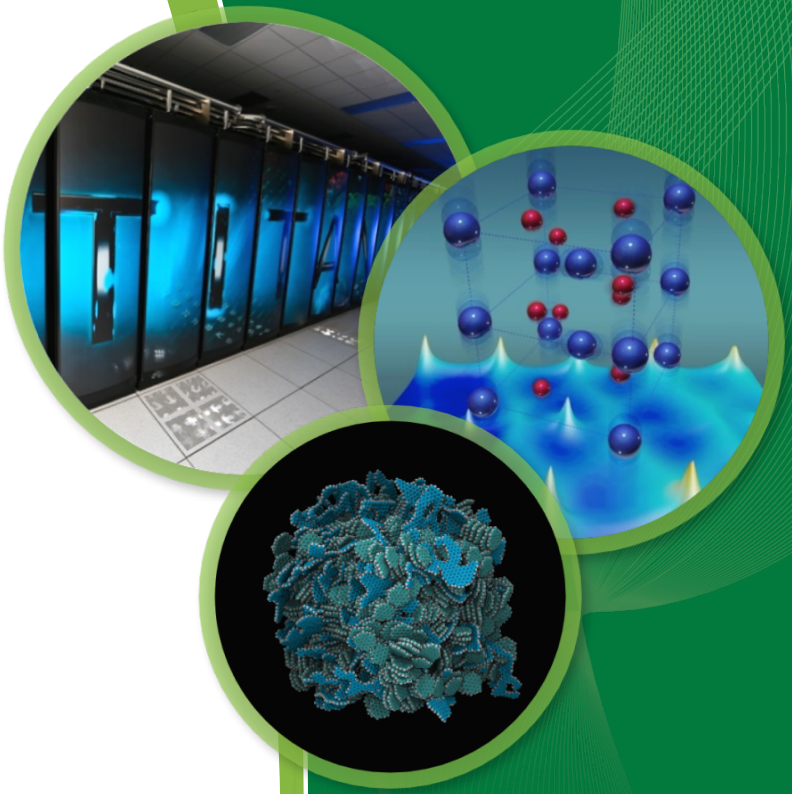
ORNL Neutron Cross Section Measurements in the Resolved Resonance Range for the US Nuclear Criticality Safety Program



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ORNL is managed by UT-Battelle
for the US Department of Energy

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Resonance region nuclear data work for NCSP

- Objective: Provide measured, evaluated resonance-region cross section data to address priority nuclear data needs for NCSP
- Vision: Address multiple nuclear data 5- and 10-year goals identified in the NCSP vision
- Final product: Rigorous ENDF/B resonance evaluations produced from cross section measurements (new/old) and analyses
- Last year's measurement work effort focused on vanadium and zirconium, which were differential nuclear data needs identified by NCSP Nuclear Data Advisory Group (NDAG)

NCSP Five-Year Plan

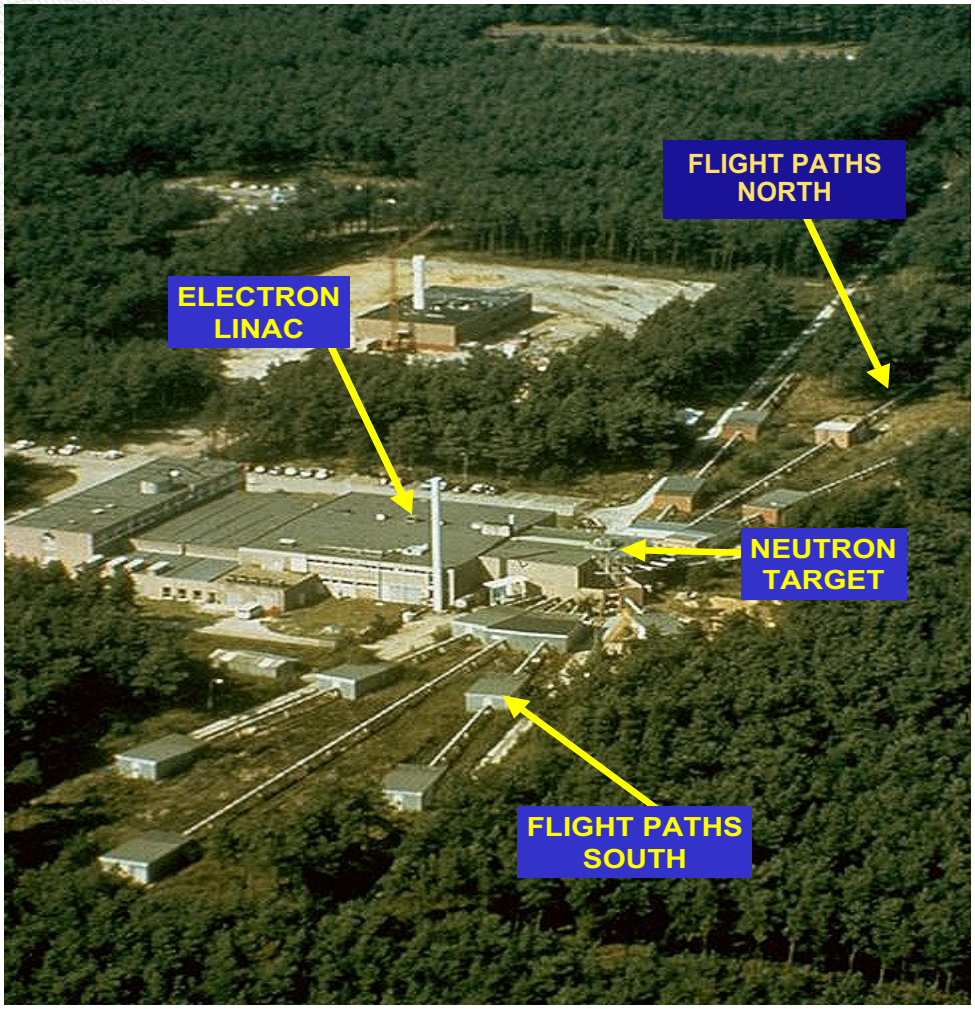
Nuclear Data

Priority Needs */ Additional Needs	Thermal scattering (BeO, HF, D_2O, SiO_2, CH_2, C_2F_4, $C_5O_2H_8$, etc.), ^{239}Pu, Cr, ^{237}Np, Pb, ^{55}Mn, Ti, ^{240}Pu / ^{233}U, Th, Be, ^{51}V, Zr, F, K, Ca, Mo, Na, La
Completed Evaluations (FY)	Minor Actinides (13), SiO_2 (12), ^{55}Mn (12), ^{180,128,183,184,186}W (14)

	Materials	Pre FY2015	FY2015	FY2016	FY2017	FY2018	FY2019	FY2020	FY2021	Post-FY2021
Measurements	Calcium (Ca)									
	Cerium (Ce)									
	Copper (Cu)									
	Iron (Fe)									
	Lucite ($C_5O_2H_8$)									
	Tantalum (Ta)									
	Strontium (Sr)									
	Tungsten (W)									
	Vanadium (V)									
	Zirconium (Zr)									
	Polyethylene (CH_2)	H_2O / CH_2								
	Materials	Pre FY2015	FY2015	FY2016	FY2017	FY2018	FY2019	FY2020	FY2021	Post-FY2021
Complete Evaluations	Calcium (Ca)									
	Cerium (Ce)									
	Cobalt (Co)									
	Copper (Cu)									
	Dysprosium (Dy)									
	Gadolinium (Gd)									
	Iron (Fe)									
	Lead (Pb)									
	Nickel (Ni)									
	Oxygen (O)									
	Rhodium (Rh)									
	Plutonium-239									
	Tantalum (Ta)									
	Strontium (Sr)									
	Tungsten (W)									
	Uranium-235									
	Uranium-238									
	Vanadium (V)									
	Zirconium (Zr)									
	Hydrofluoric Acid									
	Lucite ($C_5O_2H_8$)									
	Polyethylene (CH_2)									
		ORNL		RPI		LANL		LLNL/NCSU		

- Requests for additional IE measurements: Ni, Mo, Cr (Fe-Cr alloys), Mn in intermediate energy range (VNIITF, NCERC).
- Request for measurements and evaluation of angular distributions at high energy for Cu.
- Continuing need for thermal scattering data.

*Note: work has been completed for some priority needs (e.g., ^{55}Mn, Ti, and Cr), and these isotopes/nuclides are maintained on the list for reference. Furthermore, the table represents the list of materials that can be addressed during the next five years under the current budget target. The additional priority needs will be addressed beyond the next five years.



