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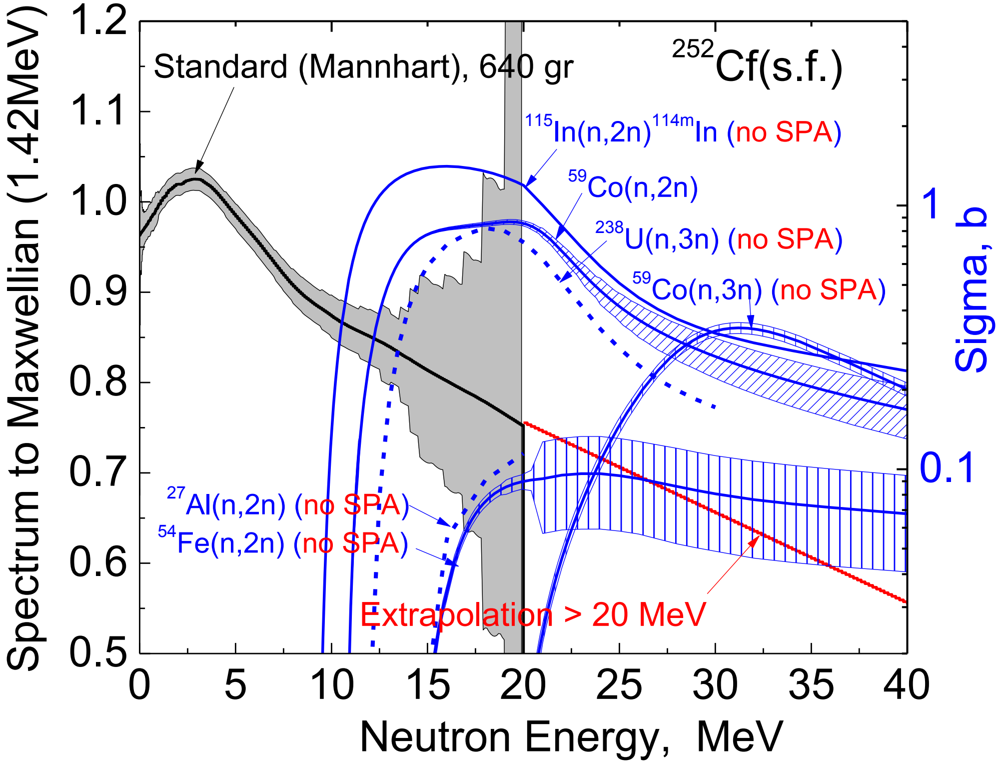
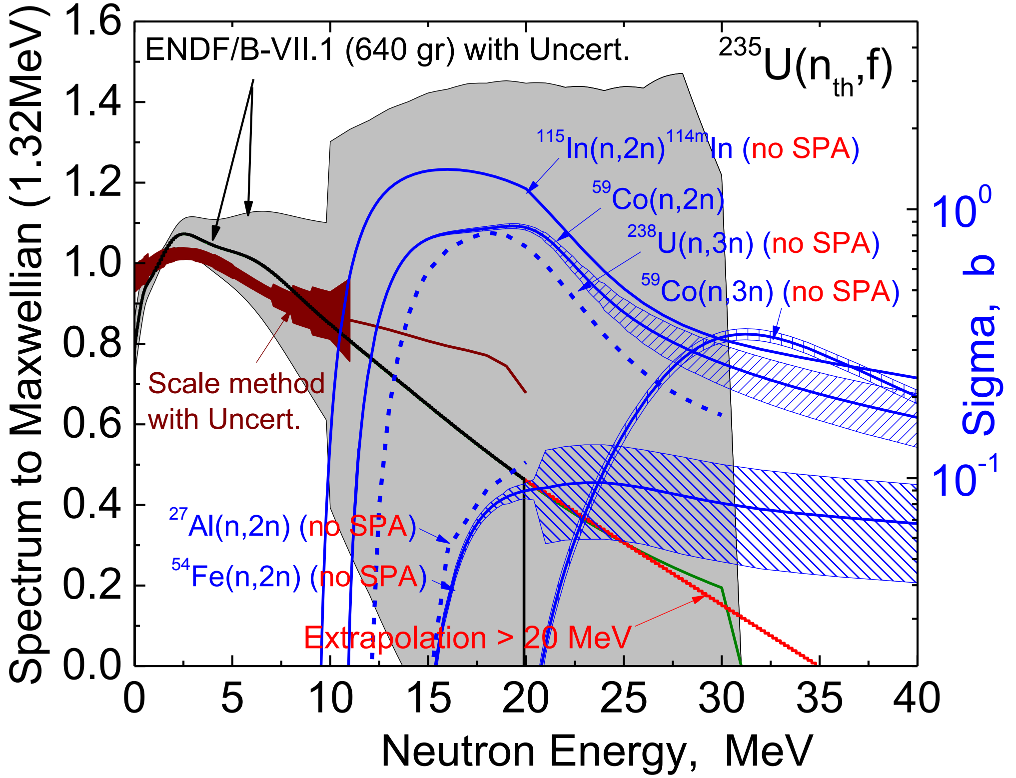
Spectrum averaged cross sections (SPA) for the high threshold dosimetry reactions: feasibility of activation and other alternative experimental techniques for SPA at level of 1 - 1000 μb

