EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

The ^{14}N(n,p)^{14}C and ^{35}Cl(n,p)^{35}S reactions at n_TOF-EAR2: dosimetry in BNCT and astrophysics.

[submission date: 30/05/2017]

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C. Abia[6], F. Arias de Saavedra[1], I. Domínguez[6], B. Fernández[3,7], M. Macías[3,7]

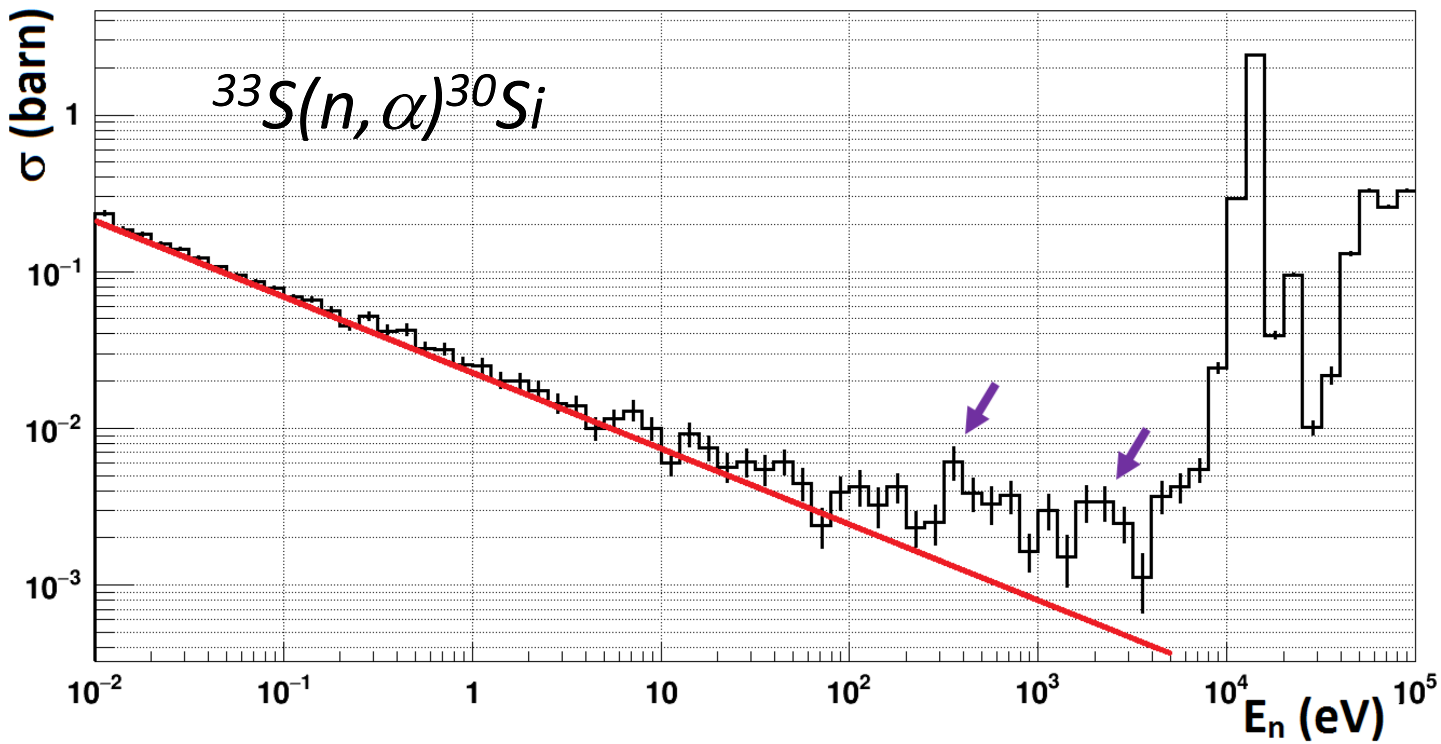
Motivations

- **Neutron capture therapy.**
  - Delivered dose to healthy tissue.
  - Limiting factor in whatever radiotherapy.

- **Astrophysics.**
  - ^{14}N neutron absorber (poison). ^{14}N, protons for ^{19}F.
  - ^{35}Cl involved in the origin of ^{36}S. ^{35}Cl/^{37}Cl deviation from solar.

- **Improving capabilities of n_TOF EAR2:**
  - Setup for (n,p) reactions with low energy protons (590 keV).
  - Continuation of ^{33}S(n,α)^{30}Si experiment at EAR2.

$^{33}S(n,\alpha)$ at EAR2 measured in 2015.

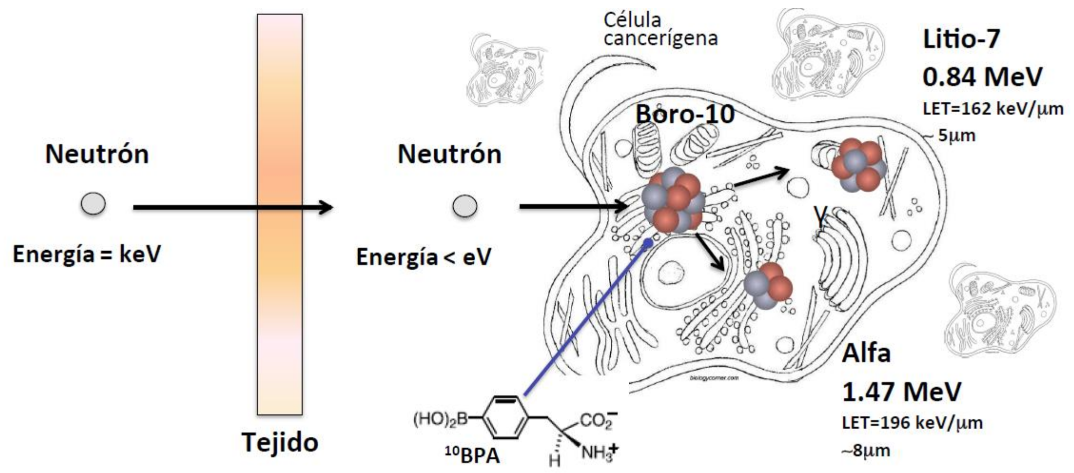


The cross-section was measured below 10 keV for the first time.

New resonances were found.

Marta Sabaté-Gilarte PhD at CERN (May 2017).

Boron Neutron Capture Therapy (BNCT).



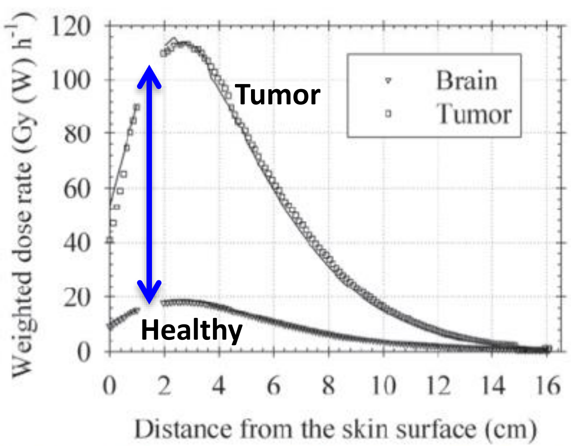


Figure 14 SERA (line) and JCDS (symbol) calculations for the total depth distributions in brain and tumor for the anterior field, using a 14 cm diameter circular FiR 1 beam. The boron dose (D_B) was calculated for 19 mg/g (ppm) of ^{10}B in brain and 66.5 µg/g (ppm) ^{10}B in tumor.

The effect of neutrons in healthy tissue is much lower than in tumor ^{10}B-load.

However the dose in healthy is the limiting factor in whatever radiotherapy treatment.

This proposal is focused on the improvement of the nuclear data for healthy tissue dosimetry.

BNCT: reference dosimetry calculations.

Kerma factor takes into account the concentration and the cross-section. K=N·σ.

They are necessary for the final simulation of the physical and biological dose delivered to the tumor and healthy tissue.

Reference dosimetry calculations for neutron capture therapy with comparison of analytical and voxel models

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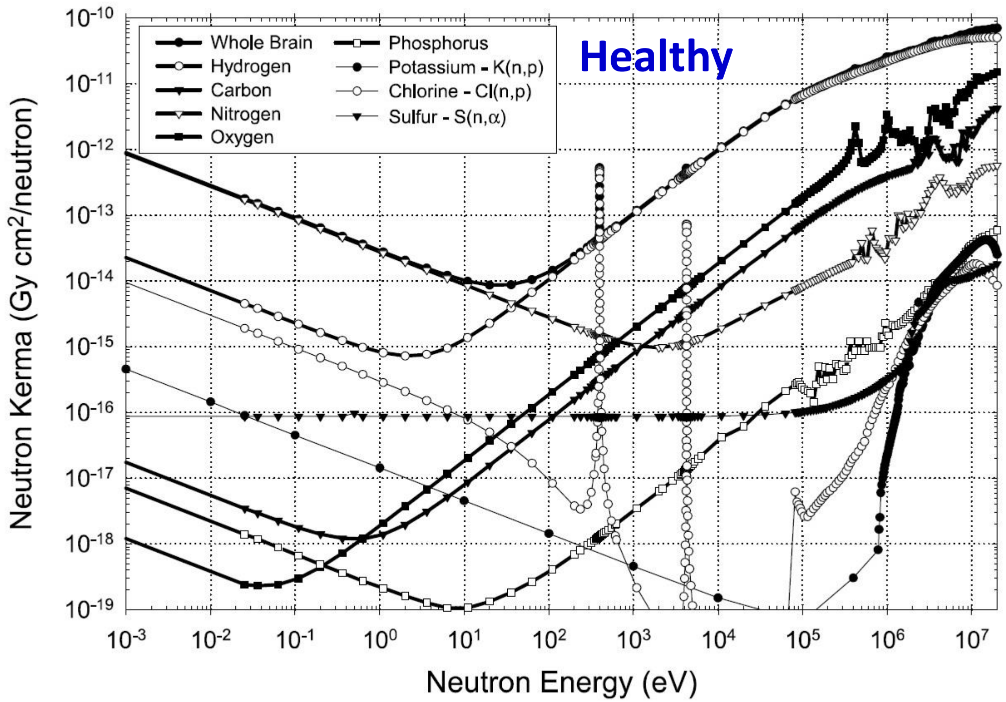
Med. Phys. 29 (2), February 2002



The $^{14}N(n,p)$ is the most important for En < 50 eV.

The $^{35}Cl(n,p)$ contributes significantly due to the resonances and more than ^{16}O and ^{12}C.