Pulse width	: 1ns
Frequency	: 40–800 Hz
Average current	: 4.7–75 μA
Neutron intensity	:1.6 10^{12}–2.5 10^{13} n/s

GELINA

- Time-of-flight facility
- Pulsed white neutron source

  (10 meV < E_n < 20 MeV)
- Multi-user facility with 10 flight paths (10–400 m)
- The measurement stations have special equipment to perform the following:
  - Total cross section measurements
  - Partial cross section measurements

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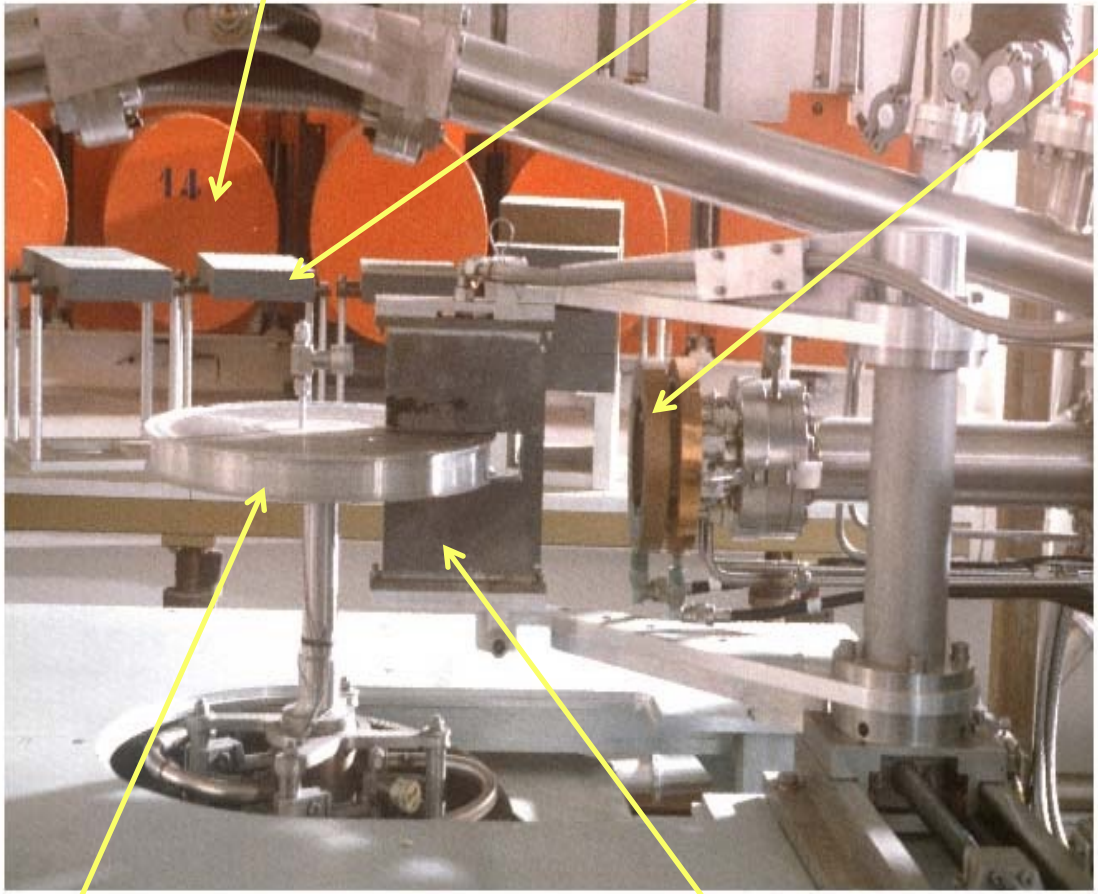
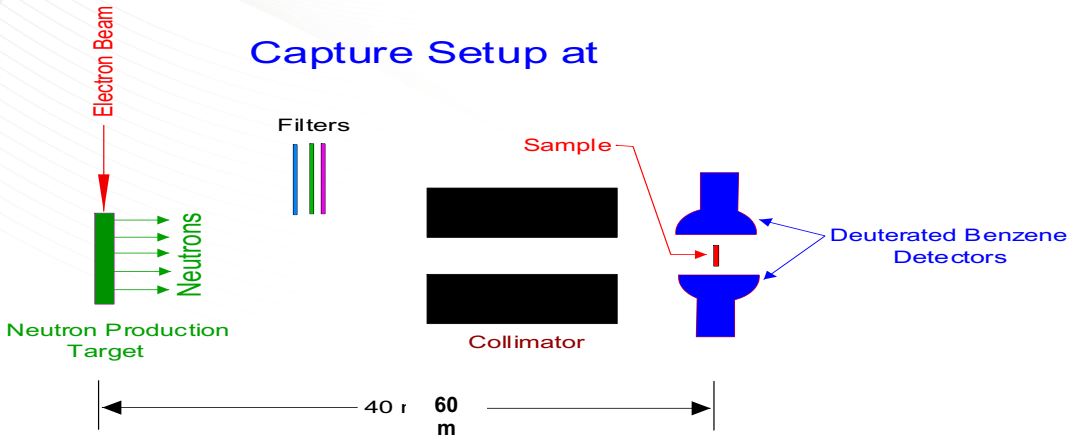
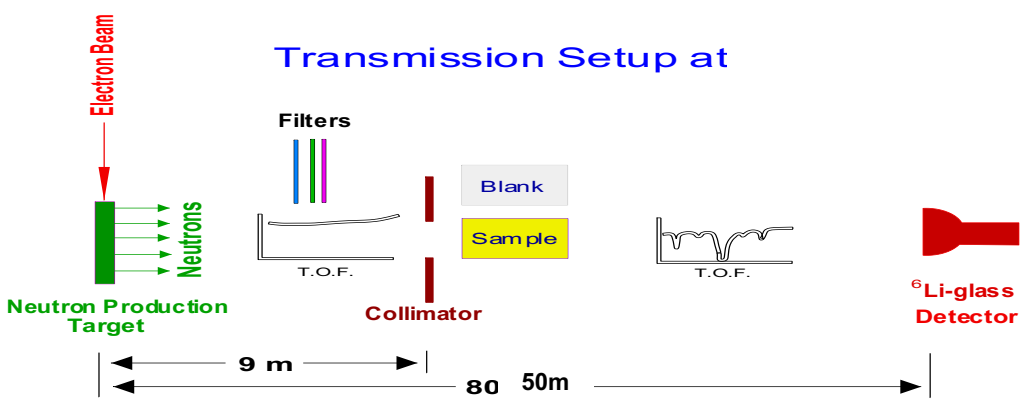
Neutron production



- e^- accelerated to $E_{e-,max} \approx 140$ MeV
- (e^-, γ) Bremsstrahlung in U-target (rotating and cooled with liquid Hg)
- (γ,n) , (γ ,f) in U-target
- Low energy neutrons by water moderator in Be-canning

Neutron capture and total cross section experiments at a white neutron source





- Time-of-flight technique used to determine incident neutron energy; "clocks" used have typically 1nsec resolution
- Pulsed electron beam of accelerator starts the clock; γ-ray or neutron detector stops the clock
  - $v_n = L/t$
  - $E_n = m_n v^2/2$
- Filters reduce frame-overlap background from low-energy neutrons, reduce γ-flash effects, and determine background