I. SPA cross sections for the high threshold dosimetry reactions

Following the recommendation of the IAEA Technical Meeting "Toward a New Evaluation of Neutron Standards", 8-12 July 2013 (INDC(NDS)- 0641):
"… assessing the possibility of using the AMS technique for the measurement of the $^{235}U(n_{th},f)$ or $^{235}U(n_{cold},f)$ prompt fission neutron averaged cross sections which can be used for validation of the prompt fission neutron spectrum at energies above 8 MeV ($<E_{50\%}> \; > 8$ MeV)"
the **spectrum averaged cross sections** (SPA) were calculated for several high threshold IRDFF reactions in ^{252}Cf(s.f.) and $^{235}U(n_{th},f)$ fields:



^{252}Cf(s.f.) PFNS (ratio to Maxwellian T = 1.42 MeV) and IRDFF cross sections (only ^{59}Co(n,2n) SPA was measured).



$^{235}U(n_{th},f)$ PFNS (ratio to Maxwellian T = 1.32 MeV) and IRDFF cross sections (only ^{59}Co(n,2n) SPA was measured).

Comments to Figures: Scale method – N.V. Kornilov, "Scale Method for Evaluation of Fission Neutron Spectra", NSE 169(2011)290

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Table 1. Dosimetry reactions, their stable products, kinematic threshold E_{thr}, effective energy $E_{50\%}$ and SPA in the **^{252}Cf(s.f.)** field, sorted by increasing $E_{50\%}$.

