Measurements of (n,α) cross sections

Khryachkov Vitaly

Institute for physics and power engineering (IPPE) Obninsk, Russia

Justification for the (n,α) reaction cross section measurement

- Reactor criticality (**structural material , O, N**)
- Standards (**^{10}B, ^{6}Li**)
- Gas production
- Astrophysics
- Dosimetry

Experimental methods for (n,α) reaction investigation

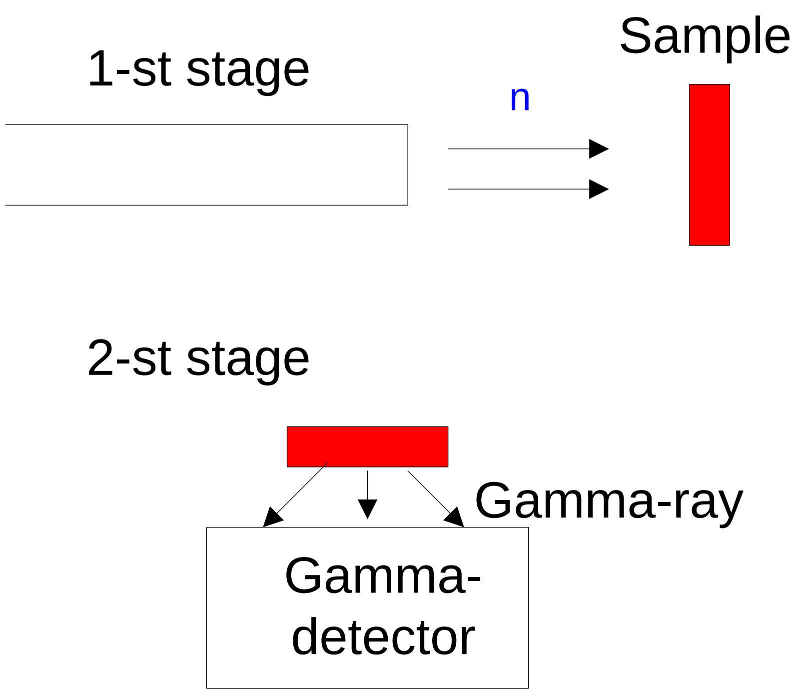
- **Activation method;**
- **Direct measurement of α – particle yield;**

Activation method

$$n + {}^{Z}_{M}A \rightarrow \alpha + {}^{Z-2}_{M-3}D$$

$${}^{Z-2}_{M-3}D \xrightarrow{\beta} {}^{Z-1}_{M-3}N^{*} + \overline{\nu} \quad , \quad T_{1/2}$$

$${}^{Z-1}_{M-3}N^{*} \rightarrow {}^{Z-1}_{M-3}N + \gamma$$

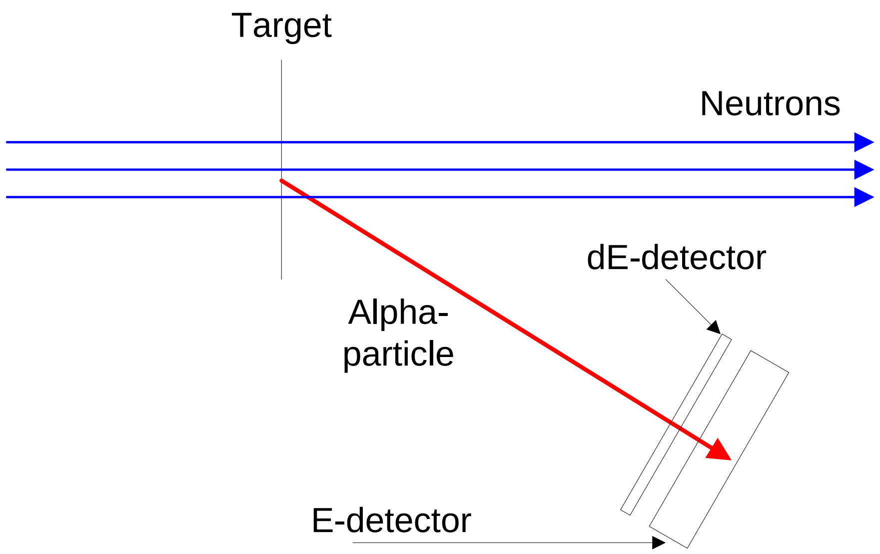


Limitations of the activation method

- Residual nuclear must by radioactive!
- Half-life time for residual nuclear must be convenient!
- Energy of gamma-ray must be convenient!
- Yield of gamma-ray must be significant!

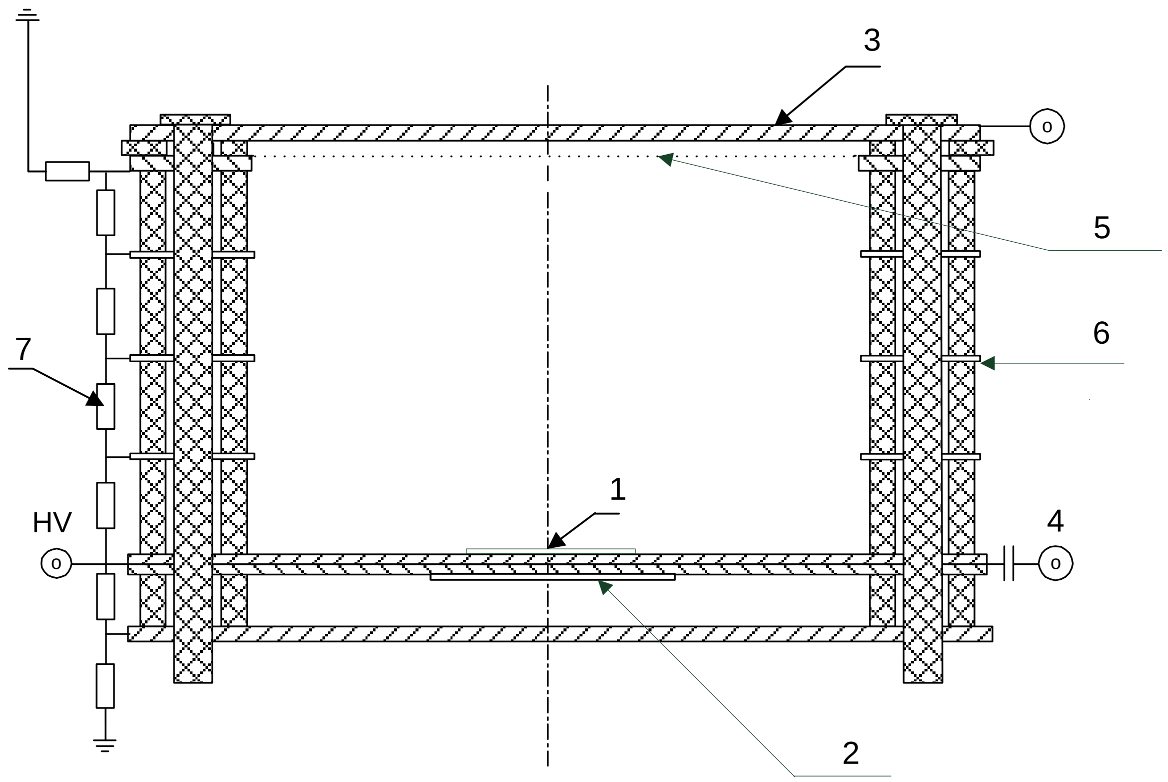
For stable residual nuclear activation measurement can not by done at all!

ΔE-E method



- Low energy particles can not pass through ΔE detector!
- High energy particles will not be stopped in E-detector!
- Low geometrical registration efficiency!
- It is needed to repeat measurements for different angles!

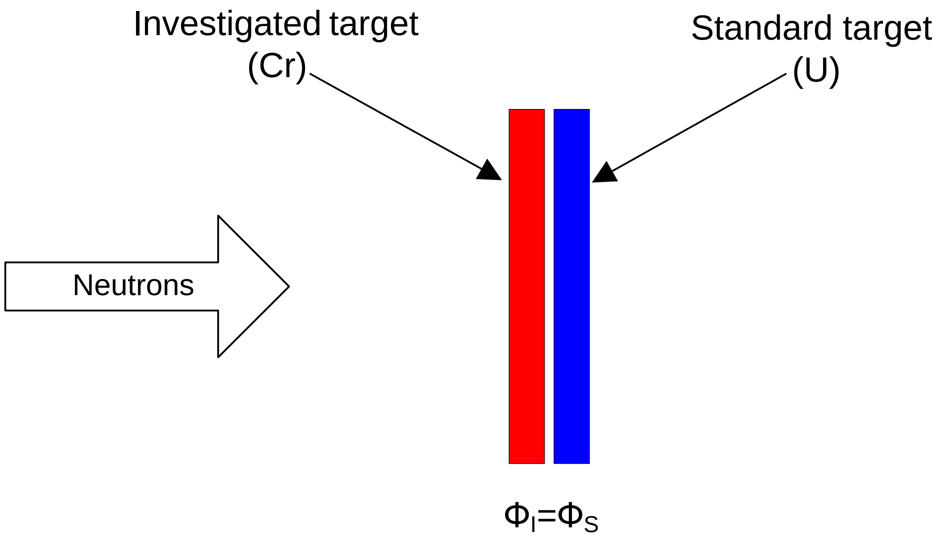
Classical ionisation chamber



1) **Target;**
2) **^{238}U target;**
3) **Anode;**
4) **Anode signal connector;**

5. **Frisch grid;**
6. **Guard electrodes;**
7. **Resistor.**

Relative method to cross section measurement



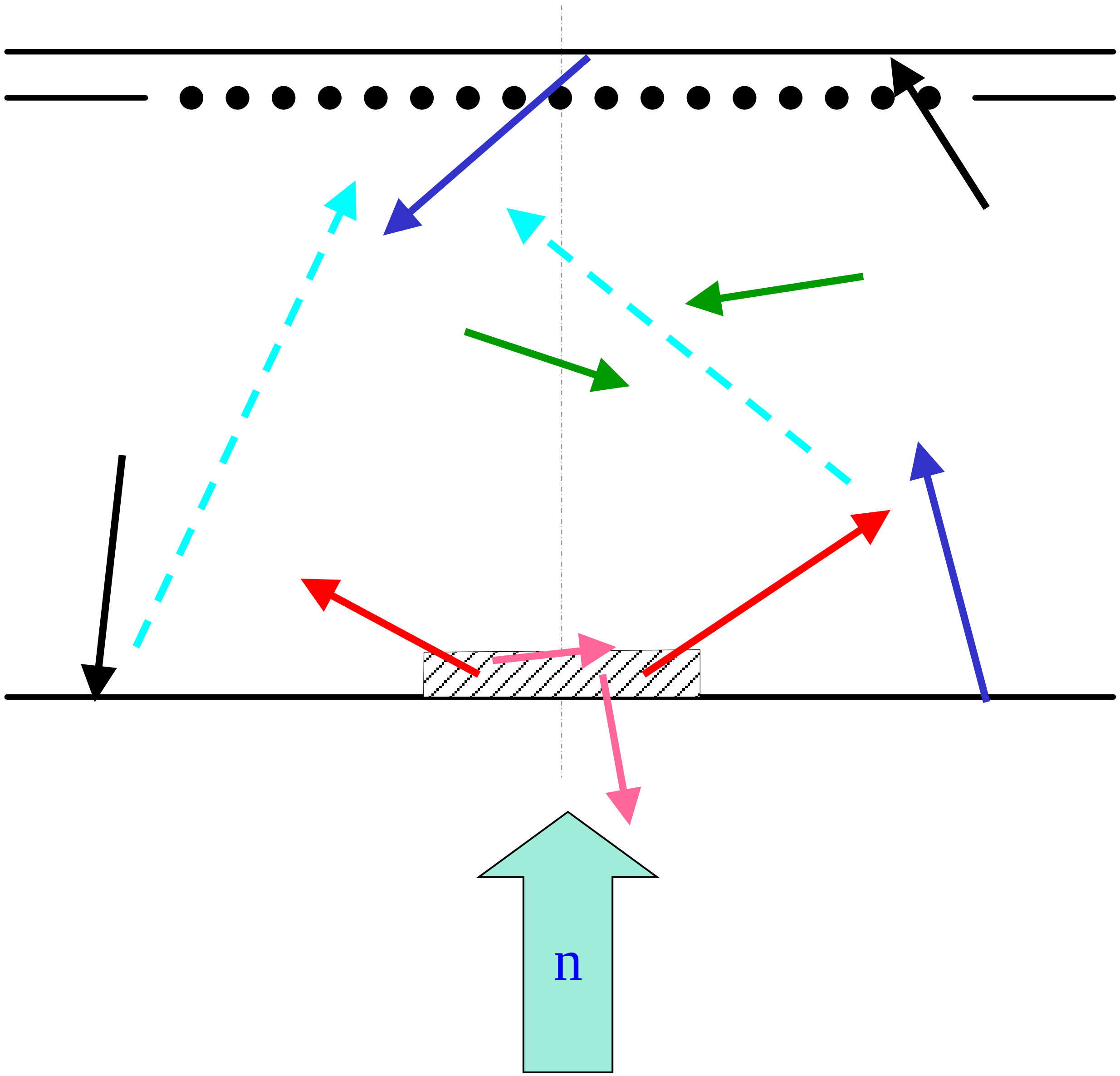
$$n_{\alpha} = N_{Cr} \cdot \sigma_{(n,\alpha)} \cdot \Phi_{Cr}$$

$$n_{ff} = N_{U} \cdot \sigma_{(f)} \cdot \Phi_{U}$$

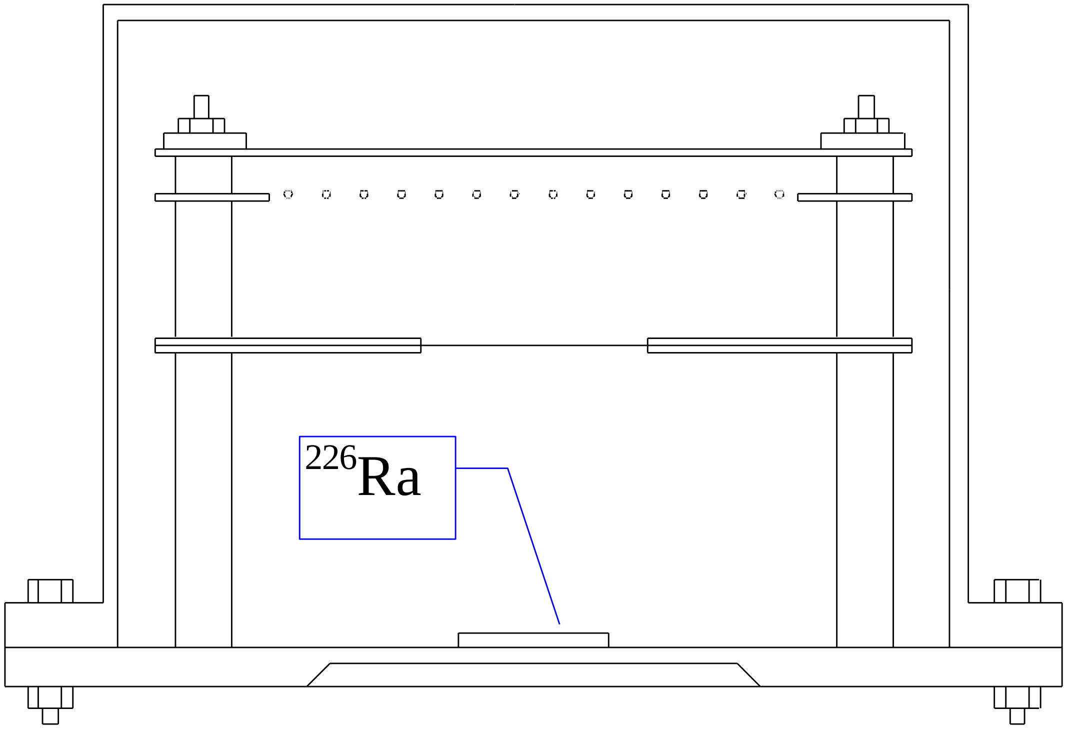
$$\sigma_{(n,\alpha)} = \frac{n_{ff}}{n_{\alpha}} \cdot \sigma_{(U)} \cdot \frac{N_{Cr}}{N_{U}}$$

Classical spectrometer events classification

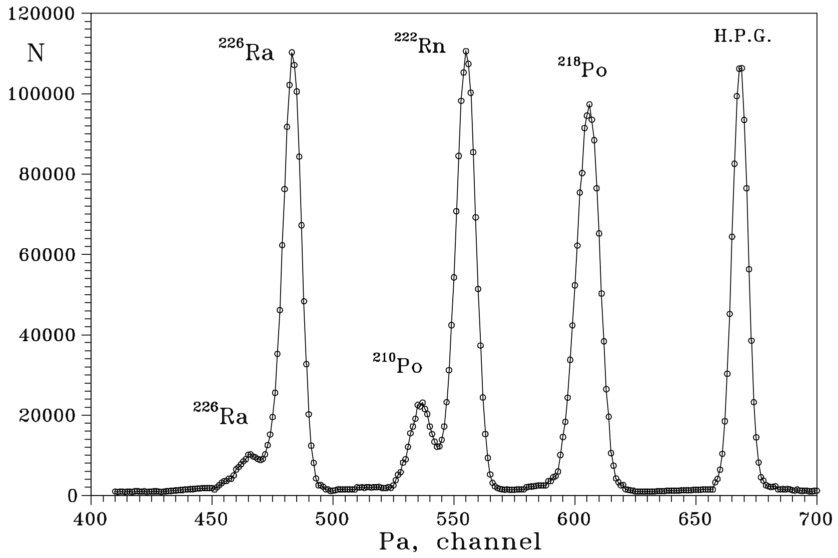
1. Target
2. Full absorption
3. Electrodes
4. Gas a-particles
5. Protons
6. Wall effect

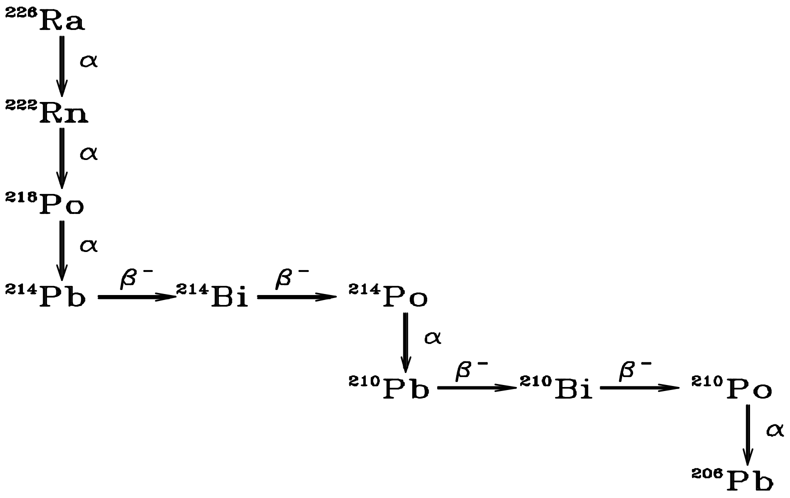


Scheme of ionisation chamber with ^{226}Ra α-particle source

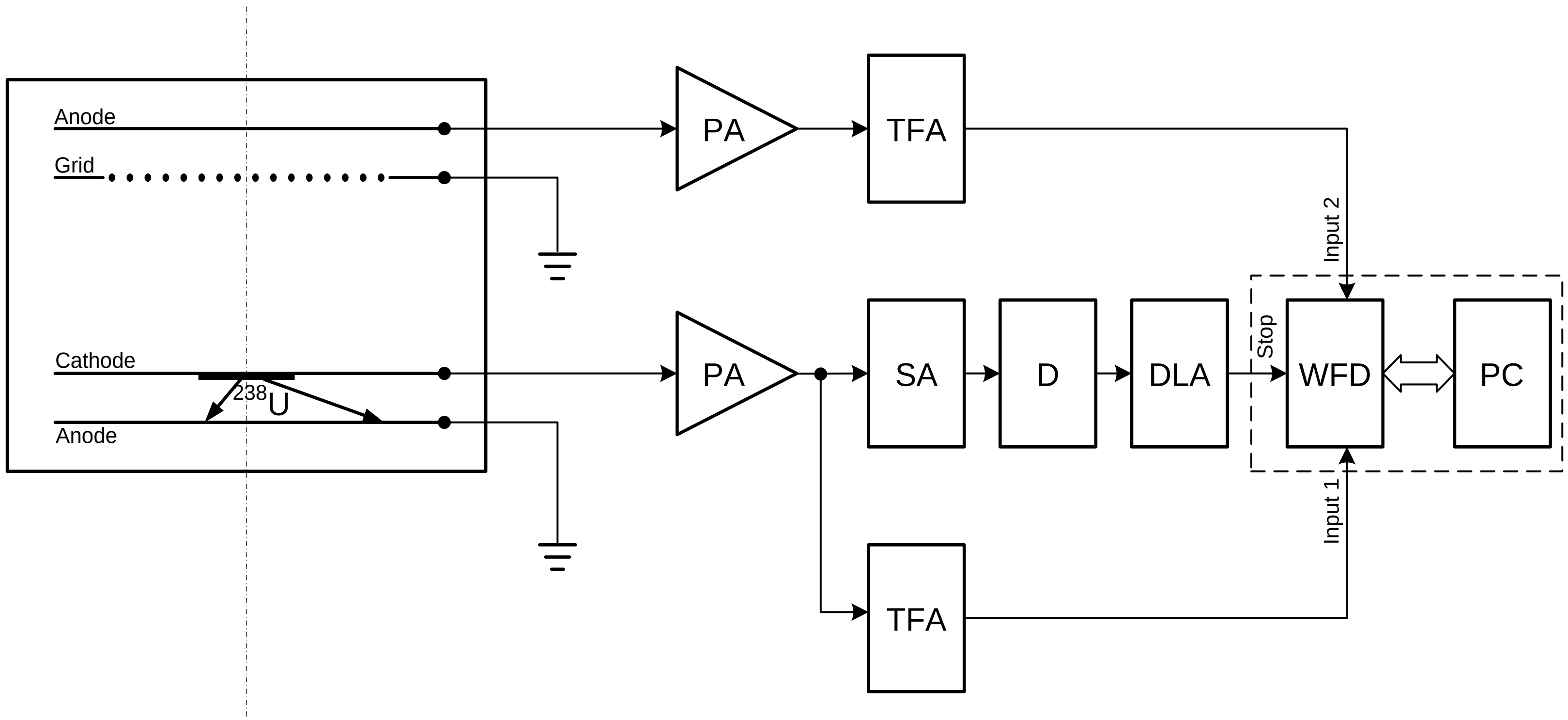


^{226}Ra decay scheme



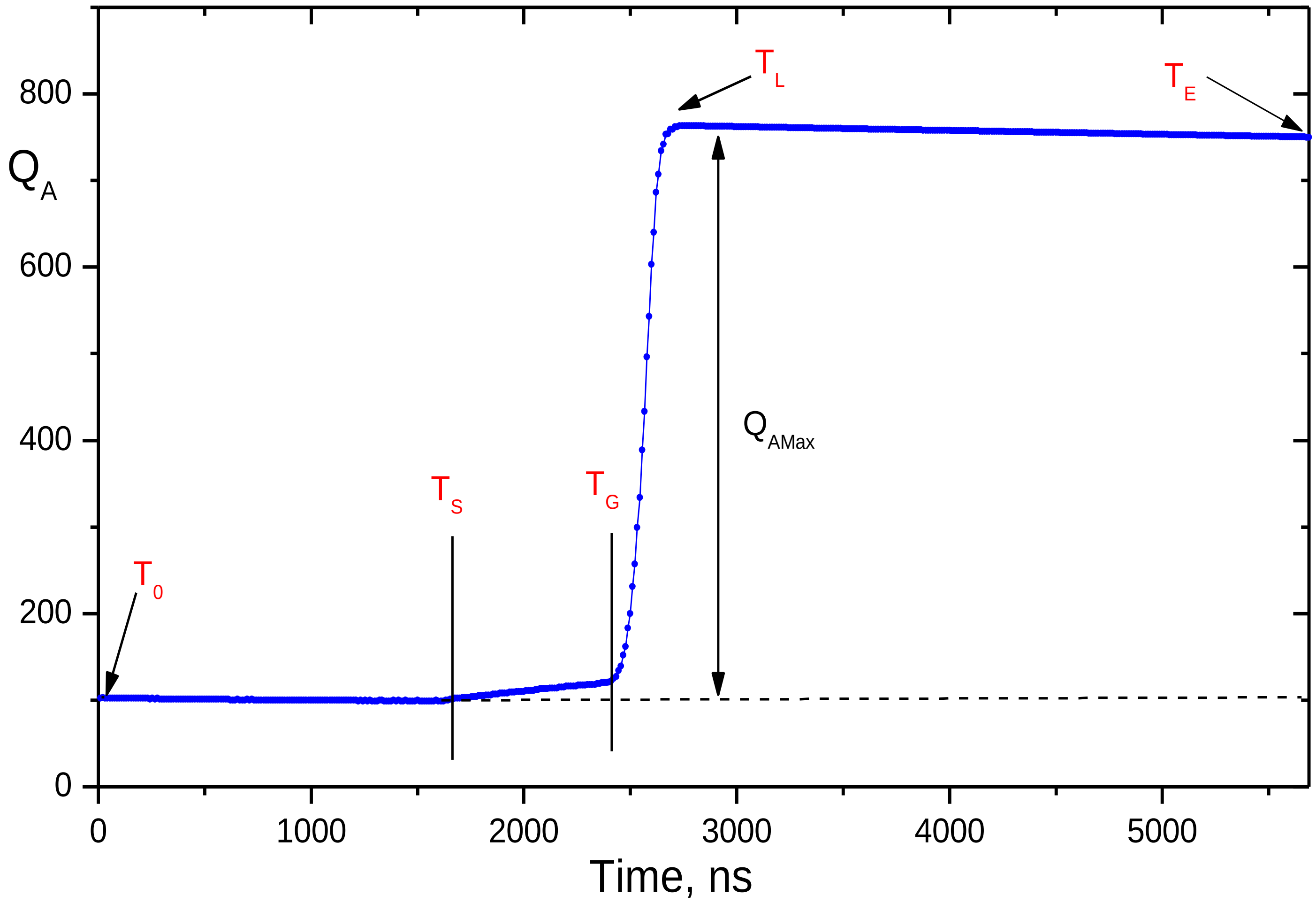


Scheme of the IPPE experimental setup

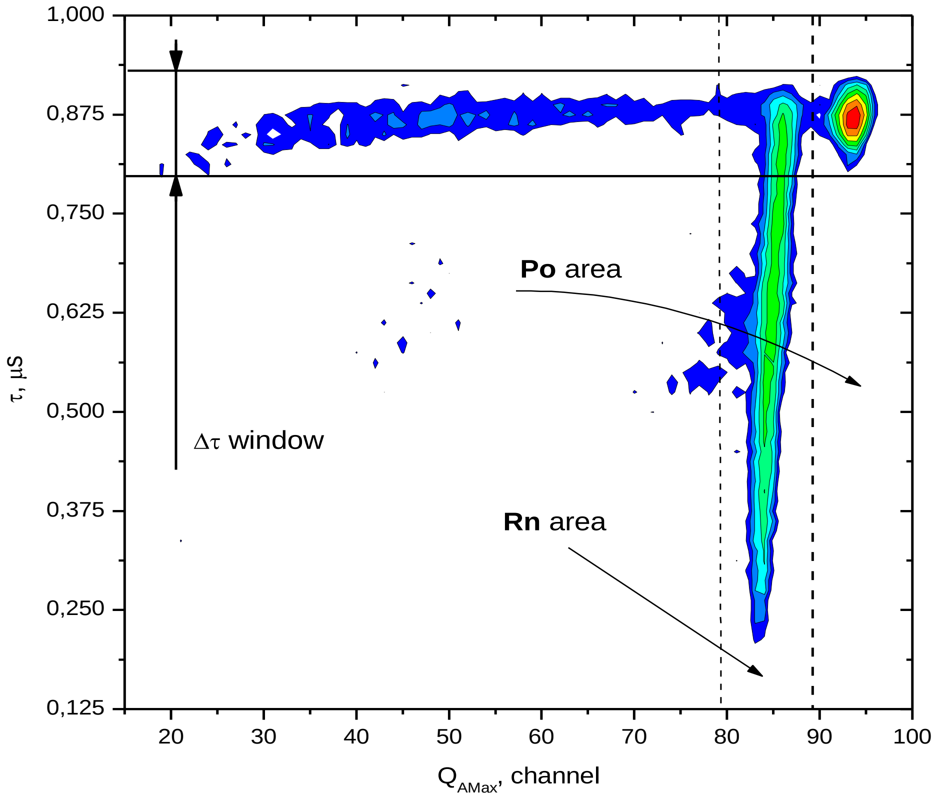


PA – preamplifier, TFA – timing filter amplifier,
D – discriminator,
SA – spectroscopy amplifier, DLA – delay line amplifier,
WFD – waveform digitizer, PC – personal computer.