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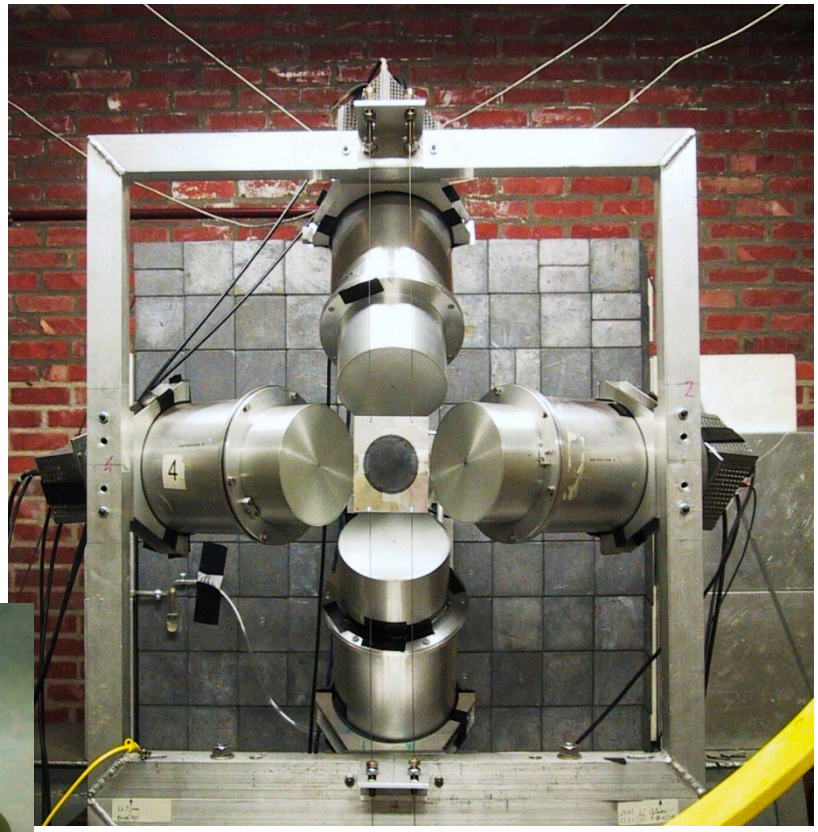
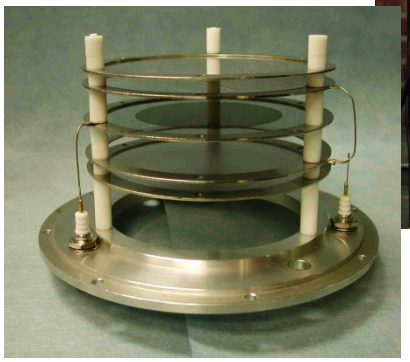
Capture cross section measurements at GELINA

Total energy detection principle

- C_6D_6 liquid scintillators
  - 125°
  - Pulse height weighting technique (PHWT)
  - Weighting function (WF) from Monte Carlo simulations
- Flux measurements (IC)
  - $^{10}B(n,\alpha)$
  - $^{235}U(n,f)$

L = 10 m, 30 m, and 60 m





$$Y_{exp} = N\sigma_{\varphi}\frac{C_w - B_w}{C_{\varphi} - B_{\varphi}}$$

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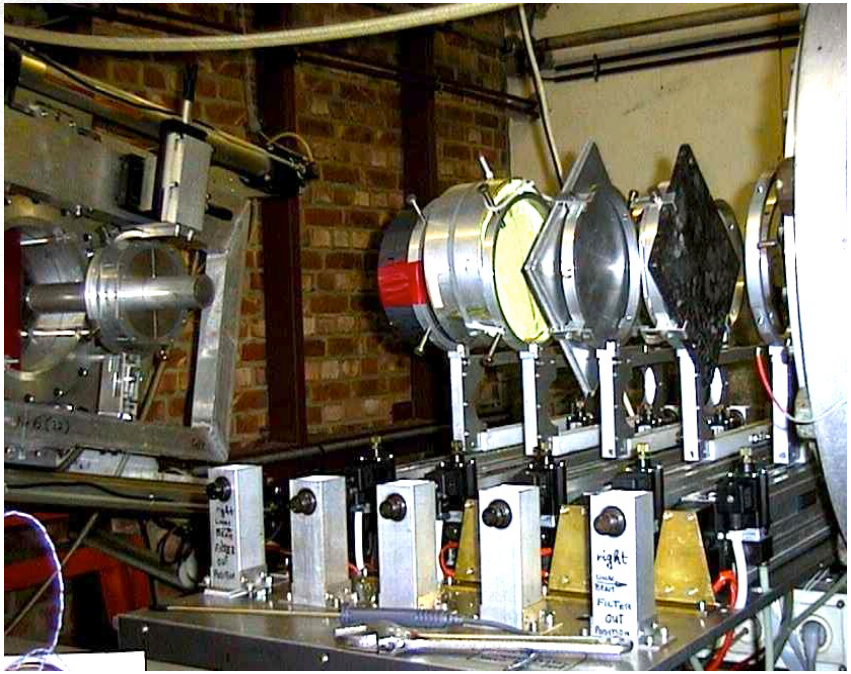
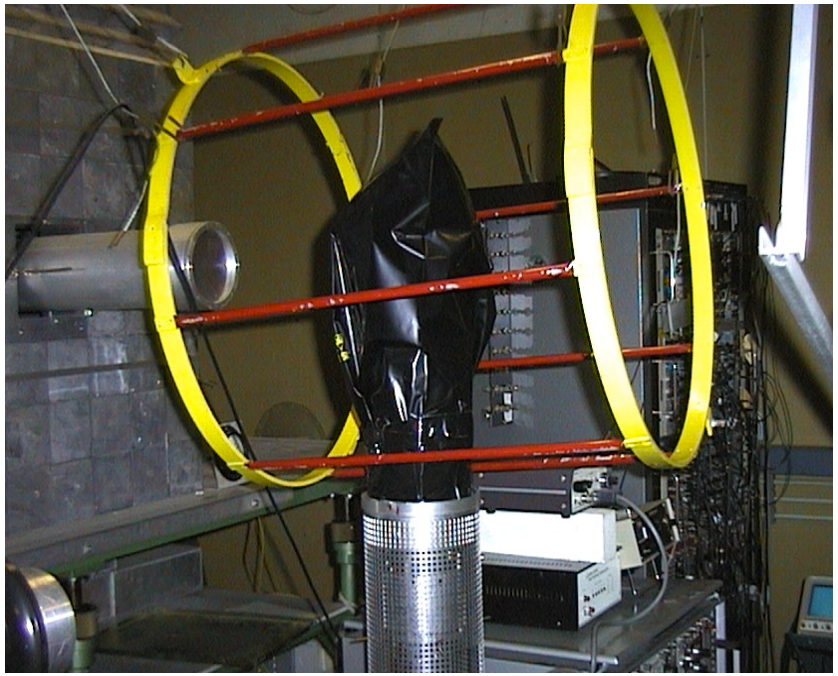
Total cross section/transmission measurements

Sample and background filters



Detector



Detector stations
Moderated: L= 30 m,50 m,(100 m,200 m)
Fast: L= 400 m

Low energy : ^{6}Li(n,t)α Li-glass

High energy : H(n,n)H Plastic scintillator

$$T = \frac{C_{in}}{C_{out}} \cong e^{-n\sigma_{tot}}$$

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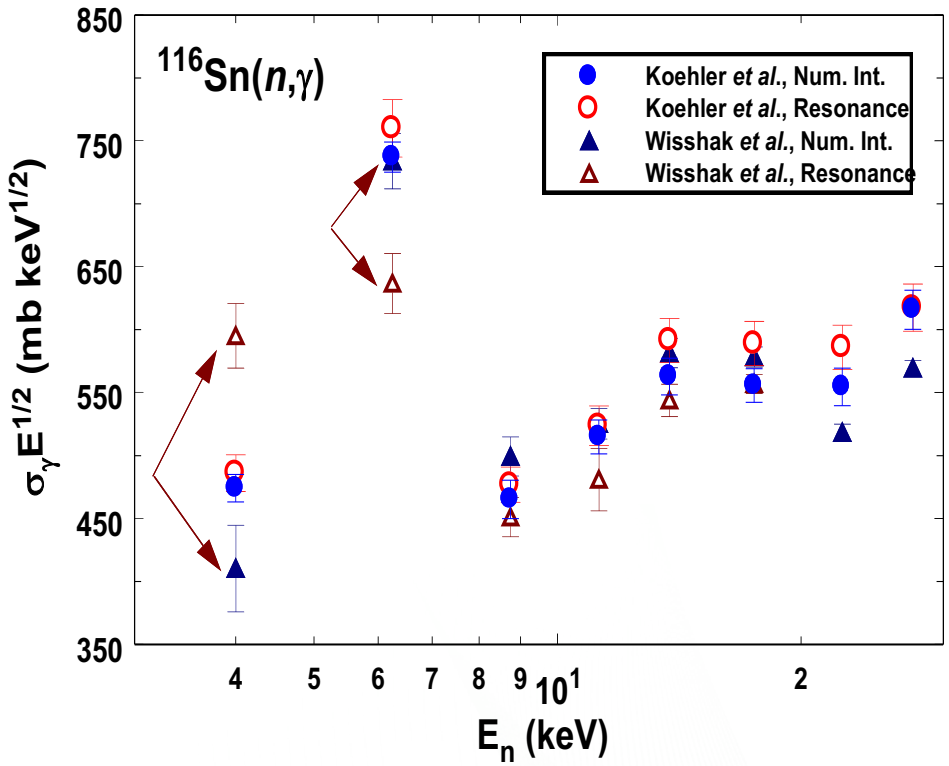
Why are total cross section (transmission) data measured?