IAEA new BNCT-TECDOC: from reactors to accelerators.

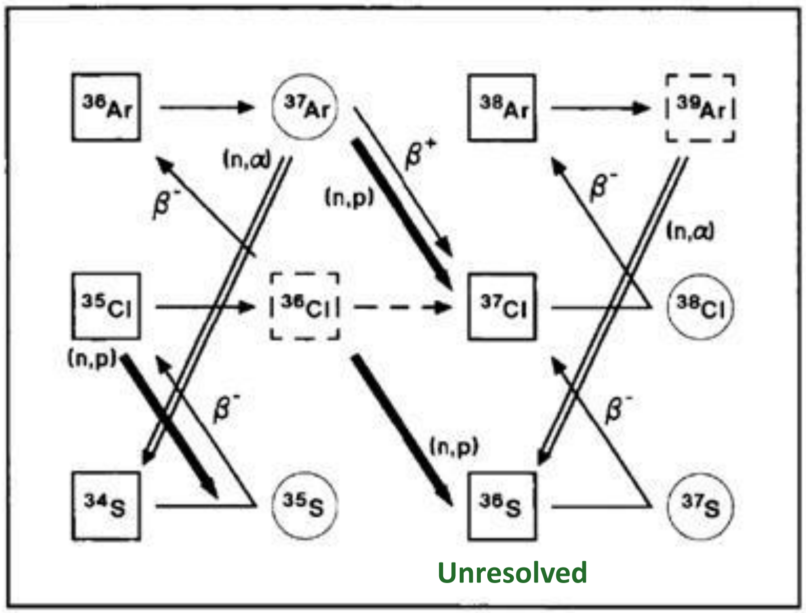
The technical document (IAEA-TECDOC-1223, 2001) IAEA established the figure-of-merit (FOM) for NCT.

FOMs and nuclear data are being reviewed for accurate dosimetry (in tumor and healthy tissue) having in mind the accelerator-based neutron sources in construction for BNCT.

The International Commission on Radiation Units and Measurements (ICRU) recommends ~5% as a deviation from the prescribed dose.

$^{35}Cl(n,p)^{35}S$ in astrophysics: ^{36}S origin.

S. Druyts et al. / Nuclear Physics A573 (1994) 291–305



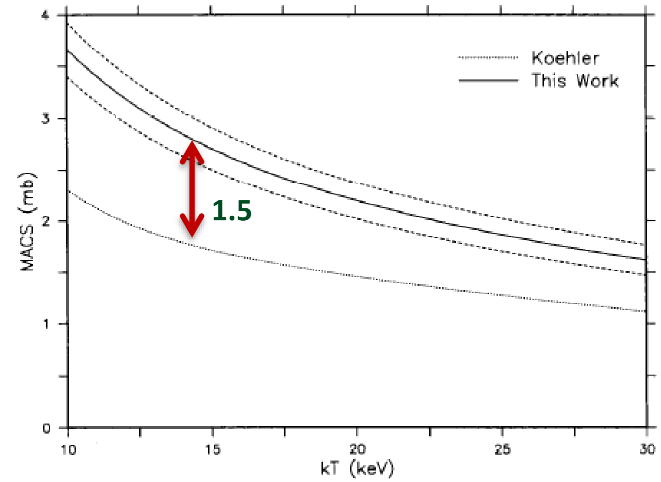


Fig. 6. Comparison of the MACS-values obtained from Koehler's measurement [12] and this work, as a function of kT. The dashed lines above and below our result indicate a one-standard-deviation uncertainty.

Important discrepancies remain in the MACS between Druyts *et al.* and Koehler.

MACS are fundamental input parameters for stellar models.

5. Astrophysical implications

Although it is clear that the MACS-values needed in nucleosynthesis calculations are only relevant for kT-values in the keV range, one should not underestimate the importance of an accurate cross-section value at thermal neutron energy. The latter value is indeed often used to normalize cross-section data at higher neutron energies, hence it has a direct impact on MACS-values, which may be very important. A very striking example are the $^{35}Cl(n,p)$ measurements of Popov et al. [10]. They were normalized to a thermal cross section of 190 ± 50 mb, which is 2.3 times lower than the value determined in this work! More recently, Koehler [12] normalized his $^{35}Cl(n,p)$ measurement to a thermal value of 489 ± 14 mb, which is 11% higher than the value reported here.

$^{35}Cl(n,p)^{35}S$ in astrophysics: its origin and isotopic ratio.

Circumstellar envelope of carbon-rich AGB.

Table 2. Observed isotopic ratios towards IRC+10216.

Ratio	Value	$1\ \sigma$	Ref.[a]	Solar[b]
$Na^{35}Cl/Na^{37}Cl$ (7-6)	2.33	0.50	(1)	
$Al^{35}Cl/Al^{37}Cl$ (15-14)	2.15	0.33	(1)	
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$Al^{35}Cl/Al^{37}Cl$ (11-10)	2.40	0.76	(3)	
$^{35}Cl/^{37}Cl$[c]	2.30	0.24	(1)	3.13

First observational evidence of significant deviation from solar isotopic ratio of elements heavier than the CNO.

→

All sources of uncertainty must be studied and removed to check the s-process calculations.

The $^{35}Cl(n,p)^{35}S$ is relevant for models.

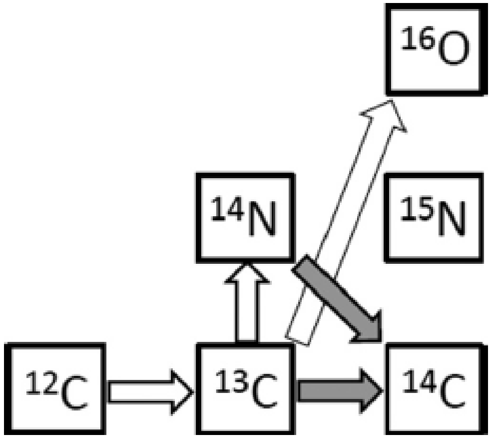
Astron. Astrophys. 357, 669–676 (2000)

ASTRONOMY AND ASTROPHYSICS

Improved isotopic ratio determinations in IRC+10216, the progenitor mass and the *s* process

C. Kahane[1], E. Dufour[1], M. Busso[2], R. Gallino[3], M. Lugaro[4], M. Forestini[1], and O. Straniero[5]

$^{14}N(n,p)^{14}C$ in astrophysics: neutron poison s-process.



CNO cycle:

$^{14}N(n,p)^{13}C(p,\gamma)^{14}N$ forms reaction cycle.

It is the most important neutron poison in s-process.

Wallner *et al.* (2015) measured by activation-AMS the MACS at kT=25 and 100 keV and they obtained values 11% and 100%, respectively, lower than the evaluations.

They pointed out that the origin of the discrepancies is in the influence of the resonance at 493 keV at lower energies.

Below 493 keV there is a lack of data as will be shown.

$^{14}N(n,p)^{14}C$ in astrophysics: ^{19}F origin and tracer.

^{19}F is easily destroyed in stellar interiors by proton and alpha particle capture reactions.
$^{19}F(p,\alpha)^{16}O$ and $^{19}F(\alpha,p)^{22}Ne$

Thus, model has to enable ^{19}F to escape from the stellar interiors after its production.

This makes ^{19}F a useful tracer of the physical conditions in stellar interiors.

In low and intermediate-mass AGB stars, C. Abia *et al.* have been found that additional sources of F are needed to explain the observed evolution of this element.

The $^{14}N(n,p) \rightarrow {}^{18}O(p,\alpha)^{15}N(\alpha,\gamma)^{19}F$ could be one of these sources.

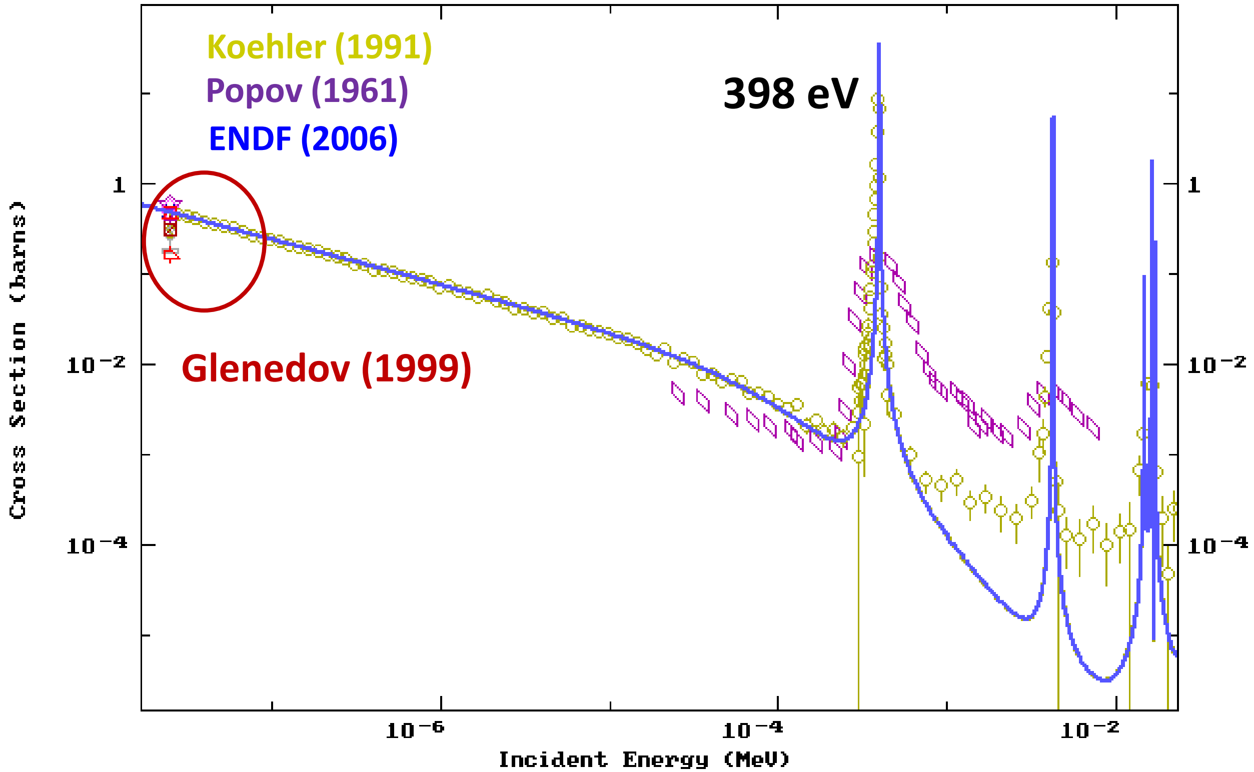
A&A 581, A88 (2015)
DOI: 10.1051/0004-6361/201526586

Astronomy & Astrophysics

The origin of fluorine: abundances in AGB carbon stars revisited

C. Abia[1], K. Cunha[2], S. Cristallo[3,4], and P. de Laverny[5]

$^{35}Cl(n,p)^{35}S$ status: evaluations and thermal.



