Search for Neutron Flux Generation in a Plasma Discharge Electrolytic Cell

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Experimental background

Claims: in a plasma ignited by high voltage in an electrolytic cell with appropriate cathode and solution

1. Thermal power generated exceeds input electric power
2. Transmutation of elements in cathode occurs
3. Neutrons are produced

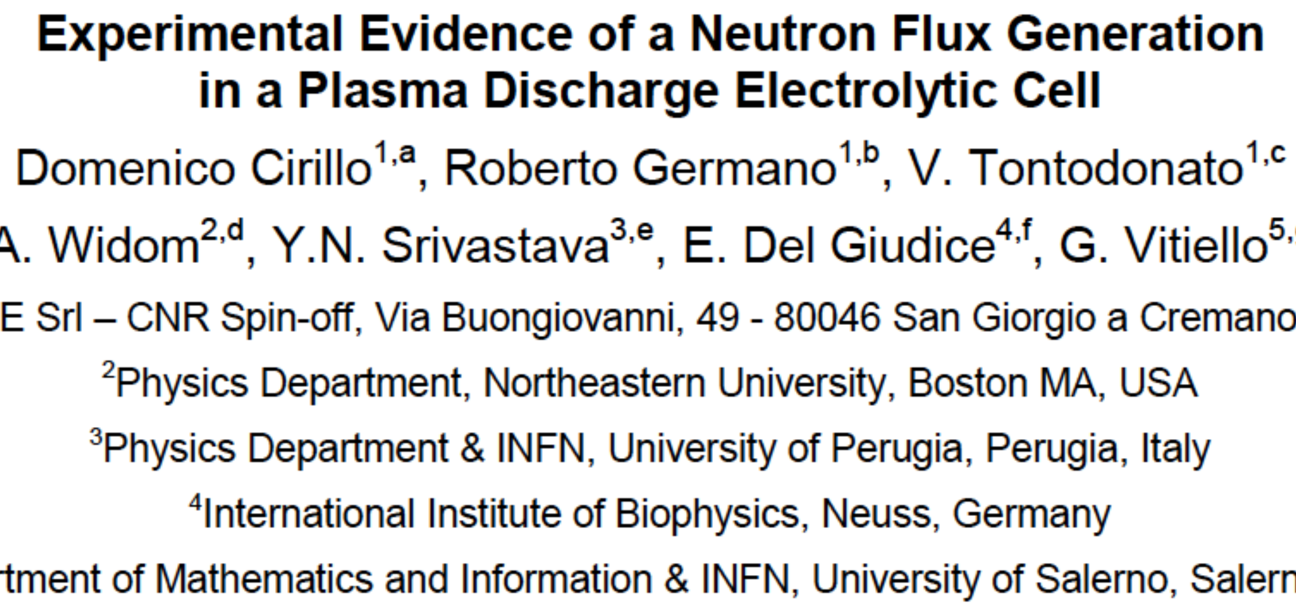
Sample References:

1. T. Mizuno et al, "Production of Heat during Plasma Electrolysis in Liquid", Jpn J. Appl. Phys., Vol.**3** (2000) 6055
2. T. Mizuno, et al," Isotopic changes of the reaction products induced by cathodic electrolysis in Pd", J. New Energy 1 (1996) 31
3. D. Cirillo et al, "Experimental evidence of a neutron flux generation in a plasma discharge electrolytic cell", Key Engineering Materials 495 (2012) 104

The "Cirillo et al experiment"

Patent US 8419919 B1, "System and methods for generating particles"
Patent WO 1999049471 A1, "Reactor for producing energy and neutrons by electrolytic reaction in light- or heavy- water solution".