- The data are indispensable for obtaining the most accurate (n,γ) reaction rates
  - The transmission data allows to see resonances that are not very visible in (n,γ) data and vise versa
  - Since experiments are not performed with infinite small samples, corrections for self-shielding and multiple scattering are needed
- Lack of good total cross section data can lead to serious errors in these corrections and hence in the (n,γ) cross sections
- More complete resonance parameter data set will help improve nuclear statistical model calculations



Example: ^{116}Sn use of incorrect neutron widths led to incorrect low-energy (n,γ) cross sections (Wisshak et al., PRC 54, 2732, 1996)

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Experiment for Cerium 142 I

- An isotopic enriched sample of 142-CeO_2 is needed (~30 g) from ORNL isotopes
- The sample is too expensive for purchasing ($22/mg); lease from ORNL isotopes is an alternative
- However, new DOE lease policy requires NO RAD ADDED, i.e. <0.6Bq/g
- The ORNL isotopes procedure is as follows:
  - For the activity check, a baseline is established with 30mg of material before the experiments
  - After returning the sample, 30mg from the sample are used to check for activation
  - The material (60mg) used must be purchased due to destructive measurements technique
  - The measurement is to check for $\alpha-$,$\beta-$ and $\gamma-$ activity
- If radiation is detected, the material must be purchased
- ORNL's ND group approach is to test and simulate the irradiation

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Experiments for Cerium 142 II

- 60mg of Cerium 142 were purchased from ORNL Isotopes
- A baseline of 30mg was established before irradiation
- The irradiation of 30mg at RPI at a 10m flight path with a flux 7E5 n/sec/cm^2 was performed for about one hour
- The total number of neutrons was similar to that found in an experiment at GELINA
- The irradiated sample was shipped back for analysis

Oak Ridge National Laboratory
Chemical Sciences Division
Radioactive Materials Analysis Laboratory

Results of Analyses for:
RMAL16110

Approved By
04/07/2016

Customer Name: Hickman, E. C.
Charge Number: 3687ANCH
Date Received: 3/1/2016 9:30:00
Project ID:
Specimen Type:

Lab Sample ID / Analysis	Customer Sample ID	Result		Uncertainty	Date/Time Sampled / Units	Filtered?	Matrix / Procedure No.
RMAL16110-001	177590				3/1/2016 7:00:00	Unfiltered	
Sample is NO-RAD by SBMS protocol with LD-Alpha = 2.5E-01 Bq/mL, LD-Beta = 5.0E-01 Bq/mL, Z-Alpha = 0.4107, Z-Beta = -5.482.							
G-Alp/Bet	G-Alpha	2.7E-01	+/-	1.1E-01	Bq/mL		CSD-EPA-900.0
	G-Beta	1.1E-01	+/-	1.4E-01	Bq/mL		
G-Scan	K-40	3.2E+01	+/-	2.0E+01	Bq/g		CSD-EPA-901.1
RMAL16110-002	177590-R				3/1/2016 7:00:00	Unfiltered	
Sample is NO-RAD by SBMS protocol with LD-Alpha = 2.5E-01 Bq/mL, LD-Beta = 5.0E-01 Bq/mL, Z-Alpha = -1.905, Z-Beta = -2.268.							
G-Alp/Bet	G-Alpha	1.6E-01	+/-	9.0E-02	Bq/mL		CSD-EPA-900.0
	G-Beta	3.2E-01	+/-	1.6E-01	Bq/mL		
G-Scan	K-40	2.4E+01	+/-	2.0E+01	Bq/g		CSD-EPA-901.1

End of data for Request Number : RMAL16110

No rad added

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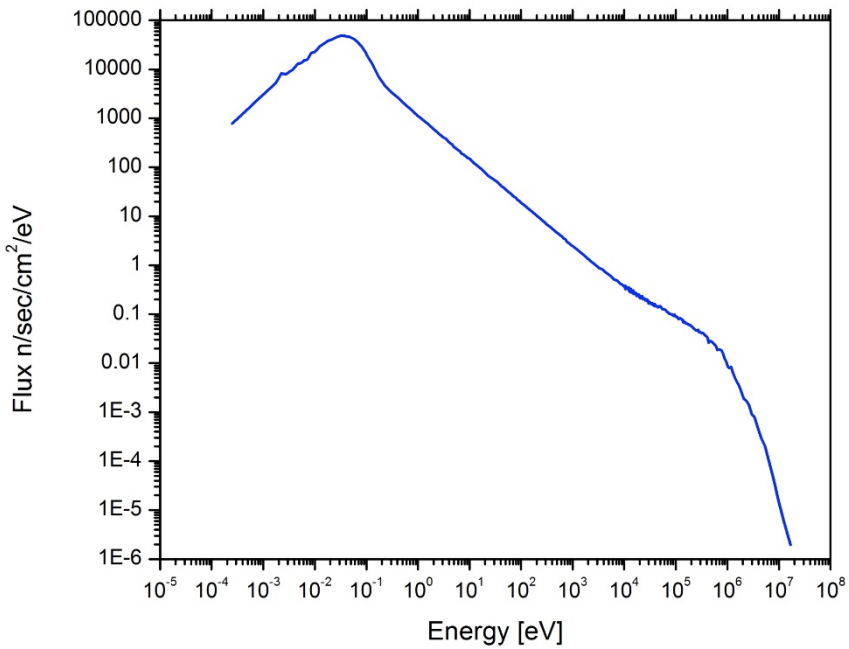
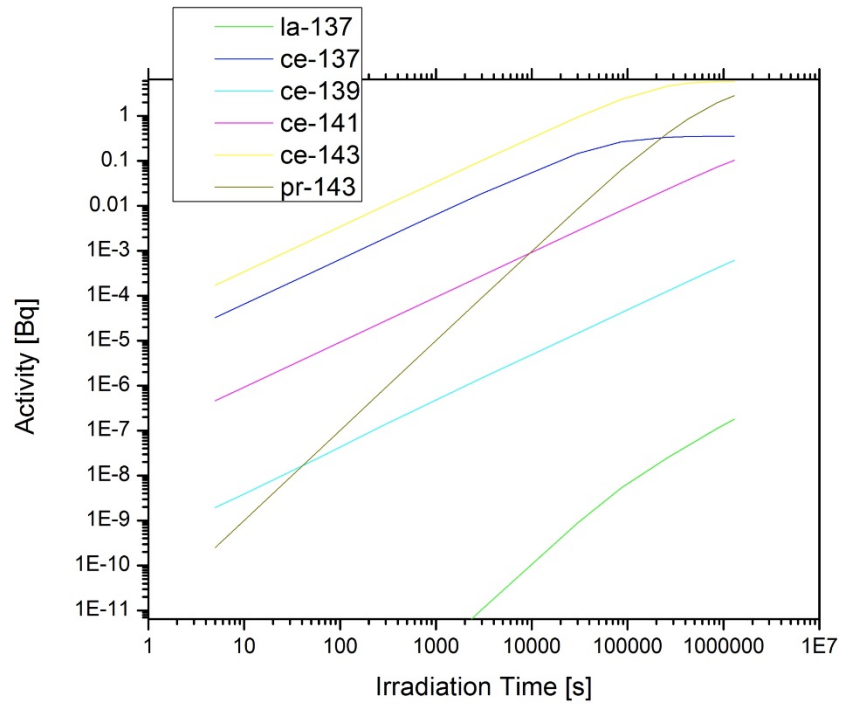
Experiments for Cerium 142 III

- A SCALE calculation was performed with the neutron flux profile from GELINA at the 60m flight station
- SCALE calculates all interactions and reactions of the neutron with the sample; output is the corresponding activity
- Irradiation time is 15 days for a typical experiment





