International Atomic Energy Agency

EXFOR Innovative Nuclear Data: response to Safeguards Needs and new Measuring Techniques

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Scope

I. Following up: what NRDC did last 2-3 years for the Safeguards relevant nuclear data:

- Neutron Multiplicities
- Nuclear Resonance Fluorescence

II. Rare or innovative data for EXFOR

- Internal Conversion Electrons (ICE) from (n,n') and α-decay
- Beta and Antineutrino Spectra from induced fission

III. Summary

I. Following up: Neutron Multiplicities used to measure the amount of fission material by coincidence technique

Several EXFOR Entries were updated to include missed data on Neutron Multiplicity:

- **12833, 12834, 12906**
  R.Gwin, *NSE 87(1984)381*
  ^{233,35}U(n,f), ^{239}Pu(n,f), ^{252}Cf(s.f.)

- **30772**
  J.Boldeman, *NSE 91(1985)114*
  ^{233,35}U(n,f), ^{239,40,41,42}Pu(n,f), ^{252}Cf(s.f.)

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REACTION    (92-U-233(N,F),NUM,NU,,AV)
ERR-ANALYS  (DATA-ERR) No information on source of uncertainty
            (ERR-1) Uncertainty due to neutron detection efficiency
            (ERR-2) Uncertainty due to pileup parameter
COVARIANCE  Correlation coefficients between multiplicities
            100
            -45
             19 -58 100
             -8  16 -71 100
             -3 -13  18 -62 100
             -1  -2  -8  23 -56 100
              0  -1   0  -5  22 -58 100
              0   0   0   1  -5  22 -59 100
EN-MIN       EN-MAX       ERR-1        ERR-2
EV           EV           PER-CENT     PER-CENT
.02          .1           .15          3.8
PART-OUT     DATA         DATA-ERR
NO-DIM       NO-DIM       NO-DIM
0            .0244        .0013
1            .15          .0034
2            .3334        .0051
3            .324         .0052
4            .1392        .0038
5            .0263        .0019
6            .0025        .0009
7            .0002        .0002
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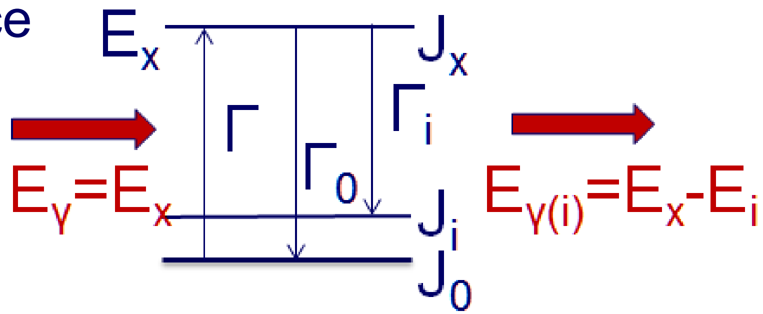
I. Following up: Nuclear Resonance Fluorescence (NRF) data

- NRF = photon' excitation of specific resonance **(scissor dipole mode)** and consequent decay by prompt γ-ray emission to ground or excited states



- firstly observed for actinides ^{238}U and ^{232}Th by R. Heil et al., NP A476(1988)39

- "exploding" number of publications in last years due to potential use in the non-destructive analysis of clandestine nuclear, toxic and explosive materials