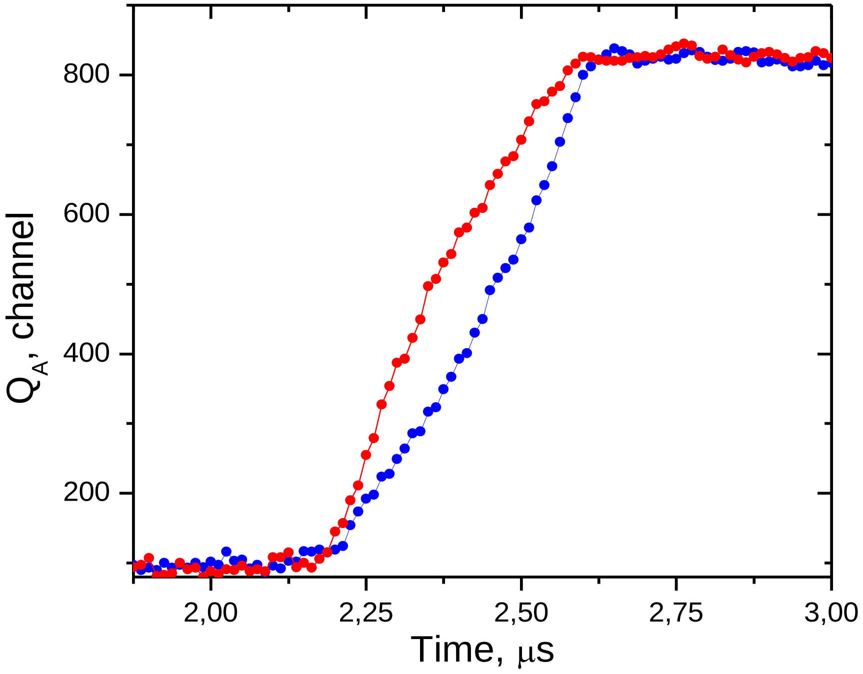
Anode pulse shape

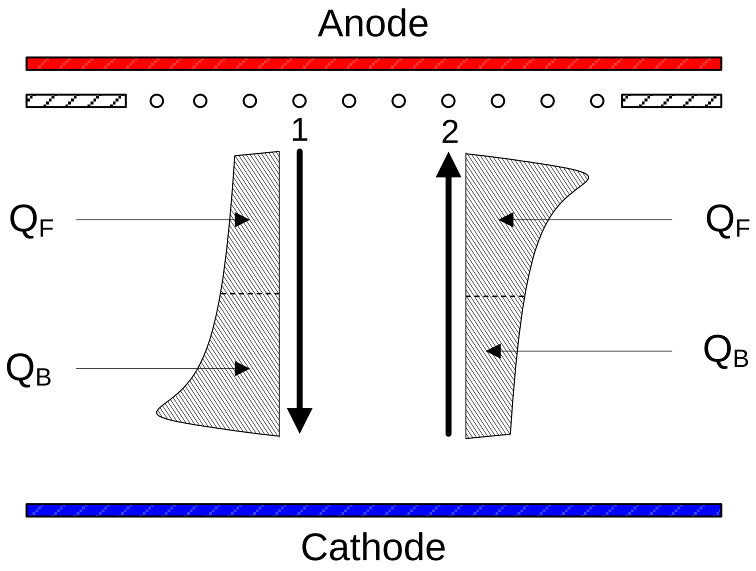


Amplitude of anode pulse vs electron drift time

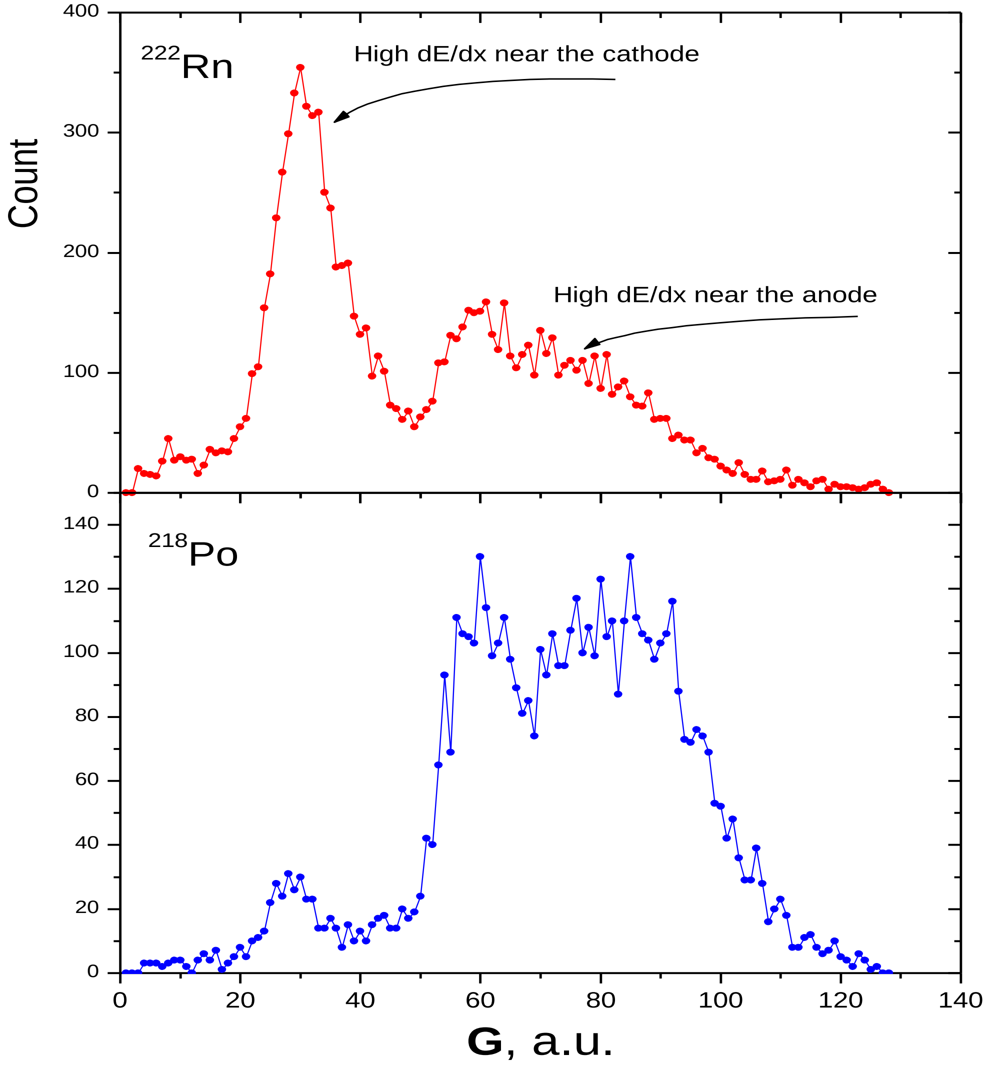


α-particle directionality determination



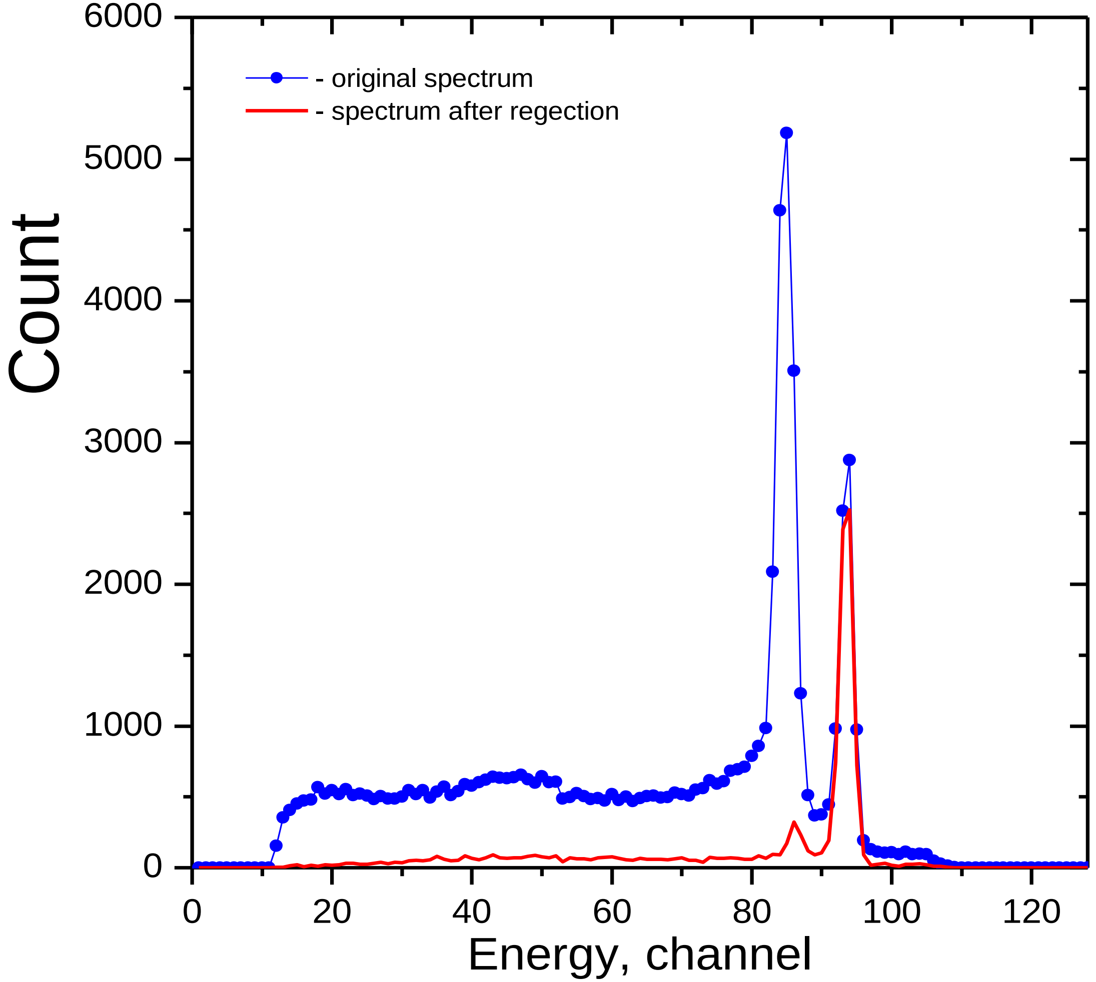


α-particle directionality determination 2



$$G = \frac{\frac{dQ_A(t)}{dt}(begin)}{\frac{dQ_A(t)}{dt}(end)} = \frac{\left(\frac{dE}{dx}\right)_{begin}}{\left(\frac{dE}{dx}\right)_{end}}$$

Result of α-particle selection

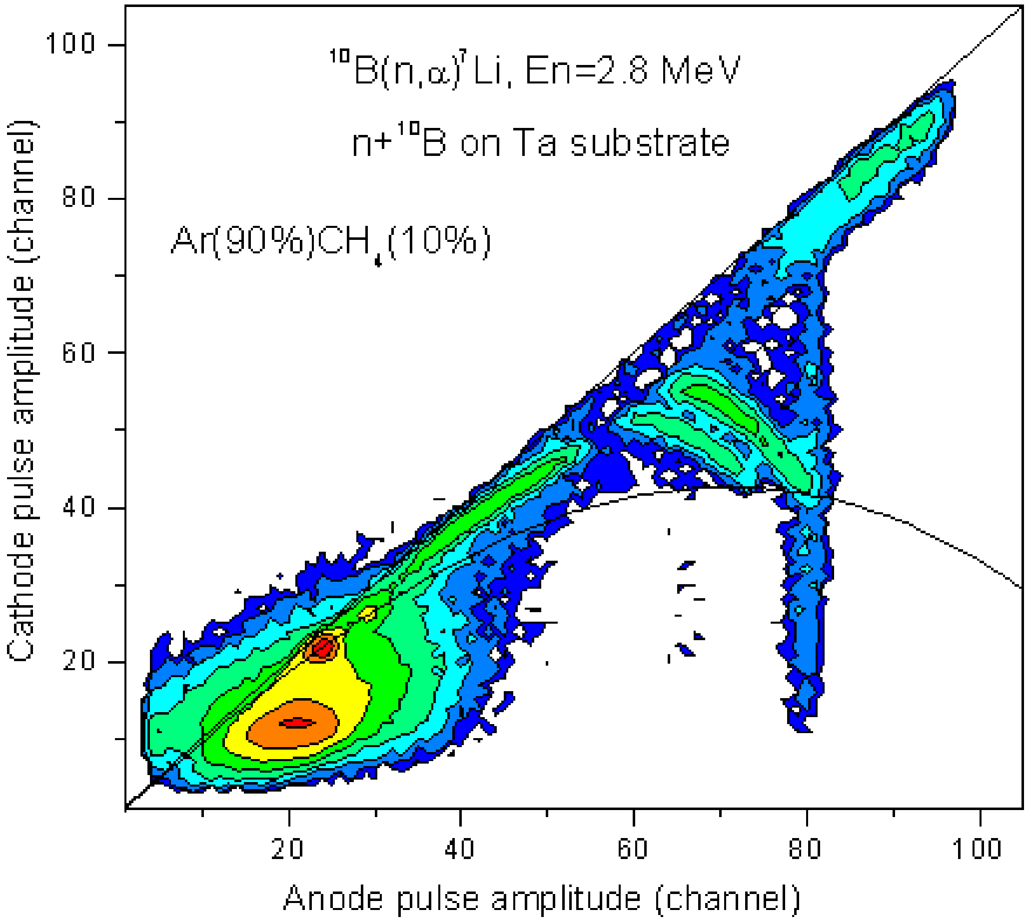


Justification for the light isotopes measurement

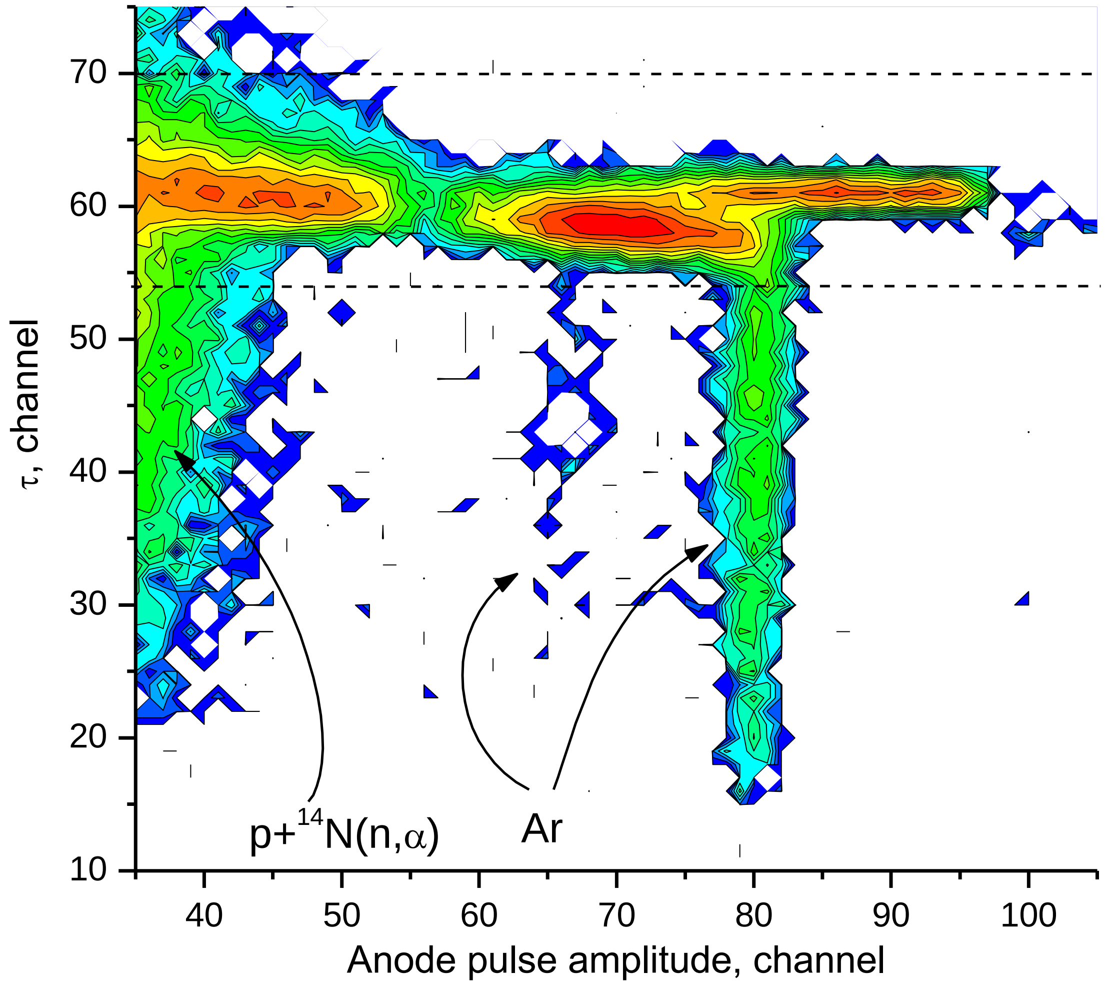
^{6}Li, ^{10}B, ^{12}C, ^{14}N, ^{16}O, ^{19}F, ^{20}Ne

- standards
- reactivity predictions of thermal and fast reactors;
- calculation of helium production in fuel pins and claddings of reactors;
- calibration of the strength of neutron sources;
- astrophysics;
- dosimetry.

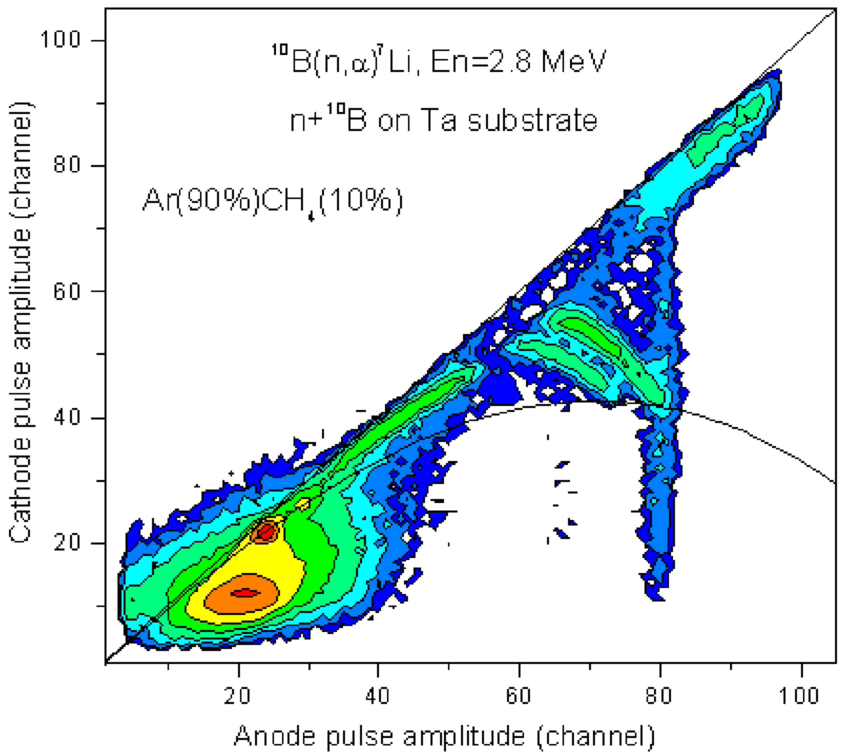
^{10}B. Two-dimensional response function

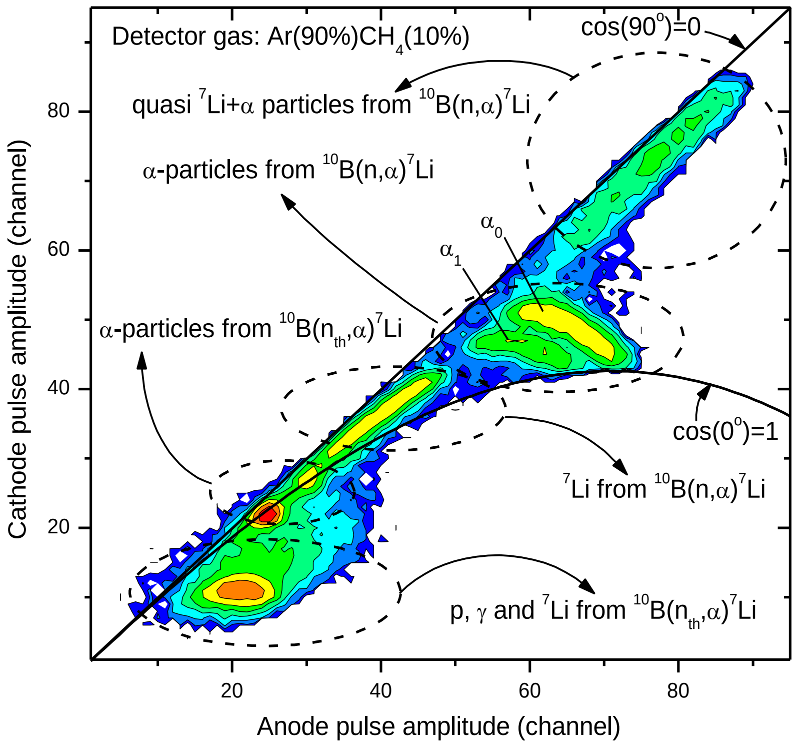


^{10}B. Anode pulse amplitude vs electron drift time

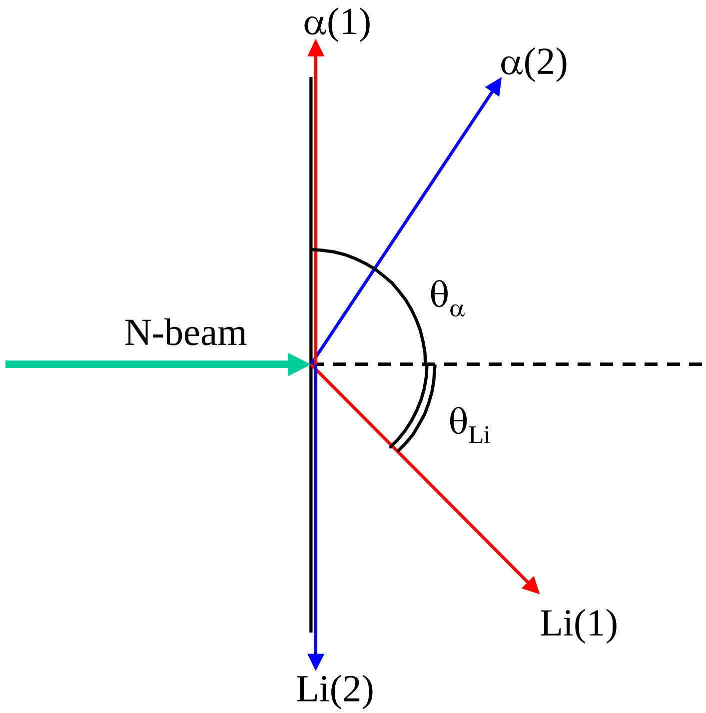


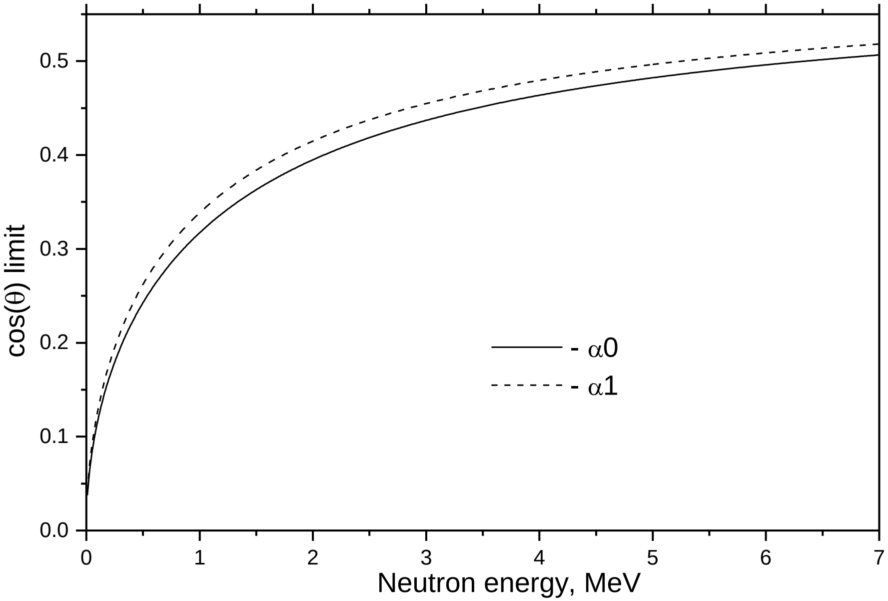
$^{10}B(n,\alpha)^7Li$. Solid target response function



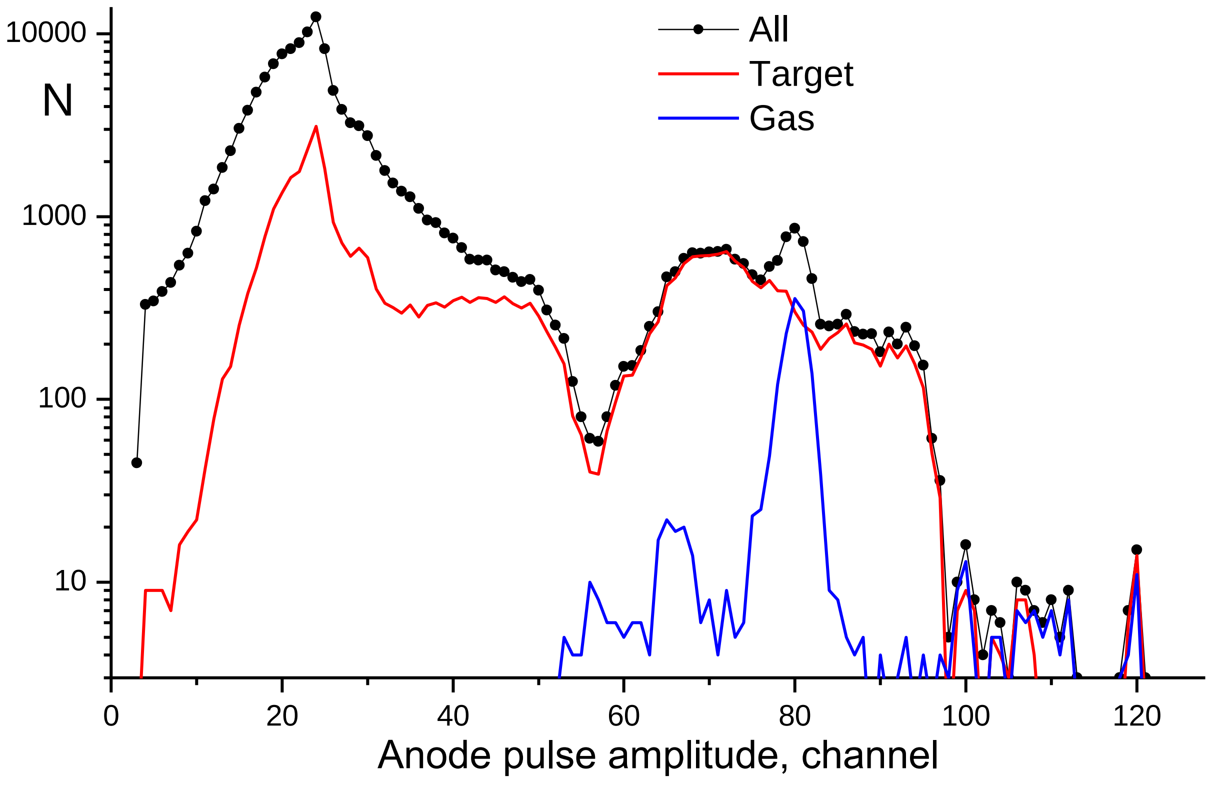


Solid target kinematics peculiarity.

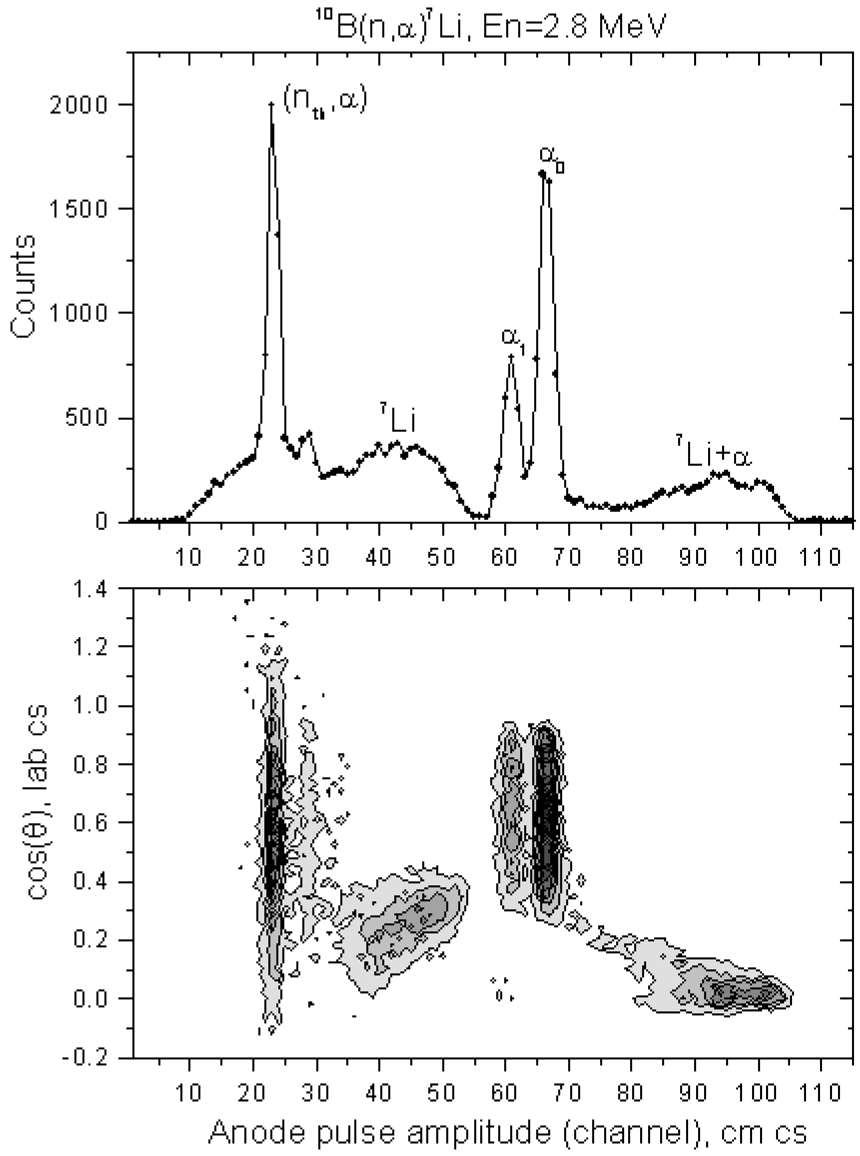




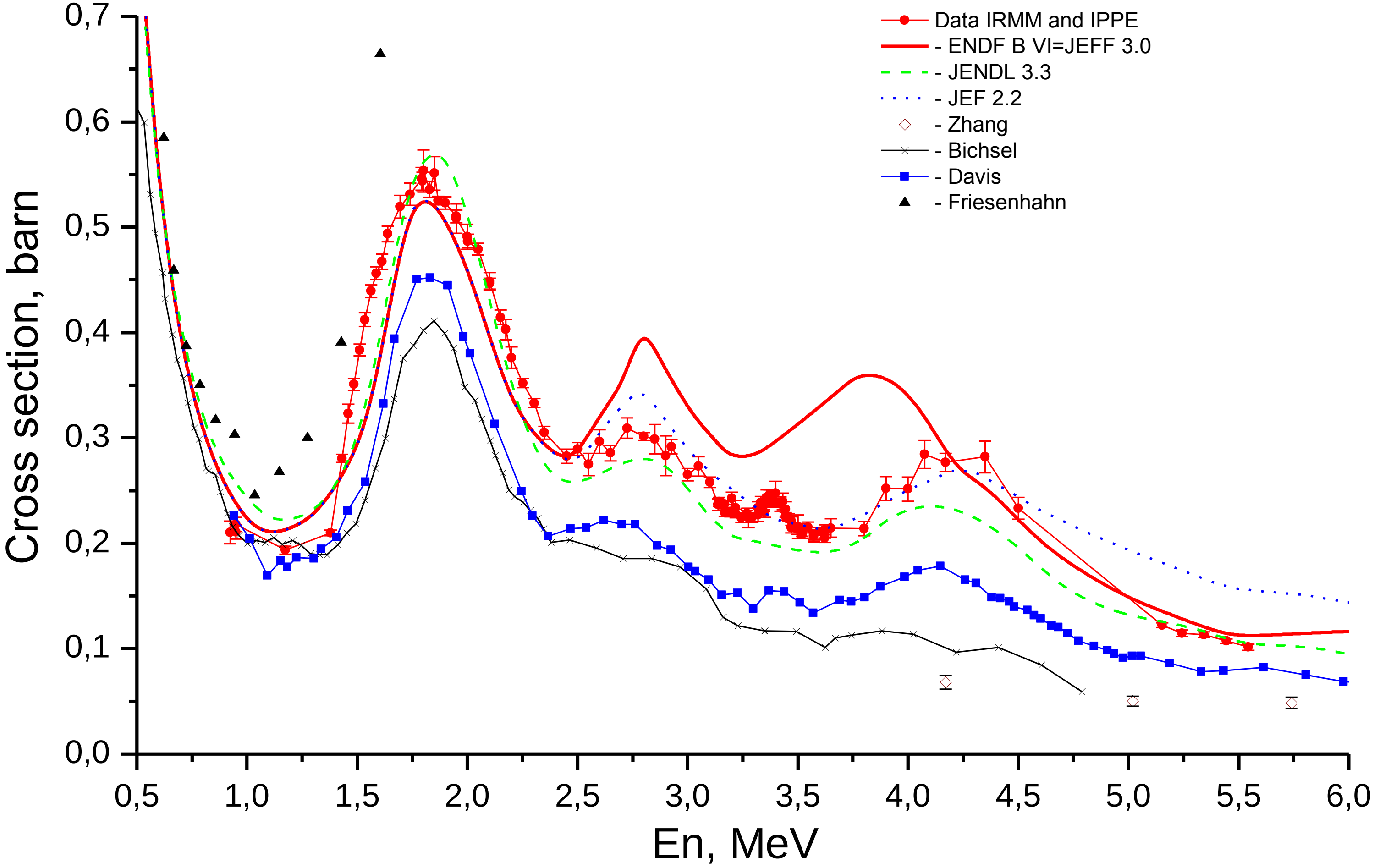
^{10}B. Background suppression.