IRDFF reactions and their products	E_{thr} MeV	$E_{50\%}$ MeV	SPA, μb IRDFF[1]	SPA, μb Experiment[2]	$N_{product}$ / N_{target} if 10^8n/cm^2/s, 1000h	Comments
^{252}Cf(s.f.) Spontaneous Fission Spectra: given ^{252}Cf produces Flux = 10^8 n/cm^2/s (i.e. at ≈1 cm from ^{252}Cf of 10^9 n/s intensity[3]) and Irradiation of sample = 1000 h = 4.17 weeks						
^{54}Fe(n,α)^{51}Cr (ε, 27.7 d) → ^{51}V (stable)	0	7.430	1113 ± 3.6%	No Exp	4007 10^{-16}	
^{238}U(n,2n)^{237}U (β$^-$, 6.75 d) → ^{237}Np (2.14 My)	6.180	8.276	20584 ± 2.4%	19200 ± 10% 12200 ± 12%	74100 10^{-16}	Blinov vs. Shani: measurements discrepant !!!
^{232}Th(n,2n)^{231}Th (β$^-$, 26 d) → ^{231}Pa (3.28 kY)	*6.448*		*24377 (B-VII.1)*	No Exp	*87757 10^{-16}*	
^{169}Tm(n,2n)^{168}Tm (ε, 93 d) → ^{168}Er (stable)	8.082	10.400	6260 ± 2.4%	6690 ± 6.3%	22536 10^{-16}	
^{130}Te(n,2n)^{129}Te (IT, β$^-$, 34 d) → ^{129}I (stable)	*8.484*		*3494 (B-VII.1)*	No Exp	***12578 10^{-16}***	AMS threshold[W] = 10^{-14}
^{141}Pr(n,2n)^{140}Pr (ε, 3.4 min) → ^{140}Ce (stable)	9.464	11.85	1990 ± 11.1%	No Exp.	7164 10^{-16}	
^{75}As(n,2n)^{74}As (ε, 17.8 d) → ^{74}Ge (stable)	10.383	12.91	621 ± 5.8%	No Exp.	2236 10^{-16}	
^{115}In(n,2n)^{114m}In (IT, 50 d; β$^-$) → ^{114}Sn (stable)	10.633	13.09	1633 ± 5.0%	No Exp.	5879 10^{-16}	
^{59}Co(n,2n)^{58}Co (ε, 70 d) → ^{58}Fe (stable)	10.633	13.09	410 ± 0.0%	405 ± 2.5%	1476 10^{-16}	
^{238}U(n,3n)^{236}U (α, 2.34 10^7 y) → ^{232}Th (stable)	*11.330*		*163 (B-VII.1)*	*No Exp.*	*567 10^{-16}*	AMS threshold[W] = 10^{-11}
^{56}Fe(n,2n)^{55}Fe (ε, 2.74 y) → ^{55}Mn (stable)	*11.40*		*170 (B-VII.1)*	*No Exp.*	***612 10^{-16}***	AMS threshold[W] = 10^{-14}
^{89}Y(n,2n)^{88}Y (ε, 107 d) → ^{88}Sr (stable)	11.612	13.90	346 ± 1.3%	No Exp.	1246 10^{-16}	
^{52}Cr(n,2n)^{51}Cr (ε, 27.7 d) → ^{51}V (stable	12.272	14.71	97 ± 2.7%	No Exp.	360 10^{-16}	
^{23}Na(n,2n)^{22}Na (ε, 2.60 y) → ^{22}Ne (stable)	12.419	15.40	8.6 ± 1.2%	No Exp.	31 10^{-16}	
^{46}Ti(n,2n)^{45}Ti (ε, 3.1 h) → ^{45}Sc (stable)	13.479	16.03	**12.2± 3.1%**	**93 ± 33% (?)**	44 10^{-16}	**C/E = 0.13 ± 33% ???!!!**
^{27}Al(n,2n)^{26}Al (ε, 7.17 10^5 y) → ^{26}Mg (stable)	*13.55*		*5.7 (B-VII.1)*	*No Exp.*	*21 10^{-16}*	AMS threshold[W] = 10^{-13}
^{54}Fe(n,2n)^{53}Fe (ε, 8.5 min) → ^{53}Mn (3.7 My)	13.629	16.48	3.5 ± 1.5%	No Exp.	13 10^{-16}	**not for AMS[W] due to impact of ^{54}Fe(n,np+d)^{53}Mn**

IRDFF reactions and their products	E_{thr} MeV	$E_{50\%}$ MeV	SPA, μb IRDFF[1]	SPA, μb Experiment[2]	$N_{product}$ / N_{target} if $10^8 n/cm^2/s$, 1000h	Comments
^{252}Cf(s.f.) Spontaneous Fission Spectra: given ^{252}Cf produces Flux = 10^8 n/cm^2/s (i.e. at ≈1 cm from ^{252}Cf of 10^9 n/s intensity[3]) and Irradiation of sample = 1000 h = 4.17 weeks						
^{209}Bi(n,3n)^{207}Bi (ε, 31.6 y) → ^{207}Pb (stable)	14.416	18.21	19 ± 6.0%	No Exp.	68 10^{-16}	
^{169}Tm(n,3n)^{167}Tm (ε, 9.3 d) → ^{167}Er (stable)	14.963	18.49	14.7 ± 5.7%	No Exp.	54 10^{-16}	
^{59}Co(n,3n)^{57}Co (ε, 271 d) → ^{57}Fe (stable)	19.352	22.36	0.097 ± 5.6%	No Exp.	0.35 10^{-16}	

Example of calculation for ^{27}Al(n,2n)^{26}Al: Ratio ^{26}Al/^{27}Al = Flux × Time × Sigma = 1.E+8 n/cm^2/s × 3.6E+6 s × 5.7E-30 cm^2 = 20.5E-16

Table 2. Dosimetry reactions, their stable products, kinematic threshold E_{thr}, effective energy $E_{50\%}$ and SPA in the **^{235}U(n_{th},f)** field, sorted by increasing $E_{50\%}$.