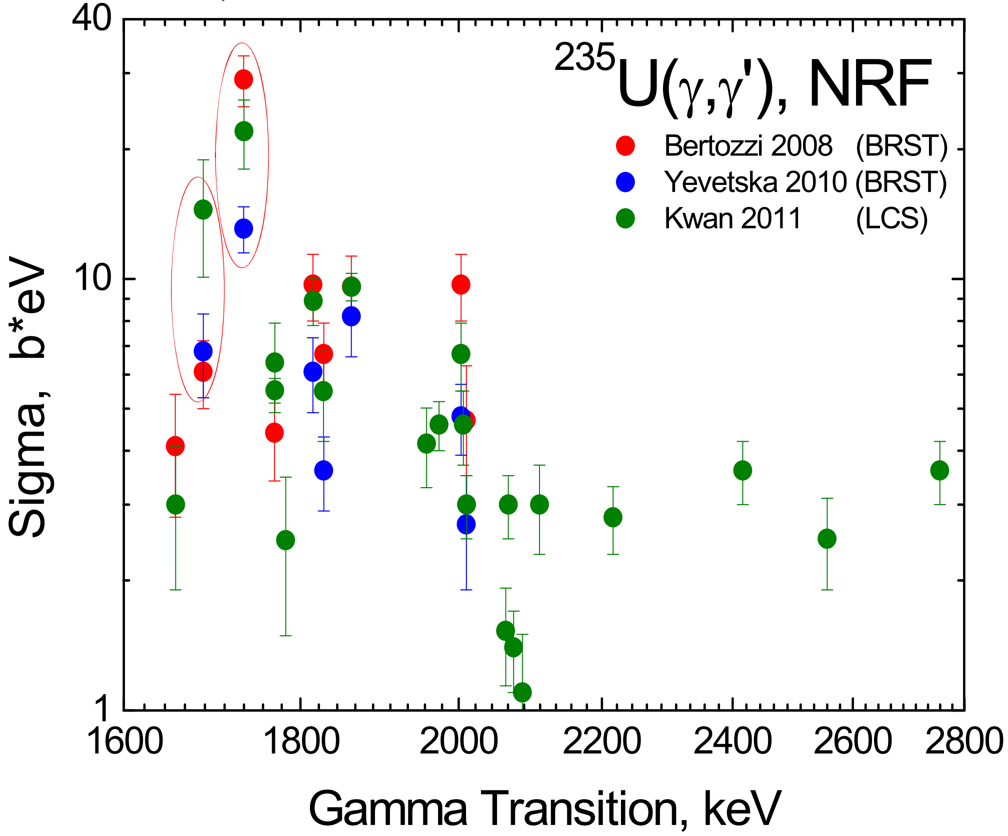
- up today 10 experiments were carried out and published

- **NDS compiled all known NRF data in EXFOR** following Memo CP-D/703 "Compilation of nuclear resonance fluorescence (NRF) data"

I. Following up: NRF experimental (EXFOR) and evaluated data

2012: all measured data are in EXFOR

S. Simakov, N. Otsuka, V. Semkova, V. Zerkin, S. Hlavac, "Experimental data for nuclear resonance fluorescence ", INMM-52, 2011



2013: Evaluated data & MC code become available

Monte-Carlo transport code COG 11.1 Beta 2 (LLNL):

which treats now NRF and has special NRF library:
COGNRF by J. Hall

(*available as package CCC-777 from RSICC since 2013*)

- the individual experiment uncertainties (even for strongest lines) ~15%, however the scatting of measurements is often larger (up to factor ~ 2)
- EXFOR promptly responds to actual nuclear data needs for Safeguards

II. Conversion electrons – measurement of Cross Sections

G. Belier, V. Meot et al. "Integral Measurement of the ^{235M}U isomer by neutron inelastic scattering"

presented at workshop:
"Experimental and theoretical problems around actinides for future reactors",
17-19 Mar 2014, CEA Saclay, http://esnt.cea.fr/Phocea/Page/index.php?id=37

Brief details of Experiment:

- irradiation of ^{235}U sample in CALIBAN, shot 60 μs, fast spectrum <E> = 1.44 MeV
- (n,n') excites 76.8 eV isomeric state of in ^{235}U ($T_{1/2}$ = 26 min), which de-excites by full conversion ($\alpha_{IEC} >> 1$) in 40-60 eV electrons $\sigma_{U(n,n')} \sim Y_{IEC} (1 + 1/\alpha_{IEC})$
- chemical separation and measurement of decay curve using detector of **electrons**
- **Result:** measured spectrum-averaged cross section **<σ> = 1.00 +/- 0.13 b**

Data, when published, will be compiled in EXFOR (*following the current rules*):

ENTRY XXXXX.YYY	*similar to* **21918.006**	*(4π gas detector of ICE was used)*
REACTION (92-U-235(N,INL)92-U-235-M,,SIG,,FIS)	**(49-IN-115(N,2N)49-IN-114-M,,SIG)**	
DECAY-DATA (92-U-235-M,26MIN,ICE, *Energy, Intensity*)	**(49-IN-114-M,49.51D,DG,190.,0157,ICE,,0.051)**	