^{10}B. Spectra after corrections.



$^{10}B(n,\alpha)^{7}Li$ cross-section result



Why is it difficult to measure light elements?

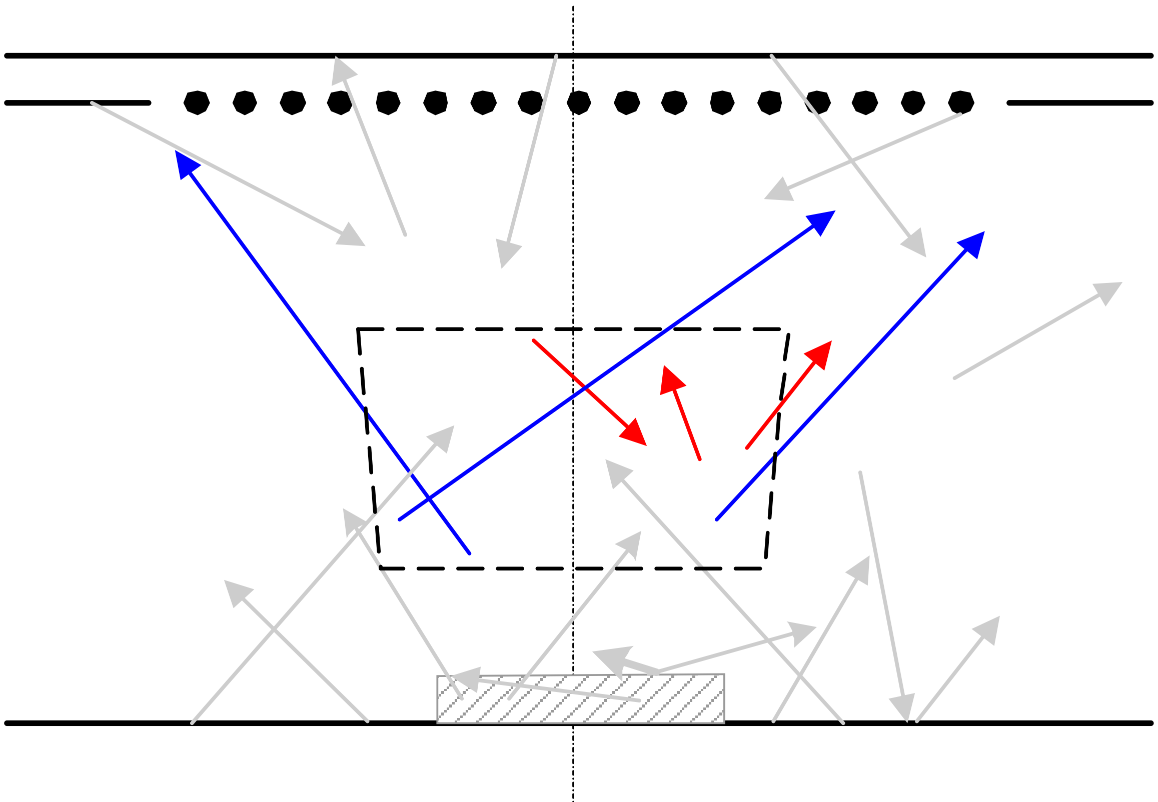
- It is difficult to prepare a clean target
- Low reaction cross section
- The kinematical effect – dependence of anode pulse amplitude from the emission angle.
- Negative Q – value. Background from (n,α), (n,p) reaction, elastic recoil at working gas.
- Background of a detector.
- Light elements from the air (O, N) are present on the electrodes surface.
- Fine structure in cross section.

Problems of a solid target

- Number of atoms is limited by self-absorption;
- No radioactive isotope number of atoms determination problem.
- Energy losses in the target;
- The particle leaking effect.
- Background from reactions on working gas components and chamber electrodes.
- Problem with solid target preparation for some elements (e.g. noble gases)

Gaseous target

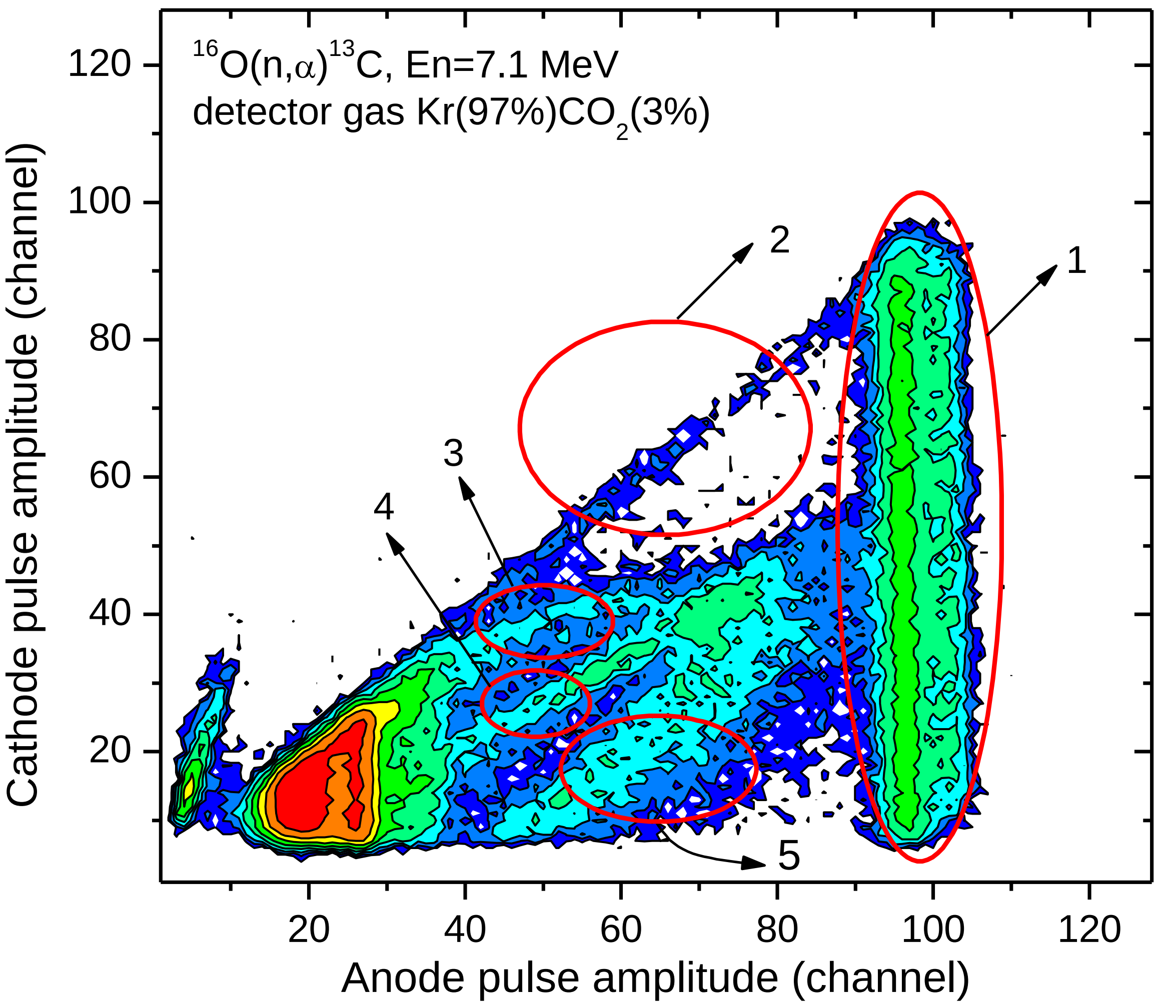
Kr+CO_2, Kr+BF_3, Kr+CH_4, Kr+N_2.....



Gaseous target properties

- Number of atoms in gaseous target is limited only by chamber size and working gas pressure;
- For events taking place on the working gas components we can register both alpha particle and residual nuclear. Signal to noise ratio will be better.
- Anode pulse amplitude is proportional to sum of alpha particle and residual nuclei energies. $P_A \sim E_\alpha + E_R = E_n + Q$.
- There are no energy losses in target.
- There are no the particle leaking effect

Two-dimensional response function of gaseous target



Identified signatures of:

$^{16}O(n,\alpha_0)^{13}C$ signal from the detector gas (1),

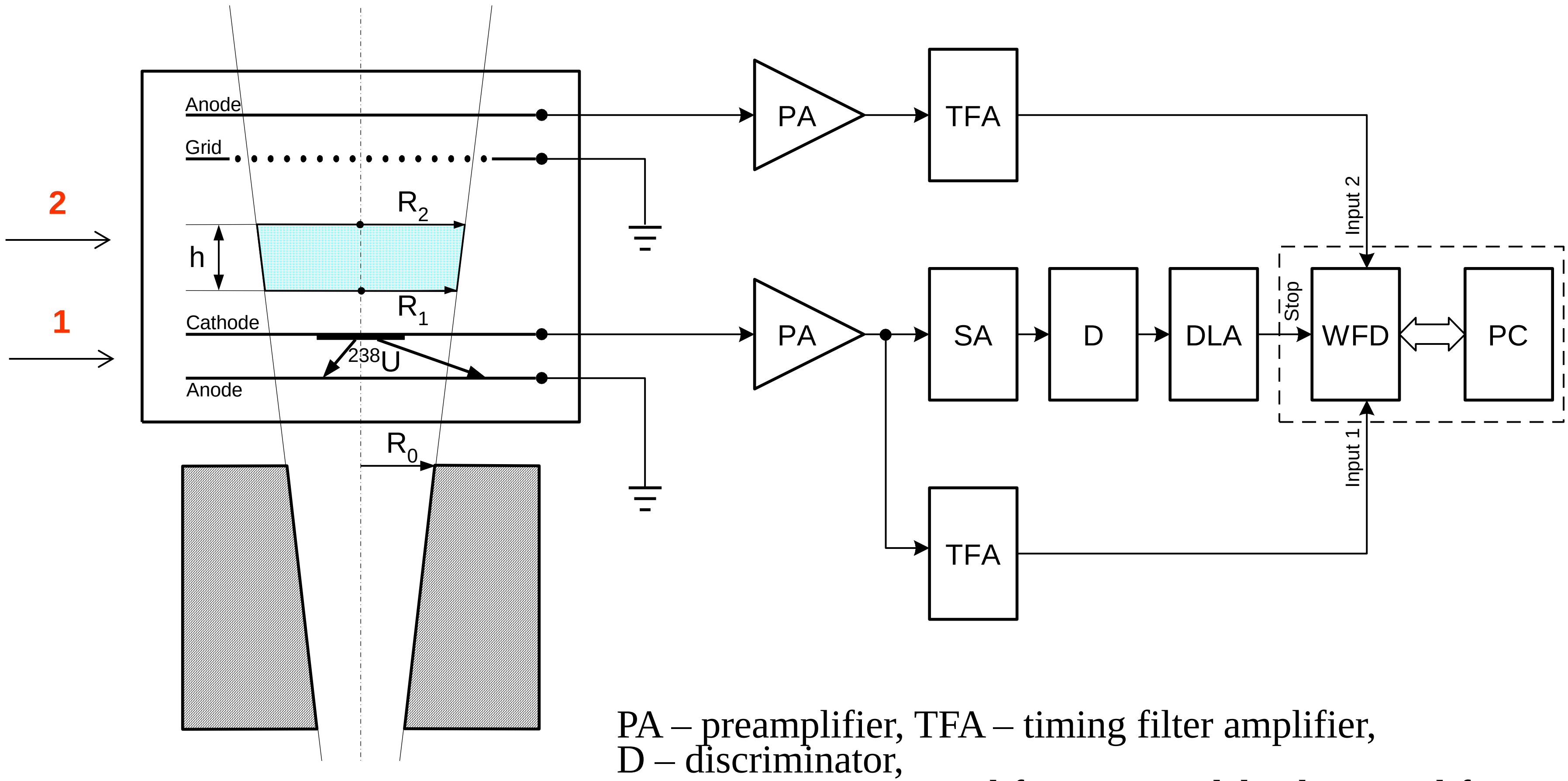
alpha particle background of the cathode (2),

background of protons emitted by the cathode that stopped in the detector gas (3),

background of protons emitted by the cathode which crossed the grid (4),

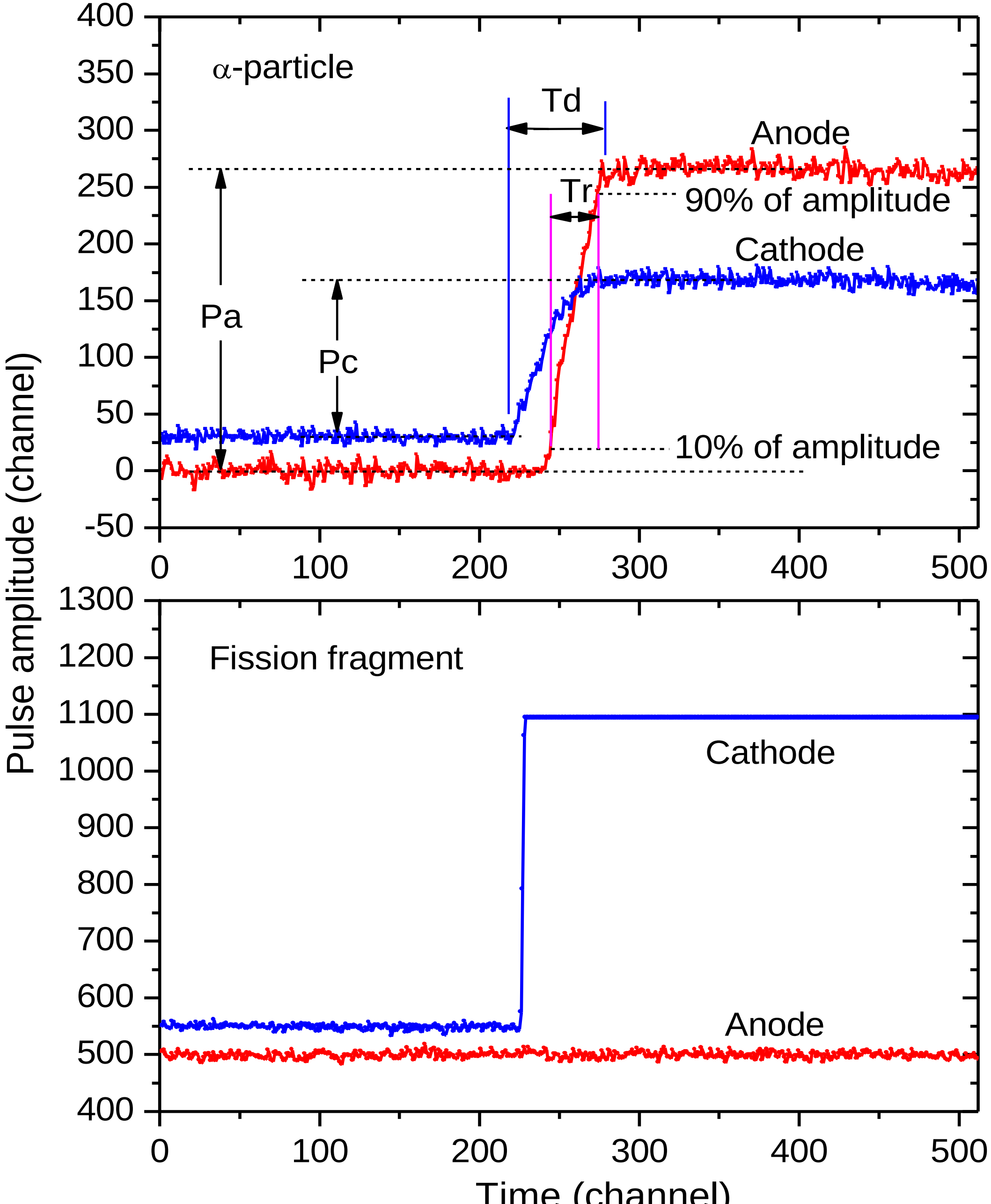
Protons background of the detector gas (5).

Scheme of the experimental setup



PA – preamplifier, TFA – timing filter amplifier,
D – discriminator,
SA – spectroscopy amplifier, DLA – delay line amplifier,
WFD – waveform digitizer, PC – personal computer.
1-monitor chamber; 2-main chamber.

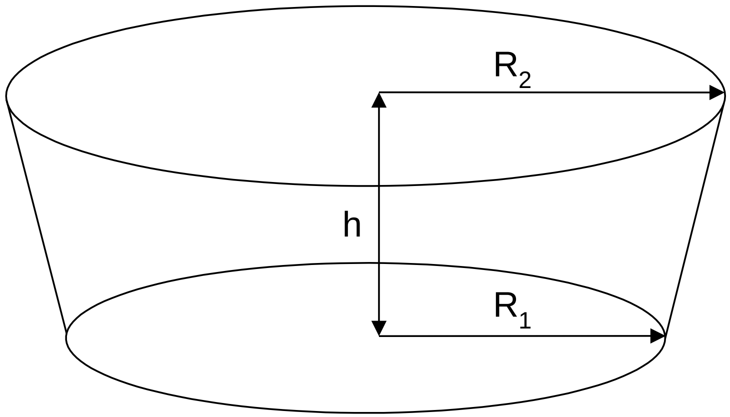
Examples of signals of the main chamber and monitor chamber



DSP allows you to analyse:

1) Amplitude of anode pulse;
2) Amplitude of cathode pulse;
3) Time when anode signal appeared;
4) Time when anode signal reached the satiation;
5) Time when cathode signal appeared;
6) Time when cathode signal reached the satiation;
7) Ionisation distribution along the particle track. (Anode signal shape).

Geometry of the gaseous target



$$V = \frac{\pi h}{3}(R_1^2 + R_1 R_2 + R_2^2)$$

One of the most crucial points of this technique was precise determination of the radii of the truncated cone's bases.

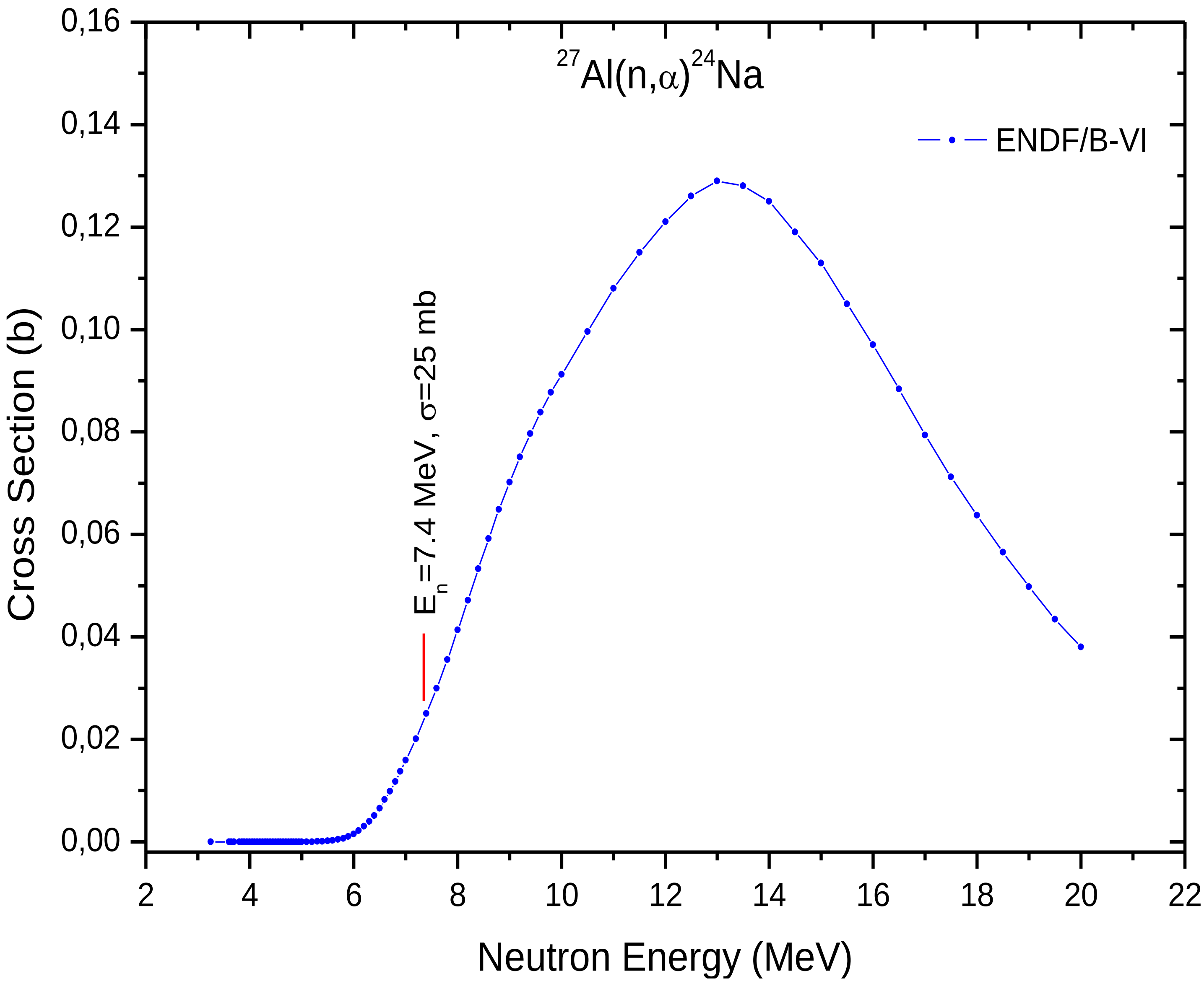
Investigation of the neutron beam profile was needed.

Indium or aluminium stripes and disks were used to measure the profile of the collimated neutron beam at energies 2.5 and 7.4 MeV, respectively.

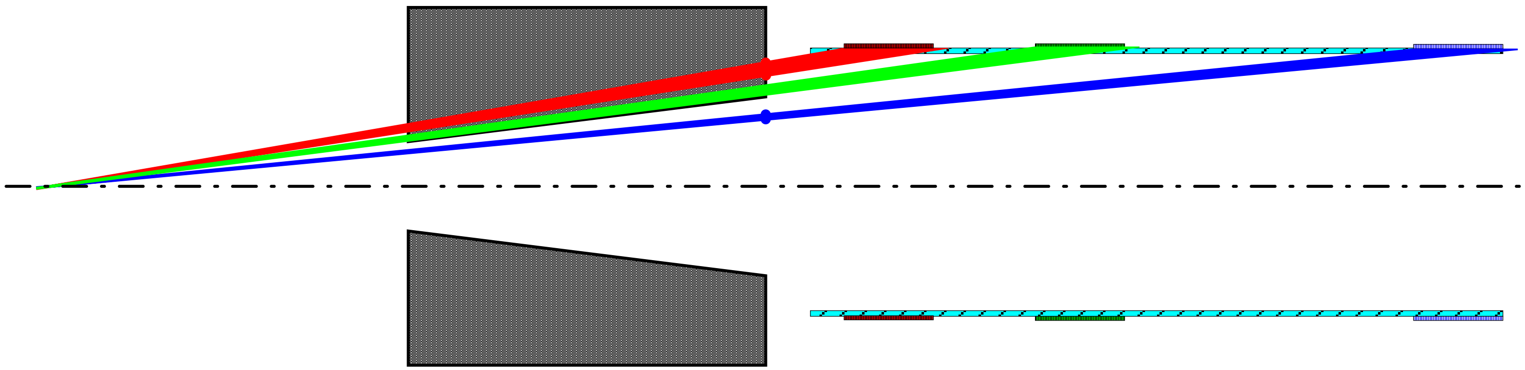
N_{Oxygen}= 2.464*10^{20} nuclei at V=5.6346·10^{-5} m^3

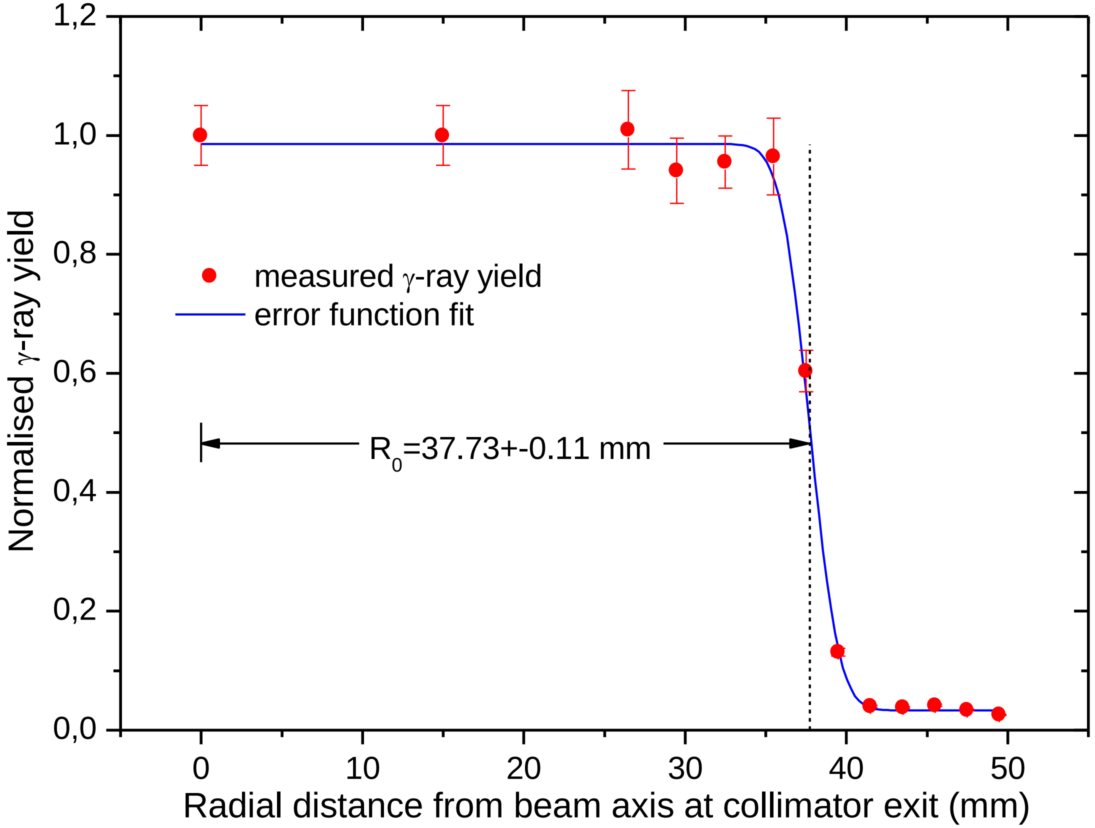
N(^{238}U) atoms in the monitor= 6.831*10^{18} (solid target ~500 mkg/cm^2)