IRDFF reactions and their products	E_{thr} MeV	$E_{50\%}$ MeV	SPA, μb IRDFF[1]	SPA, μb Experiment[2]	$N_{product}$ / N_{target} if $10^9 n/cm^2/s$, 100 h	Comments
^{235}U(n_{th},f) neutron induced Fission Spectra: given n-Source produce Flux = 10^9 n/cm^2/s (cp. 1.9 10^9 n/cm^2/s from fission plate in KUR facilty[4]) and Irradiation of sample = 100 h = 0.417 weeks						
^{169}Tm(n,2n)^{168}Tm (ε, 93 d) → ^{168}Er (stable)	8.082	10.40	3744 ± 2.6%	3735 ± 4.2%	13478 10^{-16}	
^{115}In(n,2n)^{114}In (IT, 50 d; β$^-$) → ^{114}Sn (stable)	10.633	11.60	861 ± 5.5%	No Exp.	3100 10^{-16}	
^{141}Pr(n,2n)^{140}Pr (ε, 3.4 min) → ^{140}Ce (stable)	9.464	11.65	1043 ± 12.0%	No Exp.	3755 10^{-16}	
^{65}Cu(n,2n)^{64}Cu (ε, 12.7 h) → ^{64}Ni (stable) ^{64}Cu (β$^-$, 12.7 h) → ^{64}Zn (stable)	10.065	12.46	318 ± 2.0%	No Exp.	^{64}Ni/^{65}Cu = 704 10^{-16} ^{64}Zn/^{65}Cu = 441 10^{-16}	
^{75}As(n,2n)^{74}As (ε, 17.8 d) → ^{74}Ge (stable)	10.383	12.70	295 ± 6.4%	No Exp.	1062 10^{-16}	
^{59}Co(n,2n)^{58}Co (ε, 70 d) → ^{58}Fe (stable)	10.633	13.09	191 ± 1.8%	203 ± 2.5%	688 10^{-16}	
^{238}U(n,3n)^{236}U (α, 2.34 10^7 y) → ^{232}Th (stable)	*11.330*		*682 (BVII.0)*	*No Exp.*	*2455 10^{-16}*	
^{56}Fe(n,2n)^{55}Fe (ε, 2.74 y) → ^{55}Mn (stable)	*11.400*		*739 (BVII.1)*	*No Exp.*	*2660 10^{-16}*	AMS threshold[W] = 10^{-14}
^{89}Y(n,2n)^{88}Y (ε, 107 d) → ^{88}Sr (stable)	11.612	13.90	149 ± 1.4%	150 ± 3.3%	536 10^{-16}	