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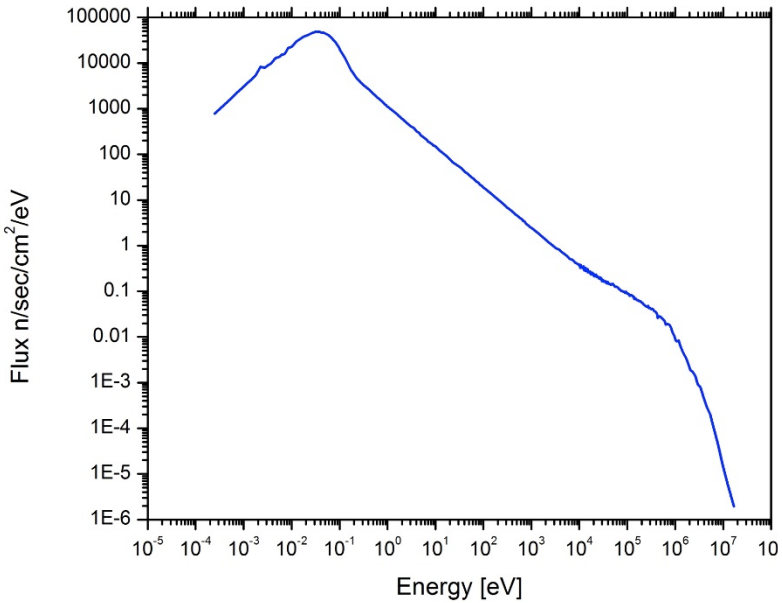
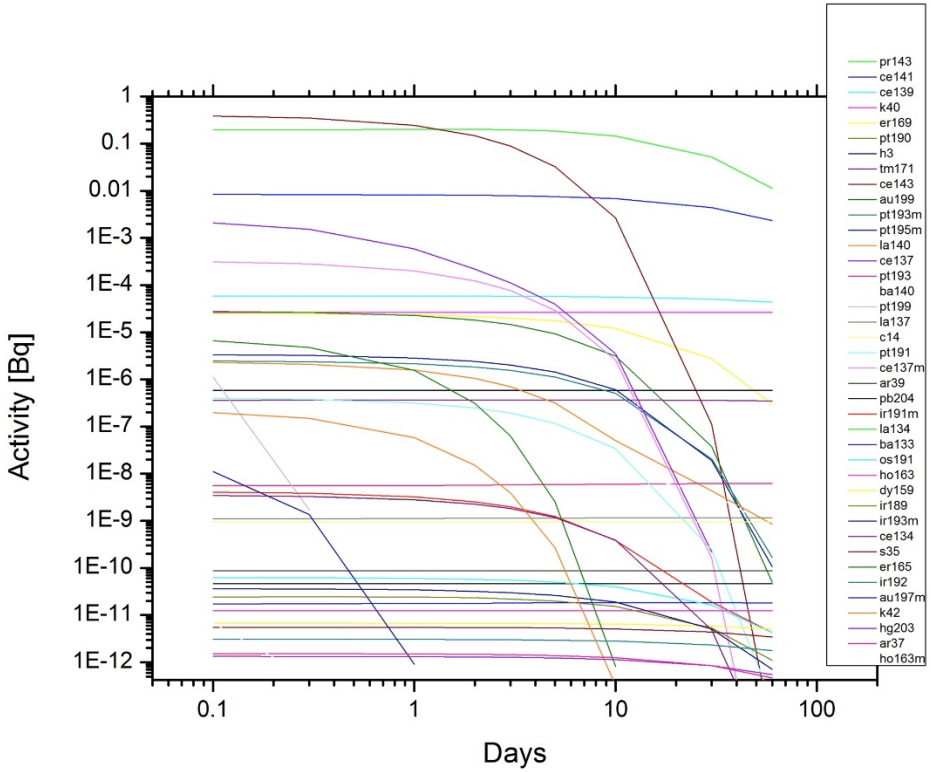
Experiments for Cerium 142 IV

- An ORIGEN calculation was performed with the neutron flux profile from GELINA at the 60m flight station
- ORIGEN calculates all possible reactions of the neutron with the sample and subsequent decay with time
- Irradiation time is 15 days for a typical experiment





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ORNL measurement activities for vanadium

- ^{51}V has a natural abundance of 99.75% and is nonreactive in air
- Measurements were made using metallic samples of different thicknesses
- Transmission experiments with different samples are performed using FP4, 50m station
- Neutron capture at FP14 is 60m
- Experiments were performed with different background filter combinations
- Capture and transmission experiments were performed in 2015/2016 and need to be finalized