Cross section of $^{27}Al(n,\alpha)^{24}Na$ reaction

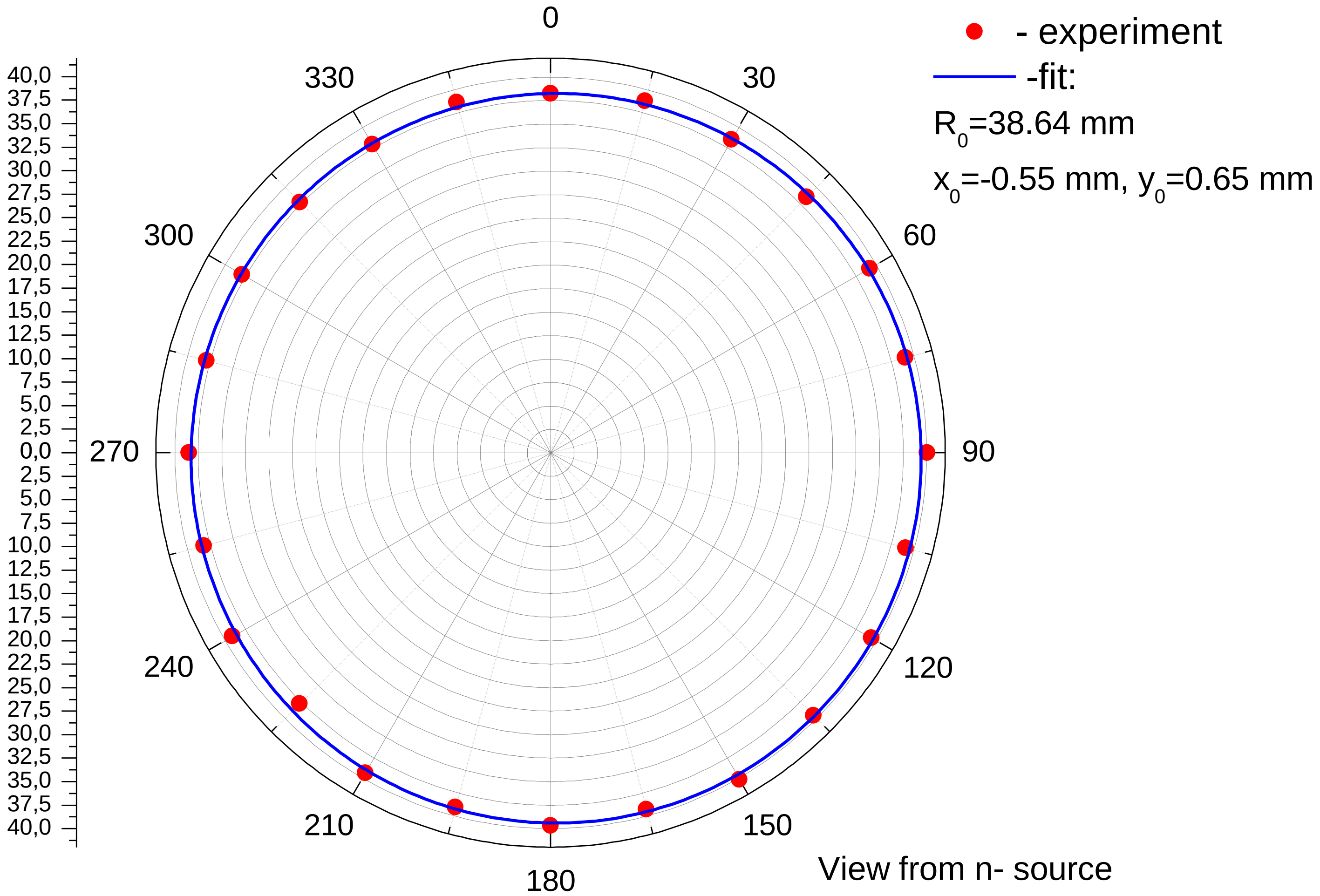


Scheme of the experiment of neutron beam profile measurement

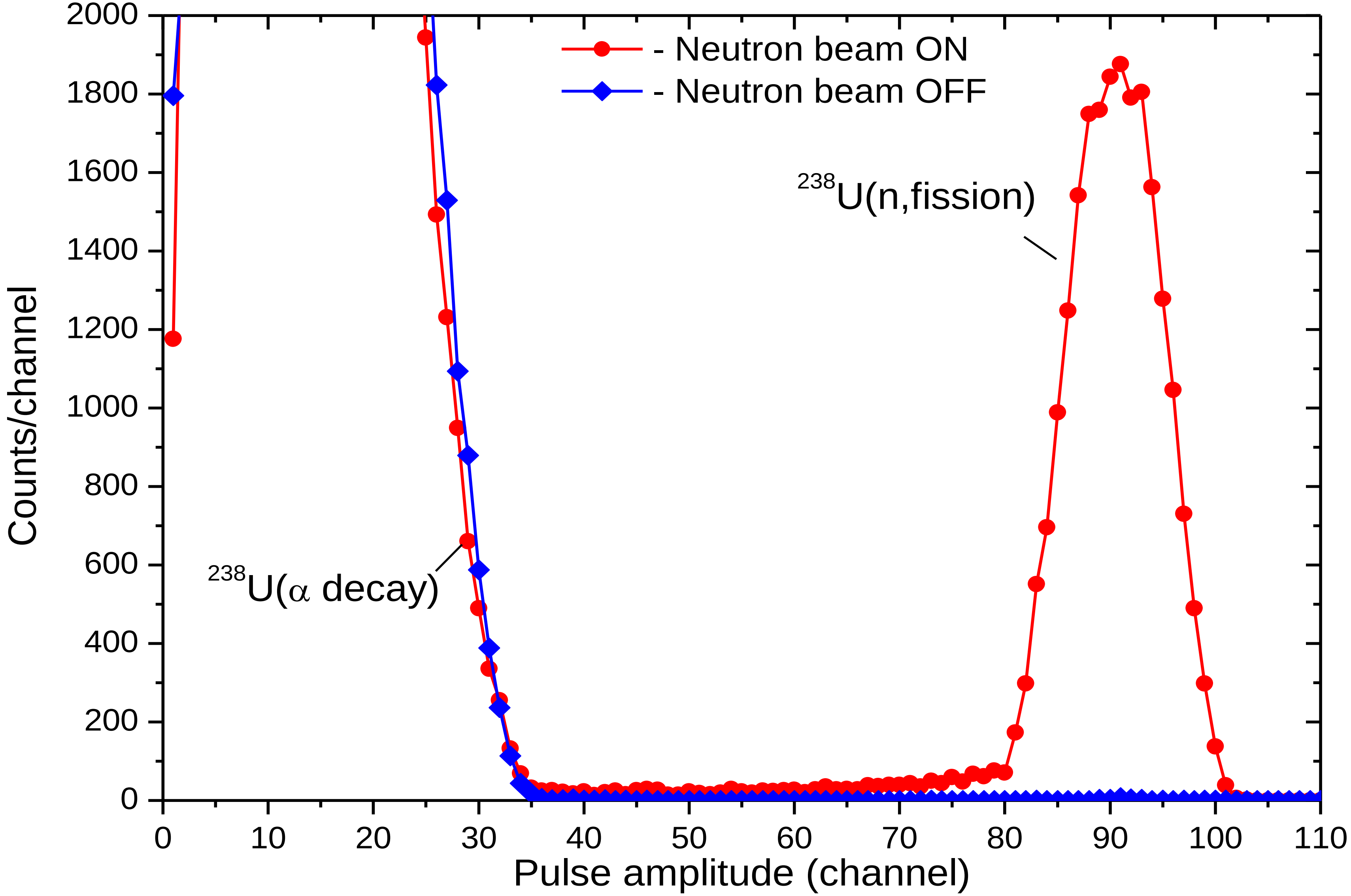




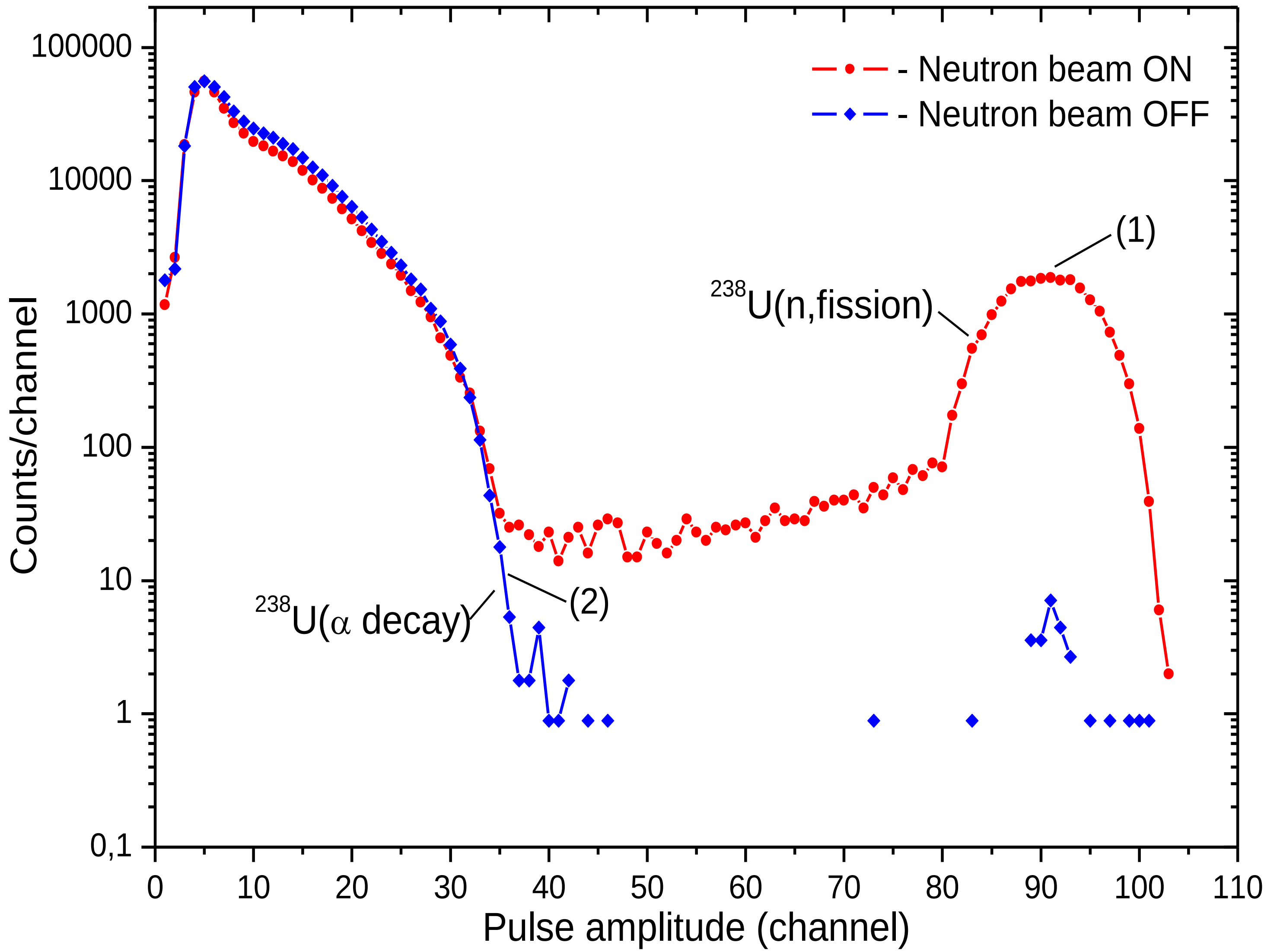
Rotational symmetry of the neutron beam at En=7.4 MeV



Pulse height spectrum of ^{238}U neutron monitor measured with: neutron beam on, and neutron beam off



Pulse height spectrum of ^{238}U neutron monitor measured with: neutron beam on (1), and neutron beam off (2)



Energy spectrum of α-particles

Nuclear Physics 48 (1963) Disintegration of ^{16}O and ^{12}C by fast neutrons. E. A. Davis, T. W. Bonner, D. W. Worley, and R. Bass

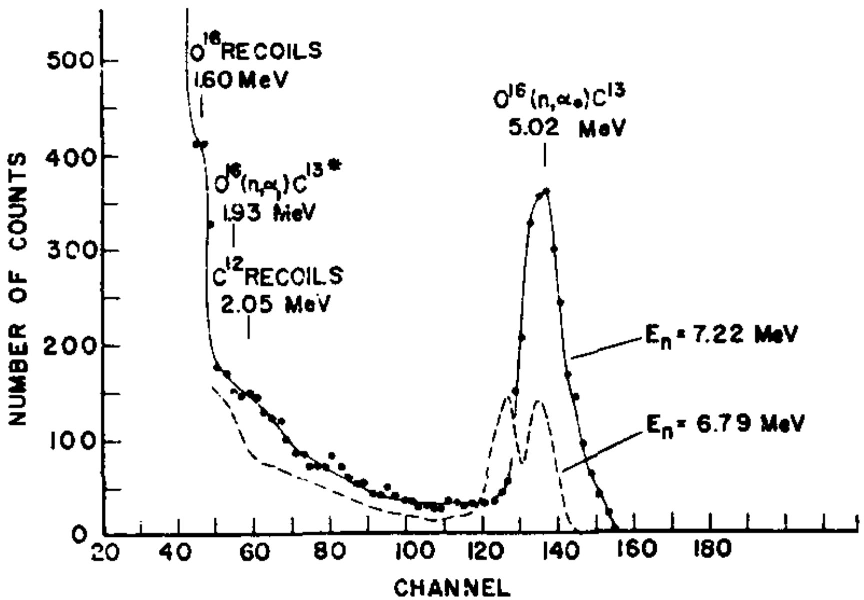
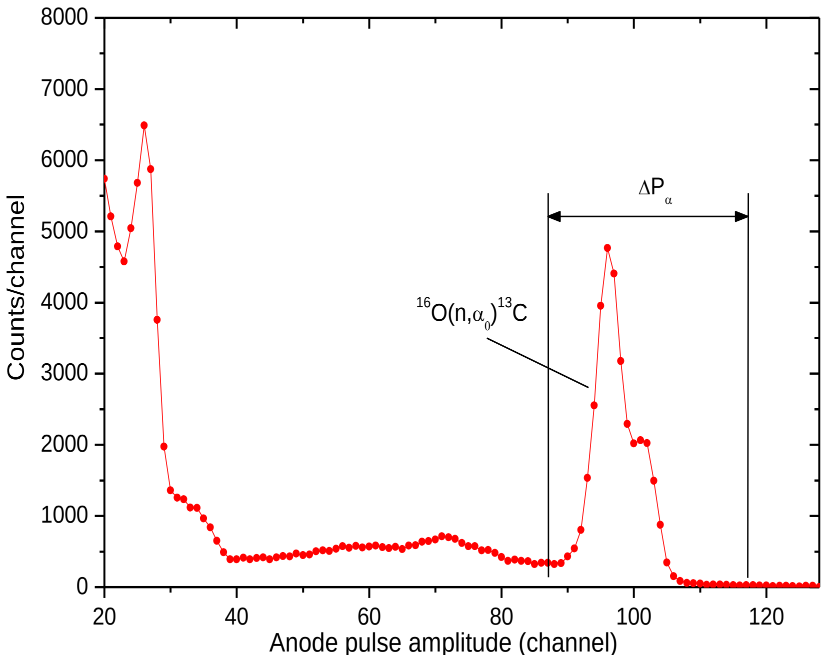
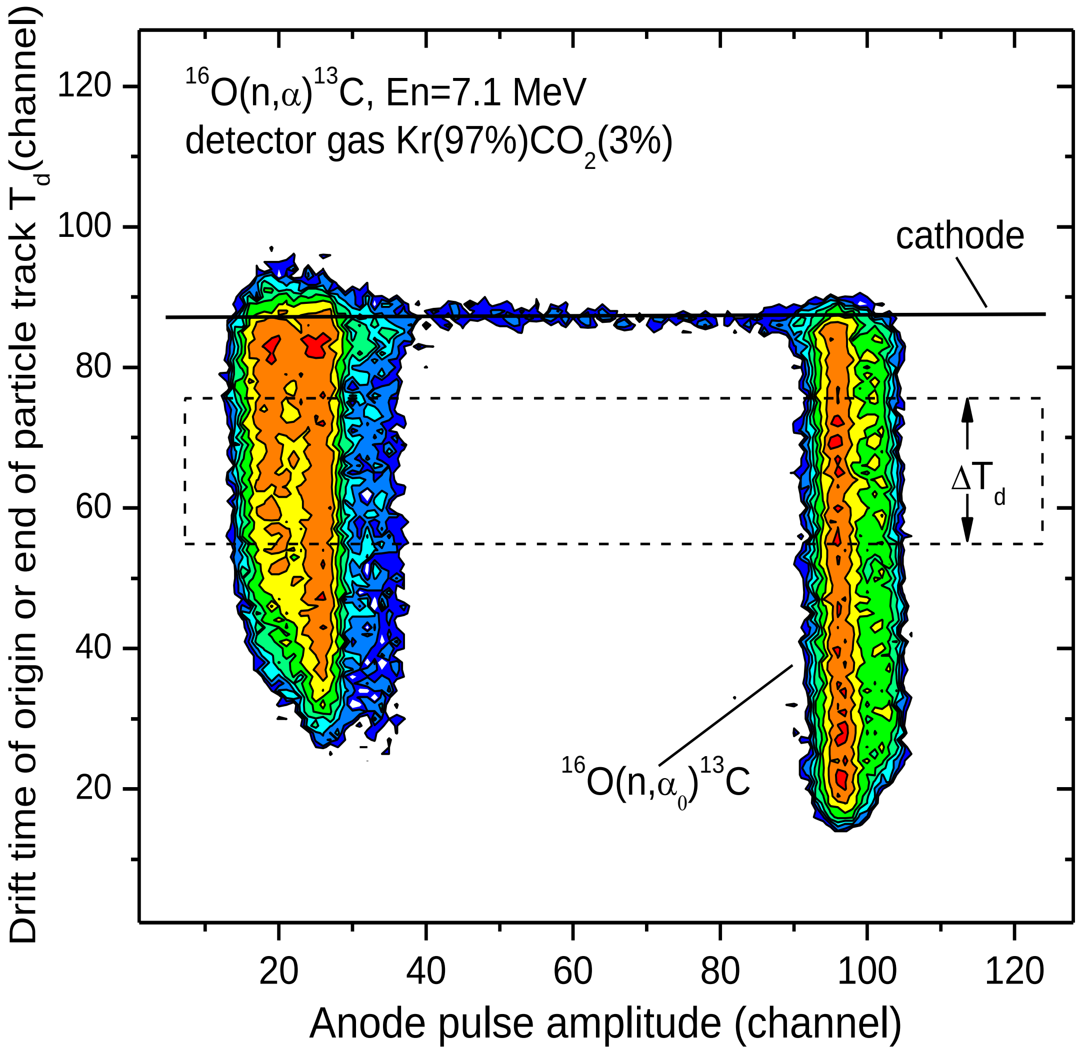


Fig. 1. Pulse-height distribution from disintegration of carbon dioxide by 7.22 MeV neutrons. The dashed curve is the distribution obtained with 6.79 MeV neutrons.

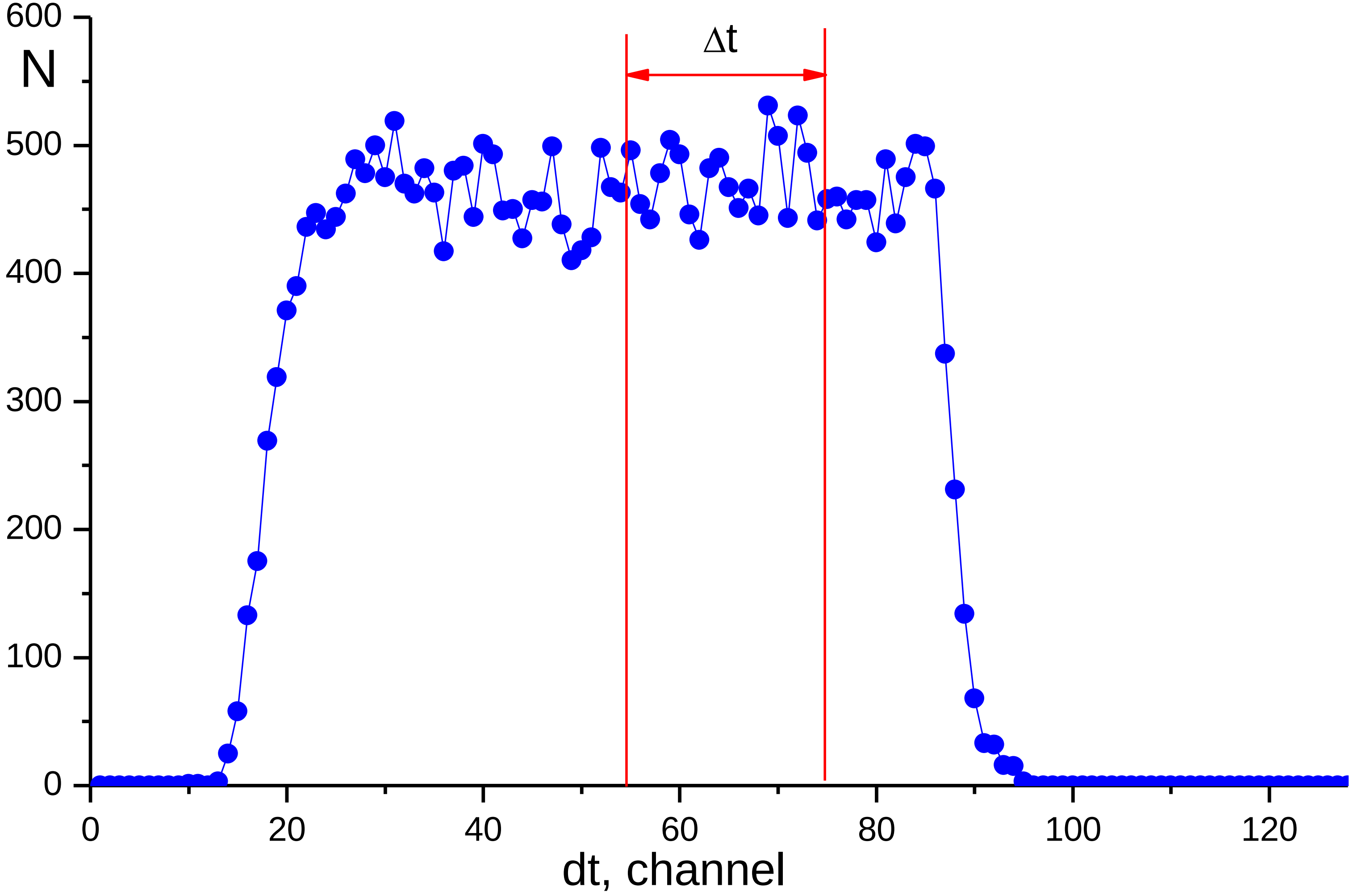
Present work



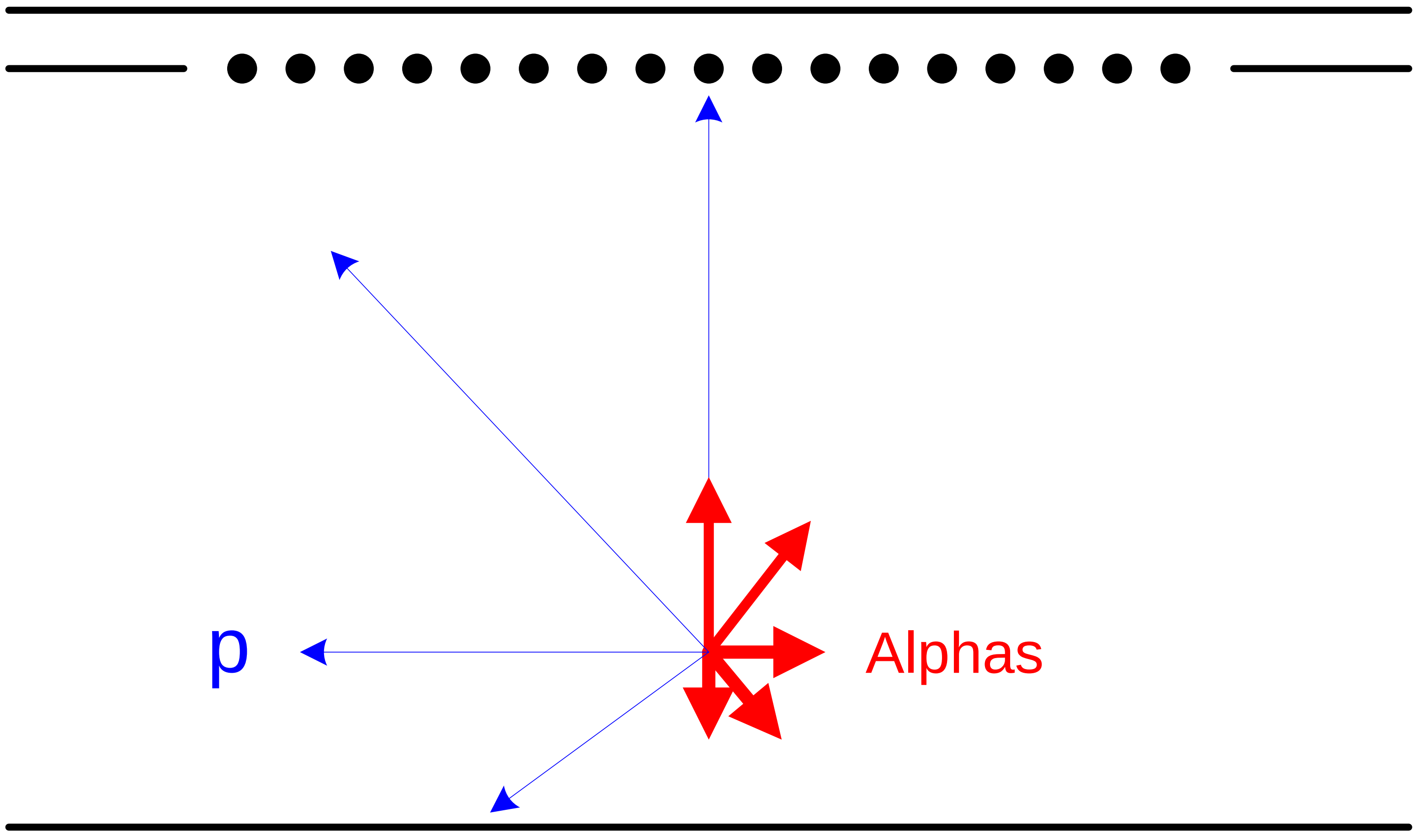
Two-dimensional spectrum of drift time of the end of the particle track versus the anode pulse amplitude. The dashed rectangle defines the region of interest for final analysis. The drift time window ΔT_d determines the height of the effective volume of the gaseous target Δx



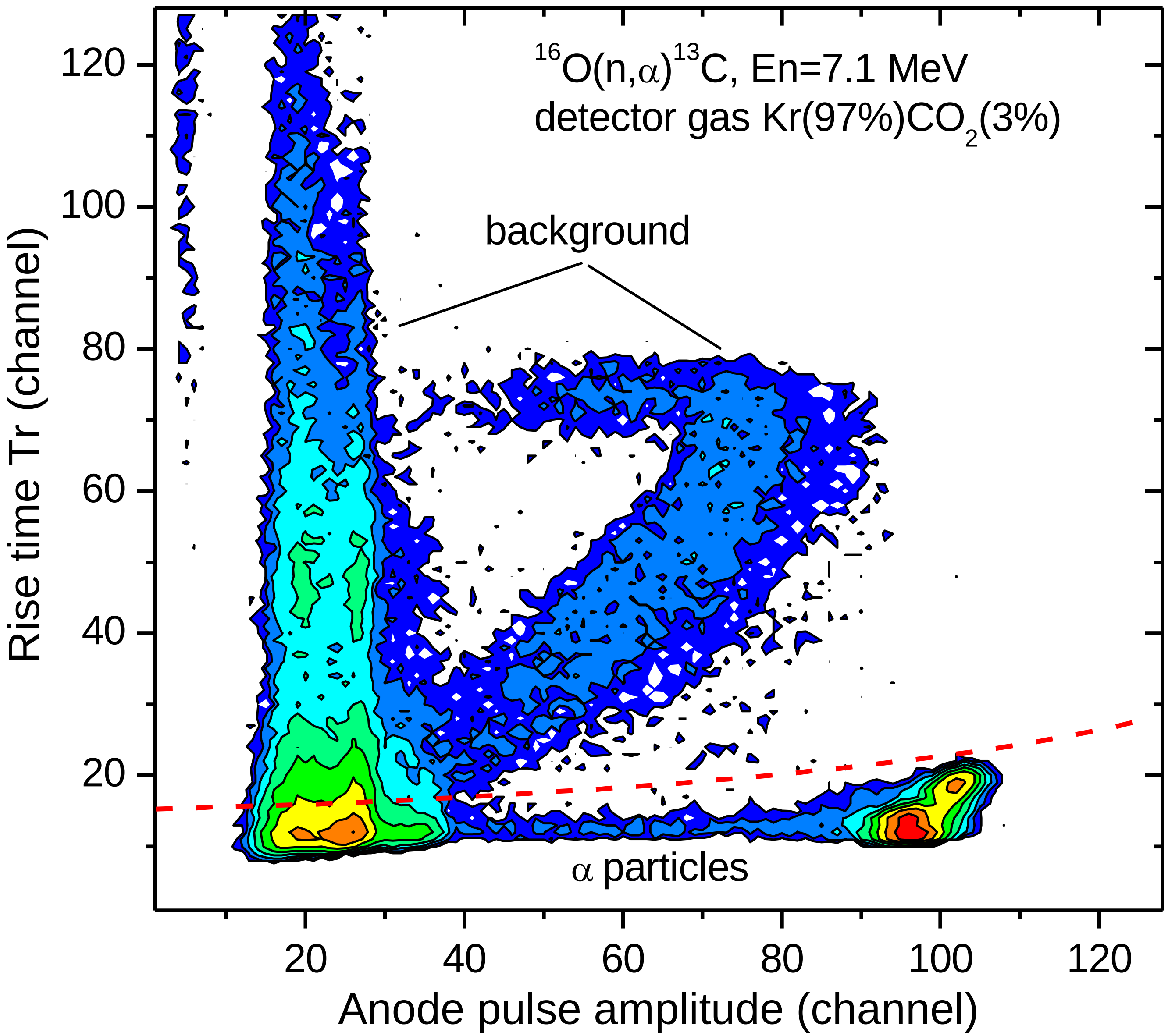
Δt distribution for $^{16}O(n,\alpha)$ α-particles



Method of type of particle determination

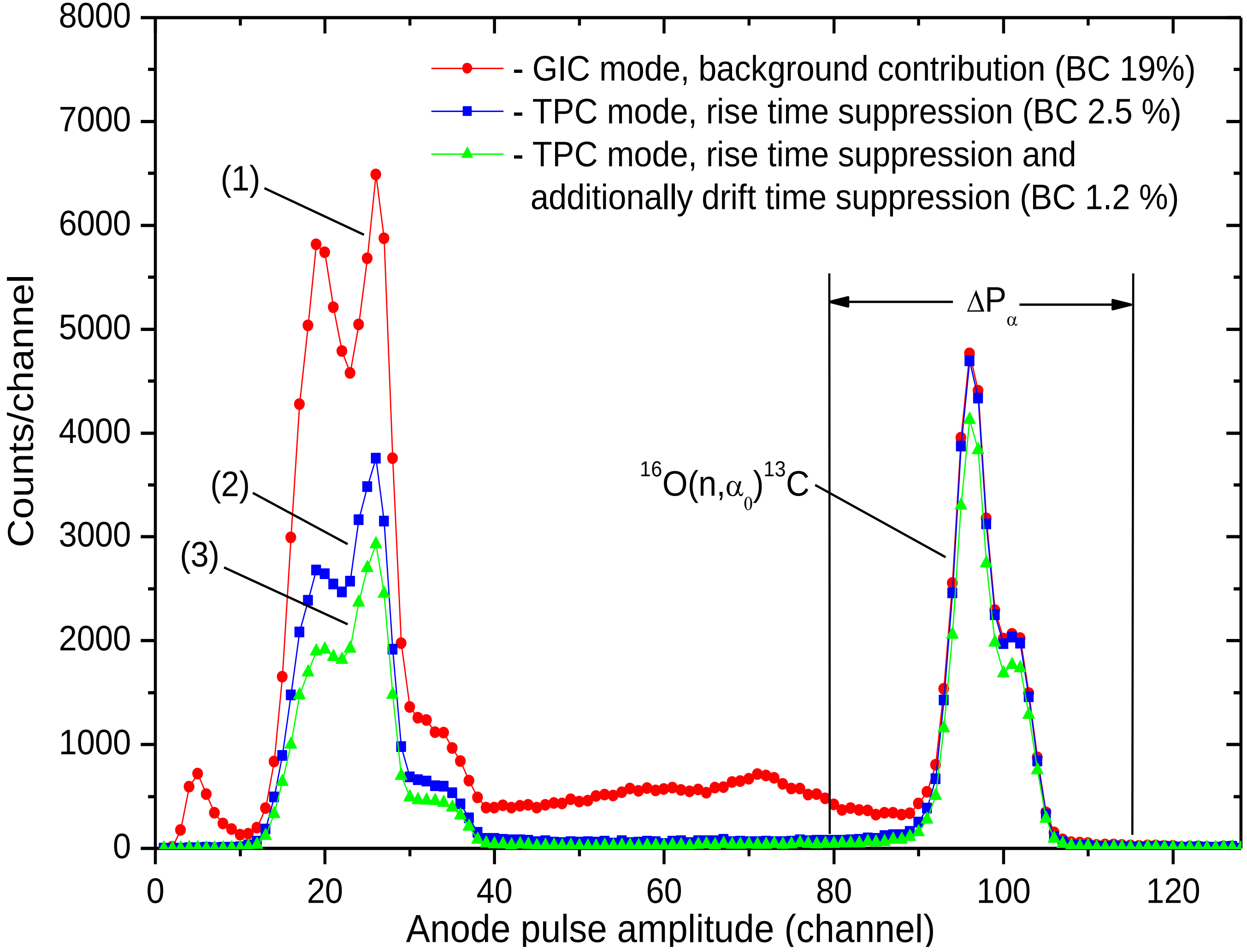


Two-dimensional spectrum of rise time versus amplitude of anode pulse with the dashed line separating α particles from background



Energy spectrum of α-particles

Pulse height spectrum in: GIC mode (1), TPC mode after rise time suppression of background (2), TPC mode after rise time and drift time suppression of background (3). The percentages in brackets show the background contribution (BC) to the α particle line within the pulse height window ΔPα for the three different operation modes of the spectrometer



Justification for the $^{16}O(n,\alpha)^{13}C$ measurement

AEN NEA Agence pour l'énergie nucléaire Nuclear Energy Agency

OECD OCDE

NEA Nuclear Data Request List

HPRL-Main Search New request template New request guidelines Related references

Request ID	2		Status of the request	High Priority request	
Target	Reaction and process	Incident Energy	Secondary energy or angle	Requested (1 sigma) Accuracy	Covariance
8-O-16	(n,a) SIG	2.5 MeV-10.0 MeV		5%	Y
Field	Subfield	Date Request created	Date Request accepted	Ongoing action	
Fission	LWR, Material recycling	21-SEP-05	11-MAY-06	Y	