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IRDFF reactions and their products	E_{thr} MeV	$E_{50\%}$ MeV	SPA, μb IRDFF[1]	SPA, μb Experiment[2]	$N_{product}$ / N_{target} if 10^9 n/cm^2/s, 100 h	Comments
^{235}U(n_{th},f) neutron induced Fission Spectra: given n-Source produce Flux = 10^9 n/cm^2/s (cp. 1.9 10^9 n/cm^2/s from fission plate in KUR facilty[4]) and Irradiation of sample = 100 h = 0.417 weeks						
^{52}Cr(n,2n)^{51}Cr (ε, 27.7 d) → ^{51}V (stable)	12.272	14.71	38 ± 2.7%	No Exp.	137 10^{-16}	
^{23}Na(n,2n)^{22}Na (ε, 2.60 y) → ^{22}Ne (stable)	12.419	15.40	3.2 ± 1.3%	No Exp.	12 10^{-16}	
^{46}Ti(n,2n)^{45}Ti (ε, 3.1 h) → ^{45}Sc (stable)	13.479	15.81	4.3± 4.4%	No Exp.	15 10^{-16}	
^{27}Al(n,2n)^{26}Al (ε, 7.17 10^5 y) → ^{26}Mg (stable)	*13.550*		*2.0 (BVII.1)*	*No Exp.*	*7 10^{-16}*	AMS threshold[W] = 10^{-13}
^{54}Fe(n,2n)^{53}Fe (ε, 8.5 min) → ^{53}Mn (3.7 My)	13.629	16.48	1.2± 5.1%	No Exp.	4 10^{-16}	**not for AMS[W] due to impact of ^{54}Fe(n,np+d)^{53}Mn**
^{209}Bi(n,3n)^{207}Bi (ε, 31.6 y) → ^{207}Pb (stable)	17.416	17.88	5.4 ± 5.9%	No Exp.	19 10^{-16}	
^{169}Tm(n,3n)^{167}Tm (ε, 9.3 d) → ^{167}Er (stable)	14.963	18.20	4 ± 6.1%	No Exp.	14 10^{-16}	
^{59}Co(n,3n)^{57}Co (ε, 271 d) → ^{57}Fe (stable)	19.352	21.92	0.017 ± 7.7%	No Exp.	0.06 10^{-16}	

Example of calculation for ^{27}Al(n,2n)^{26}Al: Ratio ^{26}Al/^{27}Al = Flux × Time × Sigma =1.E+9 n/cm^2/s × 3.6E+5 s × 2.E-30 cm^2 = 7.2E-16

Comments for Tables 1 and 2:

Italic font - reactions currently not included in IRDFF

1) Calculated SPA uncertainty includes only IRDFF-1.05 cross section uncertainty.
2) The known measurements are carried out by activation technique.
3) The most intensive ^{252}Cf sources known up to now:
   - K. Kobayashi et al. [JNST 19(1982)341] used 500 μg of ^{252}Cf which produced ≈ 1 10^9 n/s;
   - J. Czikai et al. [Antwerp (1982)418] used 40 μg (?) of ^{252}Cf which produced ≈ 1 10^8 n/s (given 1 μg = 2.3 10^6 n/s);;
   - M. Blinov et al. [Atom. Energiya 65(1988)206] used 2-3 Cf sources of 18 - 50 μg total mass or 0.4 - 1.2 10^8 n/s (given 1 μg = 2.3 10^6 n/s);
4) The most intensive PFNS source:
   KUR power fission plate: Ø27 × 1 cm, 1.1 kg of 90% ^{235}U, incident thermal n-flux = 5.8 10^8 n/cm^2 [I. Kimura and K. Kobayashi NSE106(1990)332]

W) - information from private communication with A. Wallner

$N_{product}$ / N_{target} - looks to be feasible for AMS

For other high energy reactions see: Cf-252(s.f.) http://www-nds.iaea.org/IRDFFtest/IRDFF105_MCNP_Cf.pdf
U-235(n_{th},f) http://www-nds.iaea.org/IRDFFtest/IRDFF_MCNPtest_U5.pdf .

Tables 1 and 2 show that it was impossible to measure so far some high threshold SPA by traditional activation technique with SPA below 150 - 400 μb.

SPA for these reactions, if they can be measured by activation or alternative methods, will probe the unknown high energy part (i.e. above 8-10 MeV where uncertainties ≈ 100%) of the ^{252}Cf(s.f.) and ^{235}U(n,f) spectra, since the dosimetry and some other reaction cross sections are known there with much better accuracy (≤ 10%).

II. Techniques alternative to Activation

1. The Accelerator Mass Spectrometry (AMS) was shown is feasible to measure extremely small SPA.

The method sensitivity $N_{product}$ / N_{target} ~ 10^{-12} - 10^{-16}.