MACS

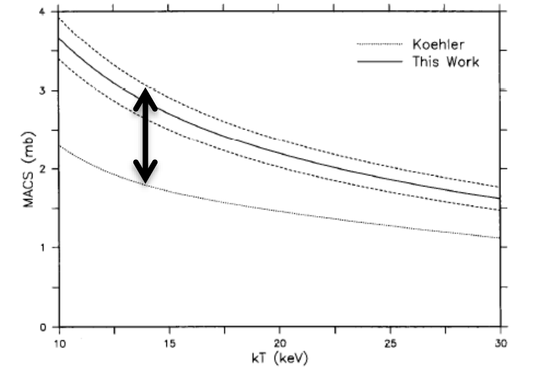


Fig. 6. Comparison of the MACS-values obtained from Koehler's measurement [12] and this work, as a function of kT. The dashed lines above and below our result indicate a one-standard-deviation uncertainty.

Evaluations are based on Koehler at thermal and in the energy range below 398-eV resonance (1/v) and in the resonances region are based on Druyts.

Discrepancies in the MACS, already mentioned, could be originated by the thermal value.

Thermal value are discrepants: Druyts *et al.* (440 mb), of Gledenov *et al.* (575 mb), Koehler (480 mb) .

$^{35}Cl(n,p)^{35}S$ status: data in the resonances.

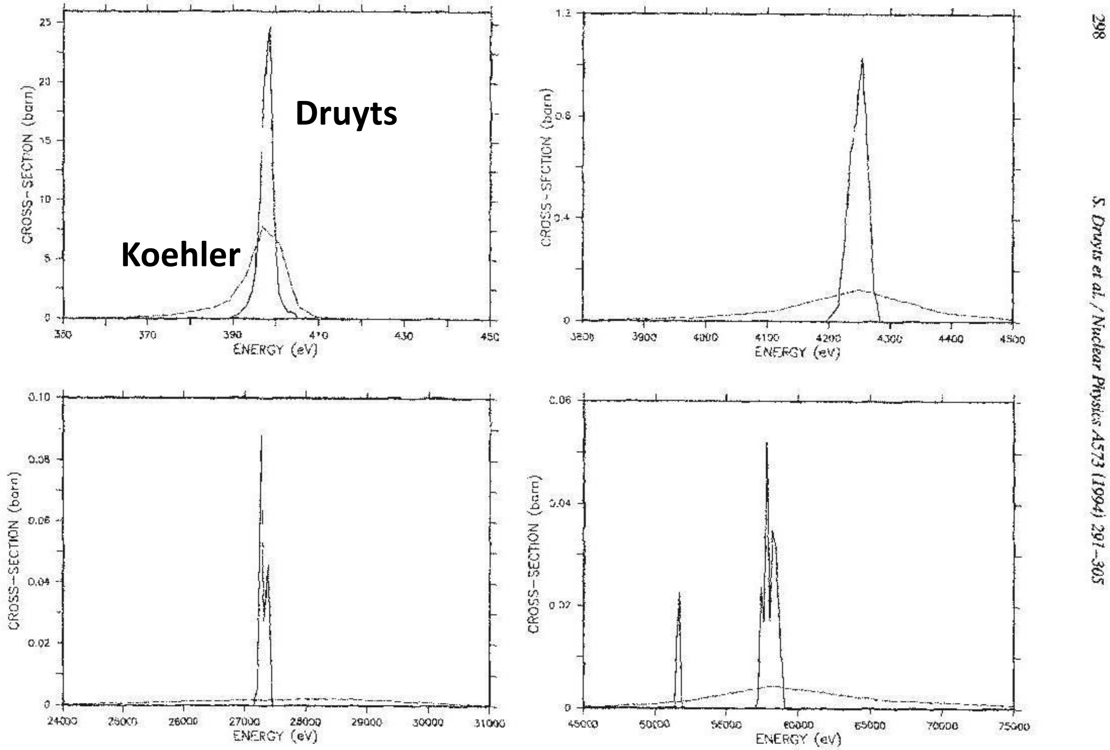


Fig. 3. $^{35}Cl(n,p)^{35}S$ reaction cross section in the region of four resonances obtained in this work (full line), as compared to Koehler's work [12] (dash-dotted line).

Druyts *et al.* (1994) and Koehler (1991).

Druyts *et al.* had higher resolution but they did not provide data between resonances.

Area of many resonances are discrepant.

^{35}Cl(n,p)^{35}S reaction status: evaluation in the resonances.

R-MATRIX ANALYSIS OF Cl NEUTRON CROSS . . . PHYSICAL REVIEW C **73**, 044603 (2006)

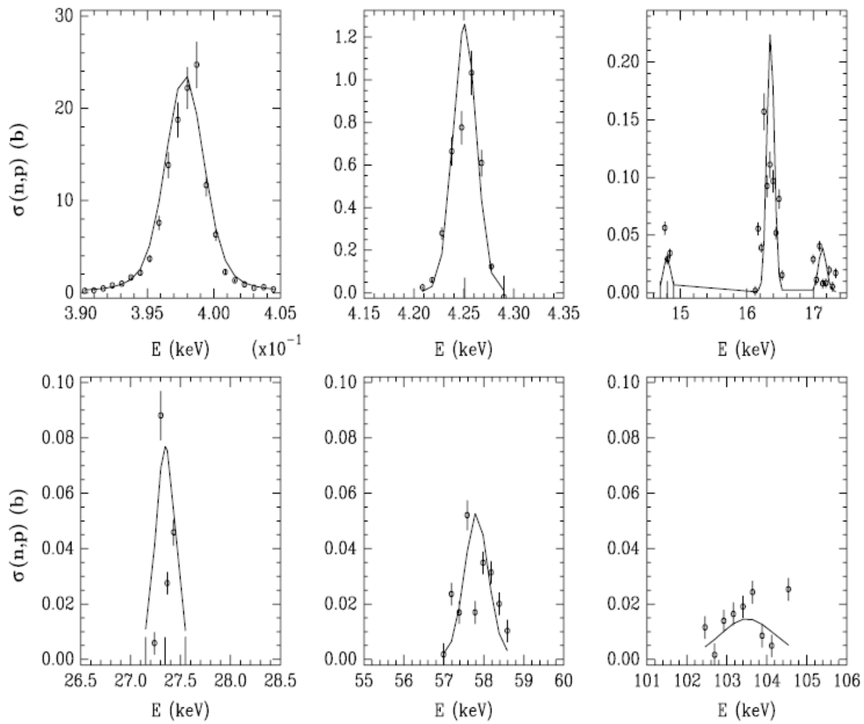


FIG. 6. Comparison of SAMMY fits (solid line) to the ^{35}Cl(n,p) cross section data of Druyts *et al*. [13] for selected resonances.

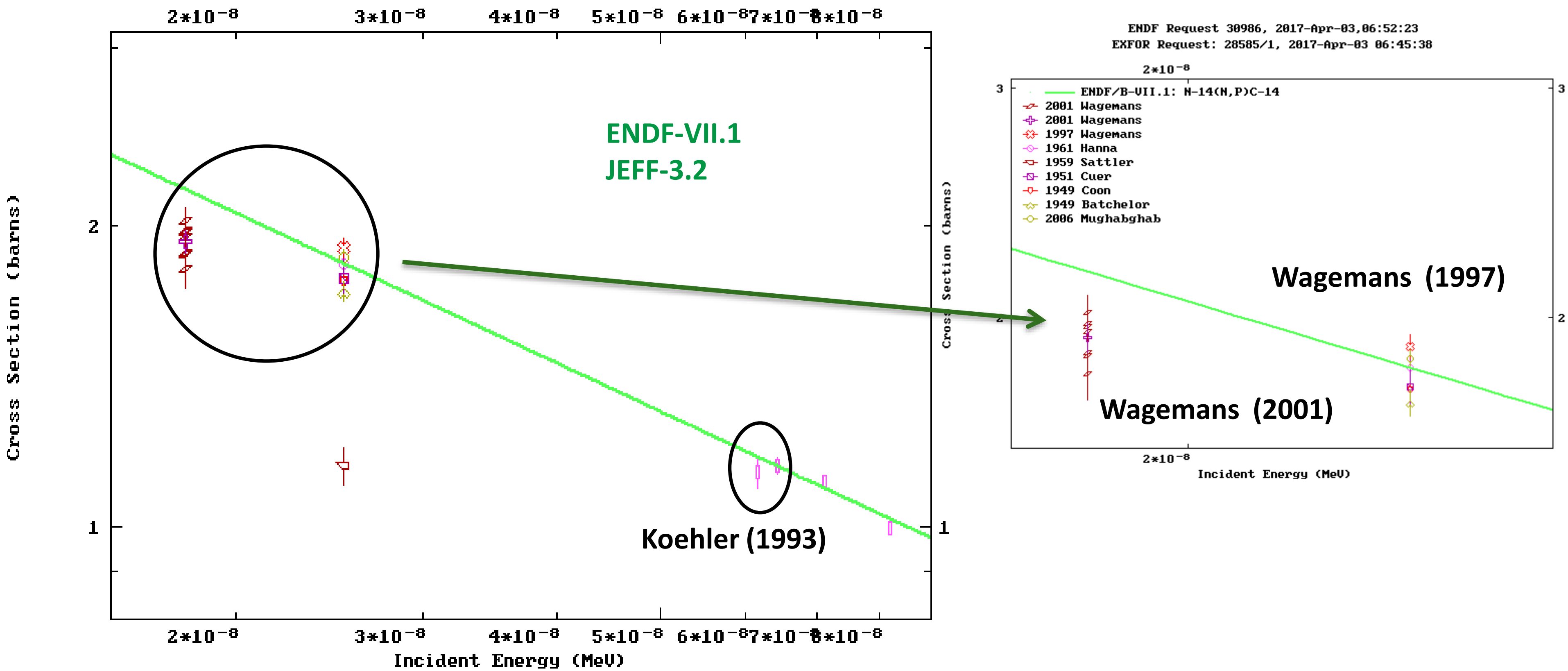
Resonances of Druyts can be only fitted using the thermal value of Koehler wich is 15% higher than Druyts.