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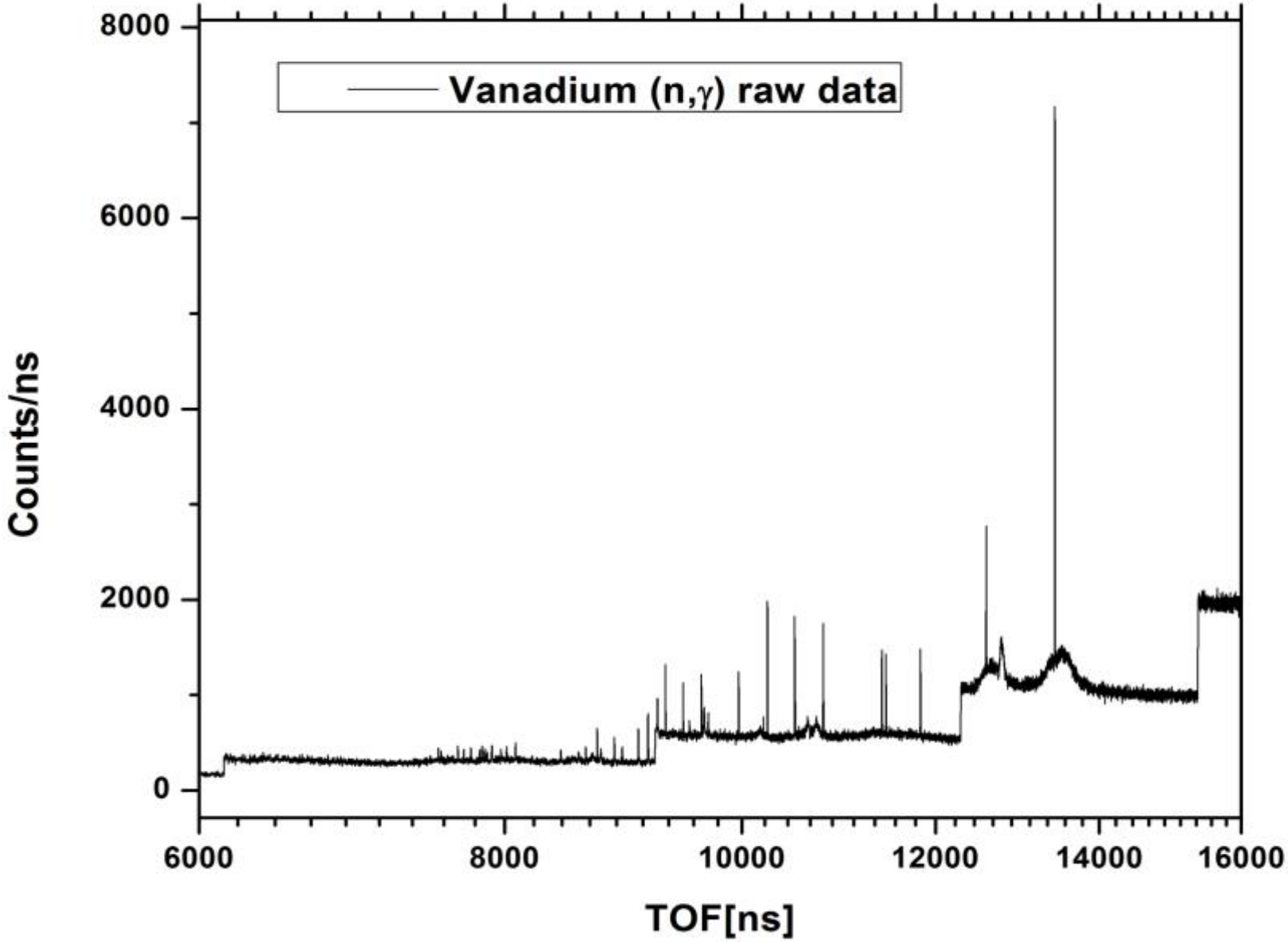
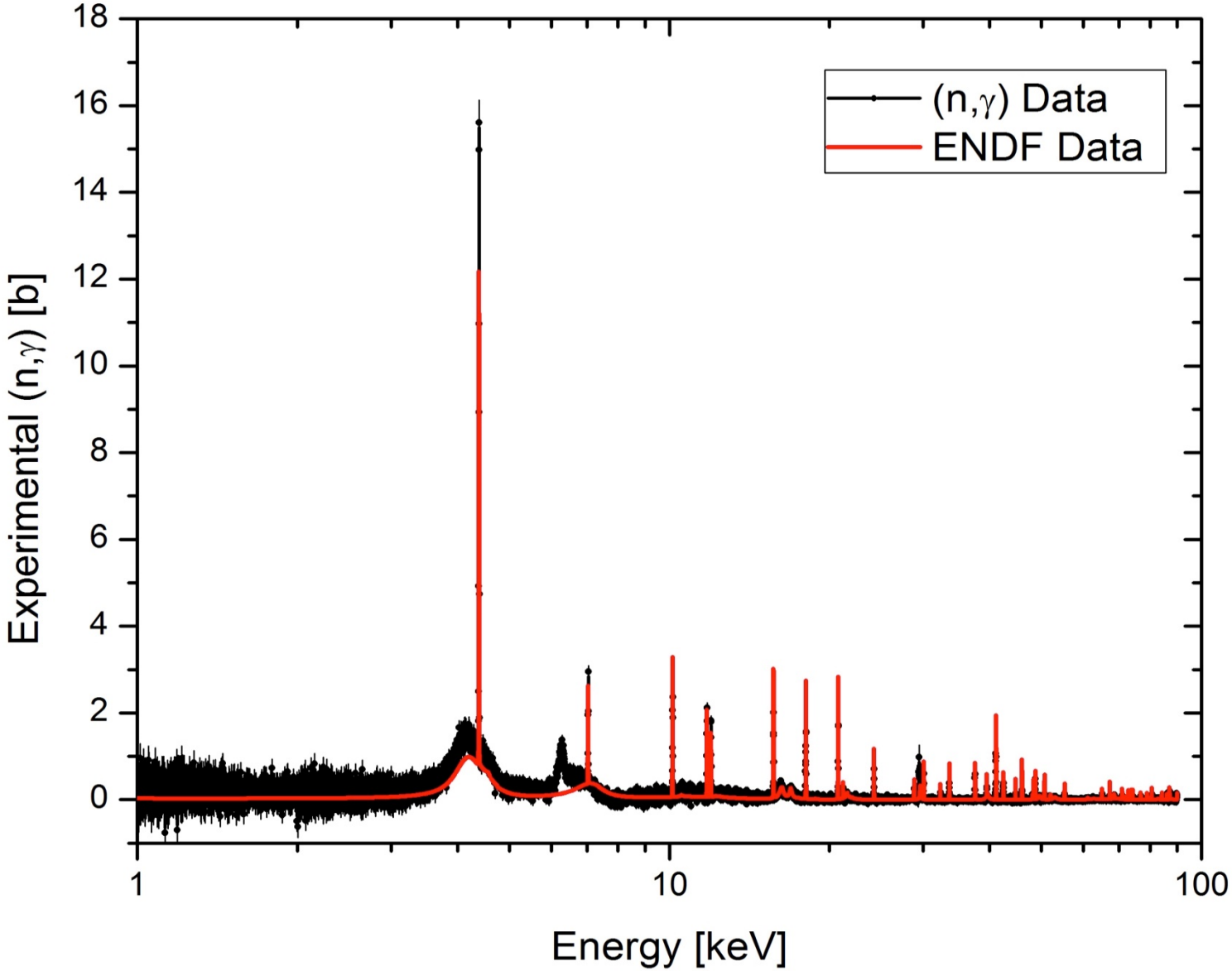
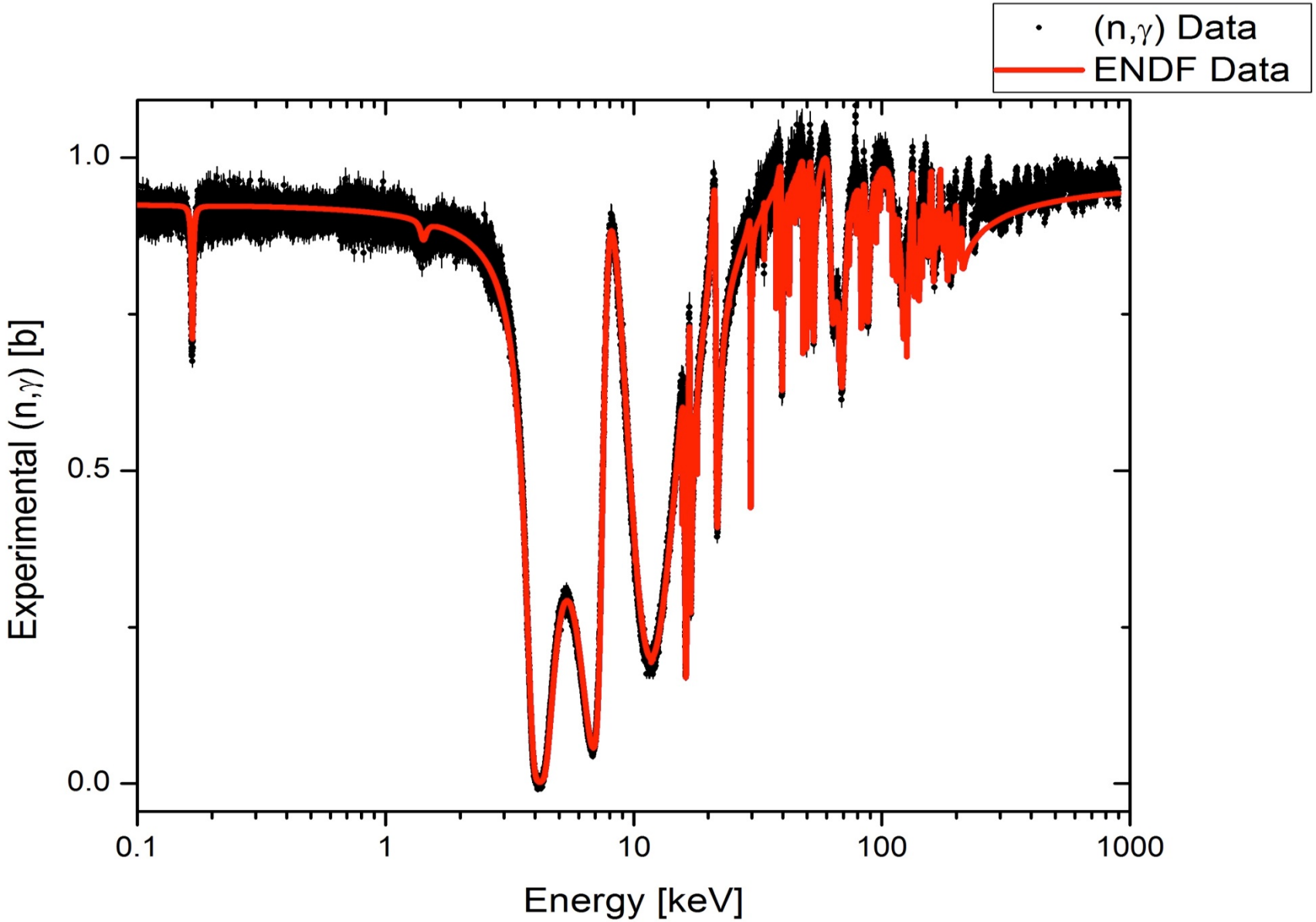
V (n,γ) raw data for thin sample



(n,γ) of 0.35mm vanadium sample



Transmission of 2mm vanadium sample



ORNL measurement activities for zirconium

- Zr has 5 stable isotopes; at least 4 major isotopes should be measured
- Good high resolution ORNL total cross section data for the separated isotopes are available, even for the long-lived radioactive fission product ^{93}Zr
- Data were retrieved from the Jack Harvey archive
- Therefore, only neutron capture experiments must be performed
- The new DOE lease policy might present a problem, especially regarding no-RAD-added with the ^{93}Zr half-life of 1.5 E6a

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ORNL measurement activities for Zr

- The current ORNL approach is to start with natural Zr neutron capture and transmission using metallic samples of different thicknesses
- The combination of the natural sample capture data with the total cross section of the separated isotopes will help to obtain already parameters for the strong capture resonances
- A natural sample is always a good check for the performance of the resonance parameters obtained from the separated isotopes experiments