Experimental Background: The "Cirillo et al" experiment



Experimental Evidence of a Neutron Flux Generation in a Plasma Discharge Electrolytic Cell

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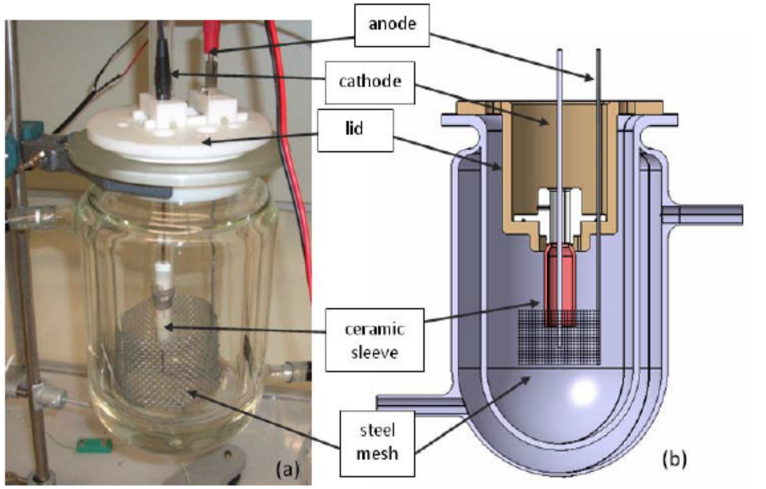
Key Engineering Materials Vol. 495 (2012) pp 104-107

The “Cirillo et al” experiment



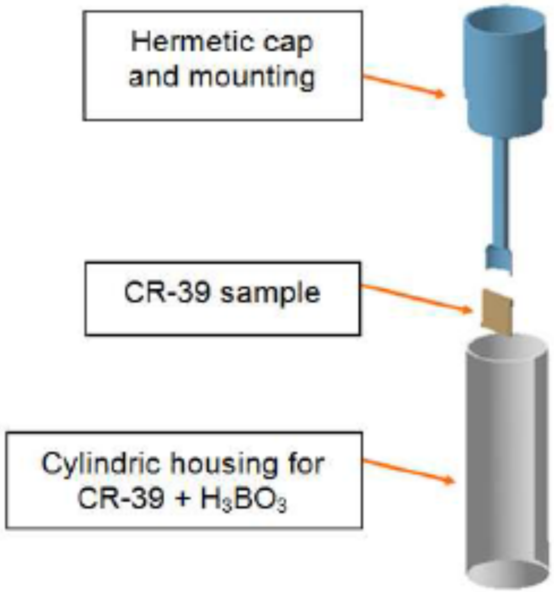
The electrolytic cell:

- Cathode: tungsten rod
- Solution: K_2CO_3
- V=290V, I=2.5A , t=500s

The “Cirillo et al” experiment

Detectors:

- Neutrons measured with CR-39 detectors in H_2BO_3.
- Observed flux $\sim 720000 n/s/cm^2$ “in proximity of the plasma”
- No discussion about the neutron spectrum: thermal? Fast?



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Our team

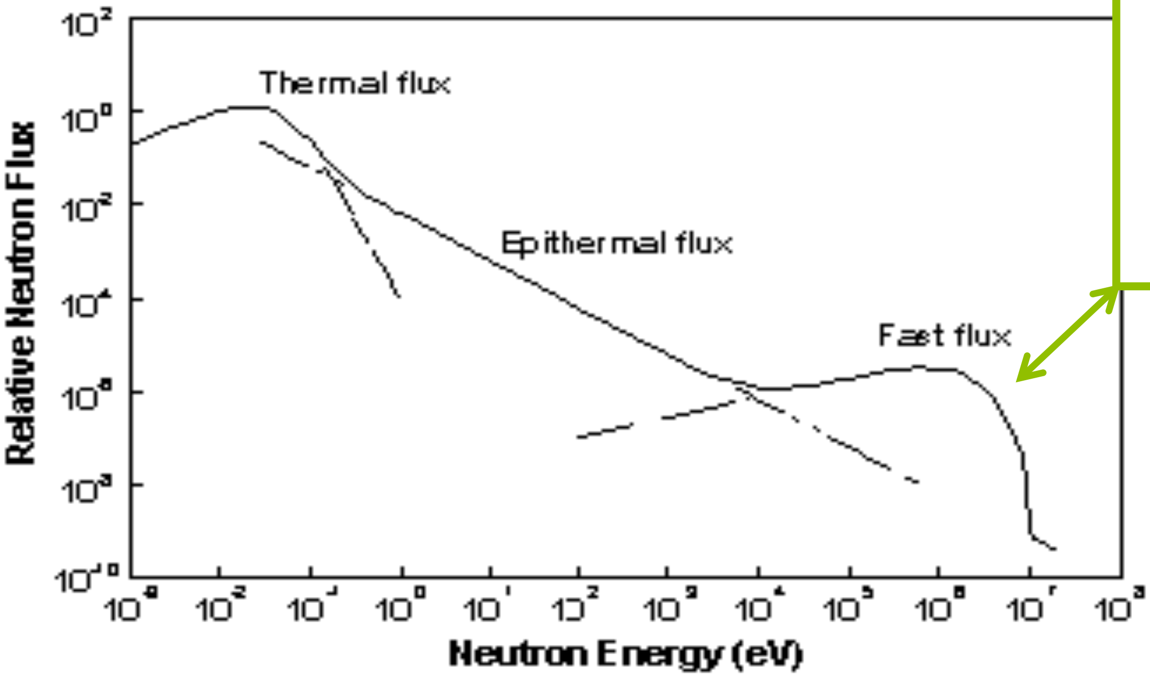
Very little scientific literature on the topic (mostly proceedings) ➔
Try and reproduce experiment [in record time…]