Send a comment on this request to NEA

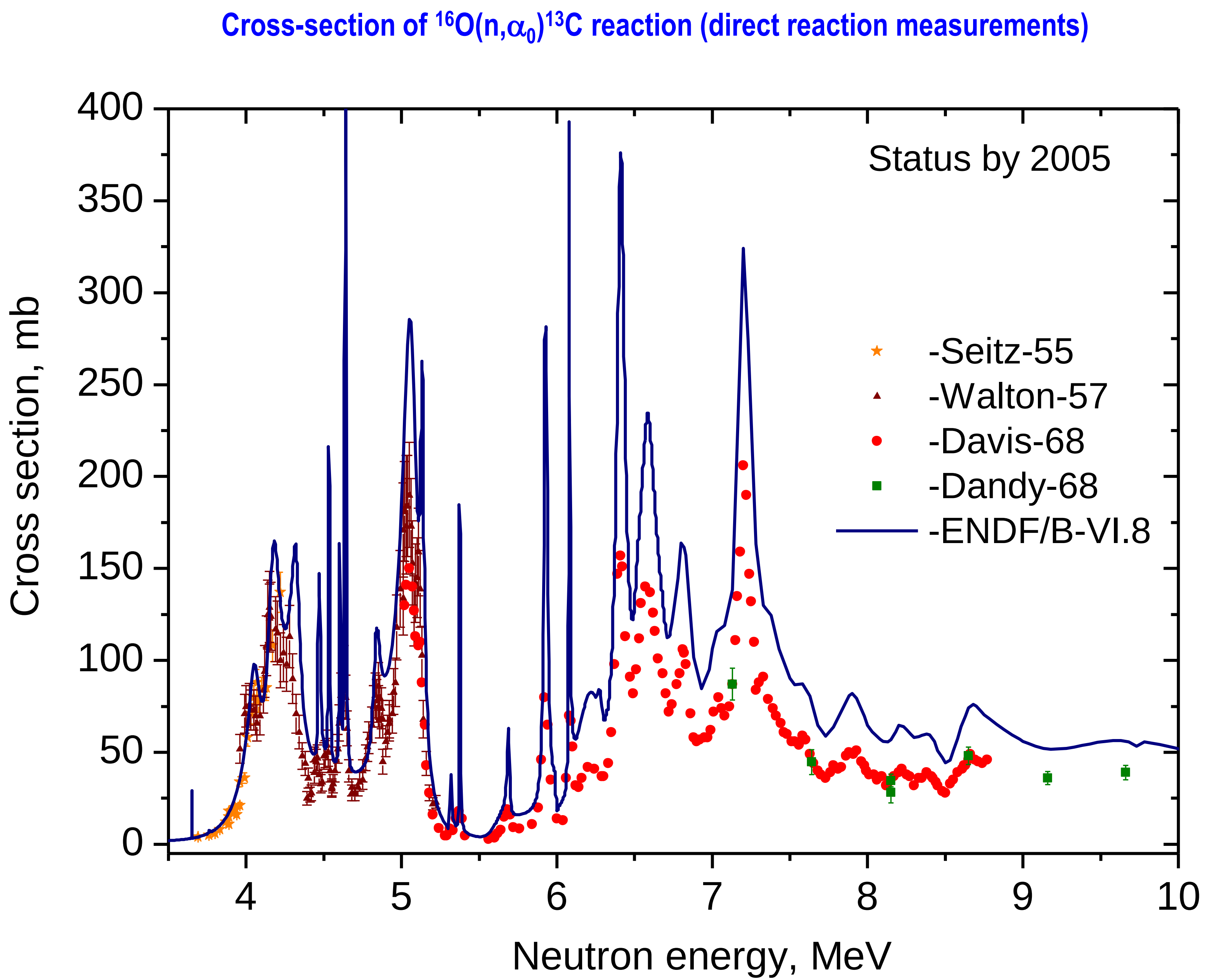
Requester: Mr. Arnaud COURCELLE at CADARACHE, FR
Email: arnaud.courcelle@cea.fr

Impact:

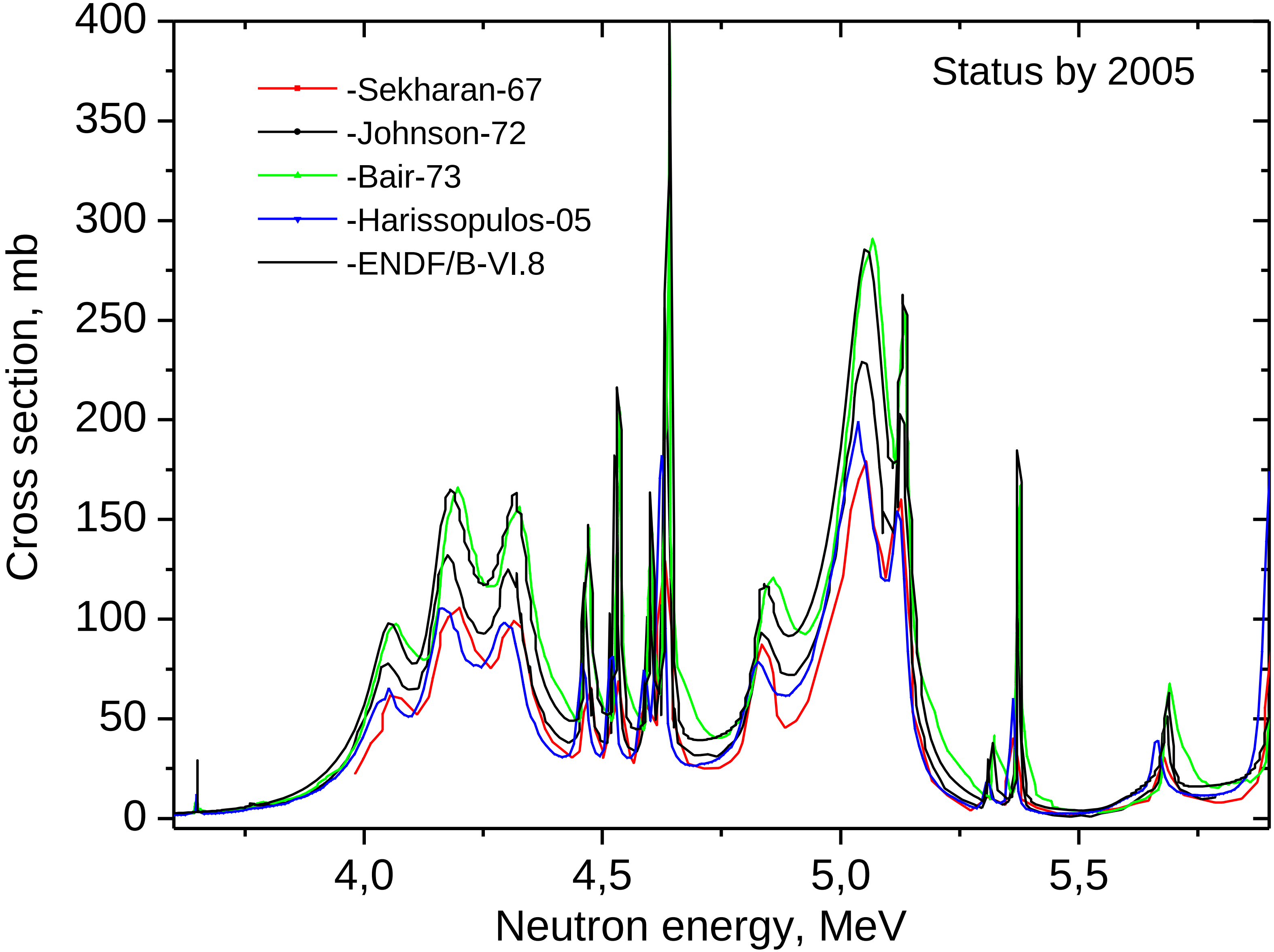
- In light water reactors oxygen is present in UOX, MOX and water. The sensitivity coefficient (dk/k)/(dσ/σ)=-3.5 pcm/%. A 30% uncertainty results in an uncertainty for k-eff of 100 pcm. This important effect was identified by Subgroup 22 of WPEC which investigated the underprediction of the reactivity of light water reactors with the most recent nuclear data evaluations.
- A second point concerns helium production which is of importance for the performance of fuel pins and clads. The O-16(n,α) reaction accounts for 25% of the total helium production and contributes 7% to its uncertainty.
- A third point concerns the calibration of neutron source strengths using the manganese-sulfate bath technique (NIST). The final requested uncertainty of 1% on neutron source calibrations is very near the 0.5% uncertainty contributed by this reaction.

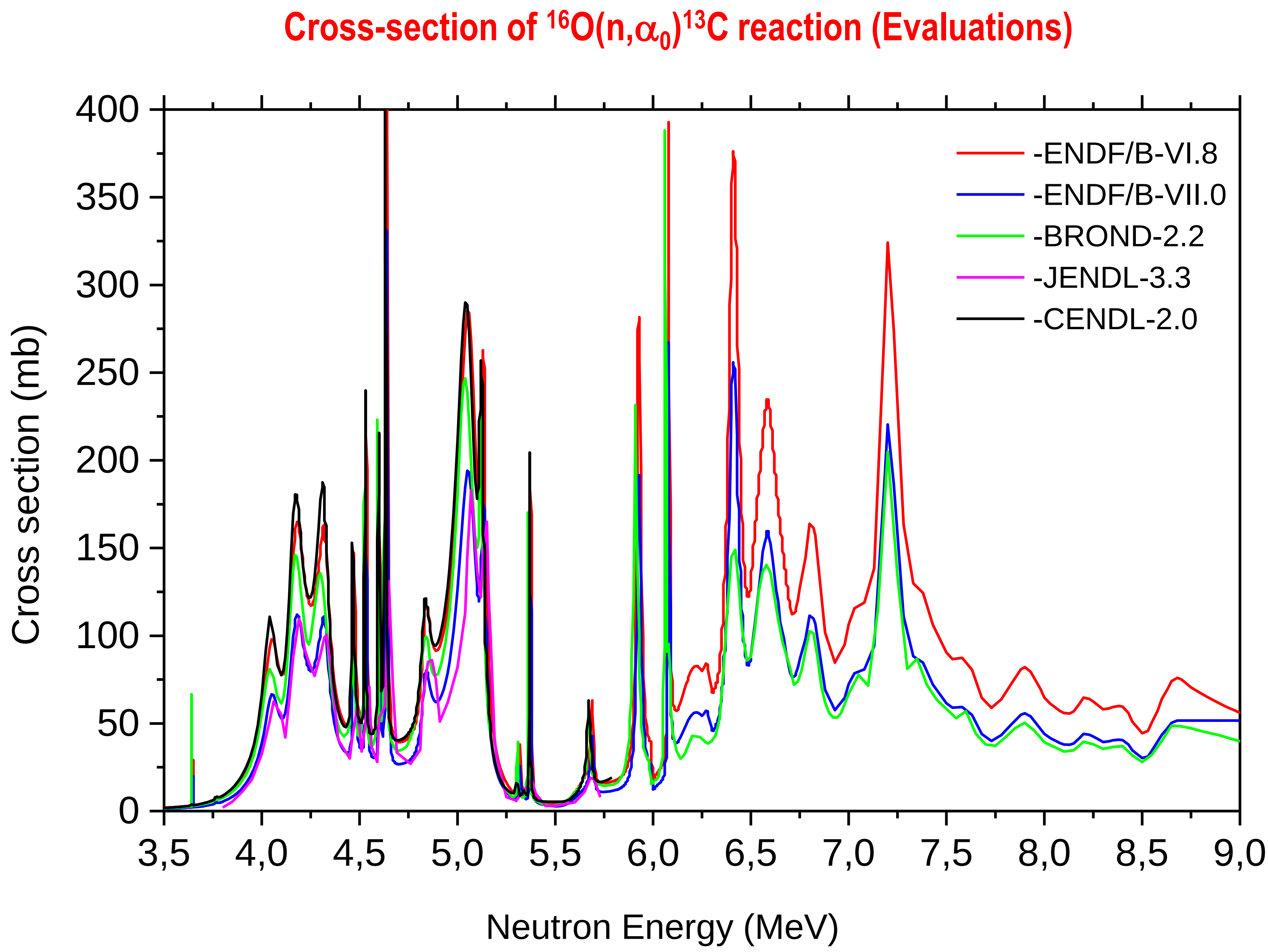
Justification document:
See: Reference 1: Need for O16(n,alpha) Measurement and Evaluation in the range 2.5 - 10 MeV A. Courcelle et al. (August 2005) and references therein.

Comment from requester:
The required accuracy concerns the normalisation of the cross section. A sensitivity analysis is also presented for k-eff in a fast reactor. Considerable differences exist among evaluations and measurements are discrepant. Recent C-13(α,n) and C-13(α,α) measurements are identified that provide detailed knowledge about the inverse reaction. Together with an R-matrix analysis this may be used to improve the evaluation for the O-16(n,α)


Cross-section of 16O(n,α0)13C reaction (direct reaction measurements)
Status by 2005
-Seitz-55
-Walton-57
-Davis-68
-Dandy-68
-ENDF/B-VI.8
Cross section, mb
Neutron energy, MeV
0
50
100
150
200
250
300
350
400
4
5
6
7
8
9
10

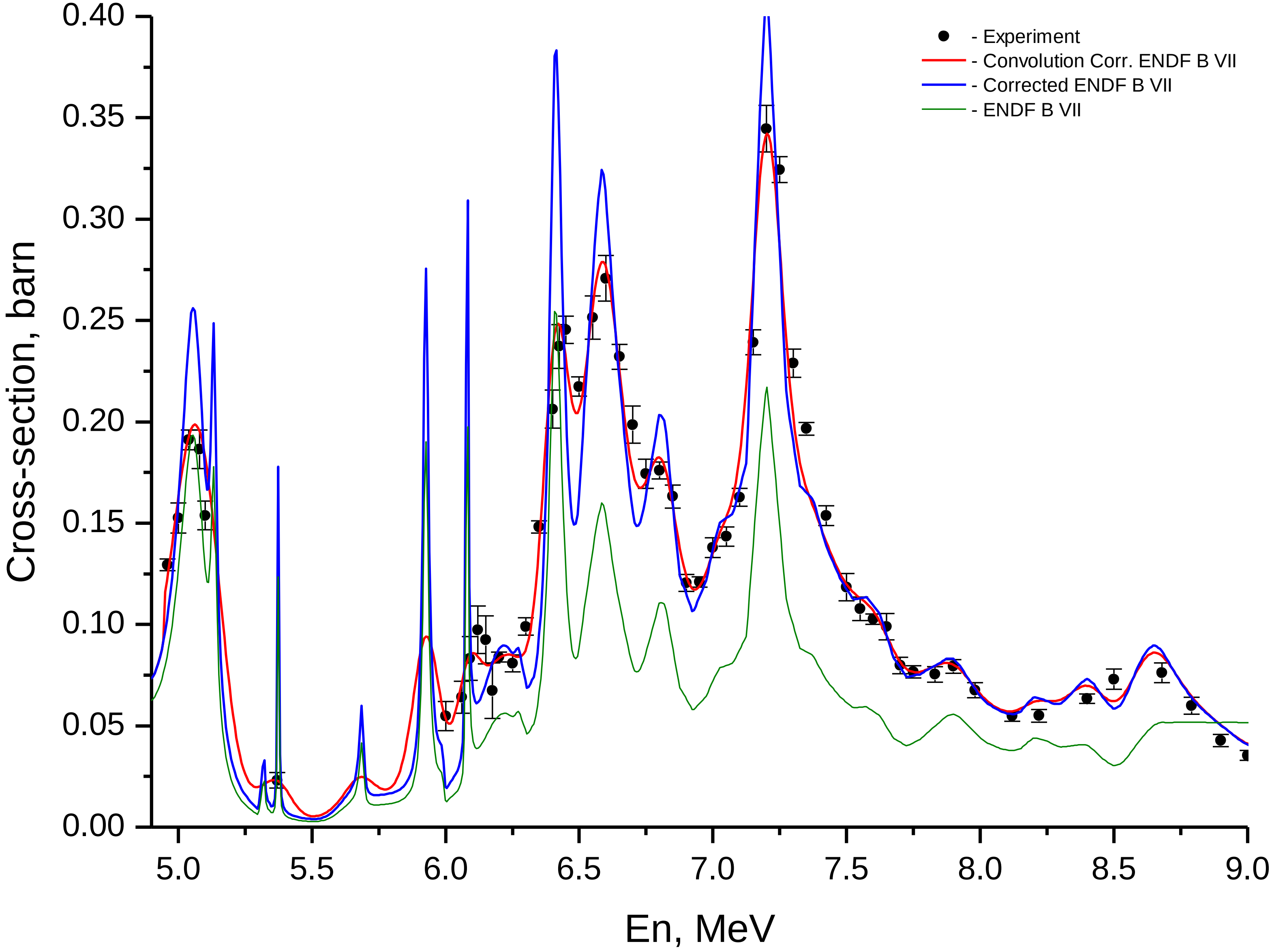
Cross-section of $^{16}O(n,\alpha)^{13}C$ reaction (obtained from an inverse reaction)





Cross-section of 16O(n,α0)13C reaction (Evaluations)
Cross section (mb)
Neutron Energy (MeV)
0
50
100
150
200
250
300
350
400
3,5
4,0
4,5
5,0
5,5
6,0
6,5
7,0
7,5
8,0
8,5
9,0
-ENDF/B-VI.8
-ENDF/B-VII.0
-BROND-2.2
-JENDL-3.3
-CENDL-2.0

Results of the cross section measurement of $^{16}O(n,\alpha_0)^{13}C$ at IRMM in compared with ENDF evaluations and experimental data



Experimental uncertainties

- Counting statistics: typical ~ 5%

- Radii of cone: 1.1%
- Height of cone: 1%
- Gas pressure: 0.1%
- Temperature of gas: 0.3%
- Content of CO_2: 2%.

- Number of oxygen nuclei: 2.5%.

- Background contributed into α-peak: 1.2%
- Number of ^{238}U nuclei: 0.7%
- Number of the fission fragments: 1.5%
- Correction for neutron background
- from neutron source 1.5%

- Total error not related to statistics: 3.6%

- **Total uncertainty: 6.1%**

Advantages of the method

- Dead time for main and monitor channel is equally
- The simple response function of the spectrometer
- Achieved a big mass of the target
- A simple method for determining the mass of non-radioactive target
- Developed numerical methods to effectively suppress backgrounds
- Wall effect is absent

$^{10}B(n,2\alpha)t$ reaction. The tritium production cross section for neutron interaction with ^{10}B nuclei

Isotopes

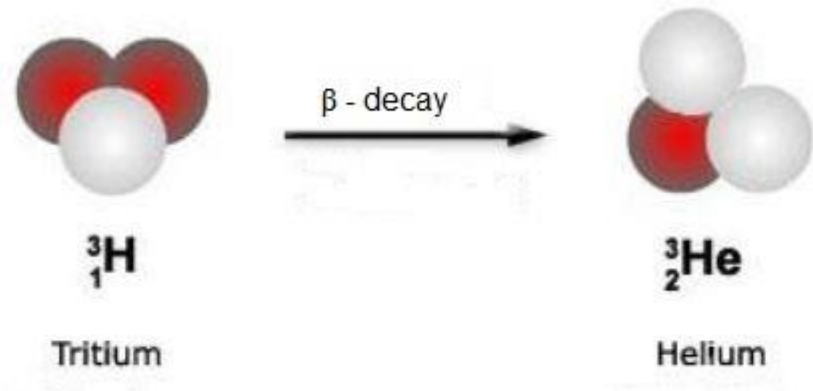
^{10}B (20%) ^{11}B (80%)

- $\sigma \sim 3000$ barn
- is used as a shielding of thermal neutrons

$^{10}B + n \rightarrow \alpha + {}^{7}Li + 2.7895$ MeV

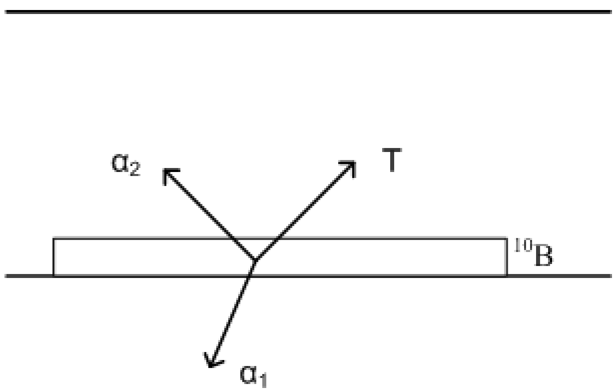
^{7}Li exited states: $E^* = 0.47761$ MeV

$^{10}B + n \rightarrow 2\alpha + t + 0.3225$ MeV

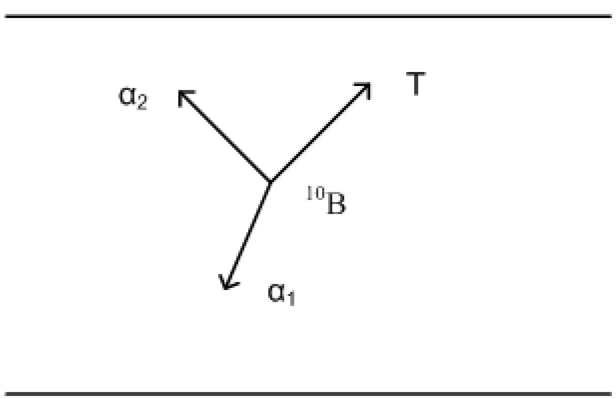


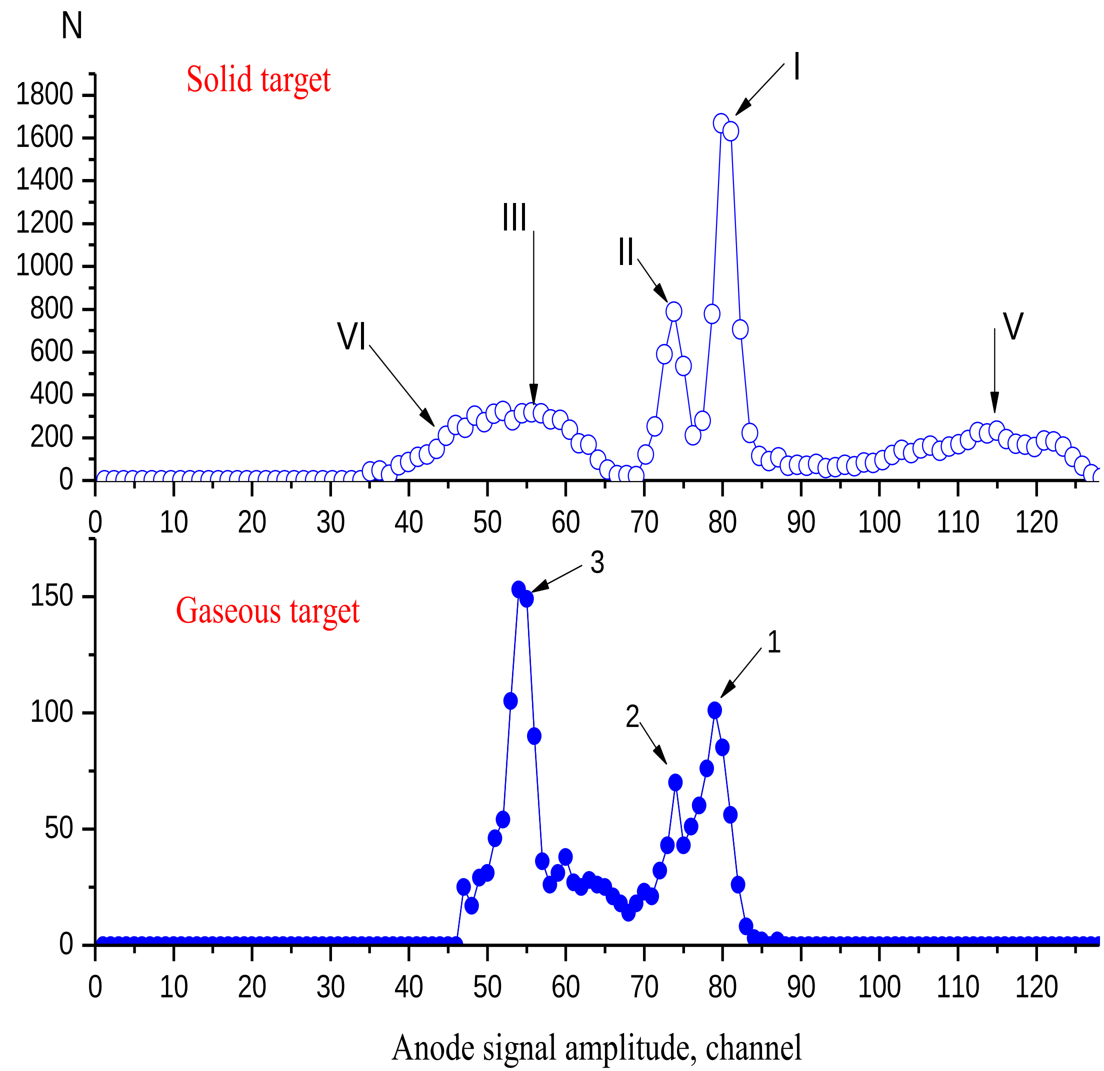
- tritium is a source of β-rays
- chemical properties of tritium are the same as those of hydrogen
- tritium can replace hydrogen in all of its compounds

Solid target



Gaseous target





Solid target:

I – $^{10}B(n,\alpha_0)$;

II – $^{10}B(n,\alpha_1)$;

III – $^{10}B(n,t)$;

VI – ^{7}Li;

V – $^{7}Li+\alpha$

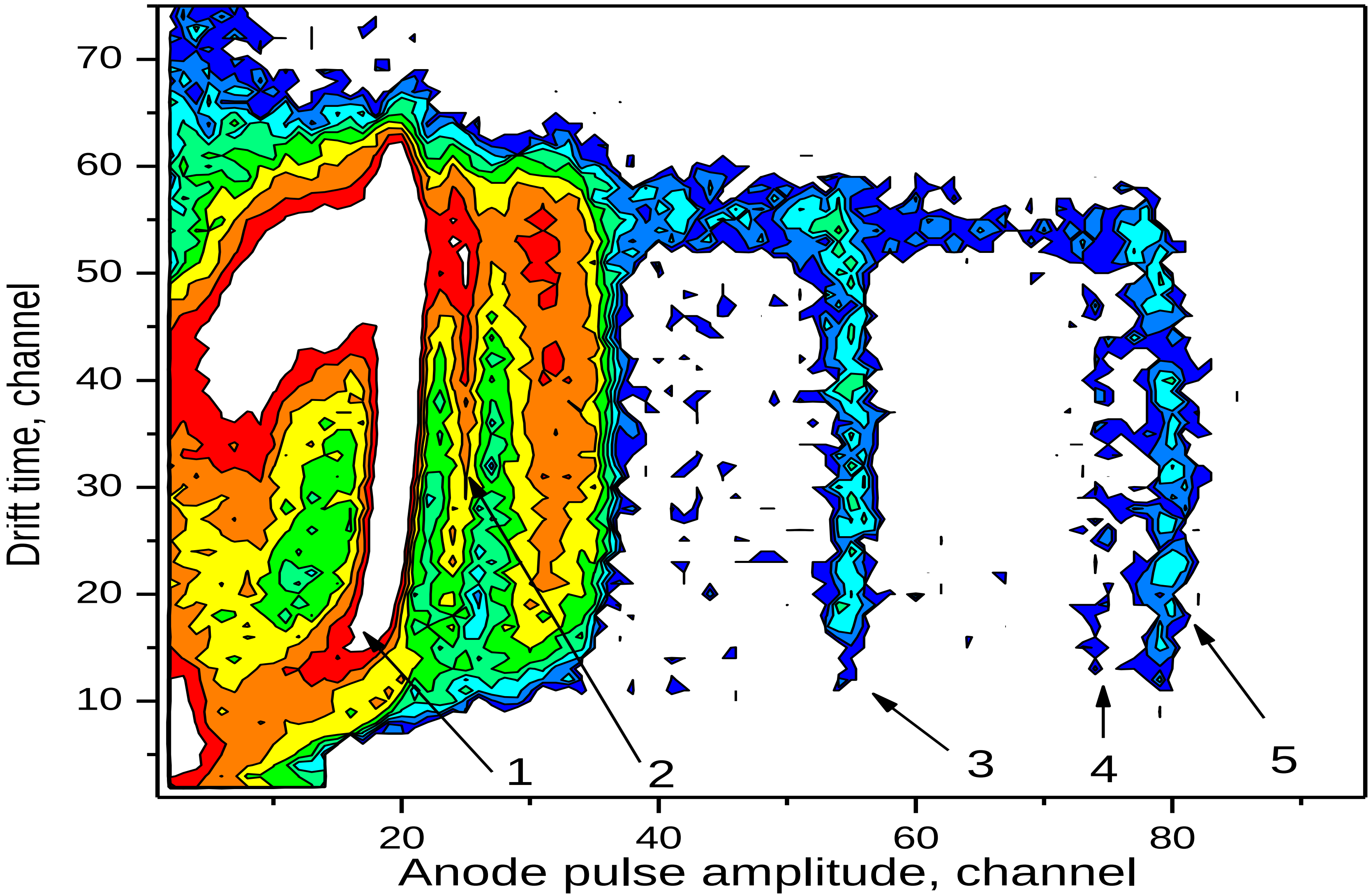
Gaseous target:

1 – $^{10}B(n,\alpha_0)$;

2 - $^{10}B(n,\alpha_1)$;

3 - $^{10}B(n,t)$

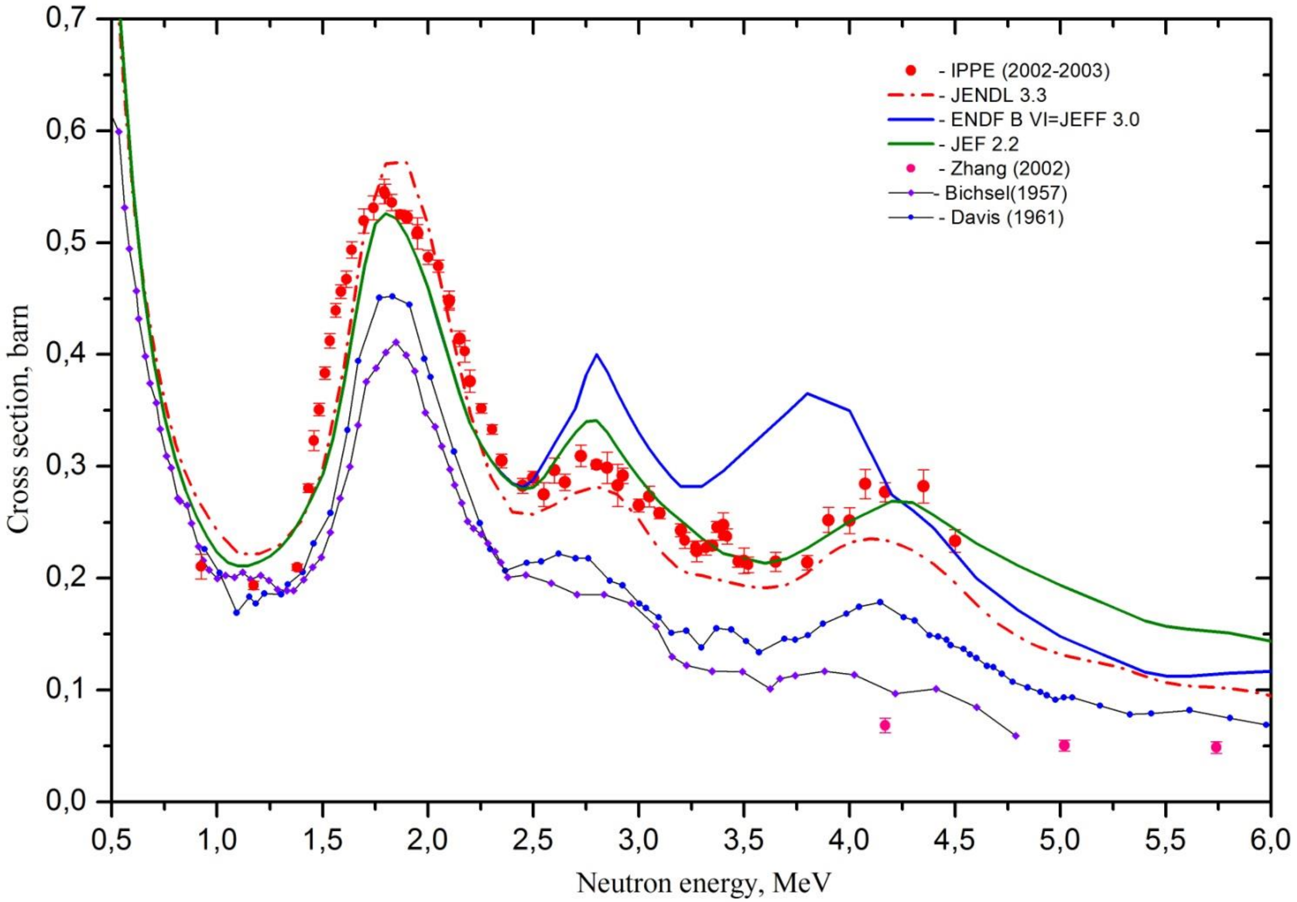
Products of $^{10}B(n,\alpha)^{7}Li$ reaction on fast neutrons



Two dimensional plot obtained for neutron energy En=5.3 MeV. Numbers mark different reaction channels: 1 – $^{10}B(n_{th},\alpha_1)^{7}Li$; 2 – $^{10}B(n_{th},\alpha_0)^{7}Li$; 3 – $^{10}B(n_f,2\alpha+t)$ (break-up reaction); 4 – $^{10}B(n_f,\alpha_1)^{7}Li$; 5 – $^{10}B(n_f,\alpha_0)^{7}Li$

Relative method of cross section determination

Measurement of $^{10}B(n,2\alpha)t$ reaction's cross section was carried out relative to $^{10}B(n,\alpha)$ reaction cross section.