For more details see A. Wallner et al.:

“Novel method to study neutron capture of ^{235}U and ^{238}U simultaneously at keV energies”, Phys. Rev Lett.112(2014)192501
“Precise measurement of the ^{27}Al(n,2n)^{26g}Al excitation function near threshold and its relevance for fusion-plasma technology”, J.Eur.Phys. A**7**, 285 (2003)
“Production of Long-lived Radionuclides ^{10}Be, ^{14}C, ^{53}Mn, ^{55}Fe, ^{59}Ni and ^{202g}Pb in a Fusion Environment” J. Korean Phys. Soc. 59, 1378
“Nuclear Data from AMS & Nuclear Data for AMS – some examples”, EPJ 35 (2012) 01003
“Accelerator Mass Spectrometry & Neutron-induced Reactions”, presentation at the IAEA TM on Standards (July 2013) here .

A. Wallner pointed out on the following **high threshold non-dosimetry reactions accessible for AMS:**

^{27}Al(n,2n)^{26}Al was measured by AMS up to 19 MeV with accuracy 10% by A. Wallner et al., Eur. Phys. A**17**, 285 (2003))
^{56}Fe(n,2n)^{55}Fe was measured by AMS around 14 MeV by A. Wallner et al. J. Korean Phys. Soc. 59, 1378);
^{238}U(n,3n)^{236}U was measured by AMS at 14 MeV by X. Wang et al. Phys. Rev. C87(2013)014612).

The status of these reaction cross sections are shown in Figs. 3-5.

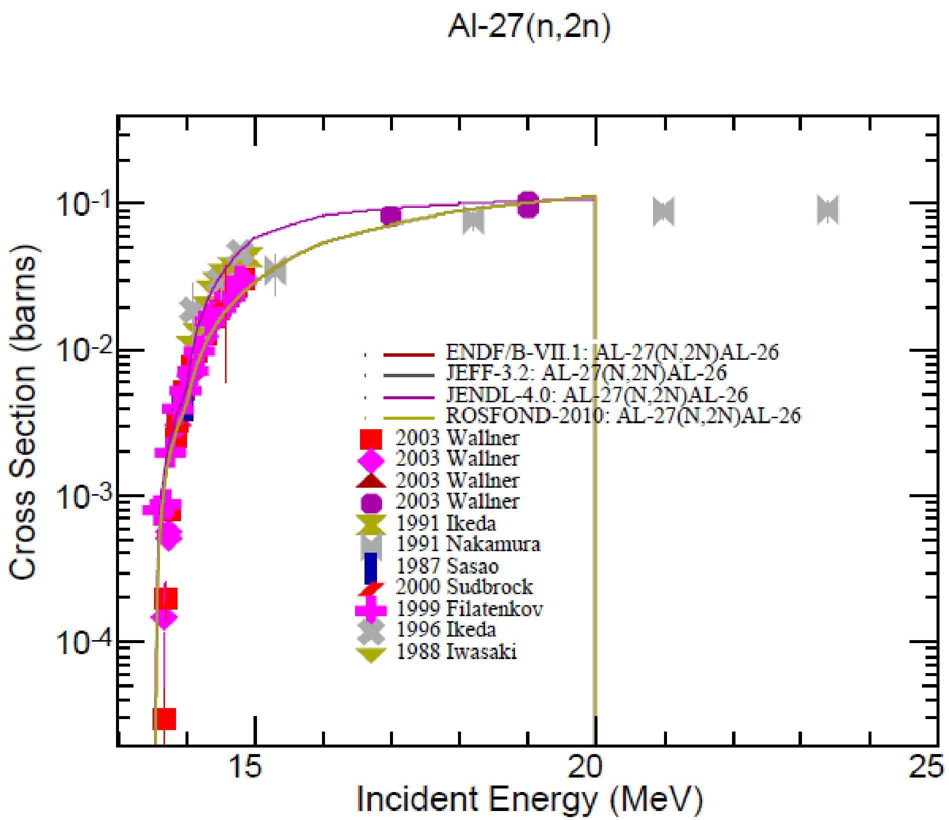

Fig 3. Available experimental and evaluated data for $^{27}Al(n,2n)^{26}Al$.

