. V.John, S.Santra, R.P.Vind, R.K.Choudhrury, S.Ganesan	J,PR/C,78,061602,2008

Sr. No	Entry No.	Title of the Paper
8.	33003	Mass yields in 229-Th(n,f)
9.	D6007	Fission fragment anisotropies for the 13-C+235-U system at near-Coulomb barrier energies
10.	33011	Angular momentum of fission fragments in low energy fission of actinides
11.	33016	Single-particle spin effect on fission fragment angular momentum
12.	33017	Fission fragment angular momentum in ODD-Z fissioning systems
13.	33018	Systematics of fragment angular momentum in low-energy fission of actinides
14.	33019	Correlations of fission fragment angular momentum with collective and intrinsic degrees of freedom
15.	G0014	Post-neutron mass yield distribution and Photo-neutron cross-section measurements in 209-Bi with 65-MeV
16	D6016	Influence of incomplete fusion on complete fusion: Observation of a large incomplete fusion fraction at E~ 5-7 MeV/nucleon

1. Measurement of the neutron capture cross-section of 232-Th at 3.4 MeV and 7.3 MeV using the neutron activation technique communicated to **European Physical Journal A after peer-reviewed**.

2. ^{233}Pa(2n$_{th}$,f) cross-section determination using fission track technique **communicated to Physics Letters B.**

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