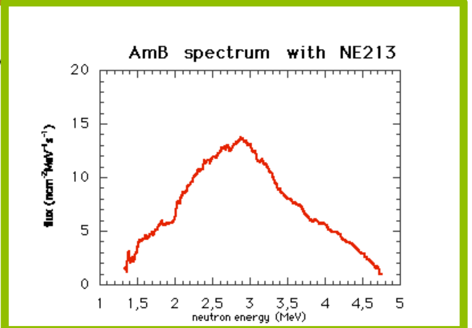
- Electrochemical experts from Enea (Violante, Castagna, Lecci, Sansovini, Sarto)
- CR-39 and neutron experts from INFN (Bedogni, Esposito)
- Fusion neutronics experts from ENEA (Pillon, Angelone, Pietropaolo)
- Data analysis and theory experts (Faccini, Polosa, Pilloni)

Neutron Detection

Neutron Spectrum

Neutron detection depends on the energy spectrum





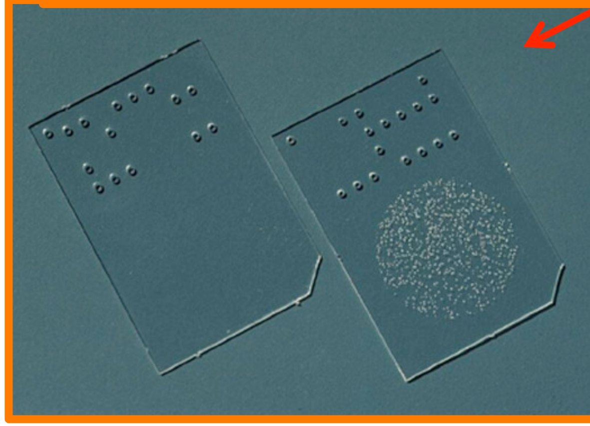
The **AmB** spectrum after traversing water gets moderated to epithermal+ thermal
➔ Partially moderated (PM) spectrum

CR-39 detectors

R. Bedogni, et al, Radiat. Meas. **43**, S491 (2008)

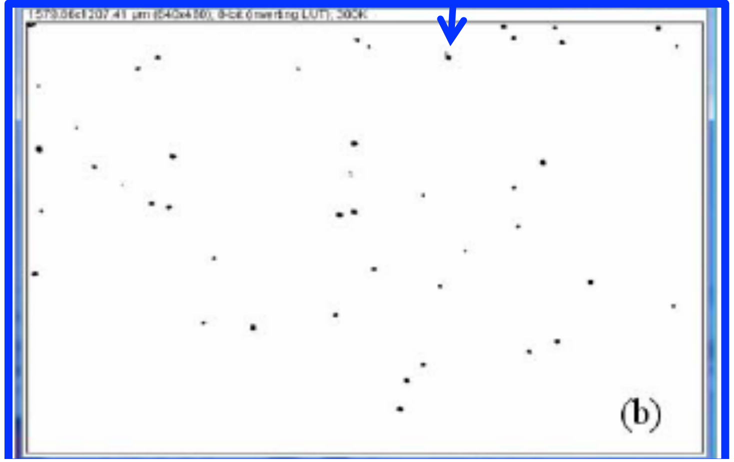
Slabs of poly-allyl-diglicol-carbonate (PADC) produced by intercast