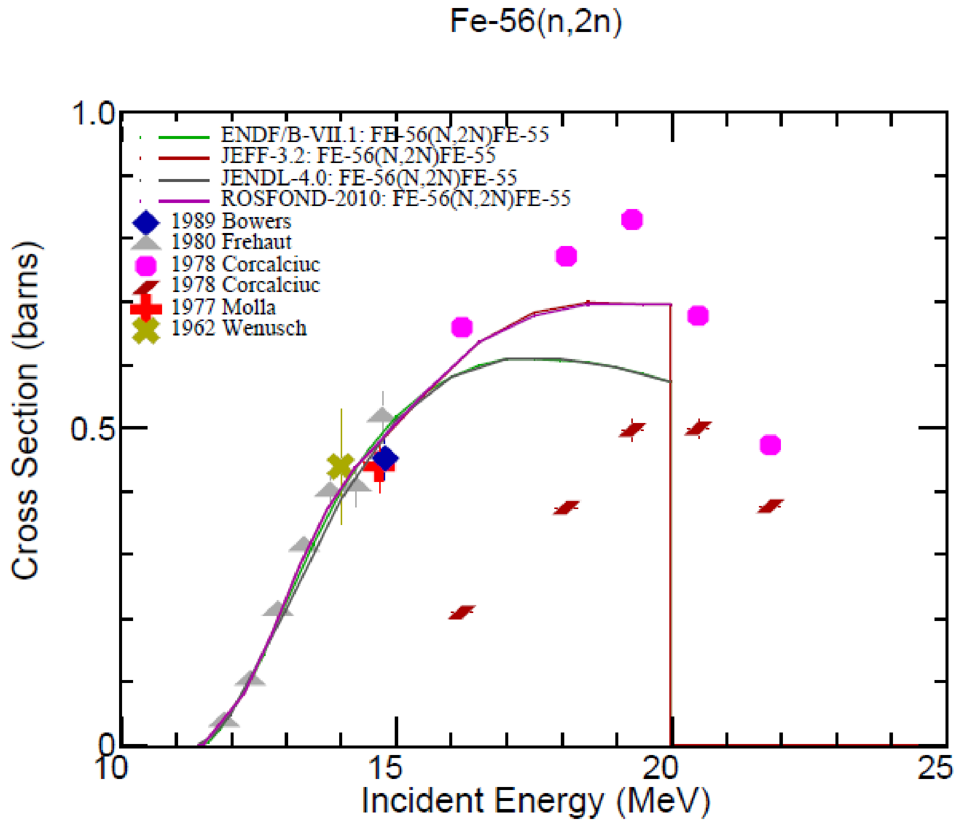

Fig. 4. Available experimental and evaluated data for $^{56}Fe(n,2n)^{55}Fe$.

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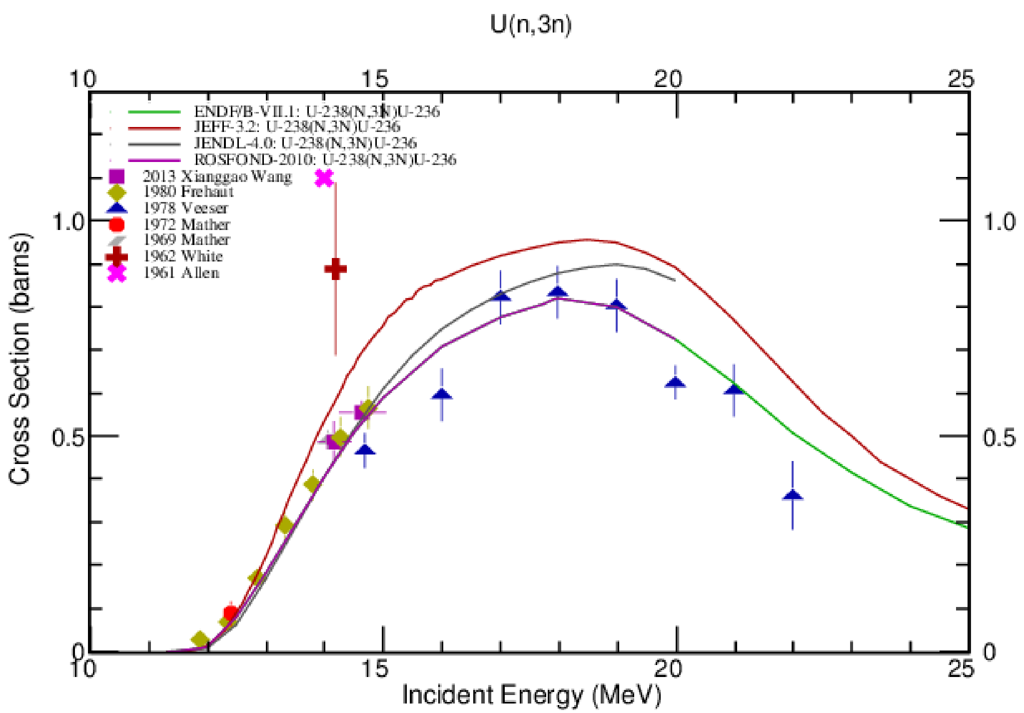