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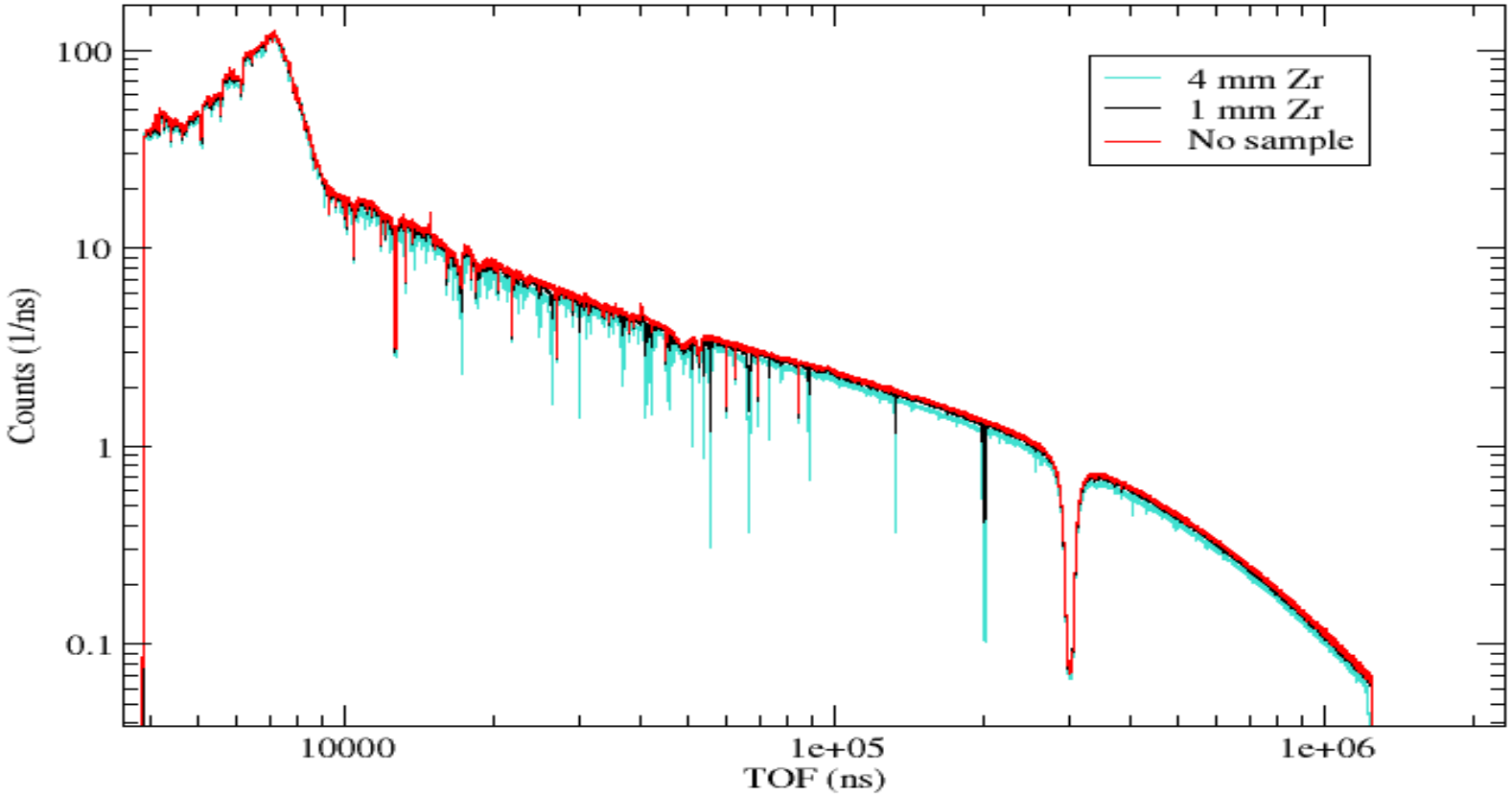
ORNL measurement activities for Zr (continued)



- Transmission experiments with different Zr samples are performed using the FP4, 50 m station
- Experiments are performed using different background filter combinations

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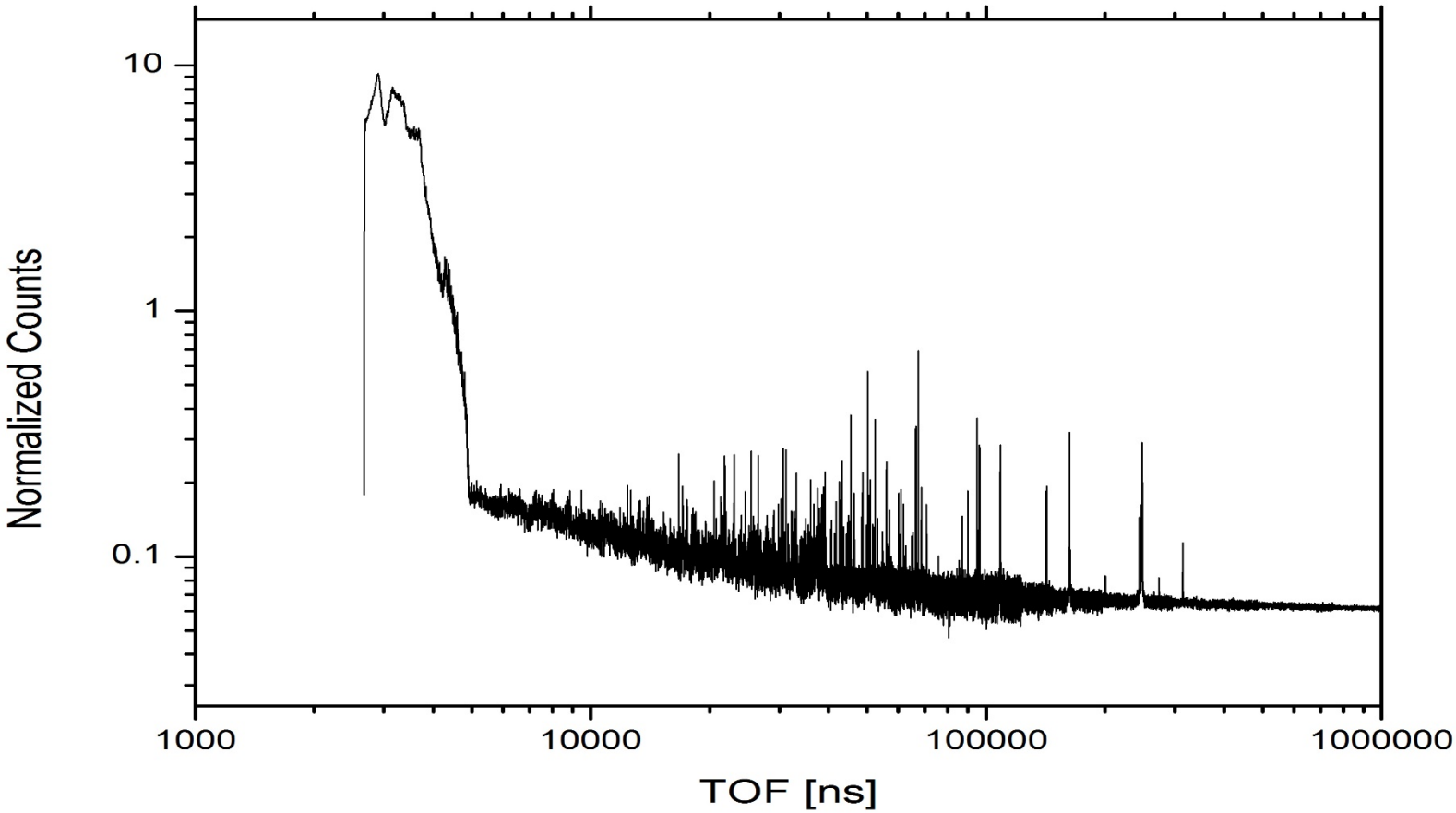
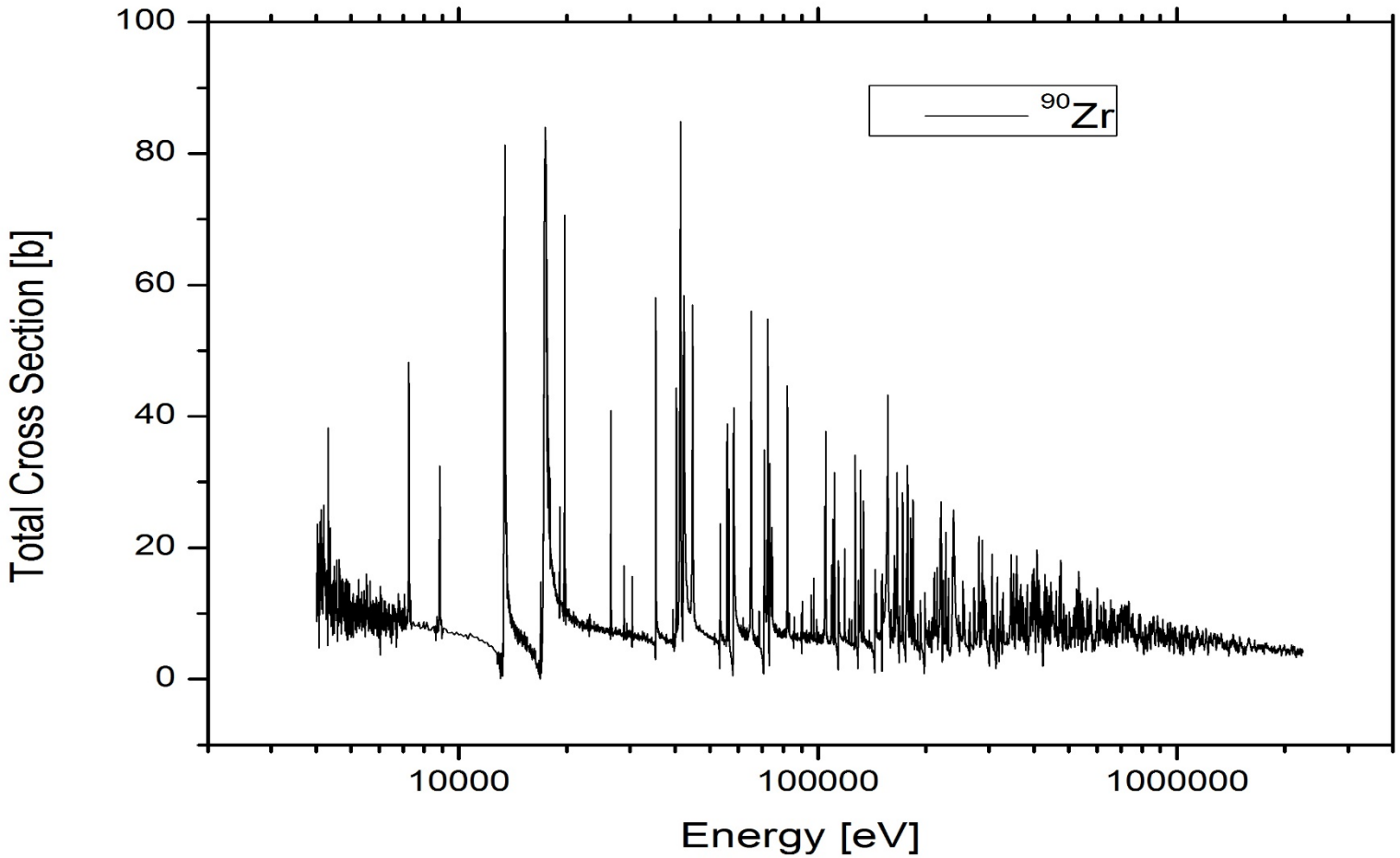
ORNL measurement activities for Zr (continued)