Significant discrepancies in the resonance strength $\omega = g\Gamma_n \Gamma_p / \Gamma$ between Koehler and Druyts were found for the most important resonances.

Many resonances had not a satisfactory description with SAMMY code.

n_TOF-EAR2 can provide new data in the whole energy range from thermal to MeV region and at the same time resolving the resonances in the keV region.

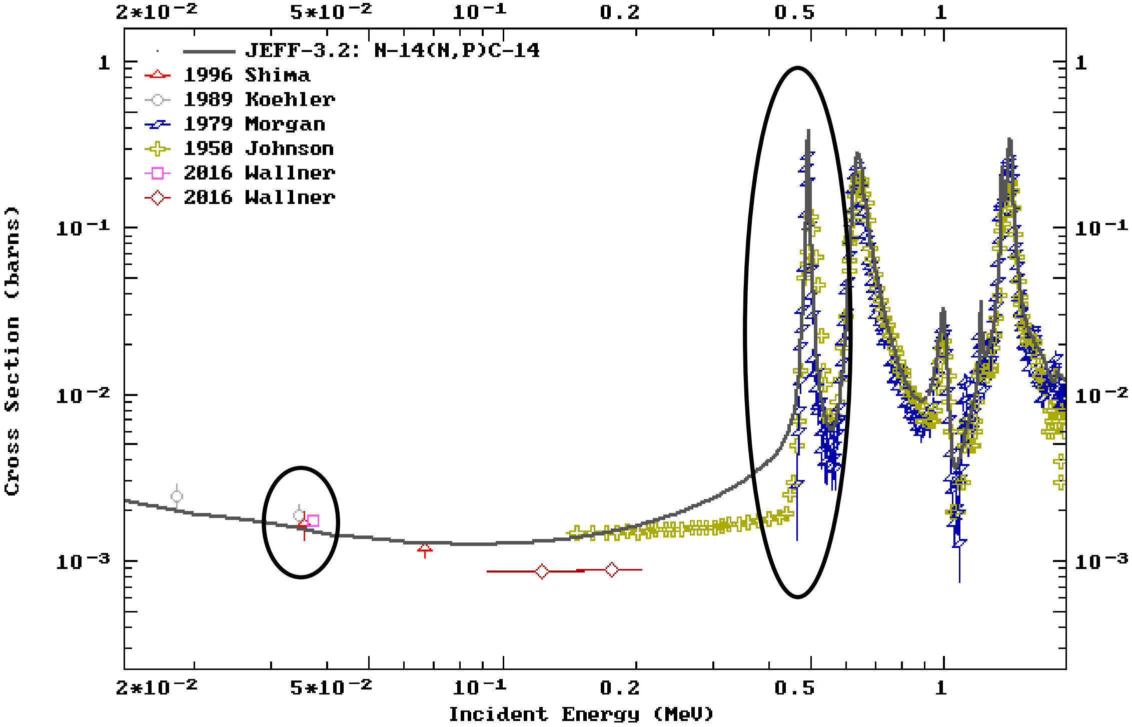
$^{14}N(n,p)^{14}C$ reaction status: thermal and 1/v.



Koehler reached near thermal value but the extrapolation to thermal does not match the most recent measurements of Wagemans (2001), "careful new evaluation" is needed.

More accurate value is very important in terms of kerma for BNCT as previously shown.

$^{14}N(n,p)^{14}C$ reaction: status at keV-MeV range.



Wallner *et al.* and Shima *et al.* are Spectrum Average measurements, no differential data.

A factor of 2 has been recently obtained by Wallner in the 100 keV region, very important in astrophysics.

Morgan (1979) measured 493-keV resonance with a strength lower than Johnson (1958).

Later, Wallner *et al* (2015) has found that is needed a further reduction in a factor 3.3 of the strength of the 493-keV resonance obtained by Morgan in order to explain the MACS from kT=10-100 keV

n_TOF-EAR2 can provide new data in the whole energy range from thermal to MeV resolving the resonances.

CERN-INTC-2014-007/INTC-I-156: samples.

Ready 4 KCl samples: 247-250 μg/cm².

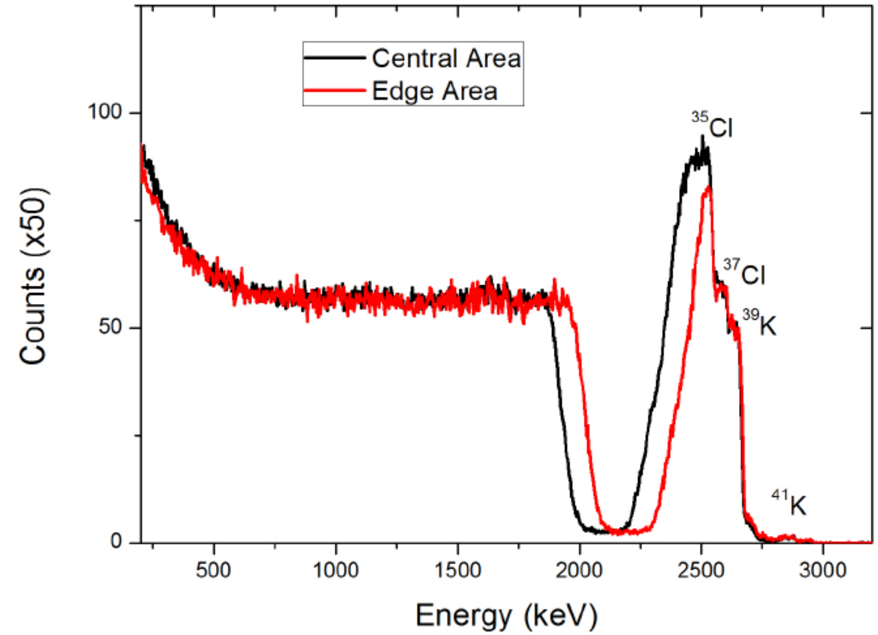
Ready 4 $C_5H_5N_5$ samples: 180-270 μg/cm².

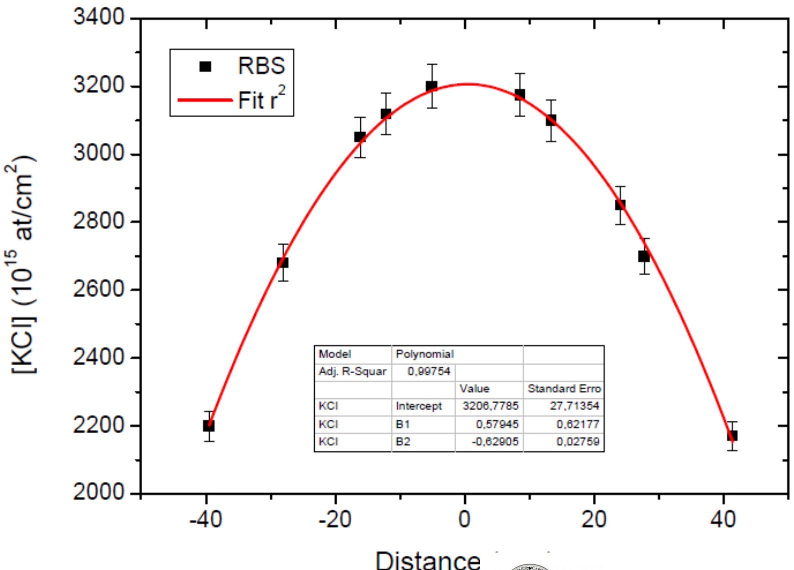
Evaporation in vacuum onto 16 μm Al backing.

«Koehler» tecnique (300-Cl35/160-N14 μg/cm²).

Druyts was evaporation of AgCl onto Pt-Al backing

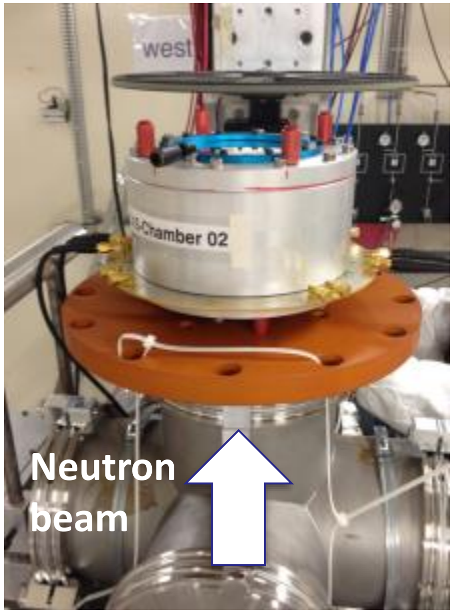
We have performed a RBS study of the sample showing a r^2 distribution (no similar studies).