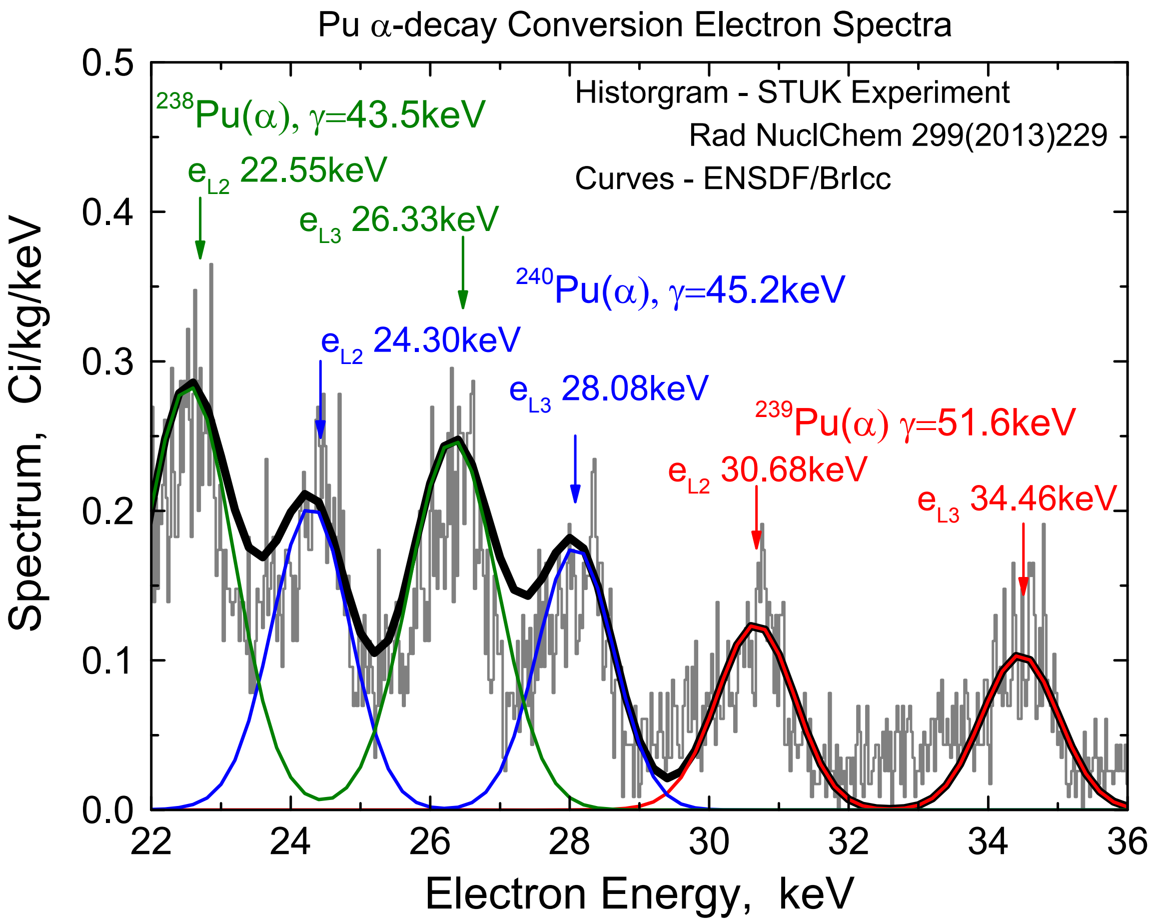
- What is an actual status of reference ICE data used for XS determination ? –> *next slide*
- Question to Experimentalists: is it feasible to use ICE detection technique for measuring prompt (not delayed) ICE, e.g. for poor known (n,n'$_i$) reactions on actinides

II. Internal conversion electrons (ICE): spectra from α-decay

- passive assay method in environmental surveillance and nuclear safeguards

ICE spectra from ^{239}Pu enriched sample:

experimental data from
K. Perajarvi et al., Radioanal. Nucl. Chem. 299(2014)229



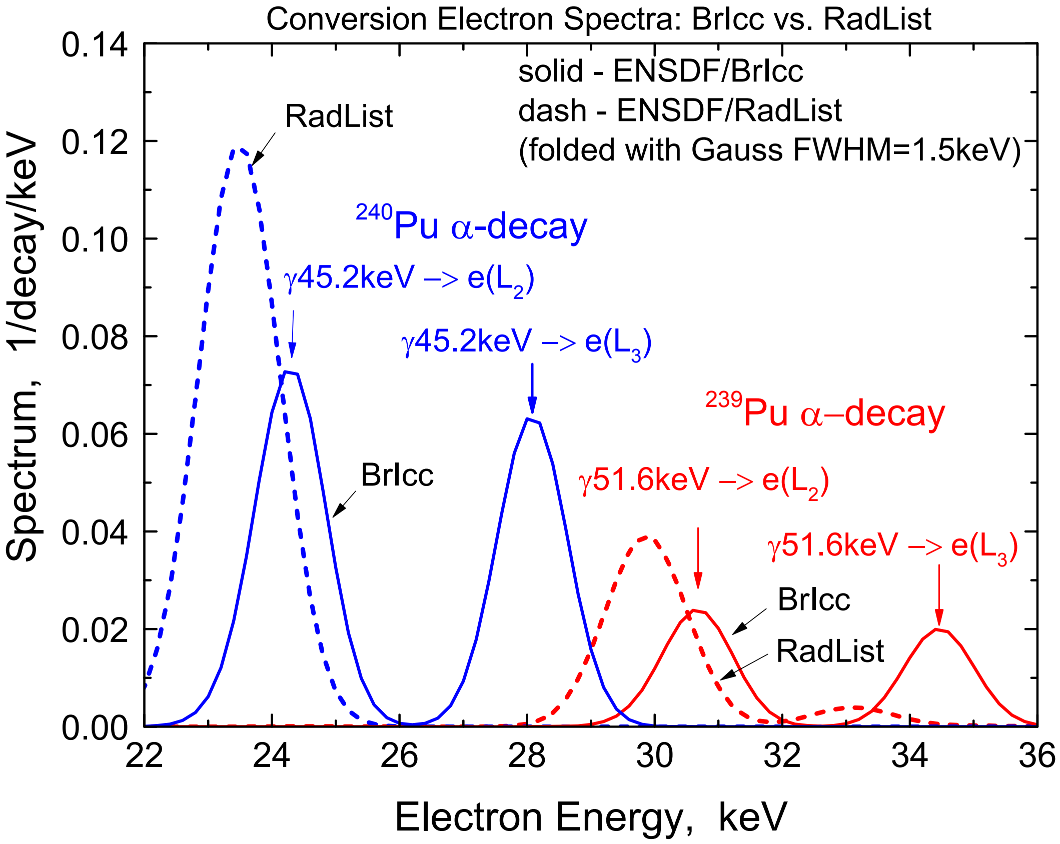
ENSDF(nuclear structure) + BrIcc (calculates yields and energy of conversion electrons)
reasonably predict measured electron spectrum

II. Conversion electrons: spectra from α-decay (cont.)

Example: comparison of BrIcc and RadList (ENSDF embedded code)

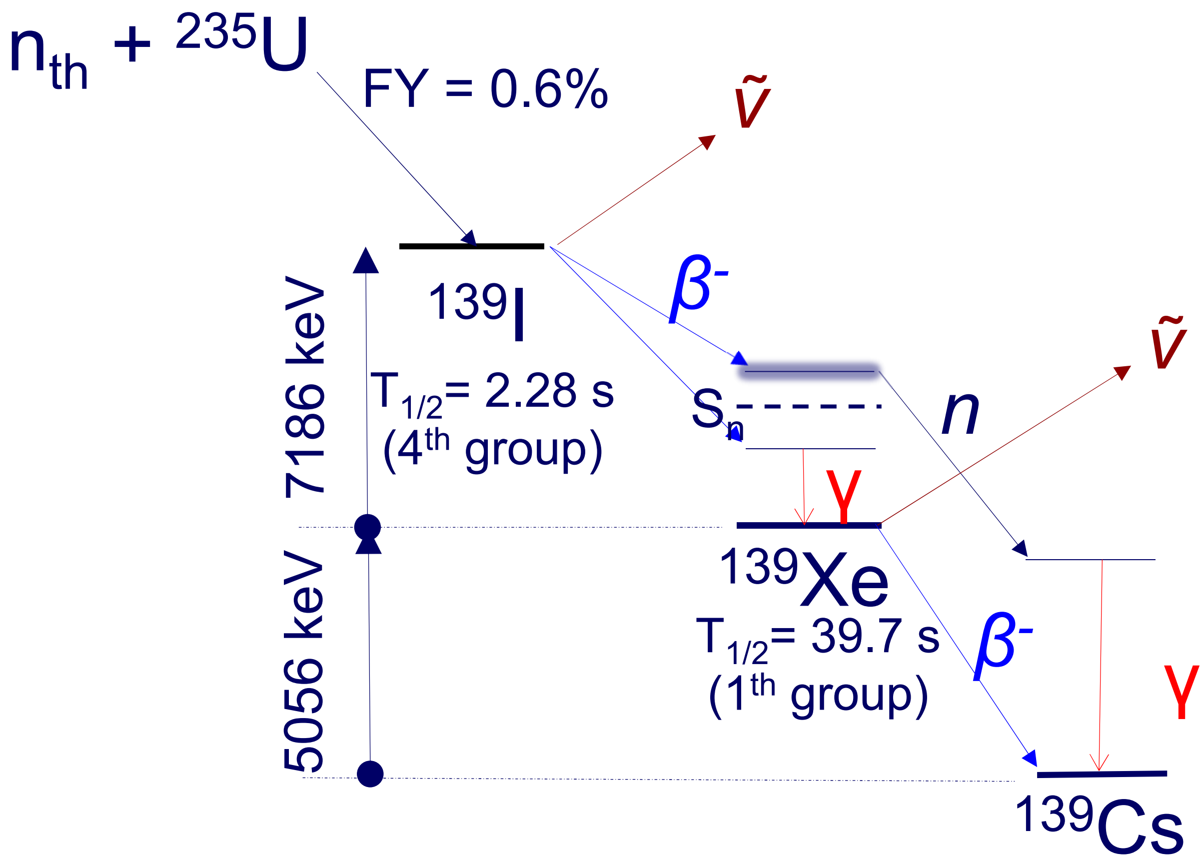
^{235}U from ENSDF: Eγ = 51.624 keV (E2)
Iγ =(0.02722 ± *22*) %/decay