Fig. 5. Available experimental and evaluated data for $^{238}U(n,3n)^{236}U$.

2. Prompt Gamma Neutron Activation Analysis (PGNAA)

The method sensitivity $N_{product} / N_{target} \sim 100$ ppm $= 10^{-4}$.

This technique was proved is capable to measure the non- threshold SPA cross sections by employing the PGNAA facility of FRM-II after Ni foil irradiation in the LVR-15 reactor (fluence rate 3.10^{14} $cm^{-2}s^{-1}$) for reactions $^{62}Ni(n,\gamma)^{63}Ni$ ($T_{1/2}$ = 101.2 y, Atlas $\sigma(n_{thermal},\gamma)$ = 14.9 b) and $^{58}Ni(n,\gamma)^{59}Ni$ ($T_{1/2}$ = 7.6 10^4 y, Atlas $\sigma(n_{thermal},\gamma)$ = 4.37 b).

For principles, first results and publications see:

V. Klupák, L. Viererbl, Z. Lahodová, J. Šoltés, I. Tomandl, P.Kudějová, "Nickel foil as transmutation detector for neutron fluence measurements", ISRD-15, EPJ Web of Conferences 106, 05013 (2016),

I. Tomandl, L. Viererbl, P. Kudějová, Z. Lahodová, V. Klupák, M. Fikrle, "Determination of trace concentrations of transmuted stable nuclides in TMD detectors using PGAA", J. of Radioanal. and Nuclear Chemistry, 300 (2014) 1141.

3. Resonance Ionization Mass Spectroscopy (RIMS) - Isotope measurements based on Laser Spectroscopy.

The method sensitivity $N_{product} / N_{target} \sim$??.
Currently under development for the trace analysis of short-lived and long-lived radioactive nuclei.

This technique was proved is capable to measure the cross section for dosimetry reaction
^{93}Nb(n,n')^{93m}Nb ($T_{1/2}$ = 16.13 y).

For principles, first results and publications see:
here: http://coe.nucl.nagoya-u.ac.jp/Measurement01_E.html and
H. Tomita, T. Takatsuka, T. Iguchi, Y. Adachi, Y. Furuta, T. Takamatsu, T. Noto, "Development of Neutron Dosimetry Technique with ^{93}Nb(n,n')^{93m}Nb Reaction by Resonance Ionization Mass Spectrometry", ISRD-15, EPJ 106, 05002 (2016)
T. Takatsuka, H. Tomita, V. Sonnenschein, T. Sonoda, Y. Adachi et al. "Development of resonance ionization in a supersonic gas-jet for studies of short-lived and long-lived radioactive nuclei" , NIM B 317 (2013)586

4. Ion Beam Analysis (IBA) technique such as PIXE, PIGE etc.

The method sensitivity $N_{product} / N_{target} \sim 10^{-4}$-$10^{-3}$.

It seems will not be possible to use this technique to measure the high threshold SPA cross sections (< 1 mb) because of its low sensitivity however it may work, as PGNAA, for the non- threshold reactions with large SPA cross sections (> 1 b).

5. Nuclear magnetic resonance (??).

The method sensitivity $N_{product} / N_{target} \sim$??.