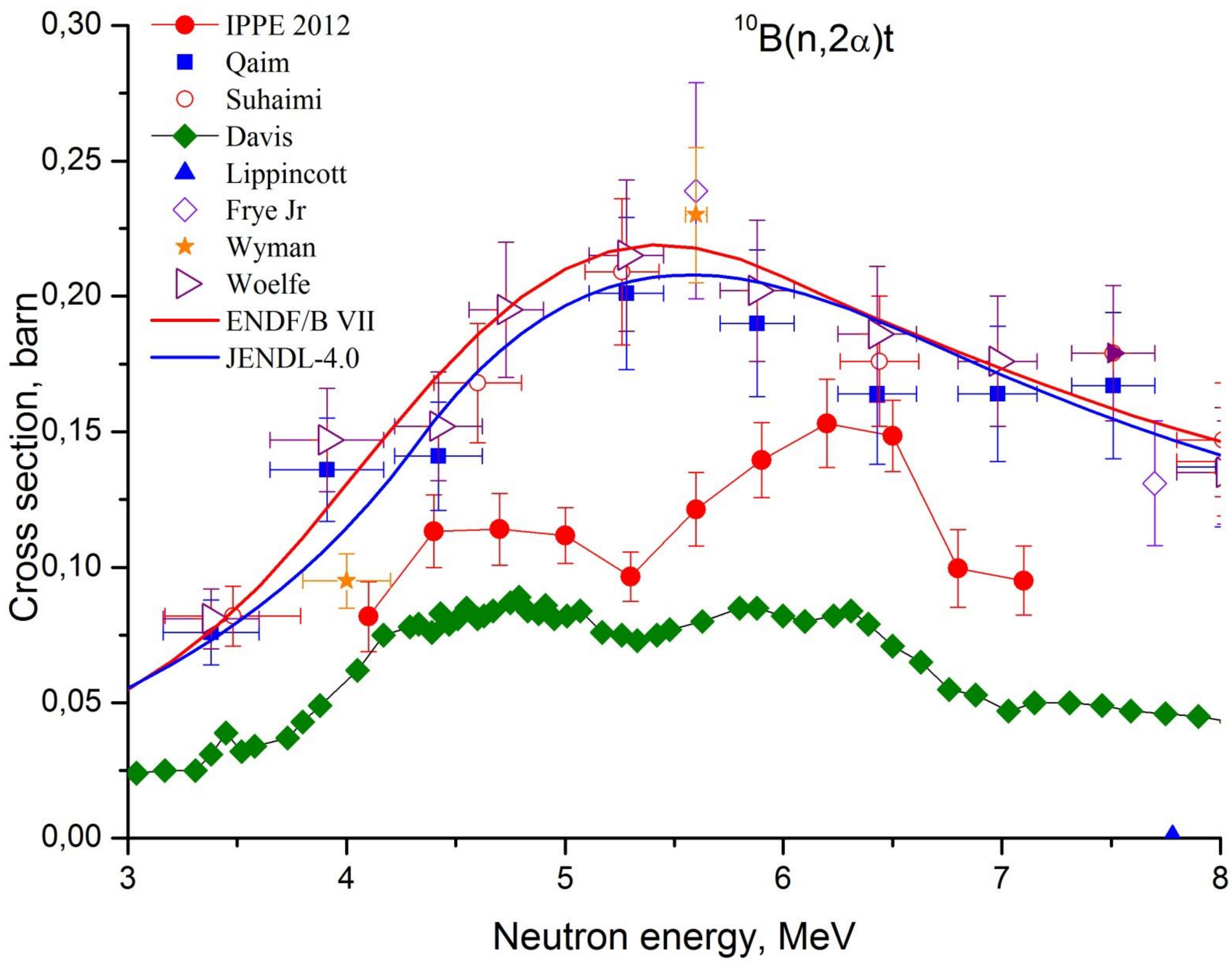


$$N_T = N_B * \sigma_T * F$$

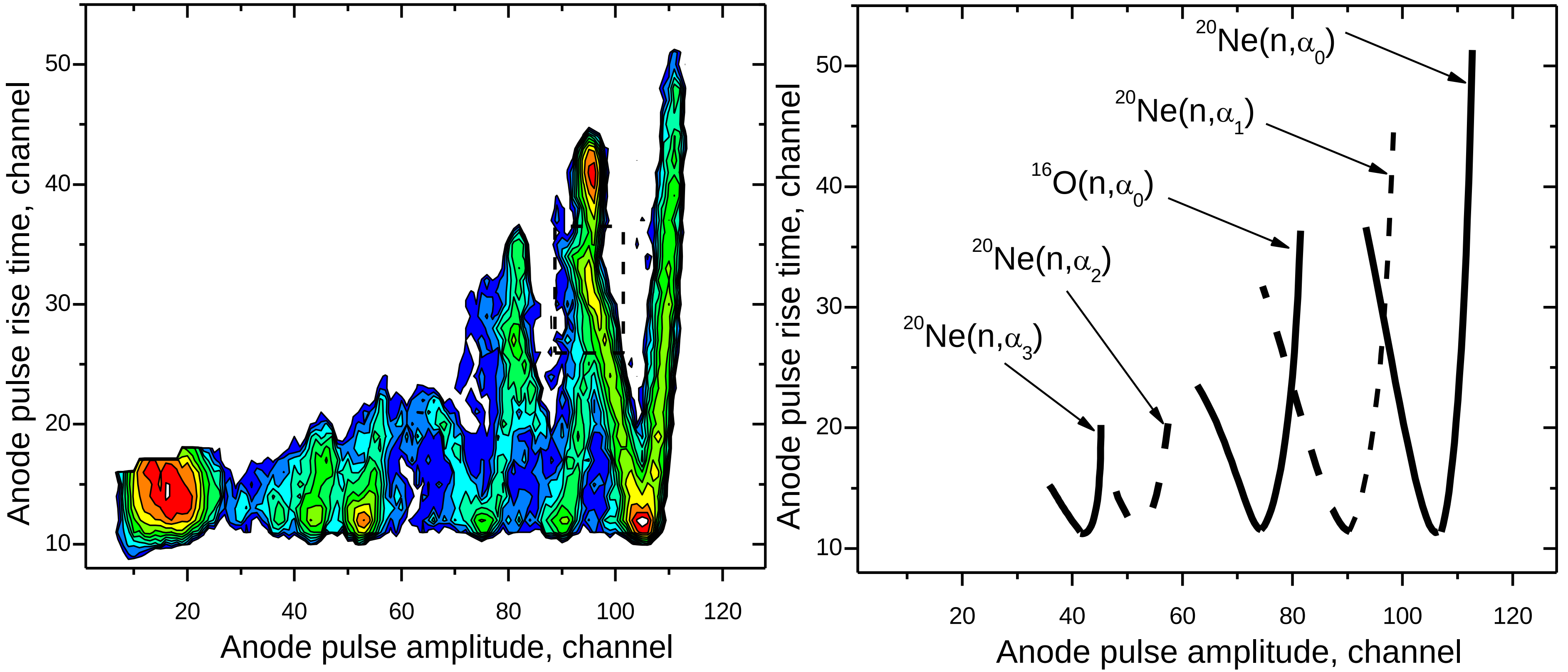
$$N_\alpha = N_B * \sigma_\alpha * F$$

$$\sigma_T = \frac{N_T}{N_\alpha} \sigma_\alpha$$

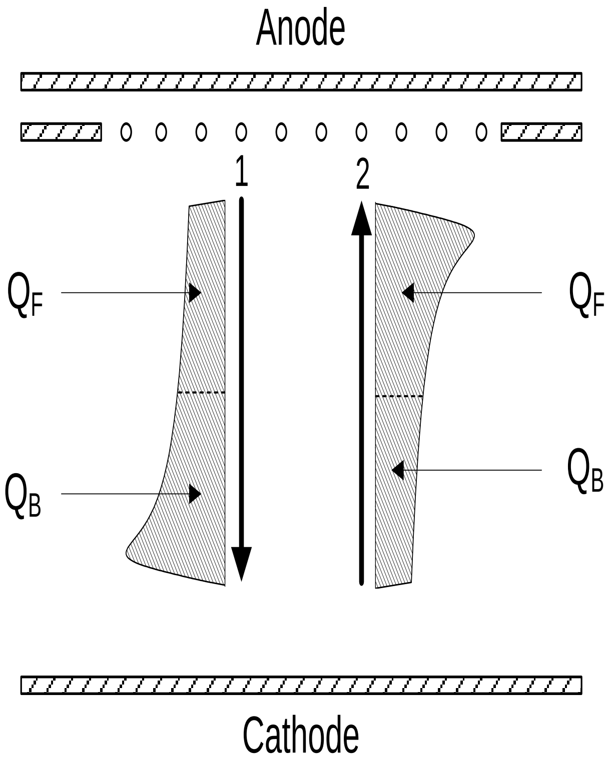
Results of the cross section measurement of $^{10}B(n,2\alpha)t$ reaction

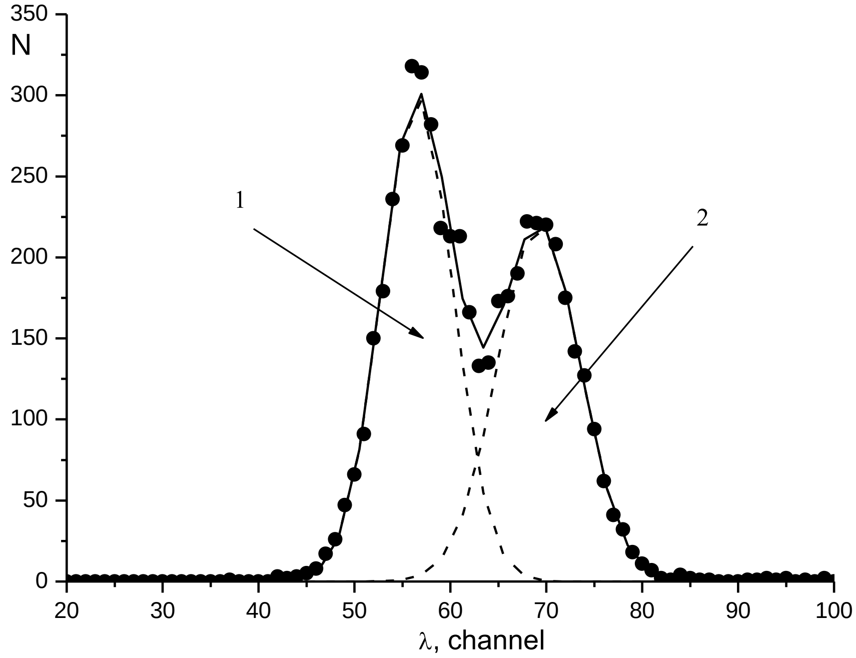


$^{20}Ne(n,\alpha)^{17}O$ cross section measurement

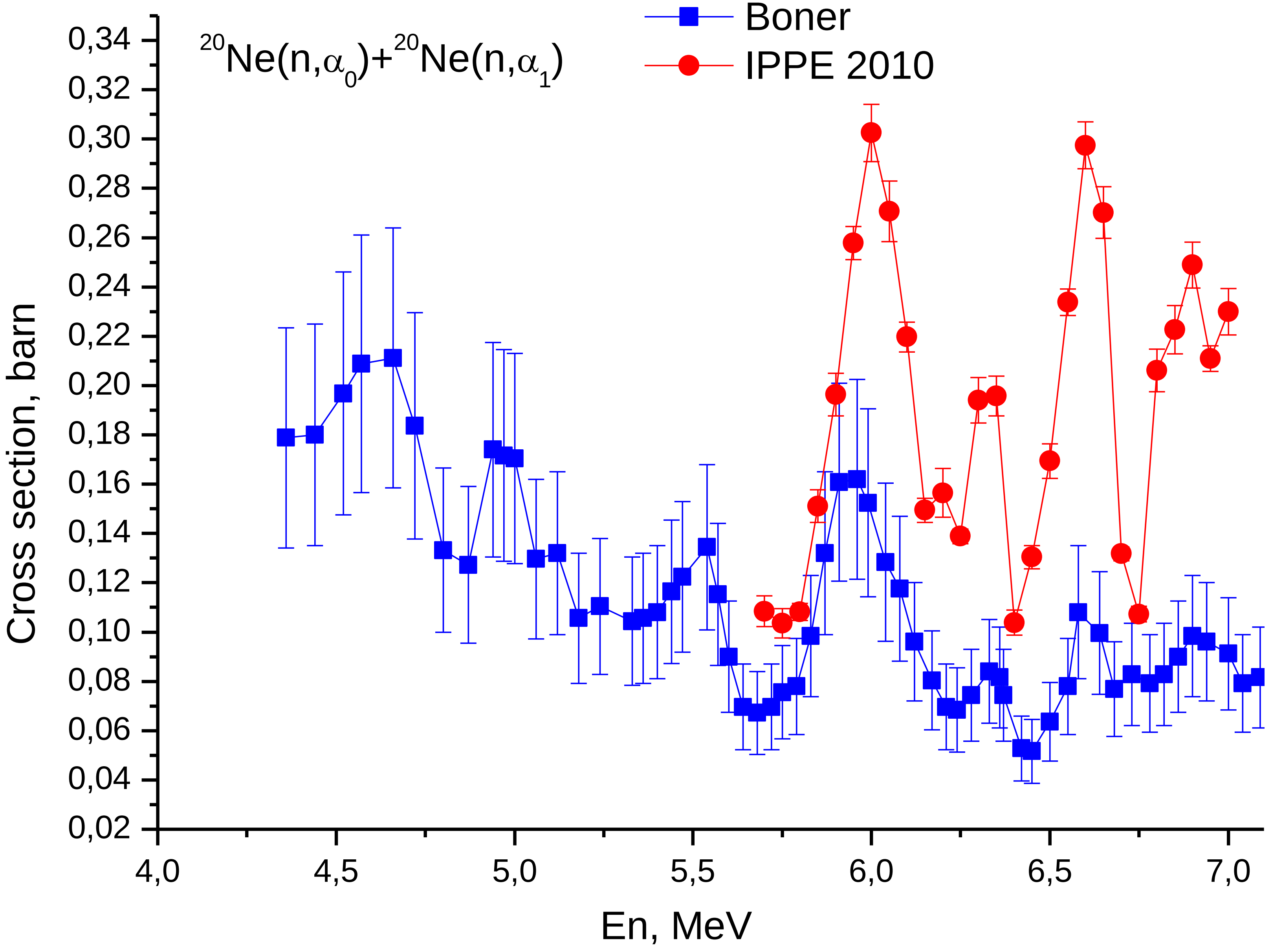


Particles separation

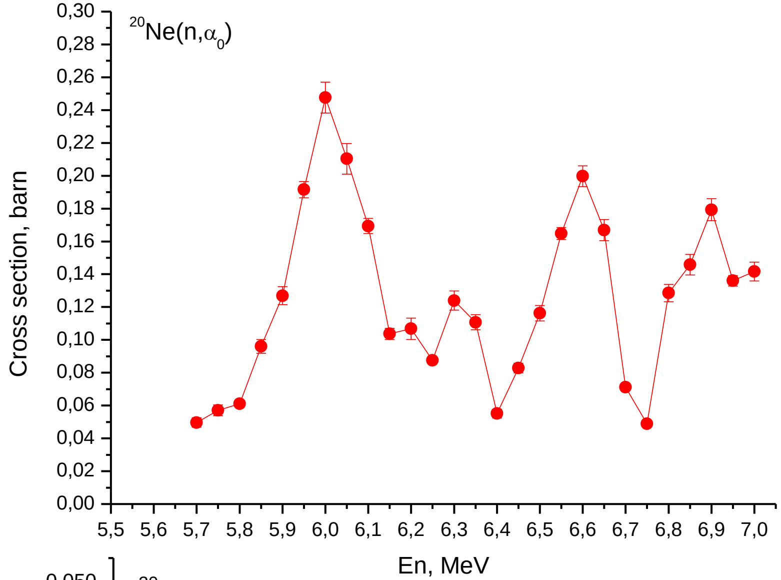


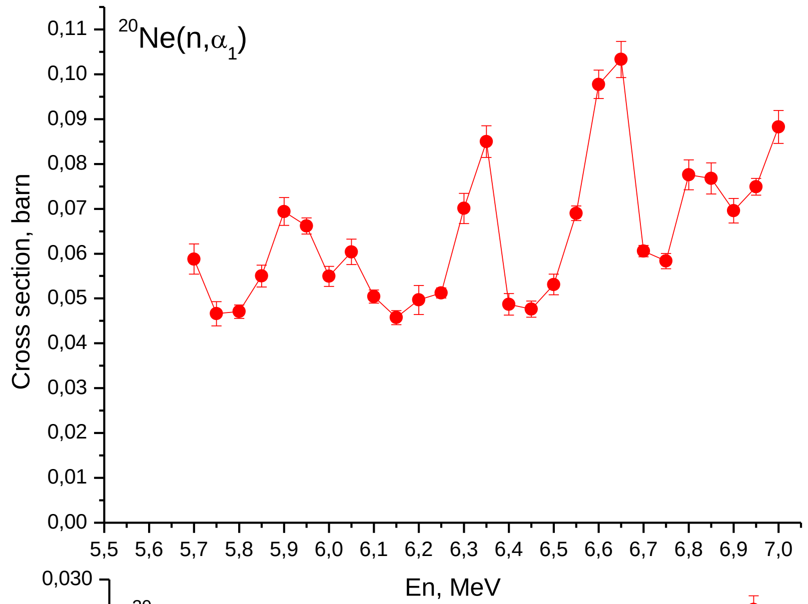


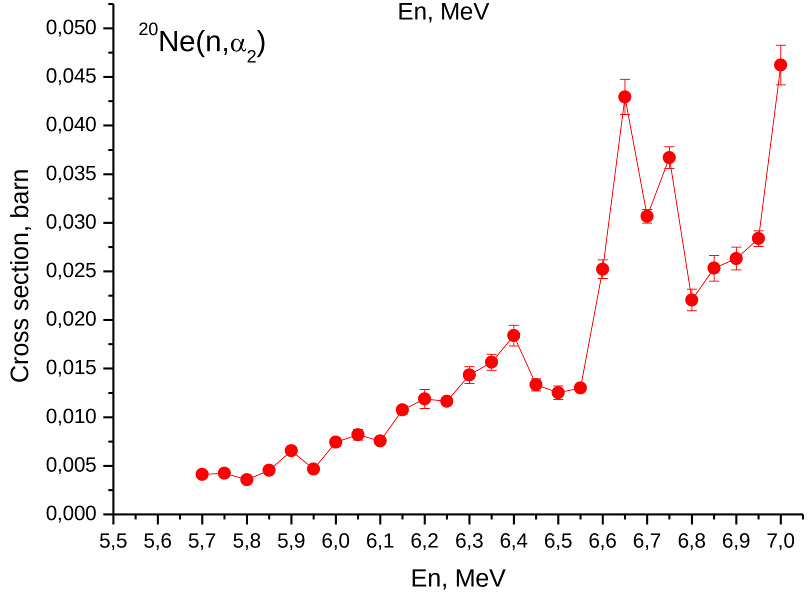
$^{20}Ne(n,\alpha)^{17}O$ cross section measurement

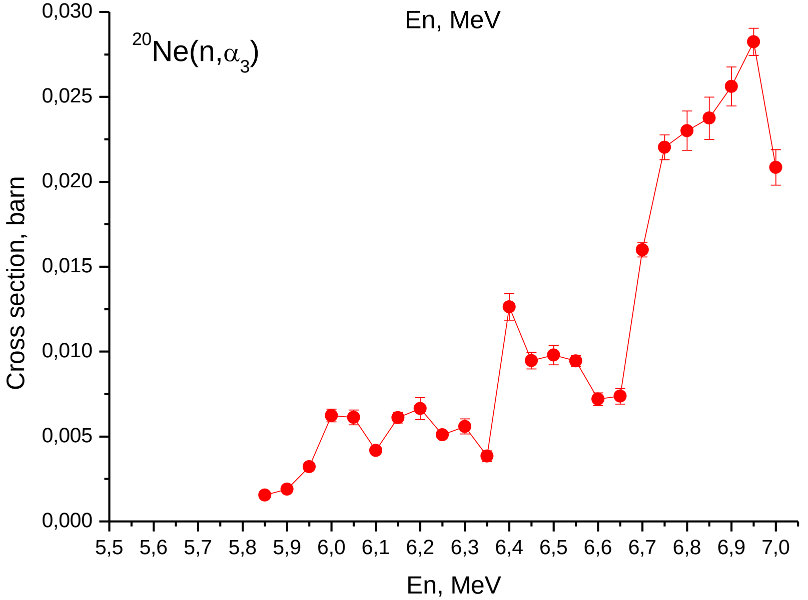


$^{20}Ne(n,\alpha)^{17}O$ cross section measurement









Justification for the structural material (n,α) reaction cross section measurement

Calculation of helium production and radiation damages in -

- **fuel-element cladding;**
- **Material of reactor vessel;**
- **reactor core;**
- **Other construction contacted with neutron flux.**

Monte Carlo calculation based on the evaluated nuclear data provided by libraries. Whether their accuracy is sufficient?

Some of structural material isotopes properties

Isotope	**Natural abundance, %**	**(n,α) reaction Q-value, MeV**
^{50}Cr, $T_{1/2}>1{,}8*10^{17}$ y, EC	4,345	+0,3213
^{52}Cr, stable	83,489	-1,2097
^{53}Cr, stable	9,501	+1,7903
^{54}Cr, stable	2,365	- 1,5466

Cr

Isotope	Residual nuclear	Stability
^{50}Cr	**^{47}Ti**	**Stable**
^{52}Cr	**^{49}Ti**	**Stable**
^{53}Cr	**^{50}Ti**	**Stable**
^{54}Cr	^{51}Ti	$T_{1/2}$=5,76 min

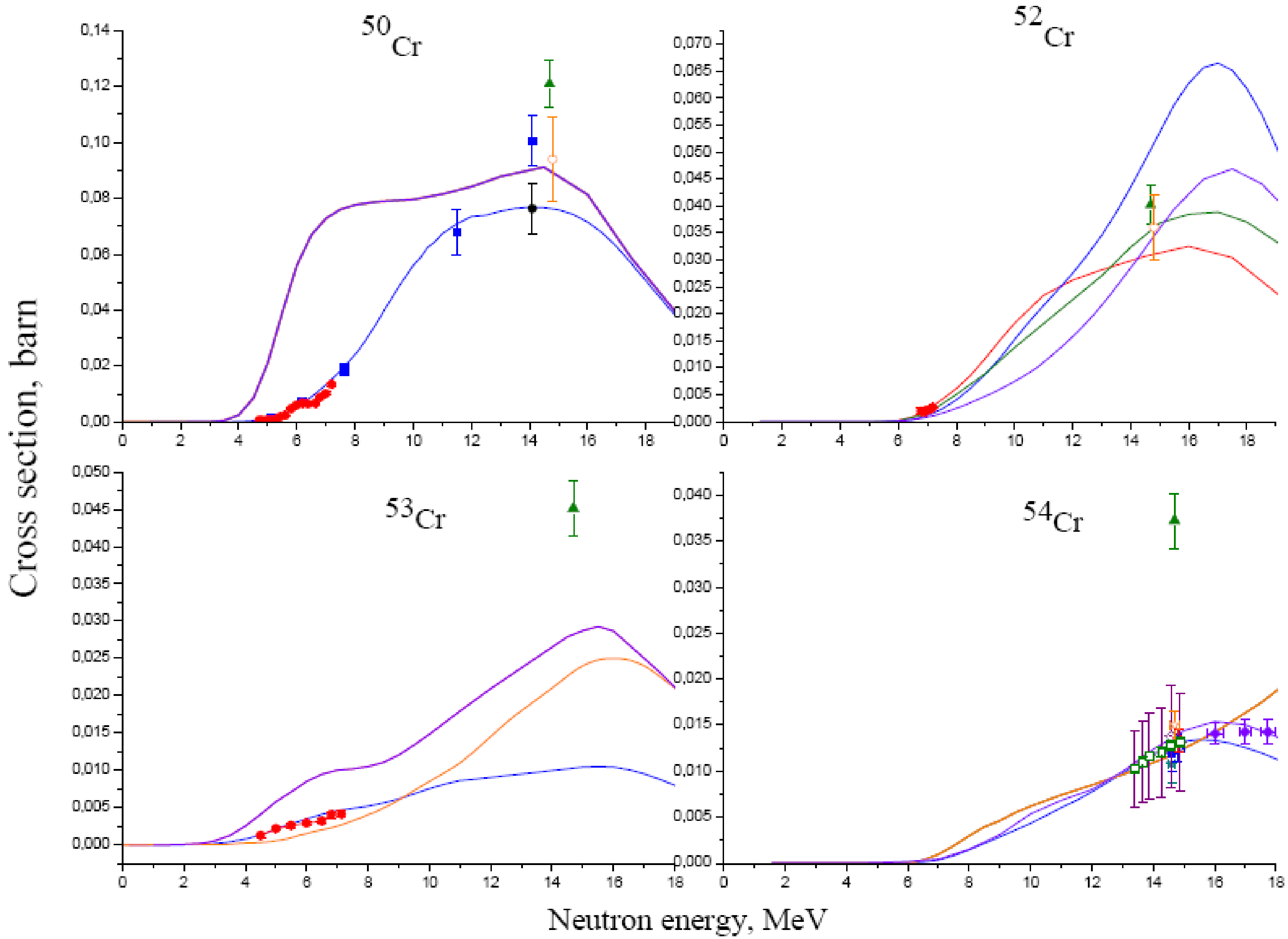
Fe

Isotope	Residual nuclear	Stability
^{54}Fe	**^{51}Cr**	**$T_{1/2}$=27,7 d, ec**
^{56}Fe	**^{53}Cr**	**Stable**
^{57}Fe	**^{54}Cr**	**Stable**
^{58}Fe	^{55}Cr	$T_{1/2}$=3,55 min

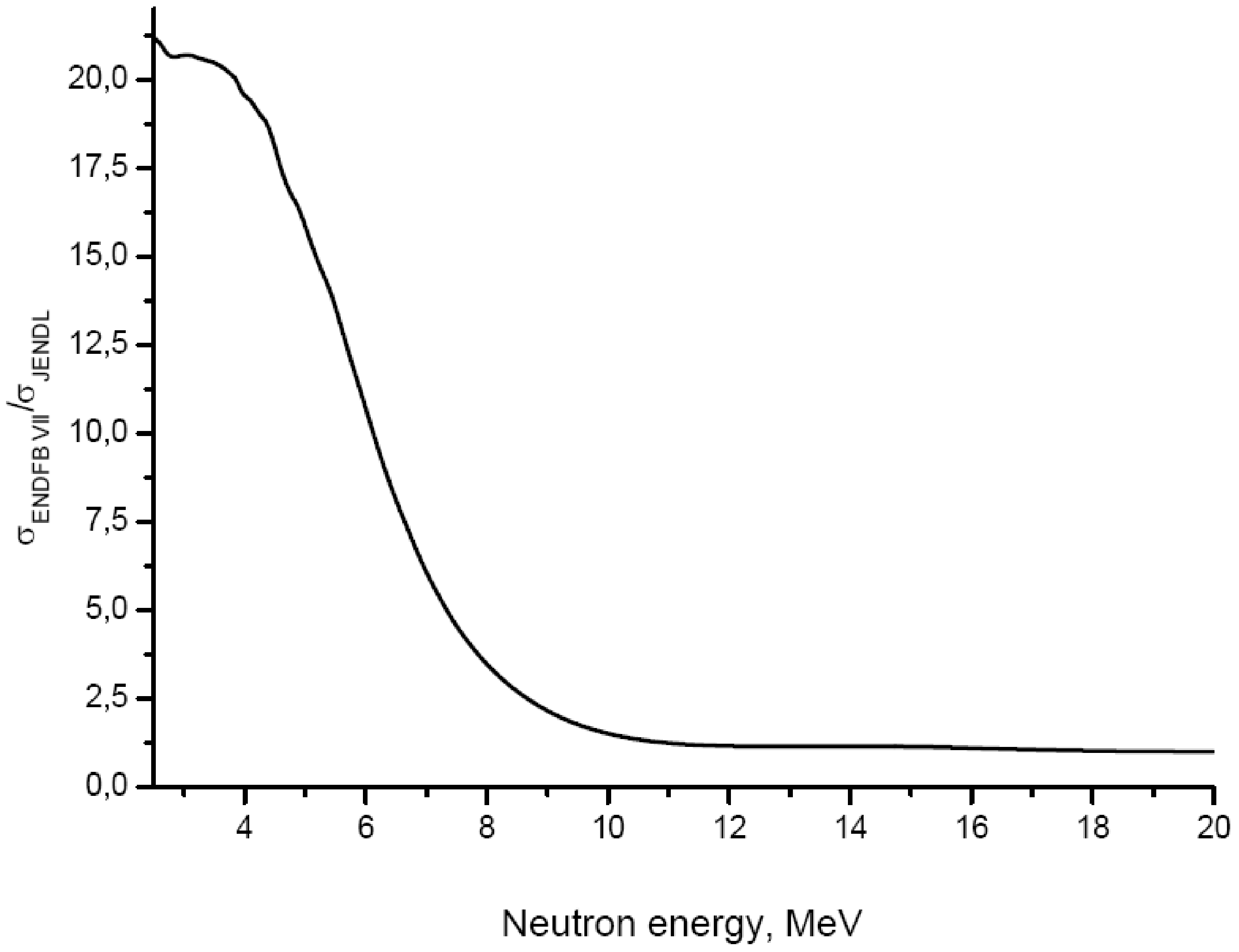
Ni

Isotope	Residual nuclear	Stability
^{58}Ni	**^{55}Fe**	**$T_{1/2}$=2,7 y, ec**
^{60}Ni	**^{57}Fe**	**Stable**
^{61}Ni	**^{58}Fe**	**Stable**
^{62}Ni	^{59}Fe	$T_{1/2}$=44,5 d