	BrIcc			RadList		
	Ece, keV	ICC 1/γ	ICC %/dec		Ece, keV	ICC %/dec
Tot		**310 *5***				**317**
L1	29.87	4.20 *6*	**0.114**	L	29.87	**6.27 *20***
L2	30.68	120.4 *17*	**3.277**	L	32.28	**0.006 *4***
L3	34.46	101.8 *15*	**2.771**	L	35.07	**0.0288**
L-tot	**31.20**	**266 *4***				
M1	46.08	1.270 *18*				
M2	46.44	32.0 *5*				
M3	47.32	28.5 *4*				
M4	47.90	0.441 *7*				
M5	47.85	0.312 *5*				
M-tot	**46.85**	**62.6 *9***				



- **Finding: disagreements for CE spectra (Energy and Intensity) between BrIcc and RadList**
- **This will be reported to the structure experts at dedicated IAEA Meeting, 10-13 June 2014**

III. Antineutrino Spectra: Physics



- every β^- is accompanied by ***ṽ*** but necessary not by ***n***
- it is essentially ^{235}U(n,**xṽ**) reaction similar to ^{235}U(n,x**n**$_{\mathbf{delay}\text{ed}}$) available in EXFOR (NB: ṽ-spectra from separated precursor will never be measured, as neutron Pn)

III. Antineutrino: Application Facts and Needs

- **Reactor anti-neutrino – most strong (anthropological) $\tilde{\nu}$-source on planet:**
  - per fission: 6 electronic anti-neutrinos ($\tilde{\nu}_e$) from β^--decay of neutron rich FF
  - anti-neutrino yield ≈ 2 10^{20} $\tilde{\nu}$/s for 1 $GW_{thermal}$ of PWR
  - dominant contributors in PWR:

    90% is from ^{235}**U** and ^{239}**Pu (**latter competes at end of fuel cycle)

    10% is from ^{241}**Pu** and ^{238}**U** amounts 10%

  Thus *β*- and *$\tilde{\nu}$-spectra* from these 4 isotopes are most important

- **Non-proliferation, Safeguards –**
  IAEA, ESARDA and many labs have interest to the antineutrino detection to monitor at the distance the power of nuclear plants, fuel amount/composition
- **Fundamental Physics** - supernova neutrino events, geo-neutrinos …
- From all of this -> **Needs for Nuclear Data on <u>Reference Isotope *$\tilde{\nu}$-S*pectra</u>**

III. Antineutrino Spectra: sources of knowledge

- **measurements of β^- *spectra***, then conversion in ***ṽ***-*spectra* relying on energy conservation $E_{\tilde{v}} + E_\beta = E_0$, where E_0 – end energy of β^- branch

- **direct measurements of *ṽ* spectra**
  employing inverse β-decay reaction $\tilde{v}_e + p \rightarrow n + e^+$ with Q = -1.804 MeV
  or $E_{\tilde{v}} = E_{e+}$ + 1.804 MeV, thus spectrum part below 1.8 MeV is not acceptable

- ***ab initio* computing approach** (or **summation method)**:
  usage of fission yields, delayed β^- probabilities and feeding branches, correction for electron screening effect, Pandemonium effect, ….