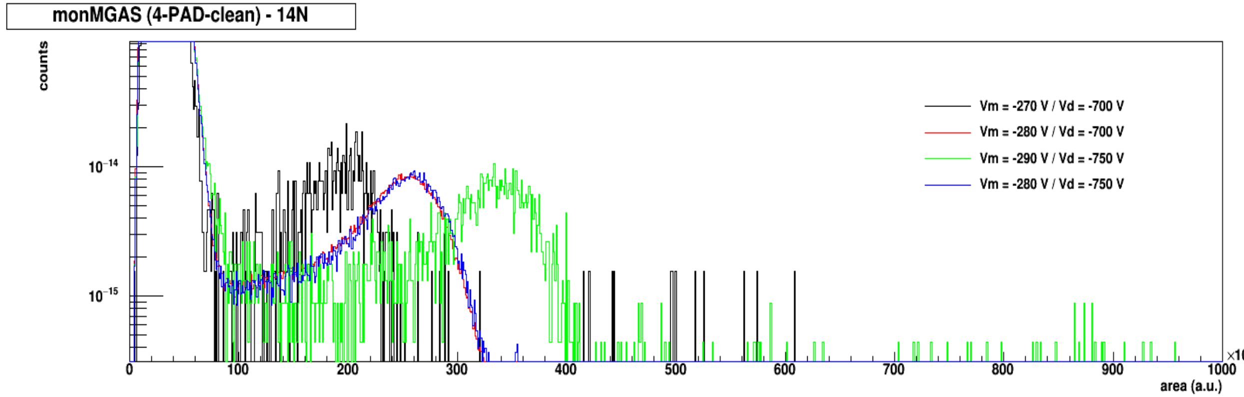


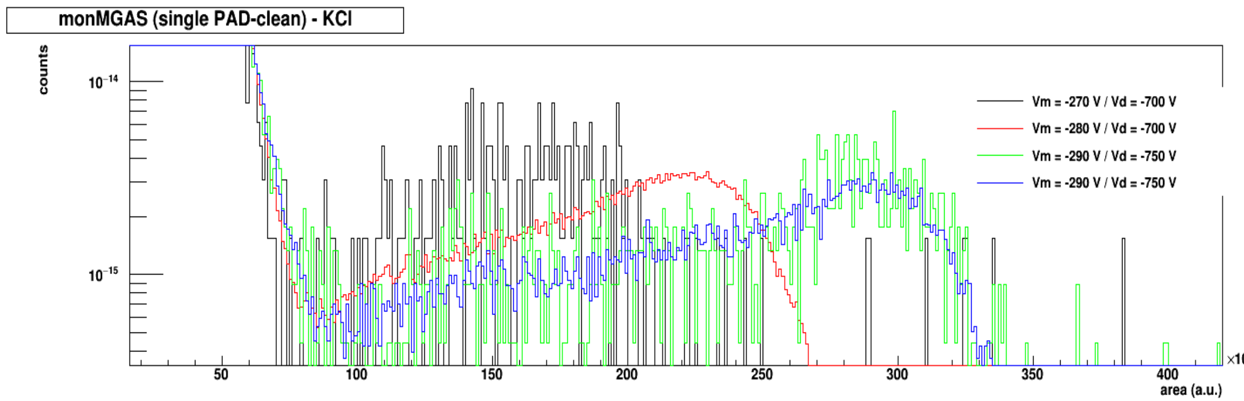


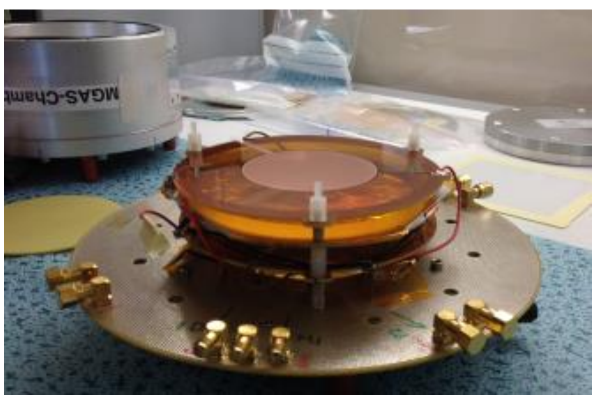
A r^2 distribution in the mass thickness could have a significant influence in the study of Druyts on the proton emission and Koehler.

CERN-INTC-2014-007/INTC-I-156: results.



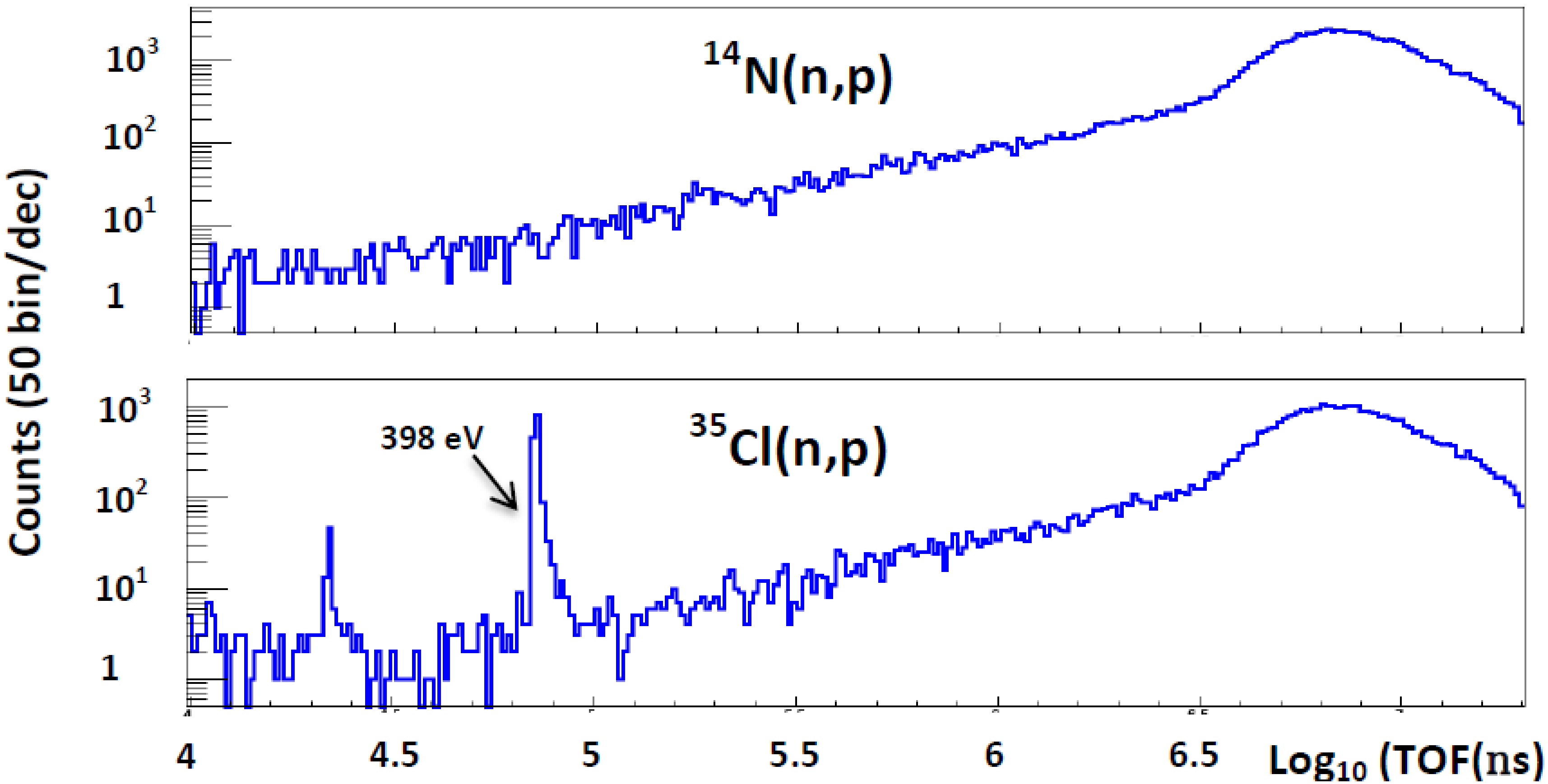






Different voltages and gains were studied in order to find a good detection of the low energy protons with a good separation from the noise.

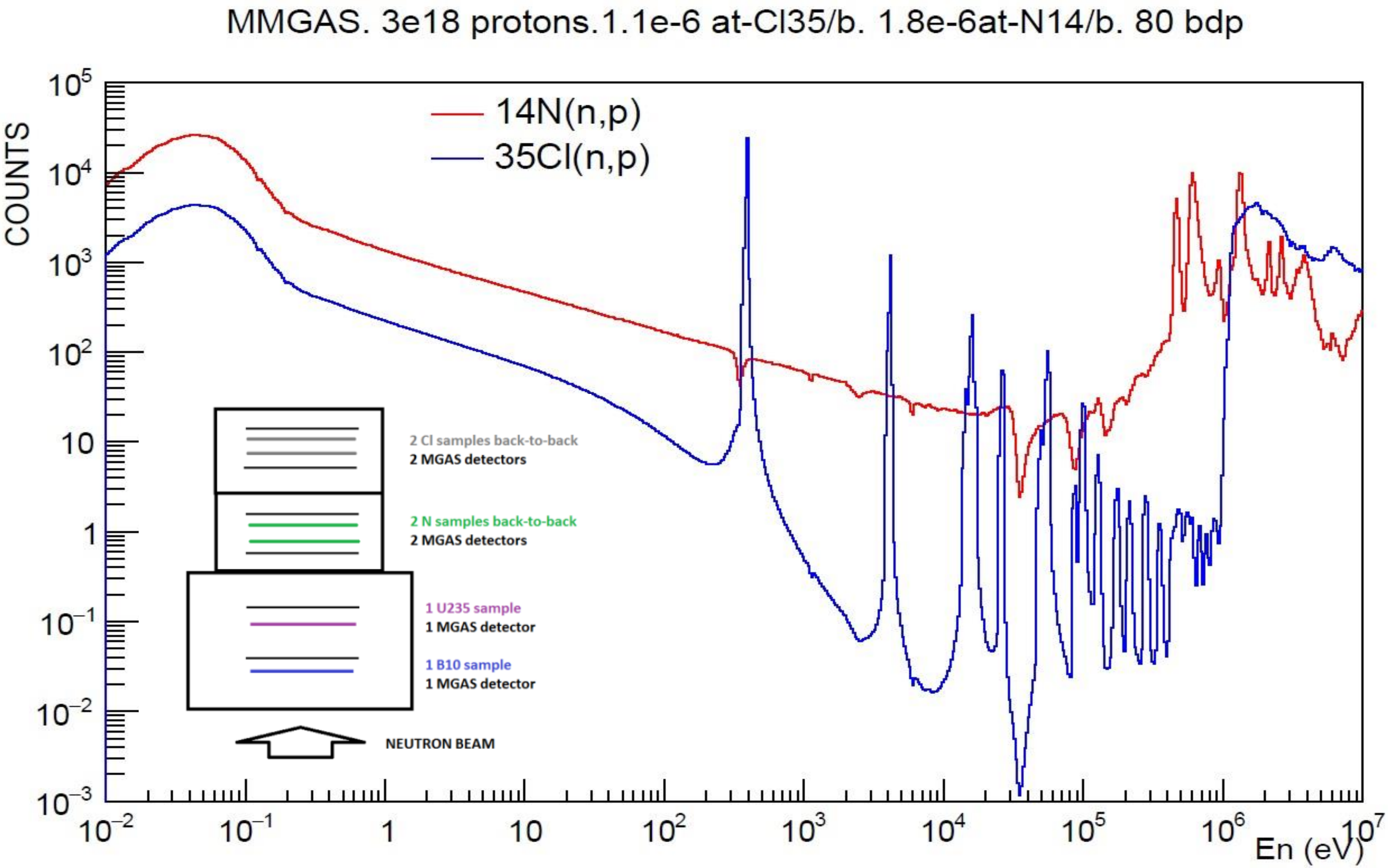
CERN-INTC-2014-007/INTC-I-156: results.