Cut in pieces (2.224cm^2)



Sensitive to **fast neutrons**:

- After n,p scattering protons leave a track
- Etching with potassium hydroxide at 70^{o}C
- Read with a custom reader



Number of tracks proportional to integrated impinging neutrons

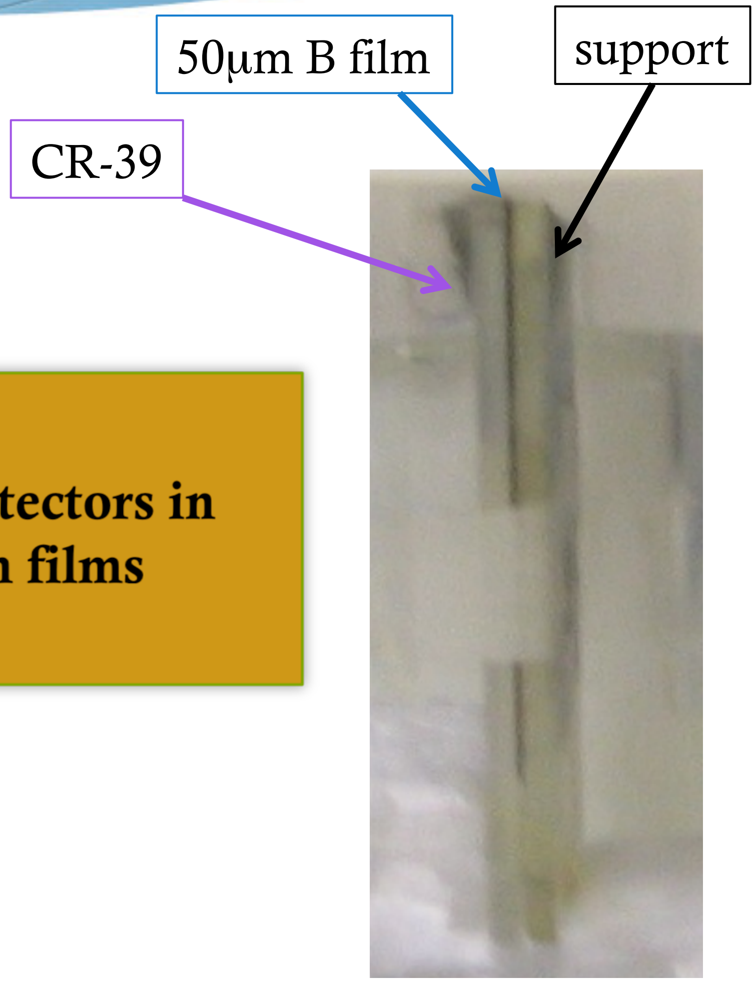
CR-39 detectors for thermal neutrons

<1.8 MeV α particles produced via

B(n,α)Li

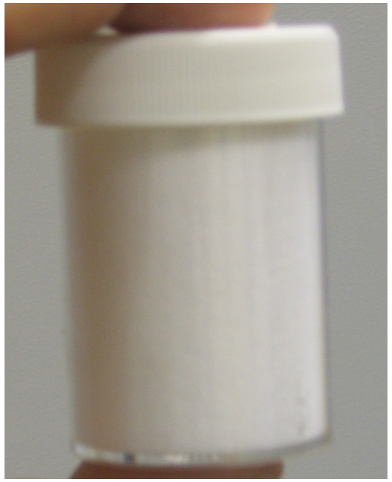
The α release tracks in CR-39 detectors

Wrap detectors in Boron films



CR-39 with copious Boron

Boron stops thermal neutrons, but also the emitted a particles → large amounts of Boron make the detectors insensitive to thermal neutrons