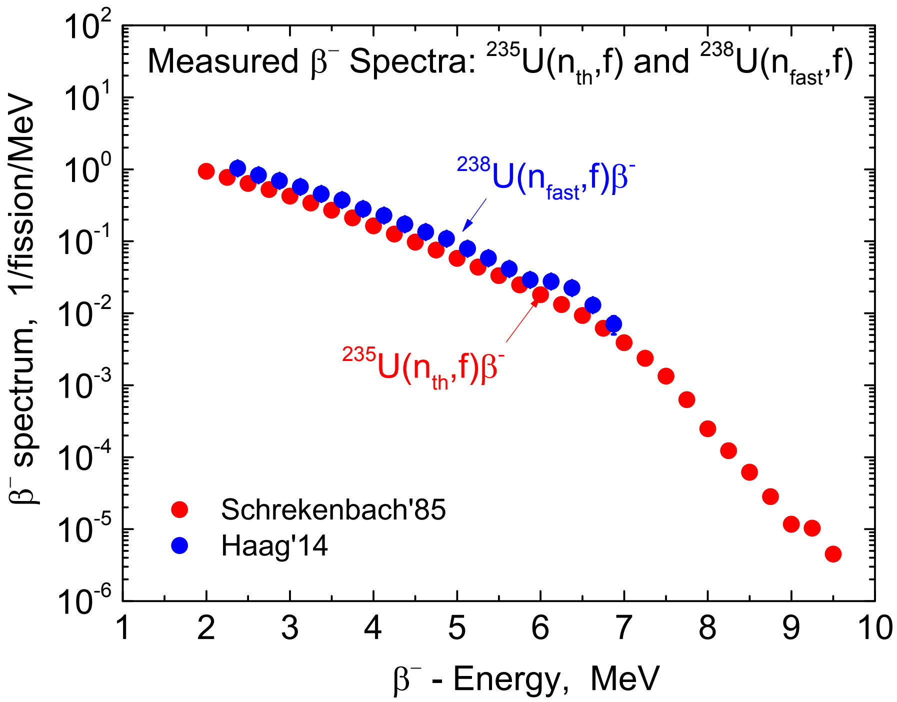
*ab-initio method may predict spectra in the whole energy range, reproduce spectrum fine details and be in such way a **reference data** … however calculations need experimental validation*

III. Antineutrino Spectra: known Spectral Measurements

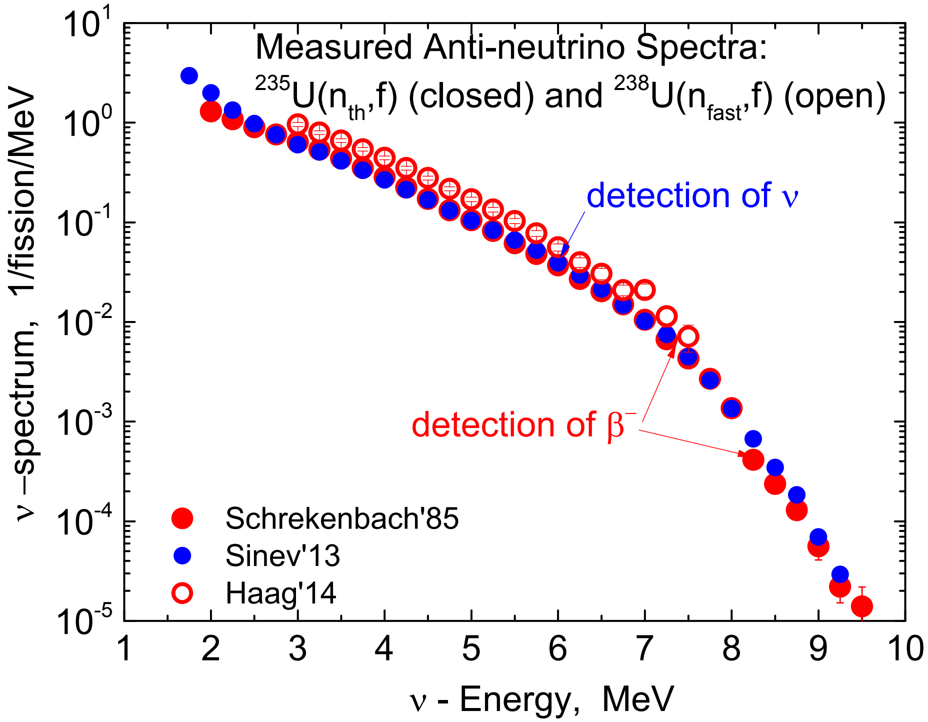
Author, Ref.	Lab, Facility	Isotope	Experimental Method	$\tilde{\nu}$- spectrum deduction details
I. Measurement of β^- spectrum, then conversion in $\tilde{\nu}$-spectrum				
R.E. Carter et al. Phys. Rev. 113(1959)280	LANL, California OWR	**$^{235}U(n_{th},f)$**	β^- scintillator with a γ coincidence	Conversion of β^- ***Data in Figure***
K. Scherckenbach et al. Phys.Lett. B160(1985)325 Phys.Lett. B99(1981)251	ILL, Grenoble HFR	**$^{235}U(n_{th},f)$** **12 h**	Magnetic β^- spectrometer	Conversion of β^- β^- ***and*** $\tilde{\nu}$ ***Spectra reported in Table***
A.A. Hahn et al. Phys.Lett. B218(1989)365 F. von Feilitzsch et al., Phys.Lett. B118(1982)162	ILL, Grenoble HFR	**$^{241}Pu(n_{th},f)$** **1.8 d** **$^{239}Pu(n_{th},f)$**	Magnetic β^- spectrometer	Conversion of β^- ***Data in Table***
N. Haag et al. Phys.Lett. 112(2014)122501	TU Garching FRM II	**$^{238}U(n_{fast},f)$**	β^- telescope with a γ supressing	Conversion of β^- ***Data in Table***
II. Direct $\tilde{\nu}$ spectrometry by inverse beta-decay (IBD) reaction $\tilde{\nu}_e + p \rightarrow n + e^+$				
V.V. Sinev et al. Phys.At.Nucl.76(2013)537 A.I. Afonin et al., SJETP 67(1988)213	Rovno NPP 1.3 GW_{th} 3-4 month	**$^{235}U(n_{th},f)$** **$^{238}U(n_{th},f)$** **$^{239}Pu(n_{th},f)$** **$^{241}Pu(n_{th},f)$**	Scintillation Detector	Decomposition of fuel $\tilde{\nu}$-spectrum in ones for isotope ***Data in Table***
F. Boehm et al. Phys.Rev. D62(2000)072002	Palo Verde Nucl. Station, Tonopah 3 PWR, 12 GW	**Fuel** **200 d**	segmented Gd-loaded liquid scintillator	fuel $\tilde{\nu}$-spectrum ***Data in Figure***