A good reconstruction of the shape of the cross-section as a function of the time-of-flight can be carried out.

In particular, the low energy resonances of the ^{35}Cl(n,p) reaction are clearly resolved.

Beam time and setup: (3+0.5)e18 protons.



SUMMARY.

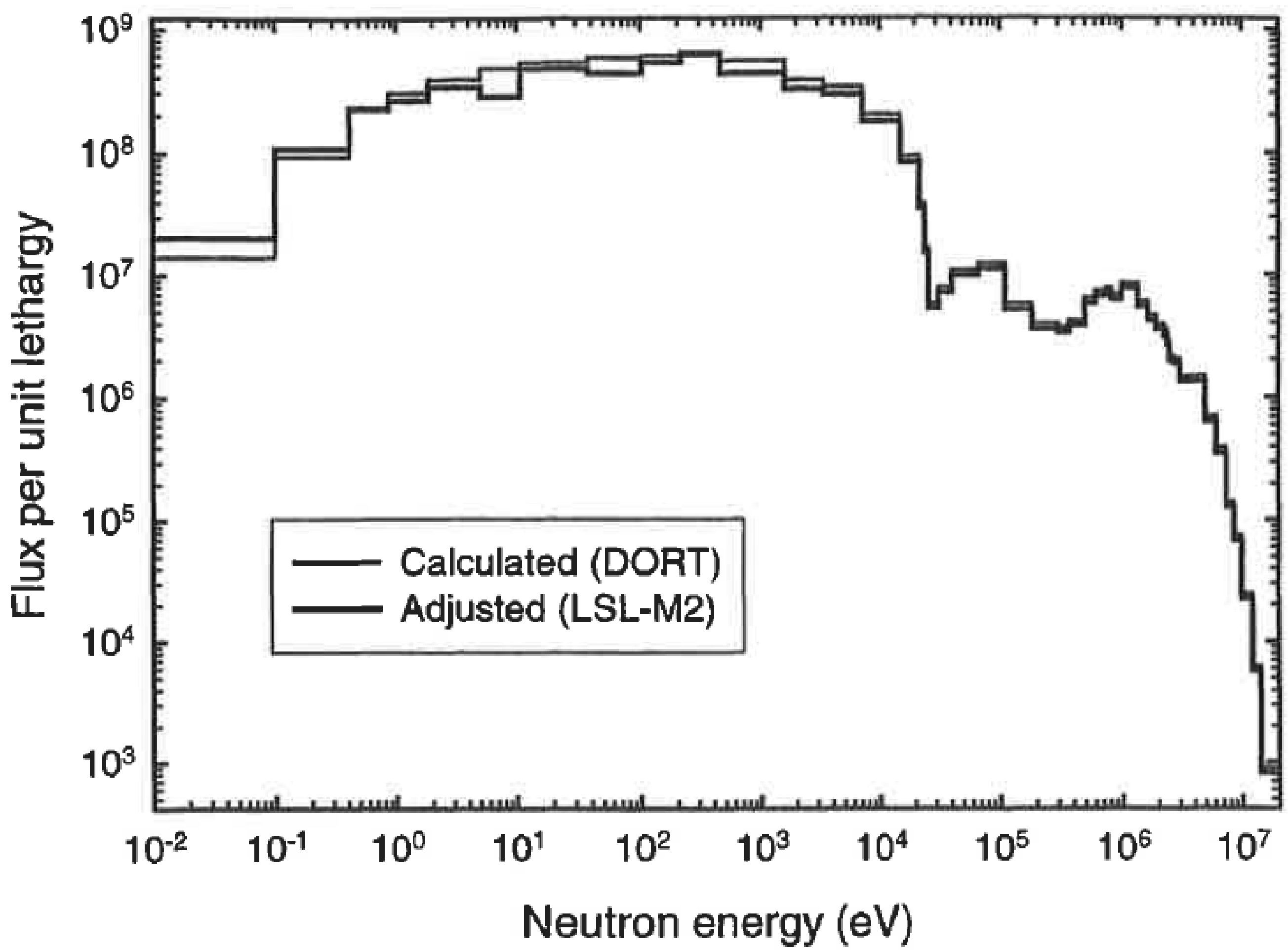
- $^{14}N(n,p)$ and $^{35}Cl(n,p)$ are important to determine the dose in healthy tissue in BNCT which is a limiting factor in a radiotherapy treatment.
- $^{14}N(n,p)$ and $^{35}Cl(n,p)$ are important in astrophysics to study the physical conditions of stars provide by models.
- EAR2 capabilities have been improved for measuring (n,p) for low energy protons following the work of Sabaté-Gilarte PhD thesis at CERN.
- The possibility to measure from thermal to MeV resolving the resonances in the keV region will provide more reliable and accurate data.
- Sample characterization by RBS shows a radial (r^2) dependency of the mass thickness, this has not been studied in previous experiments.
- 3e18 protons running in parallel are enough for achieving the goals in both experiments with MMGAS detectors.

Thank you

3e18 protons for the ^{14}N and ^{35}Cl running in parallel

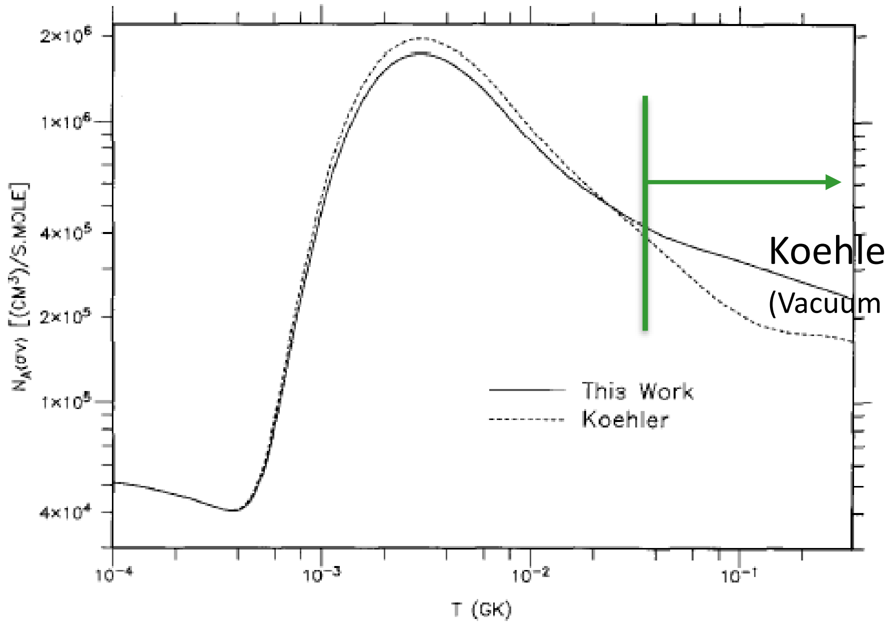
0.5e18 protons for detector calibration and setup.

Neutron beam at FiR reactor



After a few TPs, at the quenching of a thermal instability the H shell is inactive and the convective envelope penetrates in the upper region of the He intershell, bringing to the surface newly synthesized 12C and s-processed elements. This recurrent phenomenon is known as *third dredge up* (TDU)

^{35}Cl(n,p)^{35}S reaction: astrophysics. ^{36}S origin.



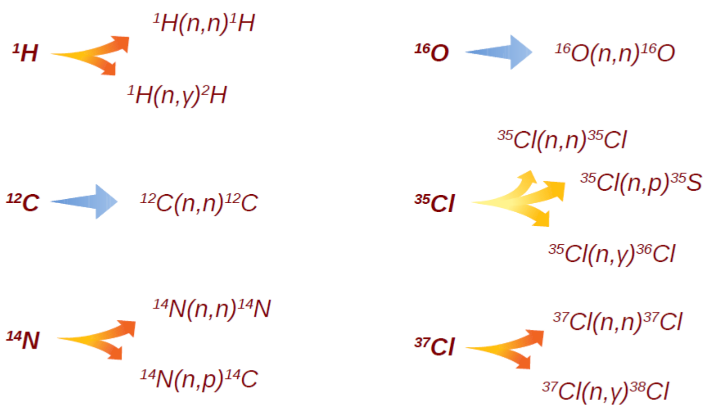
Koehler sample: Adenine ($C_5H_5N_5$), 165 μg

(Vacuum evaporation of Adenine onto 8.5- μm-thick Al b

Fig. 7. Comparison of the $N_A\langle\sigma v\rangle$ curves calculated from Koehler's renormalized data [12] and the present work, as a function of the stellar temperature.

in Fig. 7. There is agreement within the error bars below 4×10^{-2} GK, the deviation starts in the region of s-process temperatures. Surprisingly, there is also agreement between the $N_A\langle\sigma v\rangle$-values calculated by us using Koehler's (renormalized) cross-section data with the corresponding values given by Koehler [12], which are expected to be 11% higher due to this renormalization. This is probably due to the different formulas applied to calculate $N_A\langle\sigma v\rangle$. In this work it was calculated in the straightforward way mentioned above, whereas Koehler calculated this quantity by means of a theoretical fit based on the resonance parameters.

Dosimetry for neutrons (no γ) in healthy (brain) tissue.



H	C	N	O	Na	P	S	Cl	K
10.7	14.5	2.2	71.2	0.2	0.4	0.2	0.3	0.3

medium. According to Ref. [106], the dose (D) is the energy transferred to matter per unit mass by ionizing radiation; by definition, it is the energy invested in modifying the rest energy of the atoms while the emitted radiation is excluded.

1 cGy). The term *dose* can be used for any kind of particle, energy and material; nevertheless, in case of non-charged particles it is more appropriate to use the concept of *kerma*. Kerma is the *kinetic energy released per unit mass* [106] to the secondary charged particles generated as a results of the interaction of indirect ionizing particles, photons or neutrons, with the medium, regardless of where or how they release that energy:

$$K = \frac{d\overline{E}}{dm} \tag{A.2}$$

Kerma and dose are the same only when there is equilibrium between charged particles, i.e. when the net balance of particles that enter and exit any volume under consideration in the medium is zero the equilibrium condition is fulfilled.