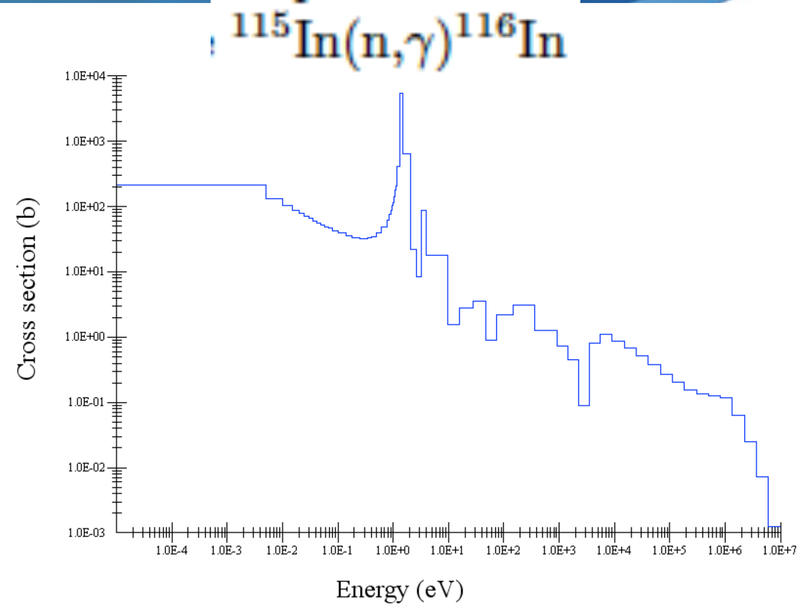
TEST: no signal observed at ENEA-IMRI thermal neutron source → **CONCLUSION**: The detector used by Cirillo et al are sensitive to fast neutrons

Activation detectors: Indium disks

5cm Indium disks (30g total)

Two steps:

- Neutrons activate Indium
  - More sensitive to thermal neutrons
- Gamma lines @1293, 1047 and 416 keV with $T_{1/2}=\log(2)/\lambda=54$ min
- The activity (α) from an irradiation lasted t_r is measured with a HPGe detector (after a time t_a)



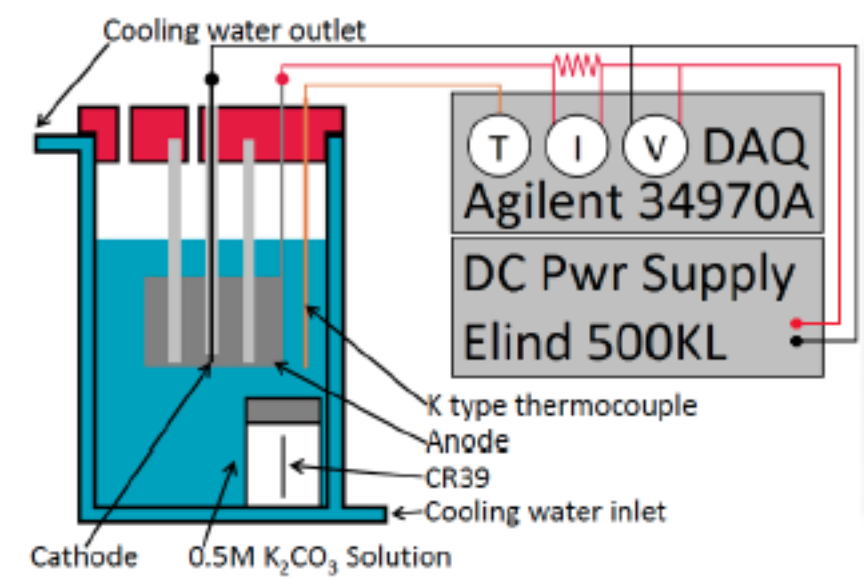
From activity to flux (ϕ)

$$\phi = \frac{\alpha}{N\sigma\left(1-\exp\left(-\lambda t_r\right)\right)\exp\left(-\lambda t_a\right)}$$