III. Beta and Antineutrino Spectra: Measured Data

$^{235}U(n_{th},f)\beta^-$ and $^{238}U(n_{fast},f)\beta^-$



$^{235}U(n_{th},f)\tilde{\nu}$ and $^{238}U(n_{fast},f)\tilde{\nu}$



Proposal to code in EXFOR both β^- (allowed) and $\tilde{\nu}$ (new code required) spectra as:

β^- (direct):

REACTION (92-U-235(N,B-),,DE,B-,FIS)

following rules in Memo CP-D/837

(33-AS-85(0,B-)34-SE-85,,PN/DE,,REL)

$\tilde{\nu}$ (direct):

REACTION (92-U-235(N,ANU),,DE,ANU,FIS,)

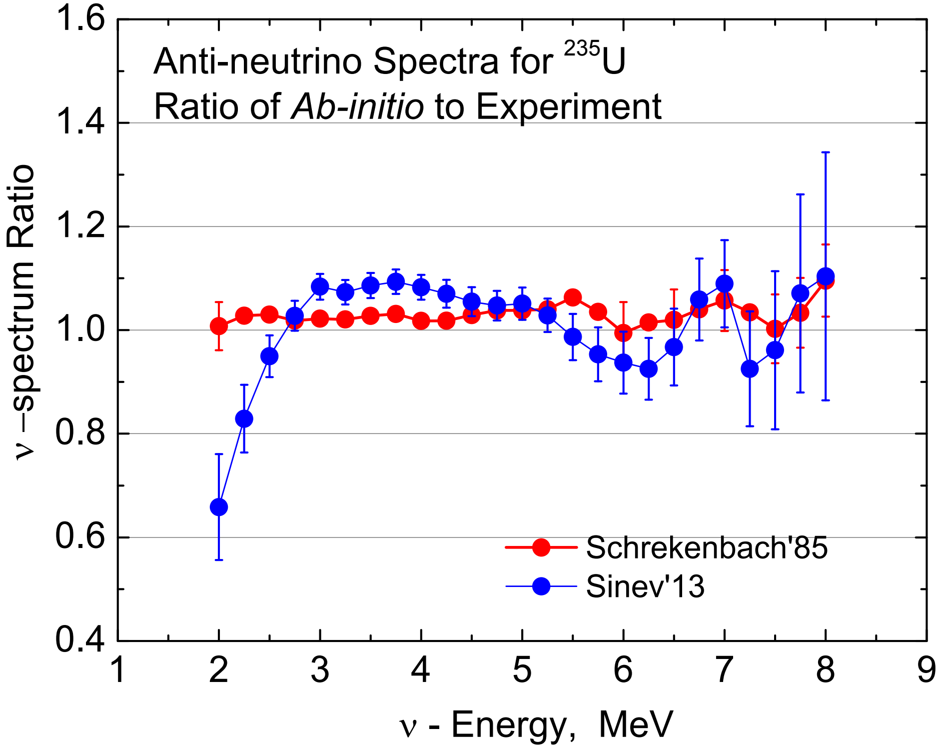
$\tilde{\nu}$ (derived):