Druyts CH_4 at 1 bar!!! <<<Signal-to-background

Ar40(n,a) 98% Th=2560 keV (n,p) Th= 6 MeV
Ar36(n,p) , 0.3365%, 72.8242 keV
Ar36(n,a), 0.3365%, 2000.72 keV , not seen in the S33(n,a)
Ar38(n,p), 0.0632%, Th=4244.2 keV
Ar38(n,a), 0.0632%, Th=228.105 keV

F19 th>1.5 MeV

88%Ar + 10%CF_4 + 2%iC_4H_{10} at 1bar

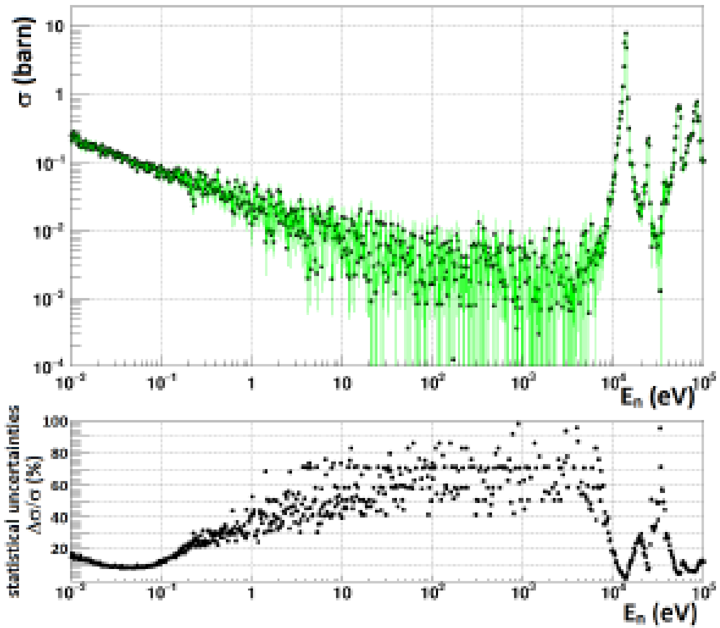


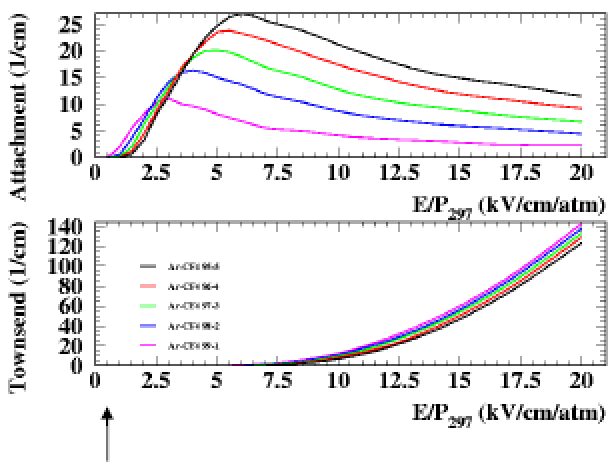
Figure 5.8: $^{33}S(n,\alpha)^{30}Si$ cross section as a function of the neutron energy with 100 bins per decade. On the bottom the statistical uncertainty associated with each bin.

- Gas gain

We obtained high gains (~5000) at modest mesh voltages, 300-350 V, except for ArCH4:10% (P10).

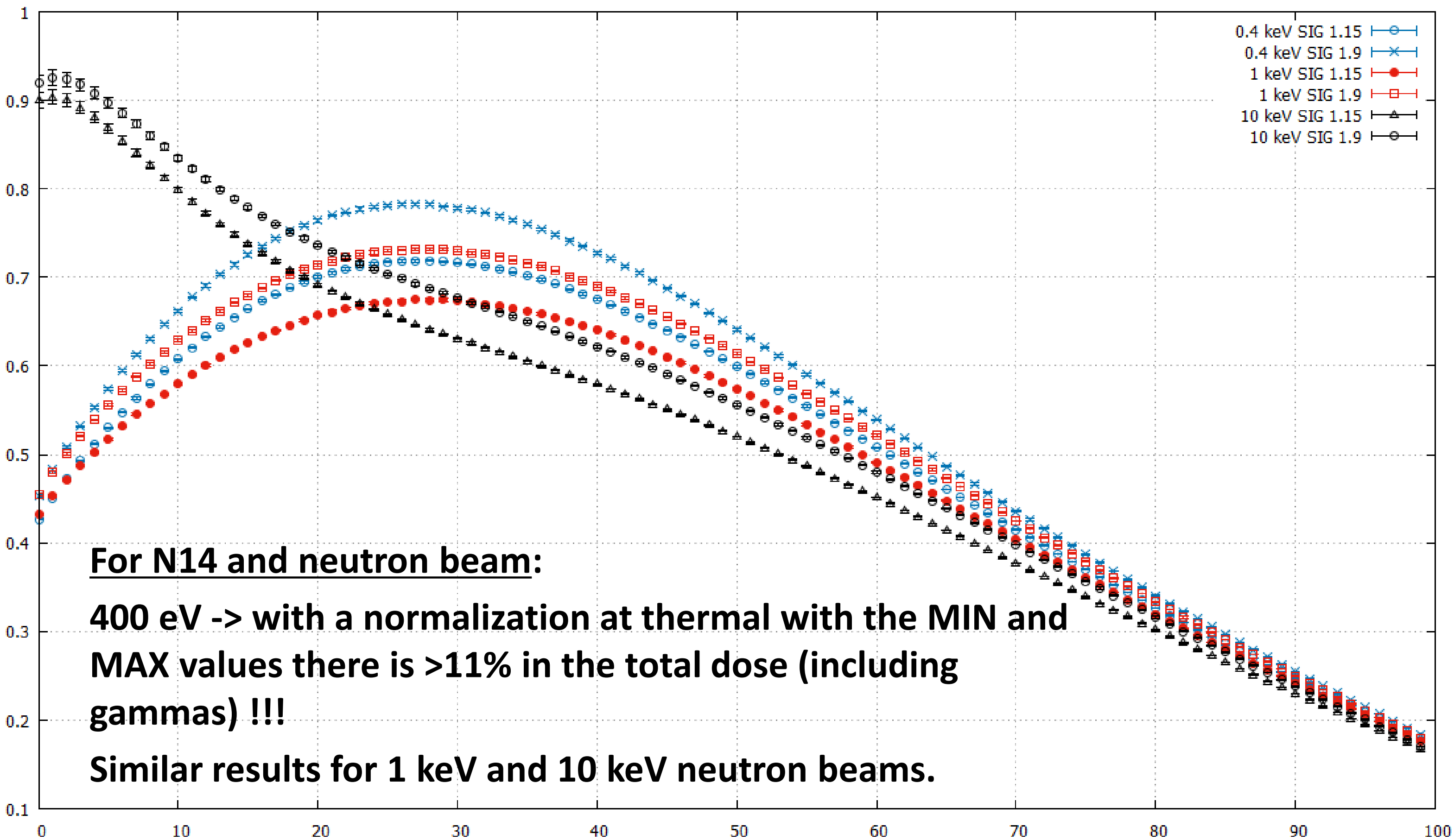
CF4 improves time resolution and total deposited energy

Ar-CF4 Attachment / Amplification



E_D~0.2V/cm

Physical dose. More important for biological dose!!!



<u>For N14 and neutron beam</u>:

400 eV -> with a normalization at thermal with the MIN and MAX values there is >11% in the total dose (including gammas) !!!

Similar results for 1 keV and 10 keV neutron beams.

<u>For Cl35 and neutron beam (400 eV):</u>

With a normalization at thermal with the MIN and MAX values there is >6% in the total dose (including gammas) !!!

The differences would be higher in case of different resonance parameters

Similar results for 1 keV and 10 keV neutron beams.

$^{35}Cl(n,p)^{35}S$ in astrophysics: its origin and isotopic ratio.

Circumstellar envelope of carbon-rich AGB.

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First observational evidence of significant deviation from solar isotopic ratio of elements heavier than the CNO.

Independent of the details of the specfic stellar code adopted (TDU, third dredge up)

All sources of uncertainty must be studied and removed to check the s-process calculations.

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ASTRONOMY AND ASTROPHYSICS

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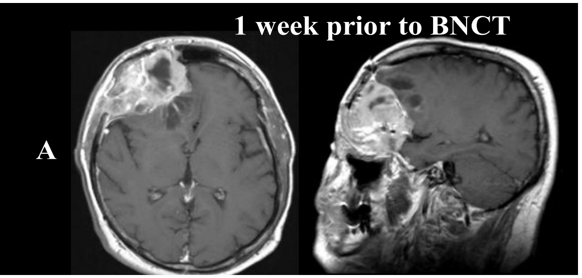
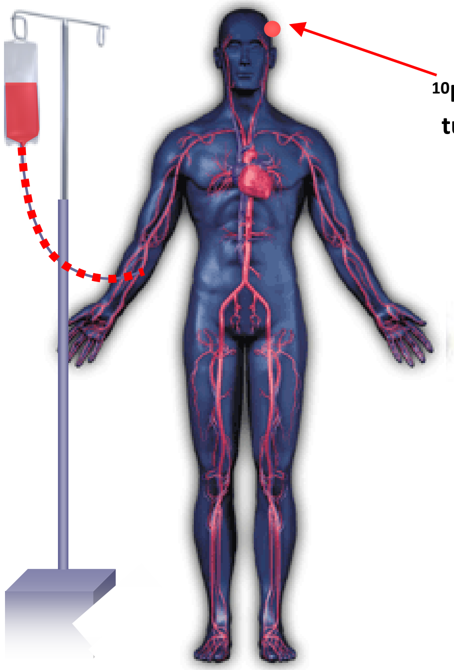
C. Kahane[1], E. Dufour[1], M. Busso[2], R. Gallino[3], M. Lugaro[4], M. Forestini[1], and O. Straniero[5]

BNCT: from reactors to accelerators.