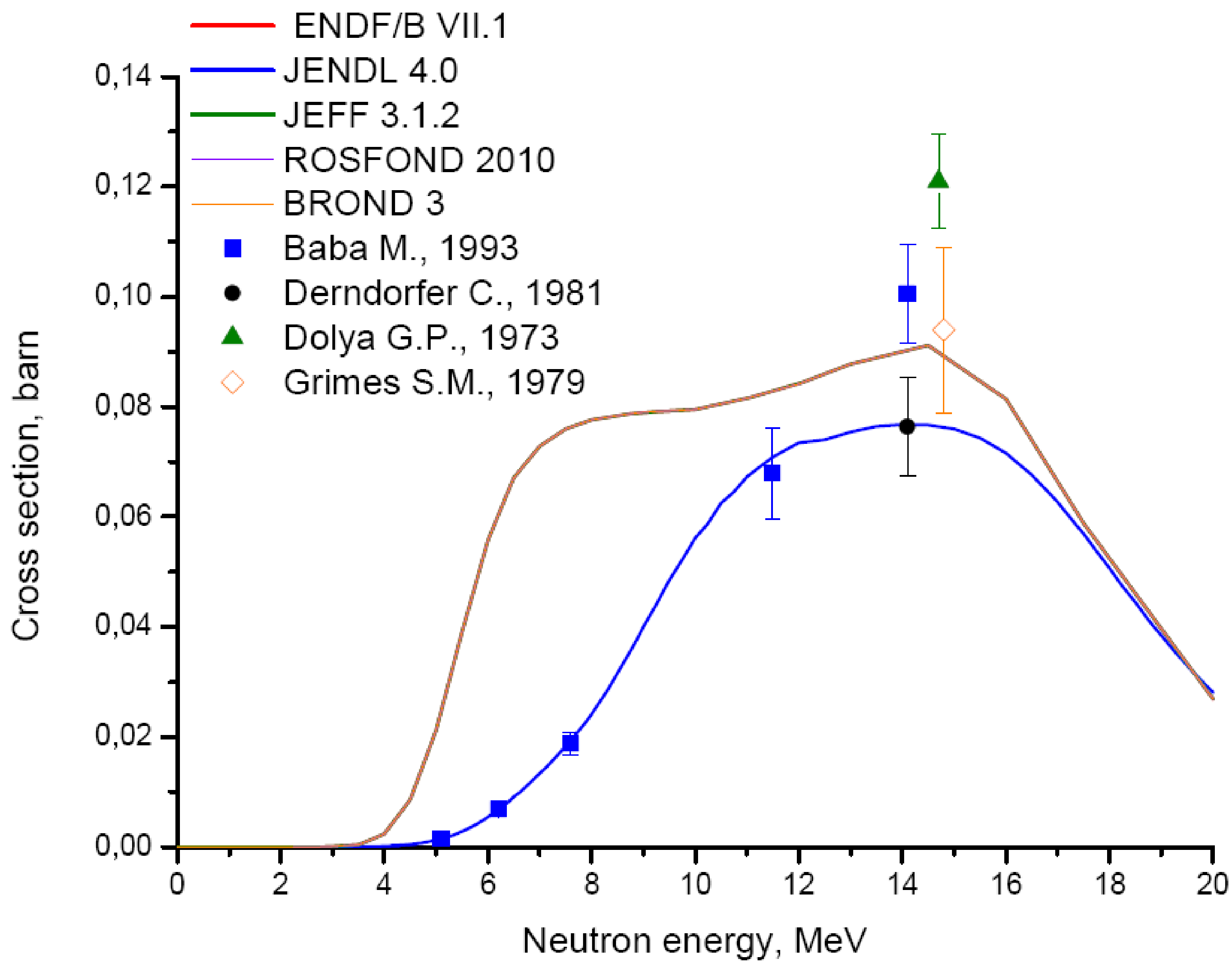
Present status of experimental data and evaluation for chromium isotopes



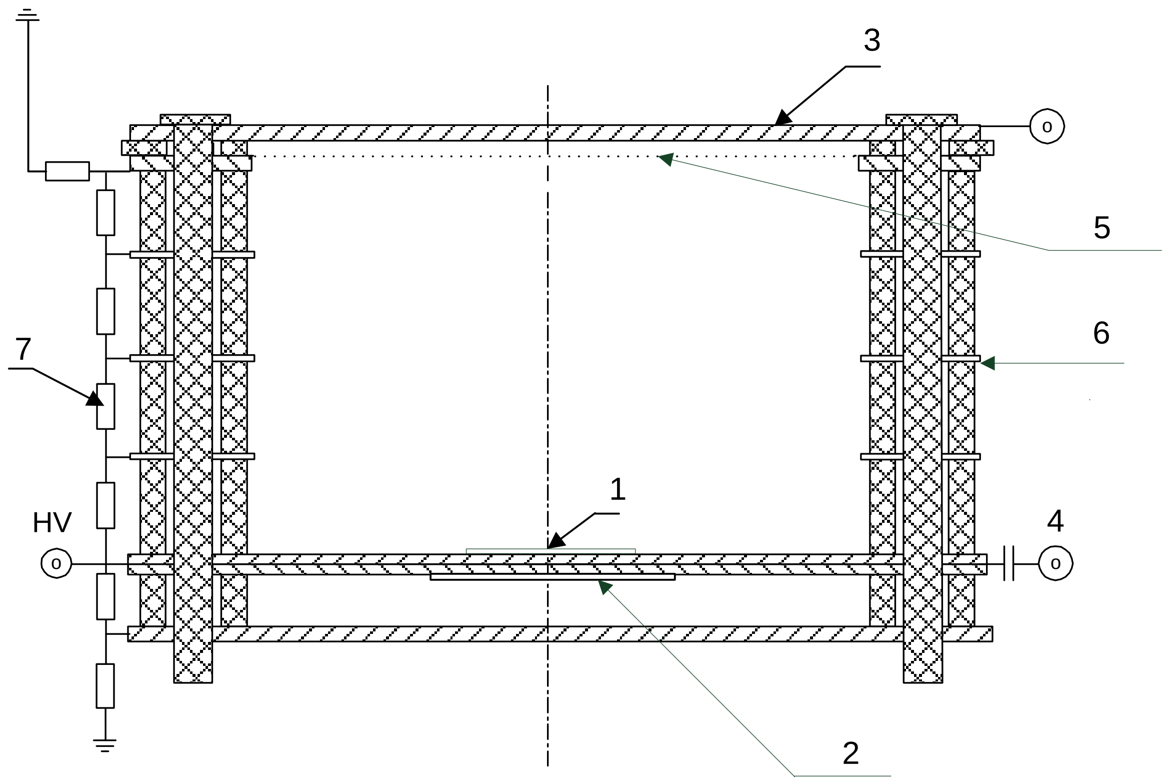
ENDF/B VII.1 to JENDL 4.0 cross section ratio



Status of experimental data for $^{50}Cr(n,\alpha)^{47}Ti$



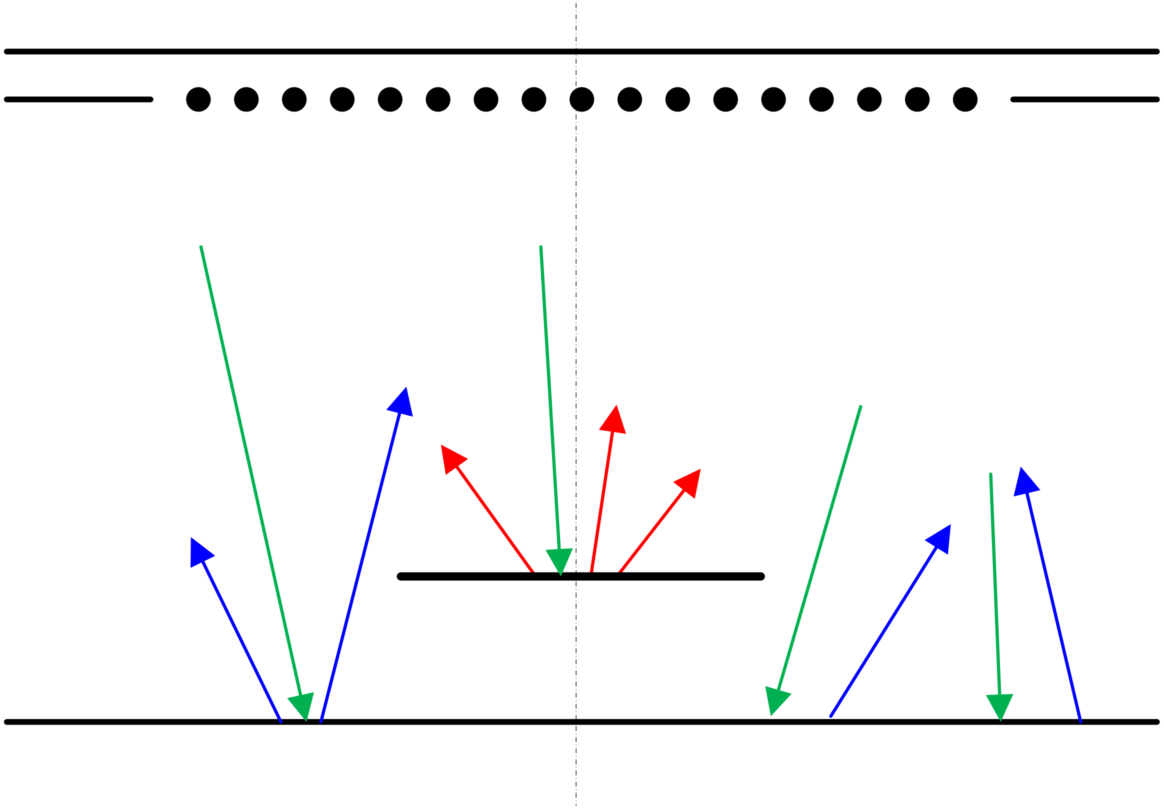
Classical ionisation chamber



1) ^{50}Cr target;
2) ^{238}U target;
3) Anode;
4) Anode signal connector;

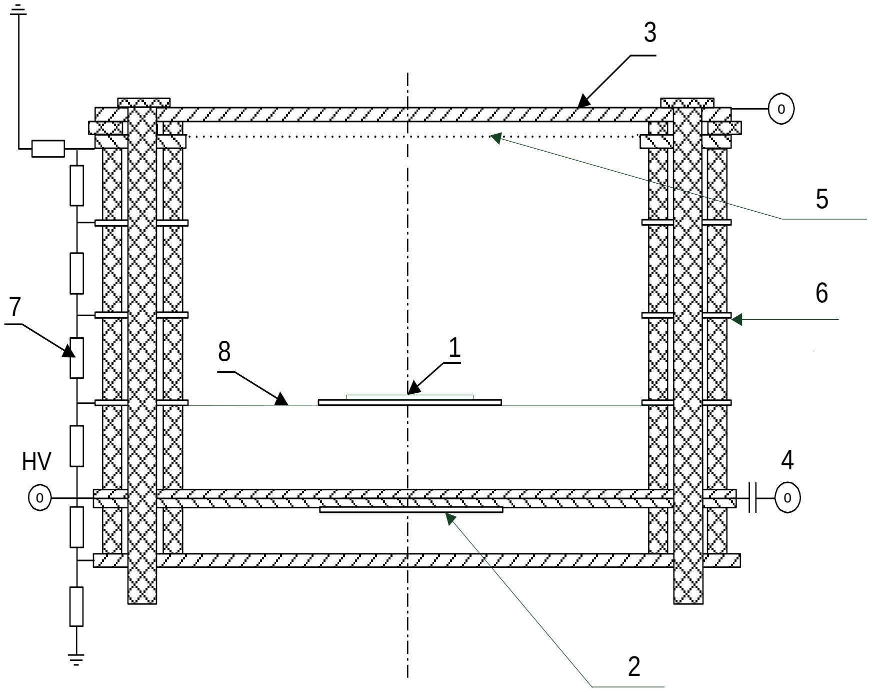
5. Frisch grid;
6. Guard electrodes;
7. Resistor.

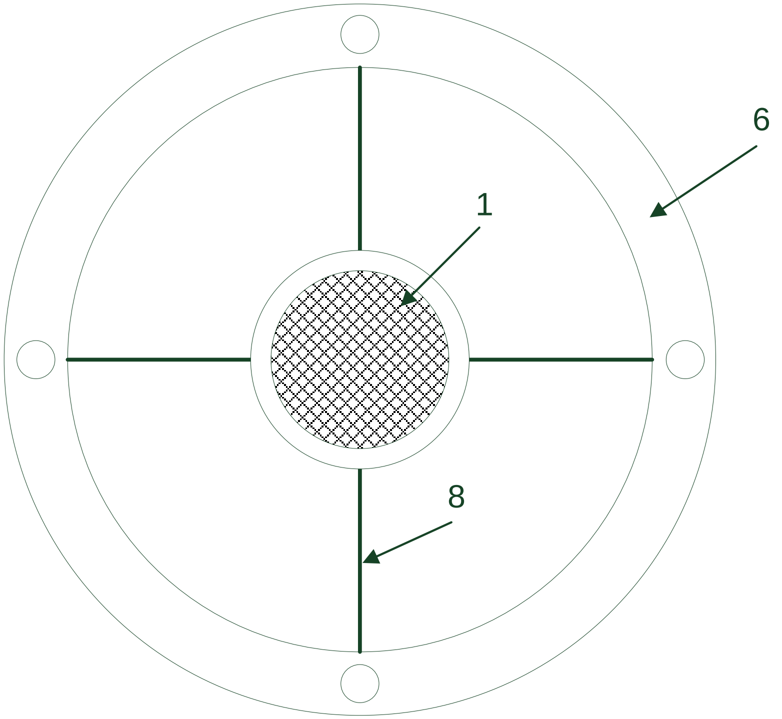
Motivations for removing solid target from cathode surface



1) Target surface 10 times less then cathode surface; Probability of gaseous particle absorption is proportional to the surface area.
2) Target material – gold. Low probability for charge particle emission;

New chamber design.

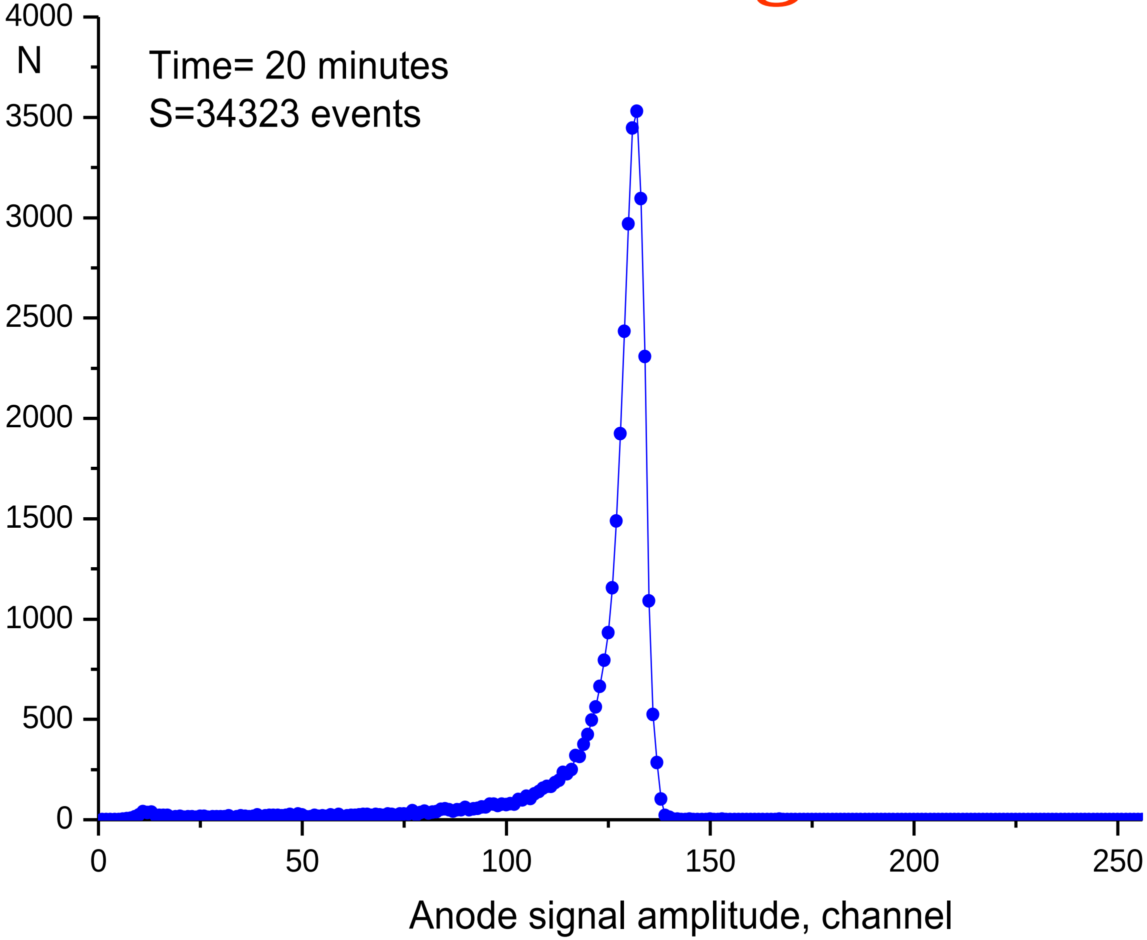




1) Cr target;
2) ^{238}U target;
3) Anode;
4) Anode signal connector;

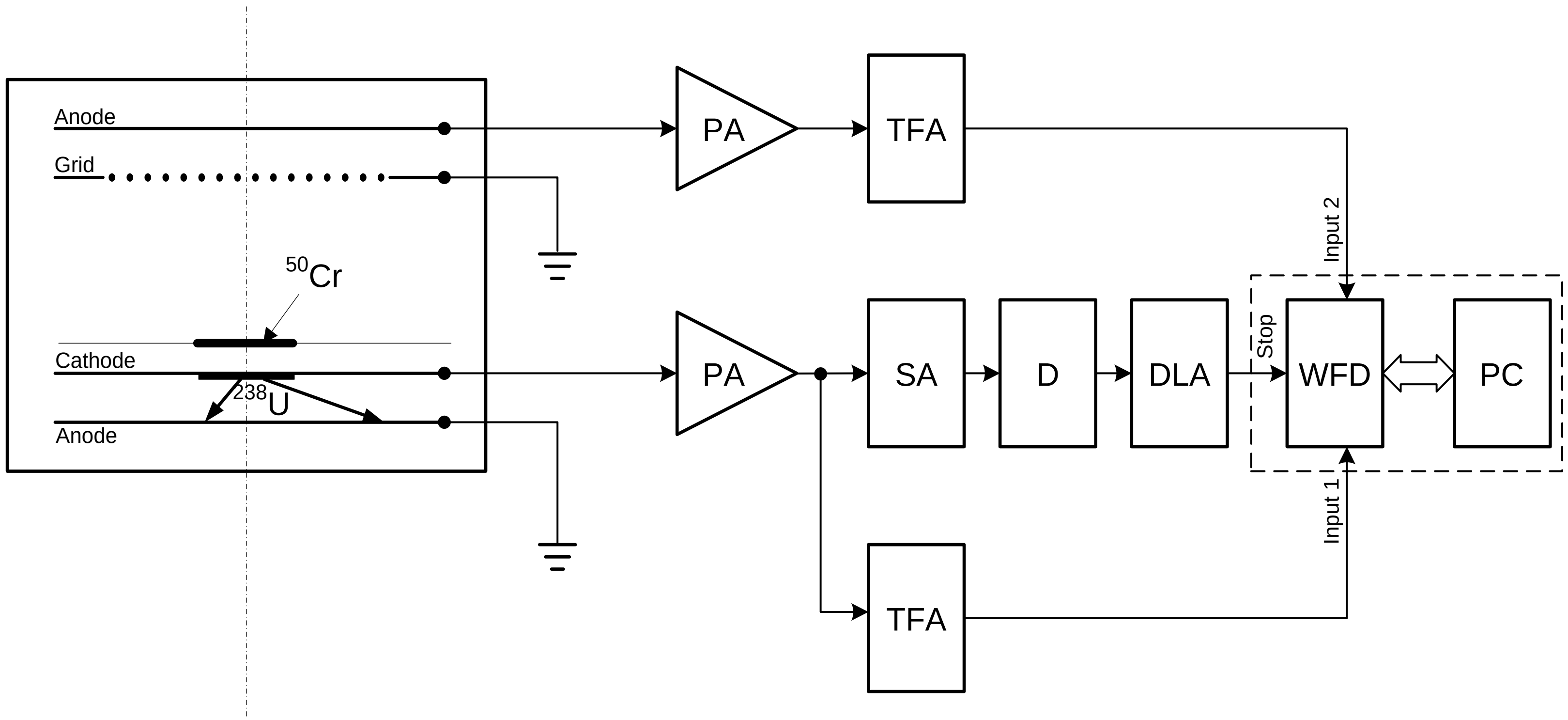
5. Frisch grid;
6. Guard electrodes;
7. Resistor.
8. Golden threads

^{238}U target



- **Stainless steel backing**
- **^{238}U enriched to 99,99 %**
- **Total ^{238}U mass – 4,598 mg**
- **Total number of ^{238}U atoms – 1,167*10^{19}**

Digital signal processing



PA – preamplifier, TFA – timing filter amplifier,
D – discriminator,
SA – spectroscopy amplifier, DLA – delay line amplifier,
WFD – waveform digitizer, PC – personal computer.

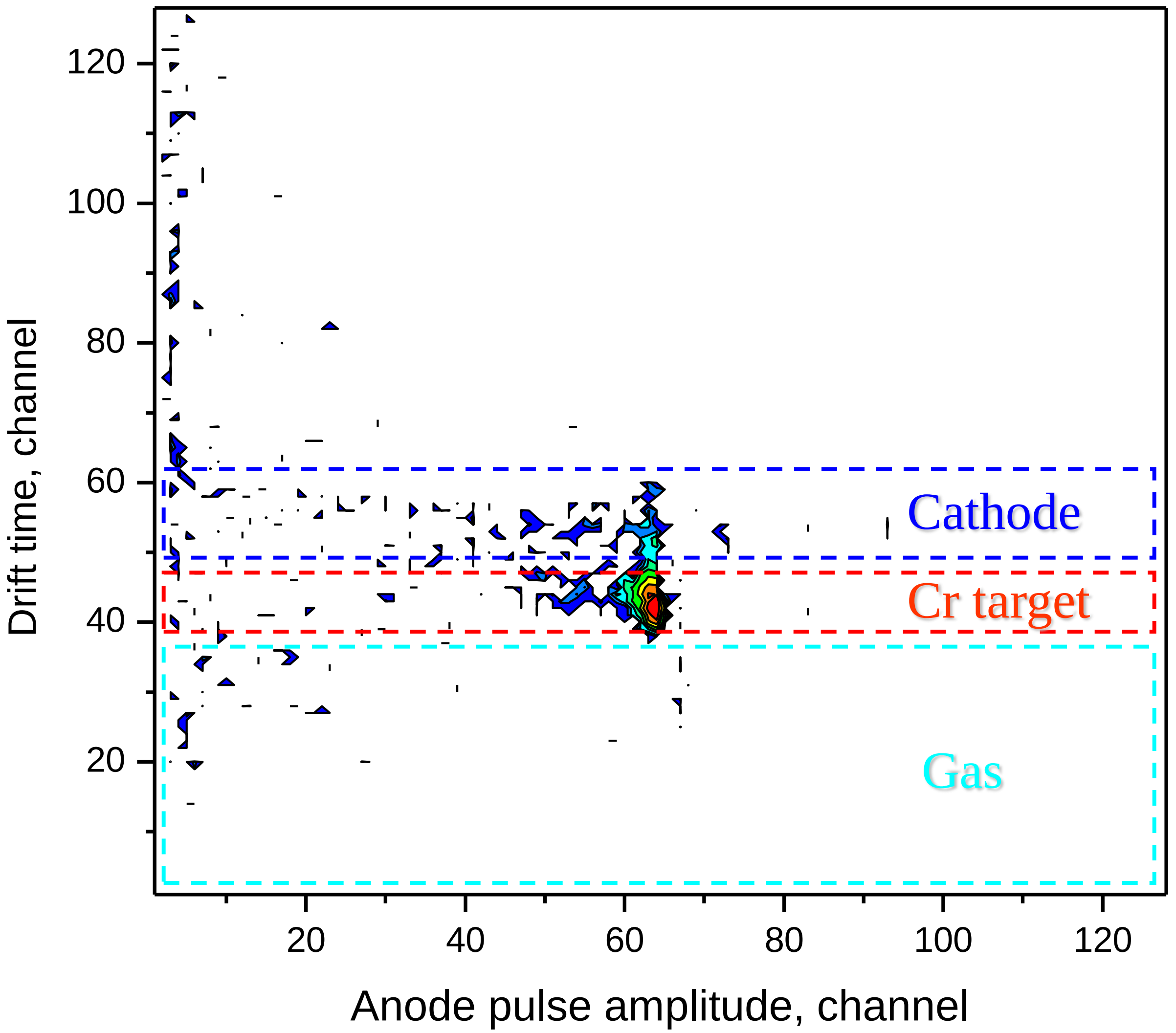
^{50}Cr target parameters

SAMPLE

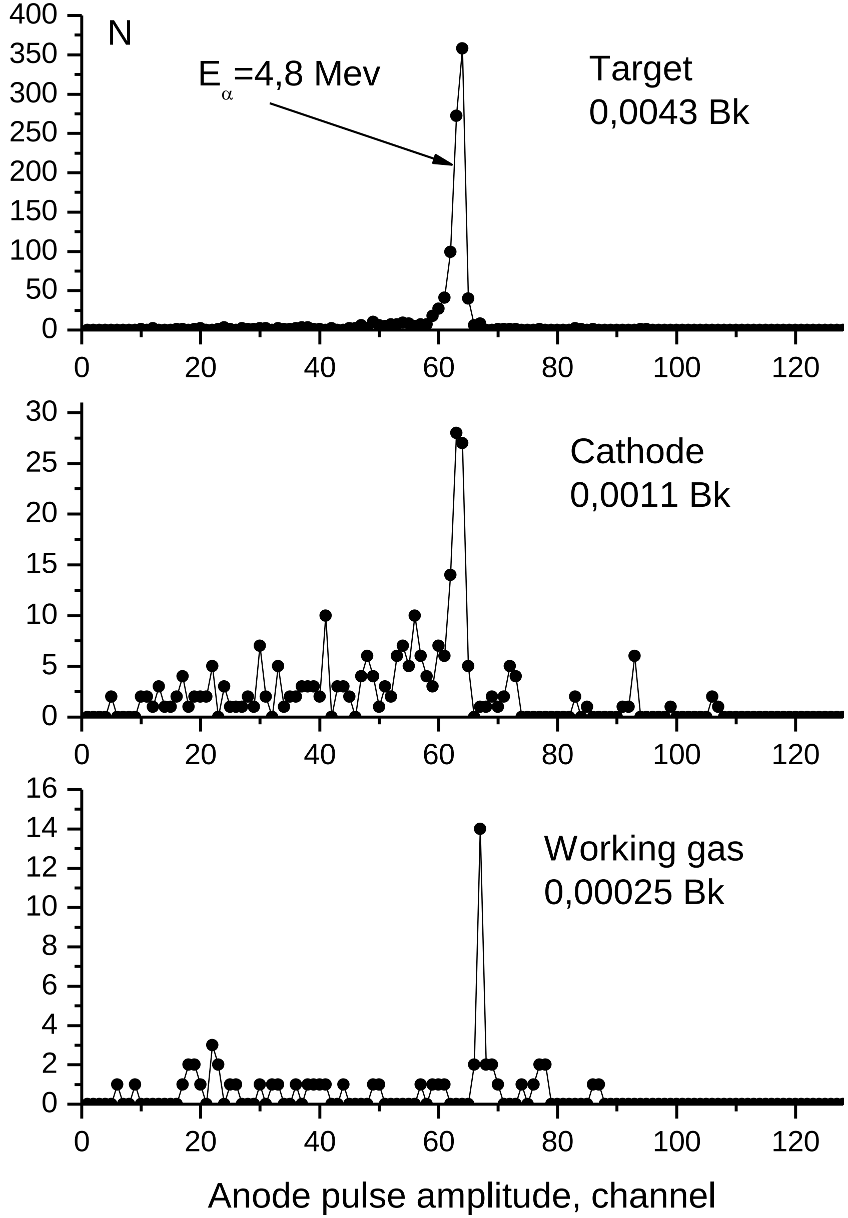
- **Gold backing of 84 mg/cm^2**
- **^{50}Cr - 365 μg/cm^2**
- **^{50}Cr enriched to 96,8 %**
- **^{52}Cr - 2.98%,**
- **^{53}Cr - 0.18%**
- **^{54}Cr - 0.04%.**

- **Target area – 14,11 cm^2**
- **Total ^{50}Cr mass – 5,15 mg**
- **Total ^{50}Cr number of atoms – 6,22*10^{19}**

Background (neutron beam off)