REACTION (92-U-235(N,ANU),,DE,ANU,FIS,DERIV)
PART-DET (B-)

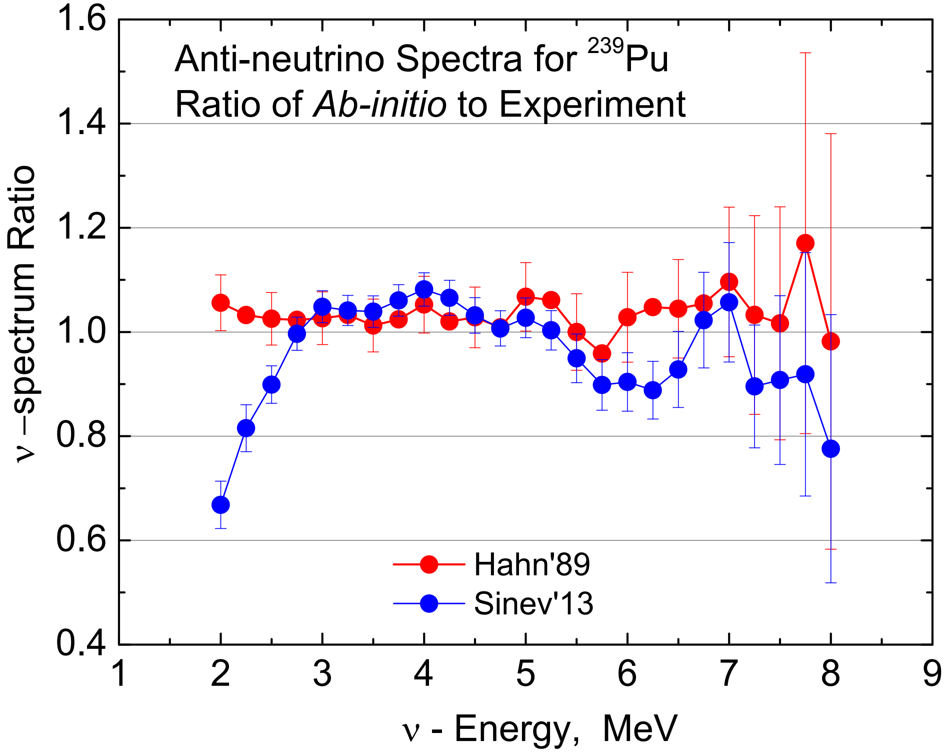
III. Antineutrino Spectra: Ab-initio (as a reference) vs Experiment

Ab-initio = A. Mueller et al., Phys. Rev. C **83**, 054615 (2011)

$^{235}U(n,f)$



$^{239}Pu(n,f)$



agreement between *ab-initio* Calculations and Measurements rather good: within ~ 10% for energies 3 to 7 MeV, up to 30% outside of this range

Summary

- NRDC tries to keep **Safeguards data up-today, complete and correct:**
  - all missing data on **Neutron Multiplicities** were included in EXFOR
  - **Nuclear Resonance Fluorescence** data were **compiled before** evaluations
  - **Conversion Electron spectra** generated by RadList code from ENSDF do not agree with BrIcc results and measurements

- EXFOR compilation of nuclear cross sections measured by detection of **Internal Conversion Electrons (ICE)** will follow *current rules* *(revision of ICE reference data is required)*

- **Proposal** to compile **β-** and **anti-neutrino** spectra (**ṽ** compliment **β^-** decay and precede delayed neutrons, which were proposed to compile in CP-D/837) :
  - such data available for neutron induced fission of **main reactor isotopes ^{235,238}U, ^{239,241}Pu**
  - published in **8 articles**, where energy spectra are mainly given **as Tables**
  - EXFOR compilation: **β^-** will follow *current rules,* **ṽ** - require *new particle code*