Neutron capture at FP14, 60 m

Old ORNL ^{90}Zr data



Total cross section of a metallic ^{90}Zr sample

Resolved resonances are visible above 1 MeV at a factor of 3 more than previous experiments

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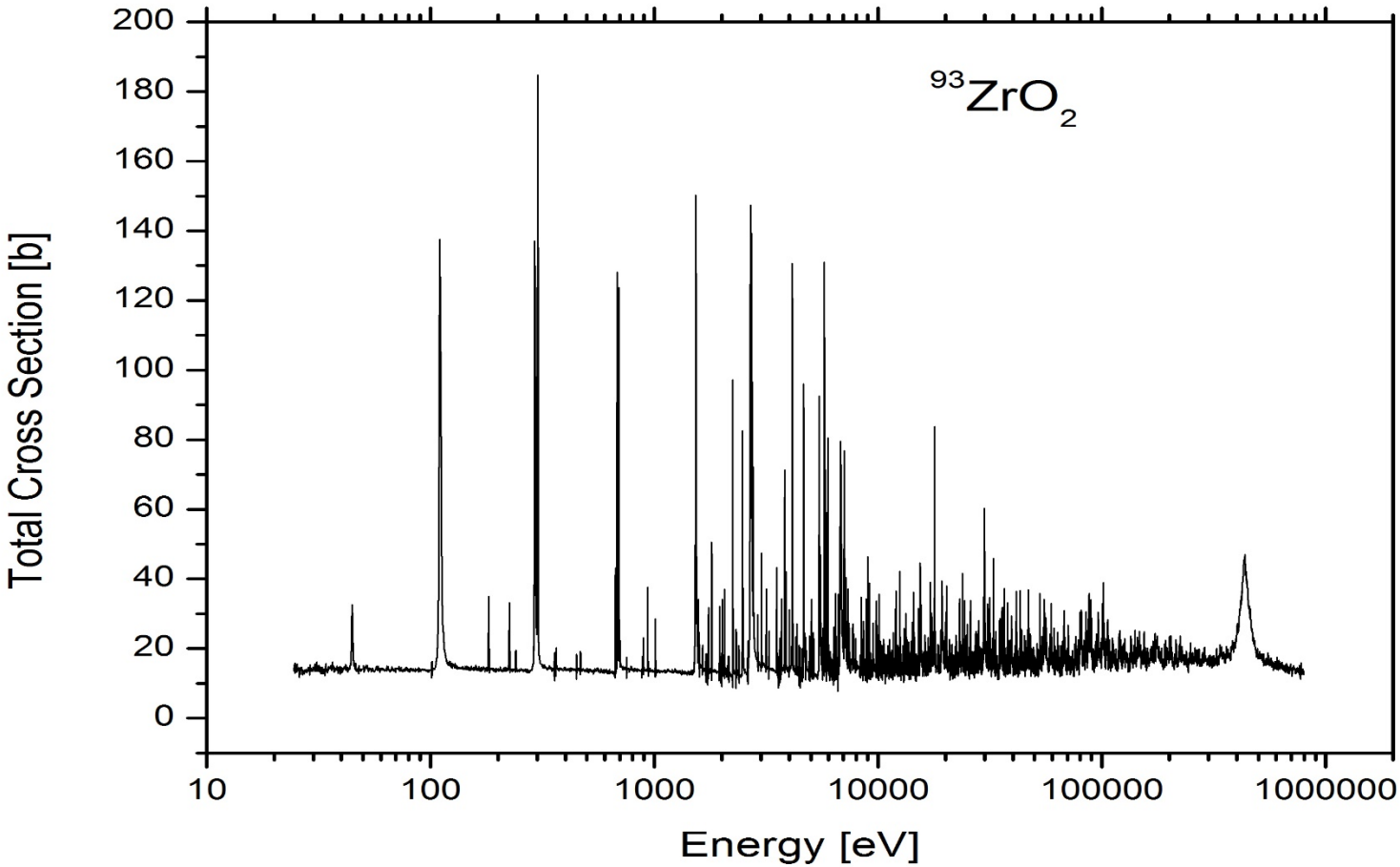
Old ORNL ^{93}Zr data



Total cross section of $^{93}ZrO_2$ resolved resonances well above 100 keV

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Status of NCSP experiments at EC-JRC Geel

	W	Cu	Ca	Ce	V	Zr
Sample	metallic disks 182,183,184,186 2009–2011	metallic disks 63 and 65 2011–2012	metallic disks nat Ca 2013–2014	metallic disks Nat Ce, Ce-142 2014–2015	metallic disks 2015–2016	Nat Zr metallic disks ^{90,91,92,94}Zr 2016
Experiments GELINA	60m, 30m (n,γ) transmission	60m (n,γ)	60m (n,γ) transmission	Nat Ce 60m (n,γ) Nat Ce transmission ^{142}Ce sample problems	60m (n,γ) transmission	Nat Zr 60m (n,γ) + transmission started ^{90,91,92,94}Zr sample problems
Data sorting	finished 60m + transmission	finished 60m	finished 60m transmission	finished for thin and thick sample	finished for thin and thick sample	
Reduced to cross section	X-section, transmission	X-section	X-section transmission 0.6, 1.0, 5 cm samples	2mm X-section 2mm transmission 10mm transmission	thin X-section 0.35 and 2mm transmission	
Data testing	Data ready for evaluation	Data ready for evaluation	Data ready for evaluation	In progress	In progress	
Analysis and evaluation	Finalized and submitted to NNDC	Finalized and submitted to NNDC	Finalizing and submitted to NNDC	Started		

People involved in the experiments

- Peter Schillebeeckx, EC-JRC Geel
- Carlos Paradela, EC-JRC Geel
- Stefan Kopecky, EC-JRC Geel
- Peter Siegler, EC-JRC Geel
- Ruud Wynats, EC-JRC Geel
- Clint Ausmus, Ian Gauld, Cihangir Celik, ORNL
- Greg Leinweber, Yaron Danon, RPI

People involved in the evaluations

- Marco Pigni, ORNL
- Vlad Sobes, ORNL
- Luiz Leal, IRSN

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Background determination in transmission using black resonance filters