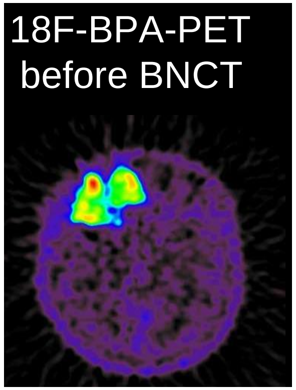


^{10}BPA: 400 mg/kg
2h

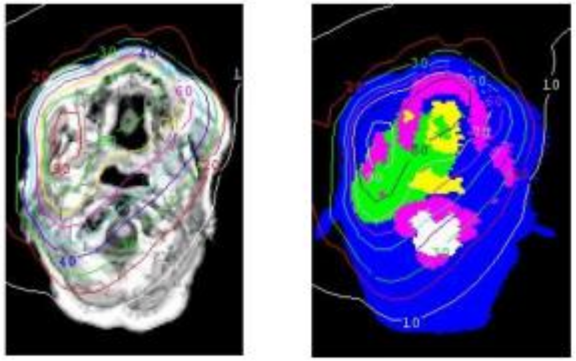


^{10}BPA absorption in tumour cell (>3.5)

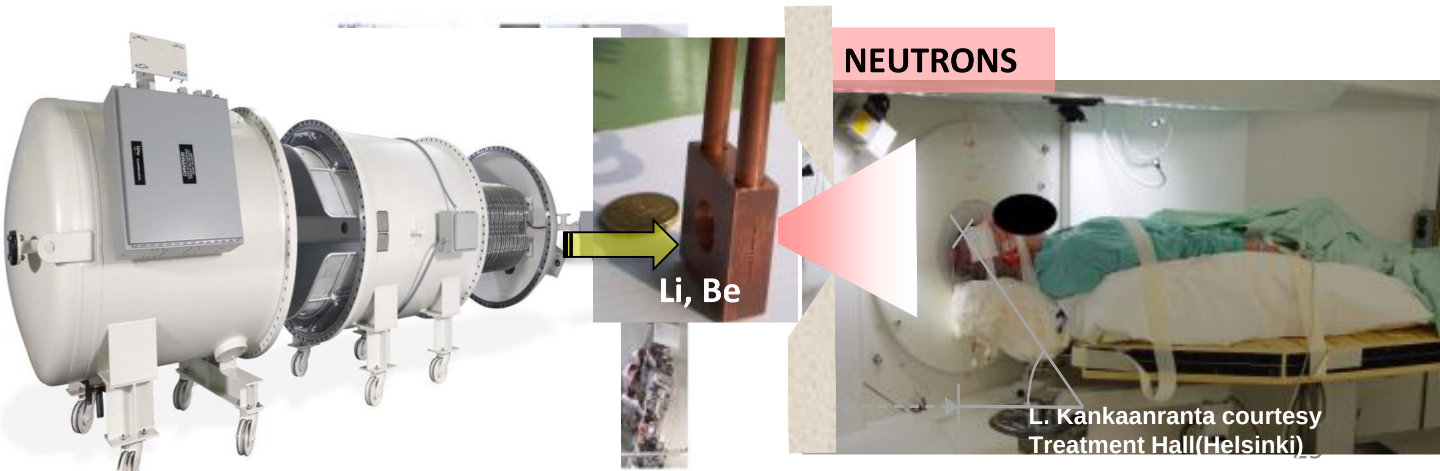
^{18}F-BPA: tumour absorption (>3.5)



Treatment planning

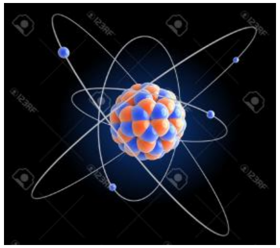


Neutron irradiation

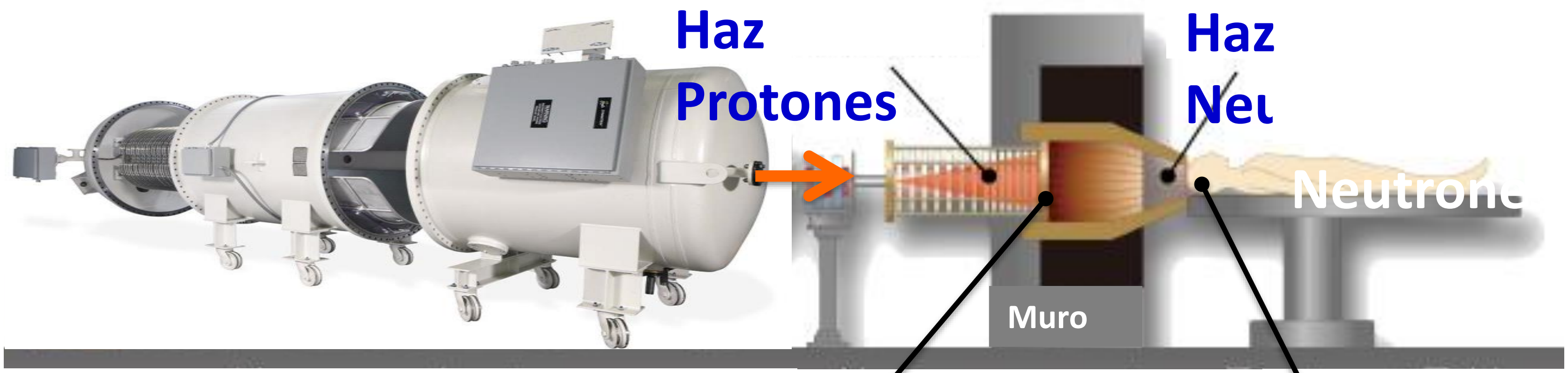


High power accelerator

Sobre los neutrones: necesidad de alta corriente.



- Componentes fundamentales de los núcleos.
- Son neutros, interaccionan menos, por ej. con los tejidos.
- Necesario una reacción nuclear que los extraiga de los núcleos.
- ¿Qué cantidad se necesita en el tumor para BNCT? 10^{10} n/s de baja energía.

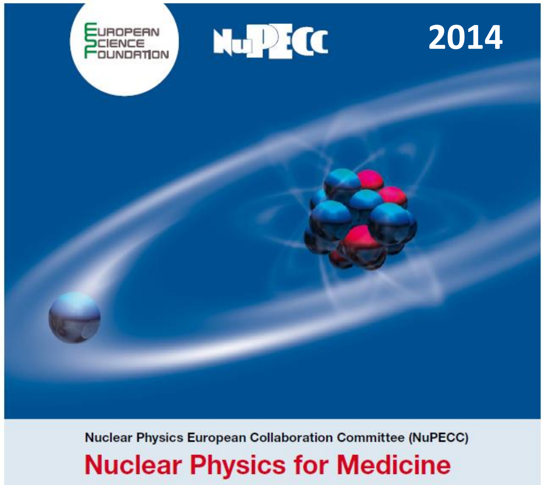


Acelerador Electroestático Lineal
15 mA de protones ≈10^{15} protones/s.

Blanco y Moderador
10^{12} neutrones/s
Media energía

Llegan al tumor
10^{11} neutrones/s
de baja

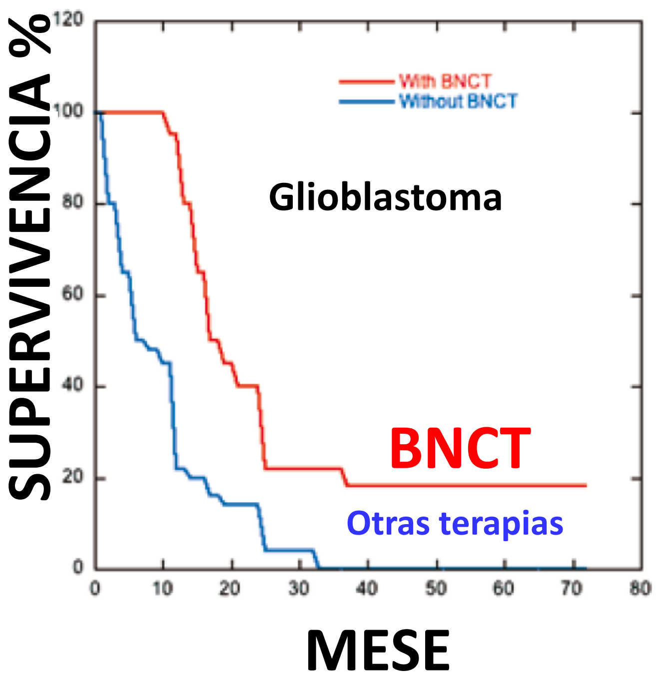
Resultados de la BNCT (www.nupecc.org)

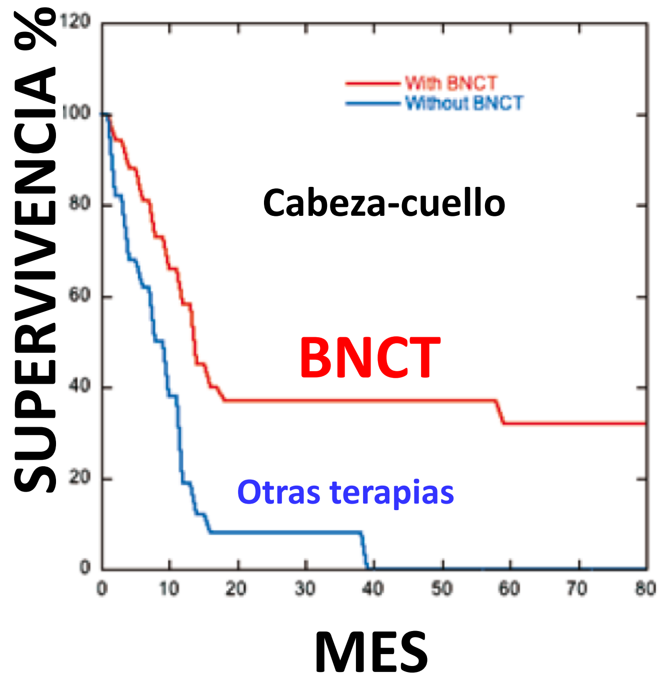


Fase II

Centre	States	Neutron source	Neoplasm	N° of treated patients*
Helsinki University Central Hospital, Helsinki, Finland	Europe	FIR-1, VTT Technical Research Centre, Espoo	GB and HN	50 GM 2 AA 31 HN
University of Tsukuba, Tsukuba City, Ibaraki	Japan	JRR-4, Japan Atomic Energy Agency, Tokai, Ibaraki	GB	20 GM 4 AA
University of Tokushima, Tokushima	Japan	JRR-4 (Kyoto University Research Reactor, Osaka)	GB	23
Osaka Medical College and Kyoto University Research Reactor, Kyoto University, Osaka and Kawasaki Medical School, Kurashiki	Japan	KURR	GB, HN, CM	30 GBM 3 AA 7 Men 124 HN
Taipei Veterans General Hospital, Taipei, Taiwan	Republic of China	THOR, National Tsing Hua University, Hsinchu, Taiwan	HN	10
Instituto de Oncología Angel H, Buenos Aires	Argentina	Bariloche Atomic Center	CM and AT	7CM 3 AT

* GM: glioblastoma multiforme; CM: cutaneous melanoma; AA: anaplastic astrocytoma; HN: head and neck cancer; Men: meningioma; AT: anaplastic thyroid cancer





UNA SOLA IRRADIACIÓN

MAYOR SUPERVIVENCIA

MEJOR CALIDAD DE VIDA