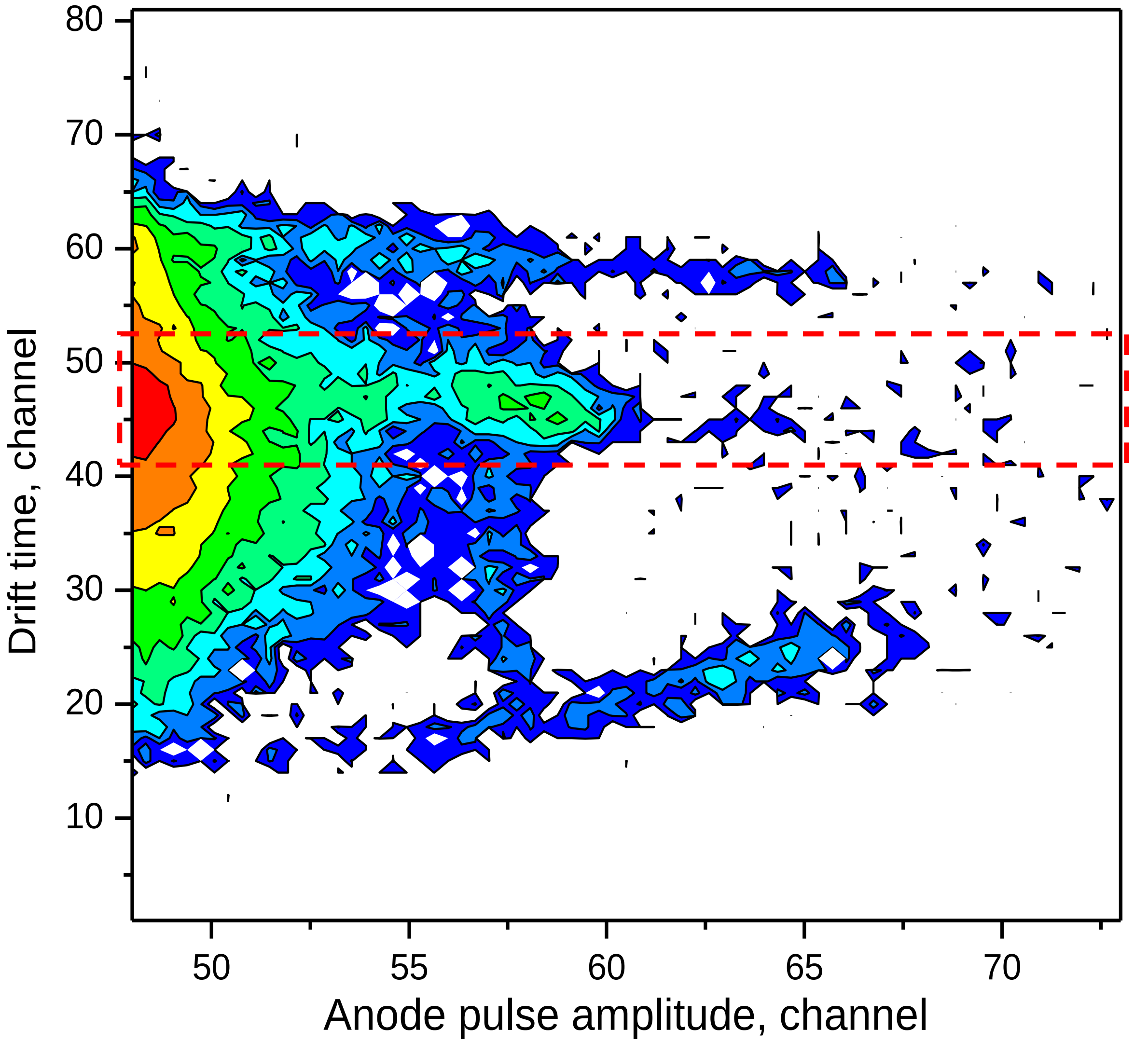


Own α - activity of the detector

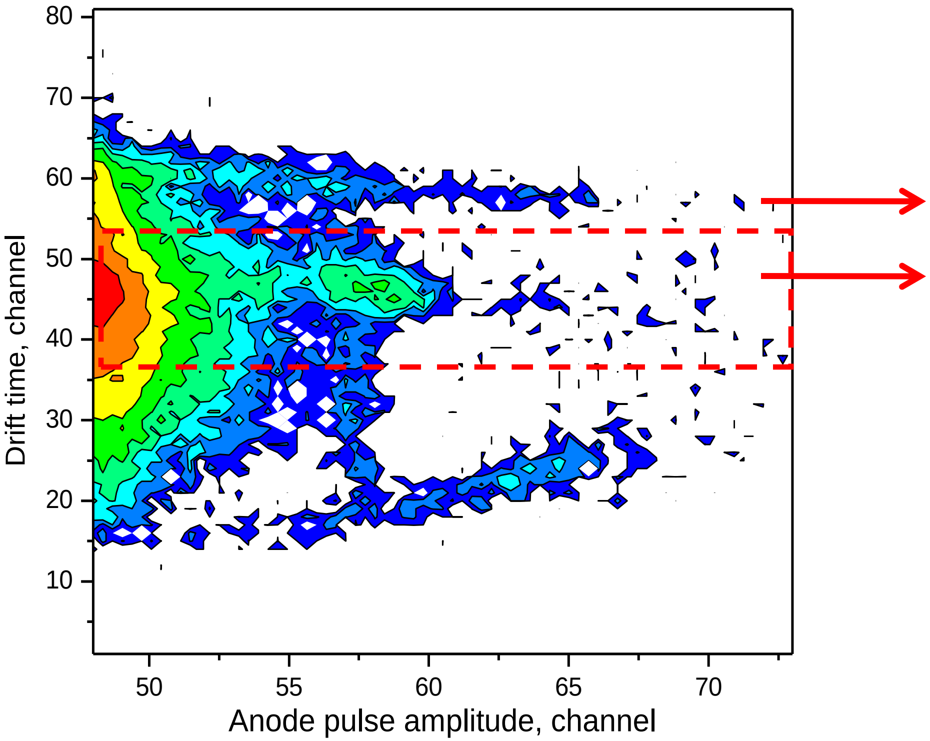


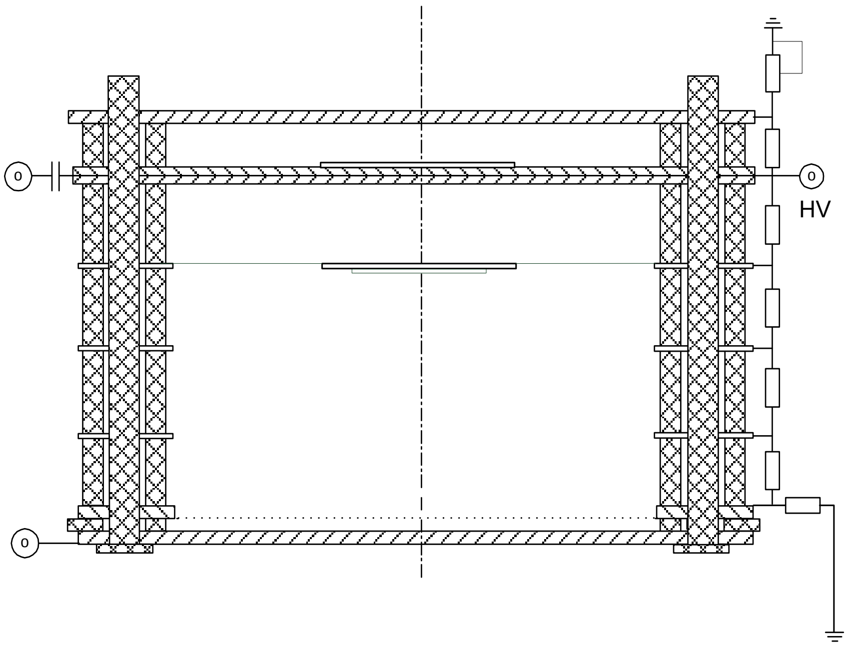
^{237}Np ??? – 4,79 (51%), 4,77 (25%), 4,65 (9%) (2,14*10^6 years).

Drift time selection for α-particles only

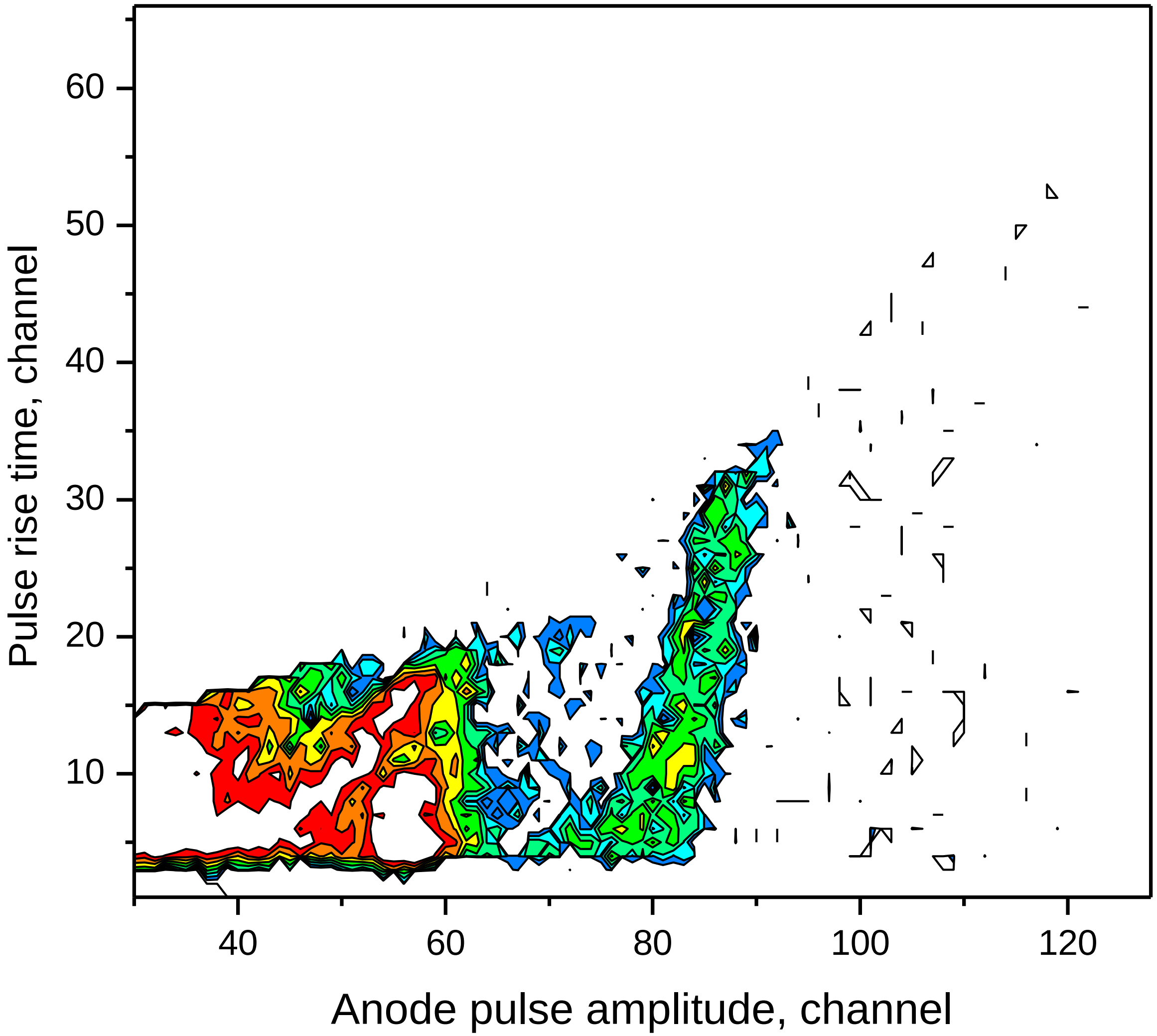


Drift time selection for α-particles only

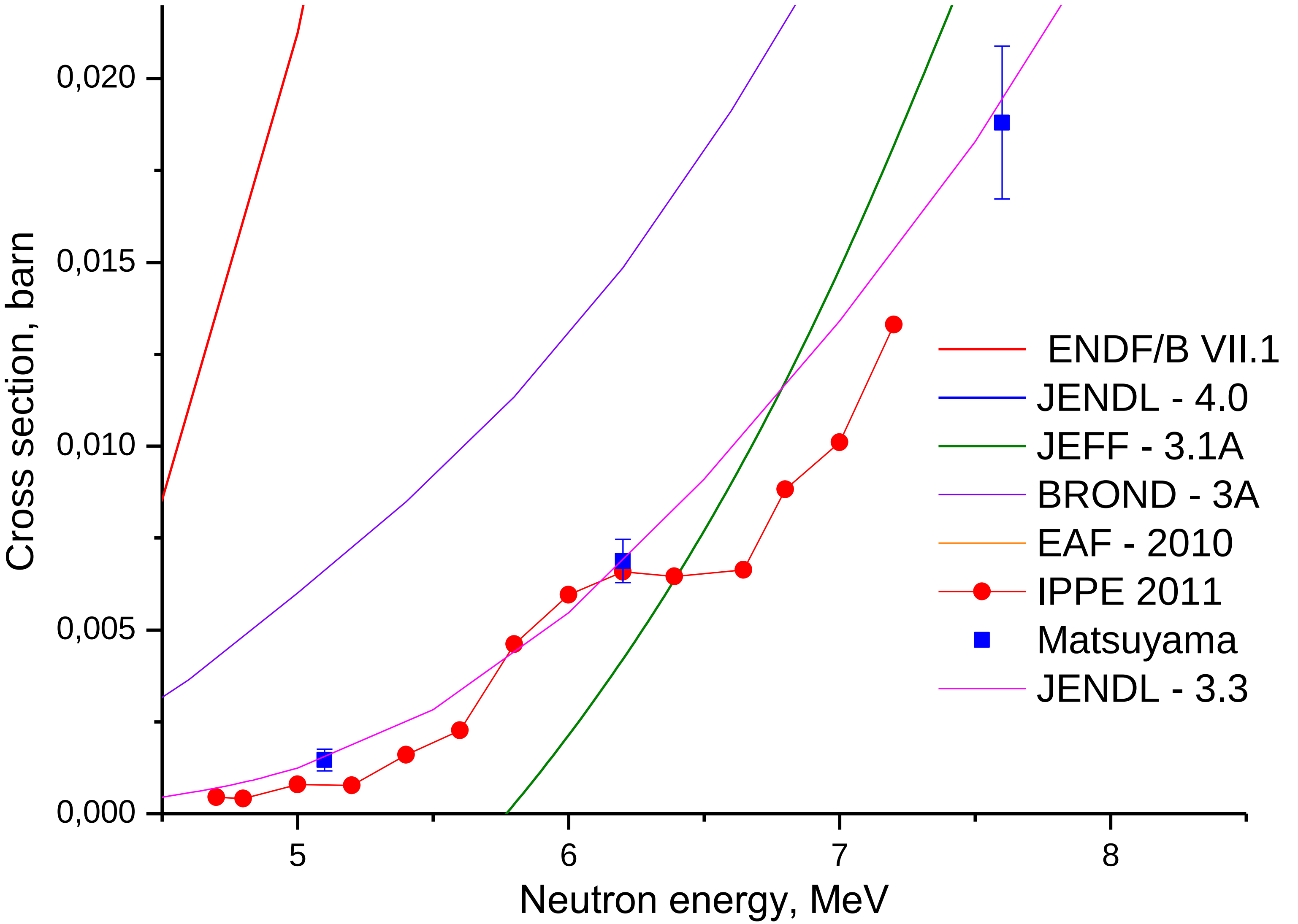




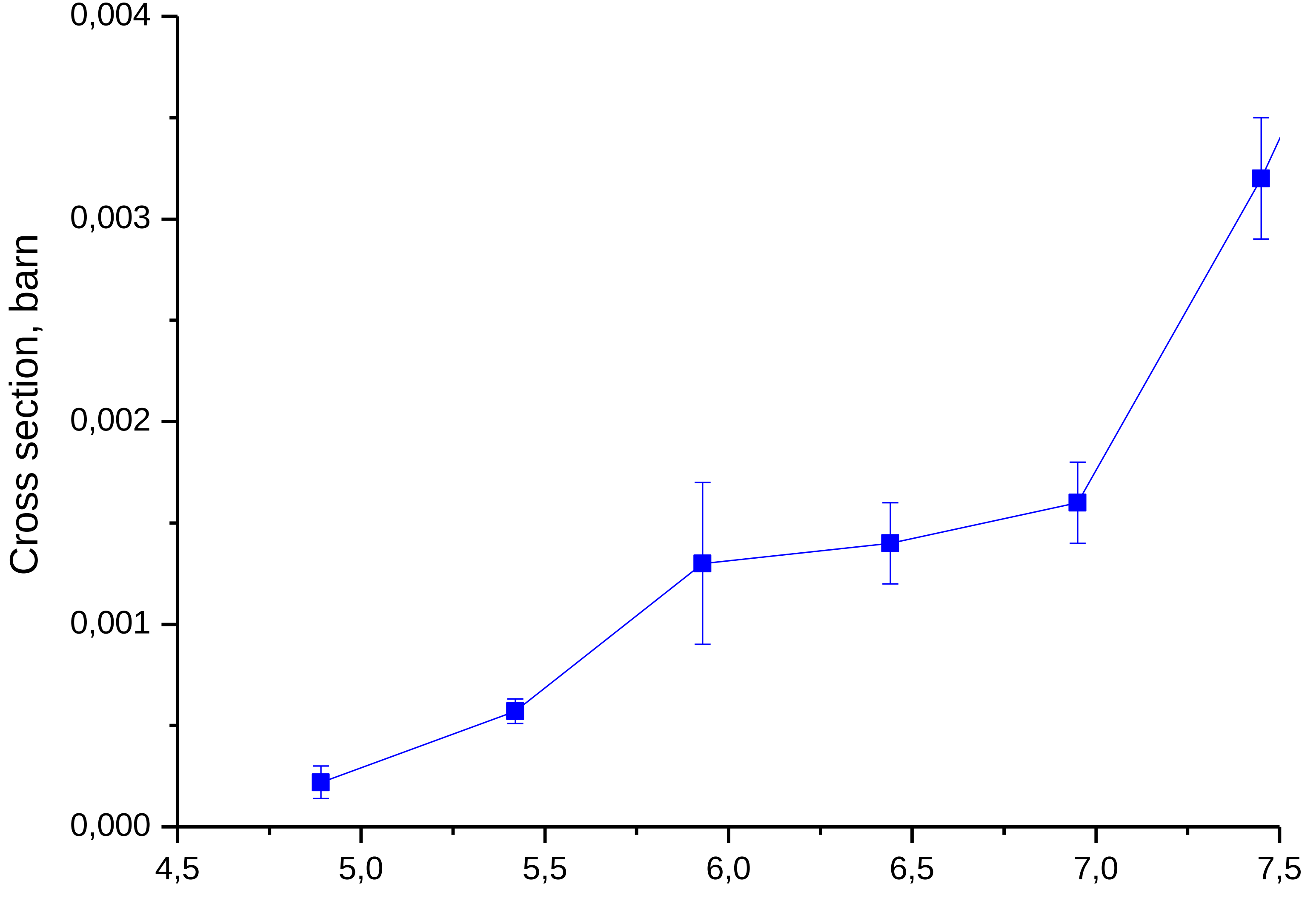
Rise time of anode signals



Result for ^{50}Cr



A.Paulsen data for natural chromium.



Experimental uncertainties

<u>Counting statistics: (статистическая ошибка)</u>

- Number of the α-particles.
- Number of the fission fragments.

<u>Errors not related to statistics:</u>

- α-particles "tail" extrapolation – 0,4%.
- Fission fragments "tail" extrapolation – 0,6%.
- Number of ^{238}U atoms – 1,5%.
- ^{238}U cross section – 1%.
- Number of chromium atoms – 3,2%.

<u>Total error not related to statistics – 3,7%.</u>
<u>Typical statistical uncertainty is from 2 to 15 %.</u>

^{52}Cr target

^{52}Cr target on the gold backing - 190 mg/cm^2.

Target diameter – 30.9 mm;

^{52}Cr target was 280 µg/cm^2.

Isotopic composition:

^{50}Cr - 0.1%,

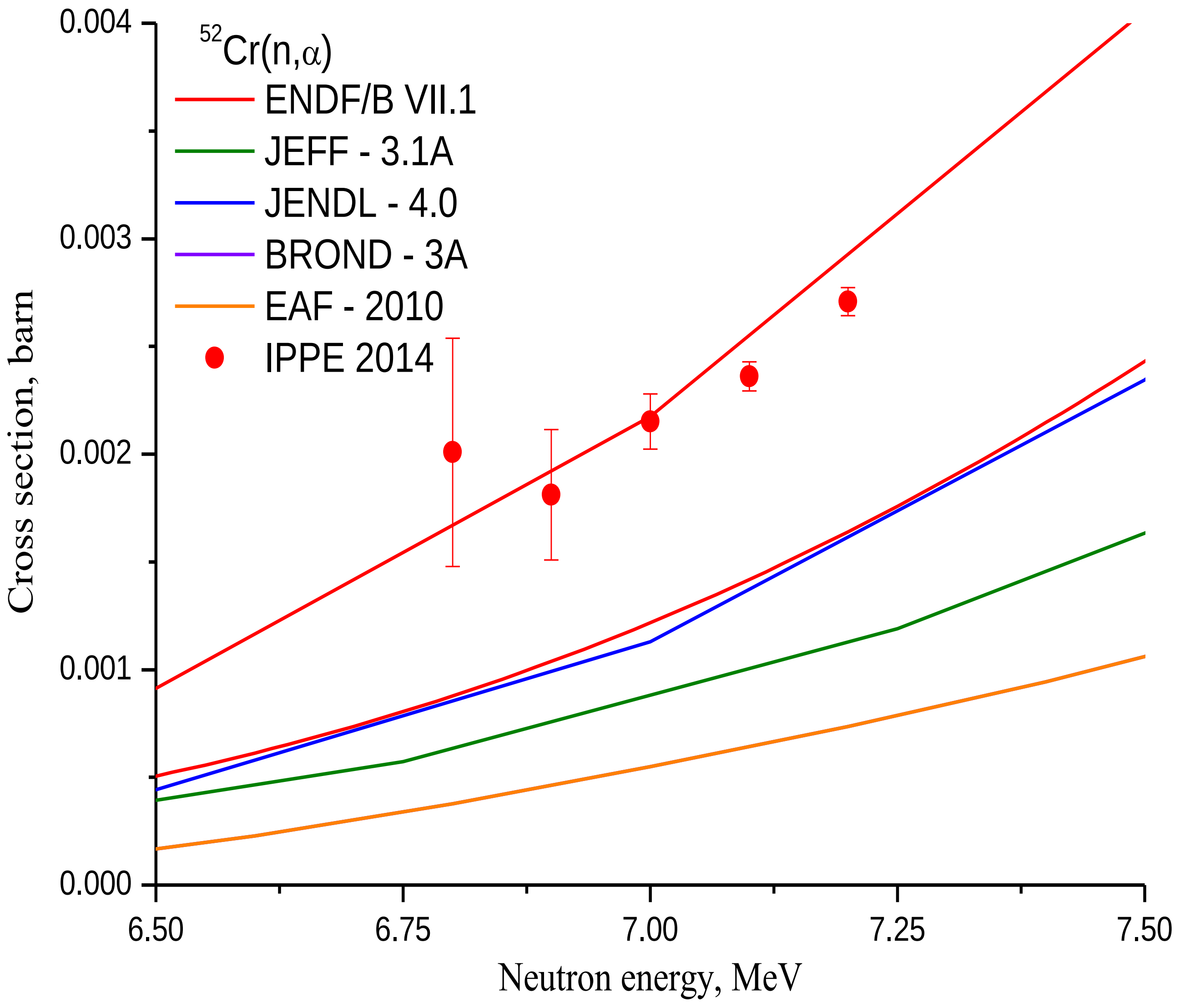
^{52}Cr-99.5%,

^{53}Cr – 0.3%

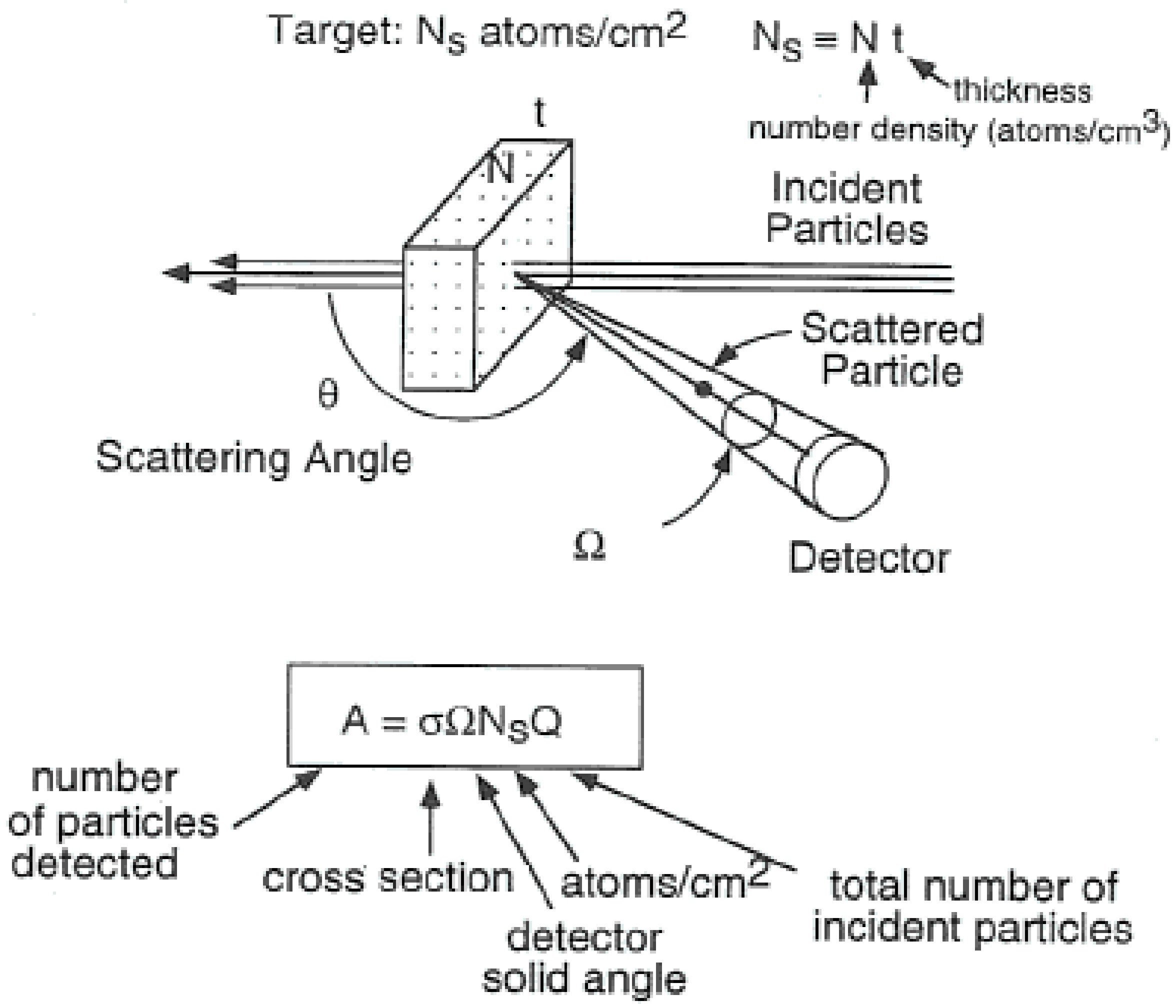
^{54}Cr – 0.1%.

Total mass of ^{52}Cr is 2.1 mg.

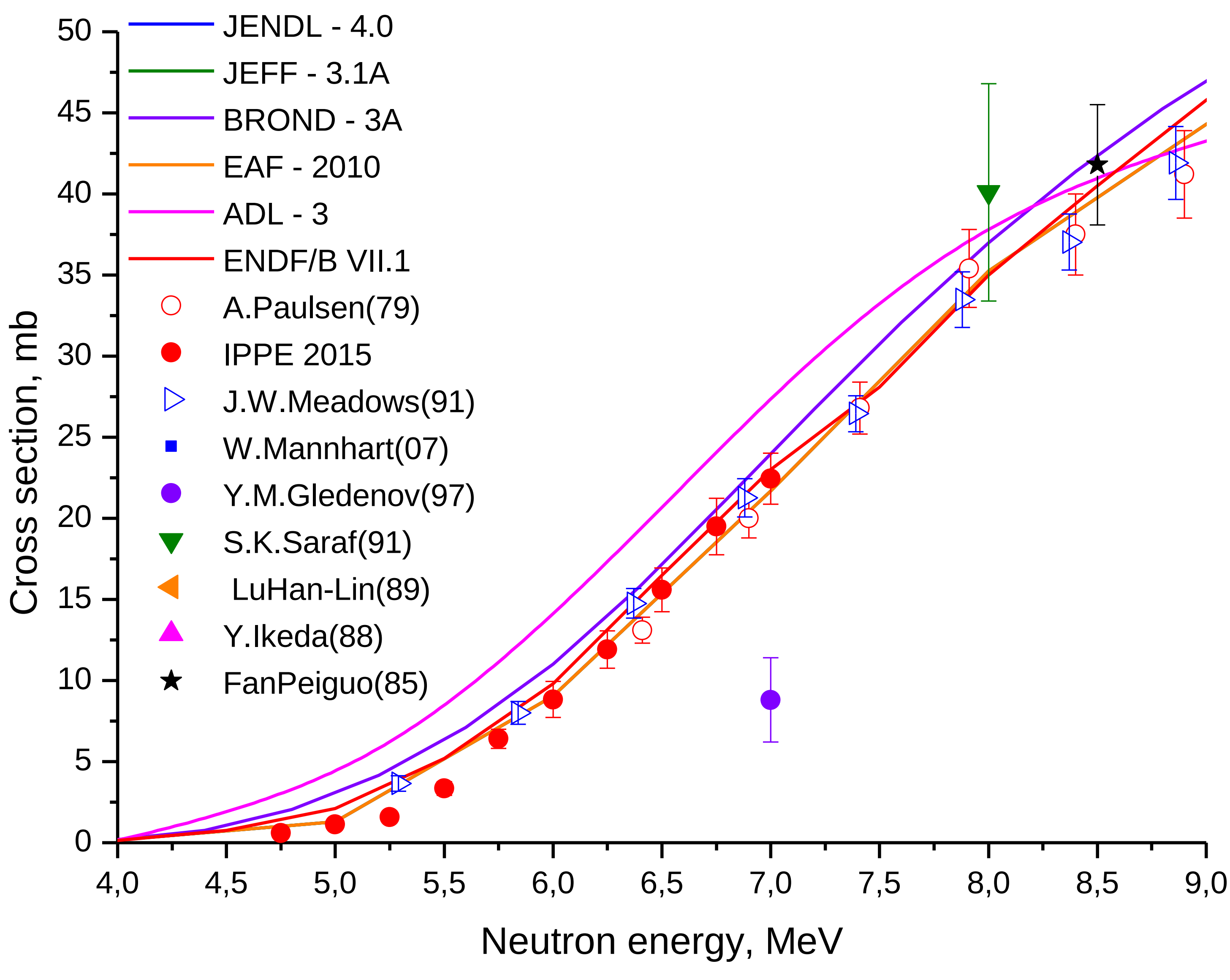
Result for $^{52}Cr(n,\alpha)^{49}Ti$ reaction cross section



Determination of the nuclei number in the self-supporting target



Result for $^{54}Fe(n,\alpha)^{51}Cr$ reaction cross section



Using digital spectrometer was measured:

- $^{10}B(n,\alpha)$ α_0 and α_1 channels
- $^{10}B(n,2\alpha t)$
- $^{14}N(n,\alpha)$ α_0, α_1, α_2 and α_3 channels
- $^{14}N(n,t)$
- $^{16}O(n,\alpha)$ α_0 channel
- $^{19}F(n,\alpha)$ α_0, α_1, α_2 and α_3 channels
- $^{20}Ne(n,\alpha)$ α_0, α_1, α_2 and α_3 channels
- $^{36}Ar(n,\alpha)$ α_0, α_1, α_2 and α_3 channels
- $^{40}Ar(n,\alpha)$ α_0 and α_1 channels
- $^{50}Cr(n,\alpha)$
- $^{52}Cr(n,\alpha)$
- $^{53}Cr(n,\alpha)$
- $^{54}Fe(n,\alpha)$
- $^{57}Fe(n,\alpha)$

Conclusion

- A few modification of digital spectrometry for (n,α) reaction products registration was developed.
- Background of different nature can be efficiently suppressed.
- Measurements for set of light and structural material isotopes was done.