Experimental setup

The electrolytic cell

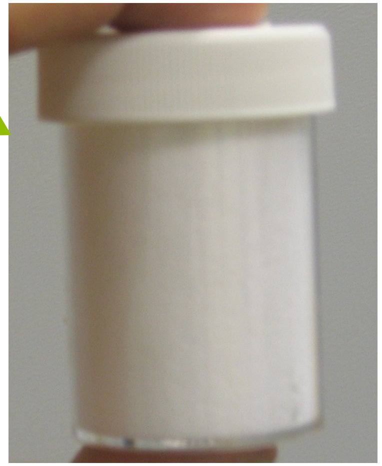
- Tungsten cathode
- Quartz sheath around it, with only tip out (increases impedance and strengthens plasma)
- Anode: grid of titanium plated with platinum
- V=150-300V, I~1-2A, variable t



CR-39 detectors

CR-39 detectors have been used in **several configurations** for systematic studies:

- Bare
- with a 50μm ^{10}B film
- with a large amount of boric acid grains
- with ^{10}B film but shielded with Cadmium (against thermal neutrons)
- with ^{10}B film but shielded with Aluminum (against electromagnetic noise)



Data Taking Campaigns

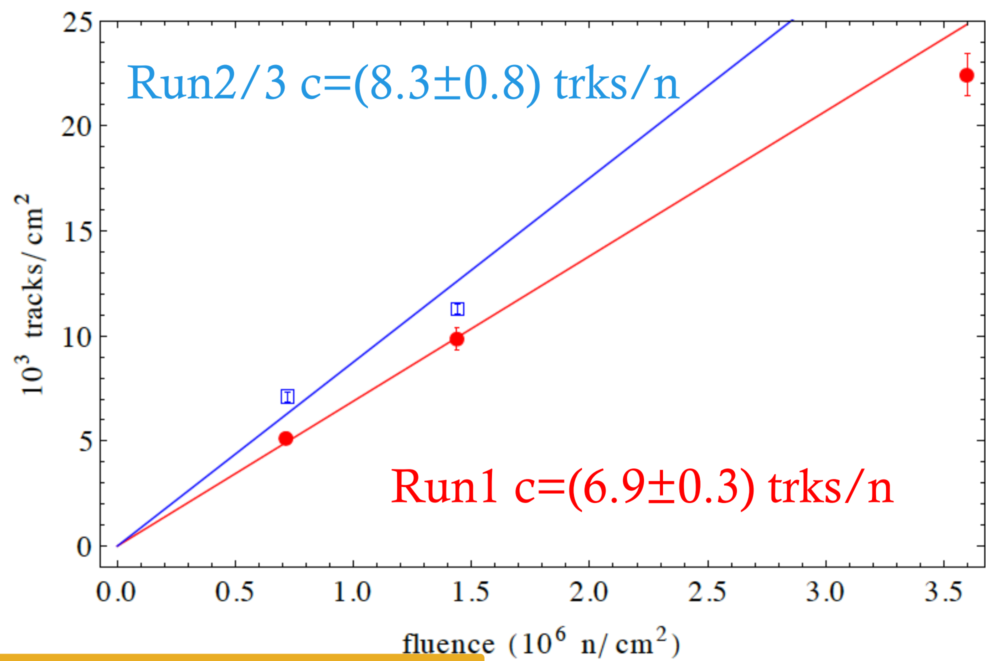
	WHEN	CR-39
Run1	March 2013	Set A
Run2	June 2013	Set B
Run 3	September 2013	Set B

Note CR-39 detectors used for the first campaign come from a different set than those used in the other two.

CR-39:Calibration

Notes:
- D is background subtracted (D_{bkg})
- The two sets of CR-39 need to be calibrated separately

- Conversion between the observed track density (D) to number of neutrons that impacted the detector (integrated)
- Calibrated using the thermal neutron fluence produced at ENEA-IMRI thermal facility ($F = 1.2\ 10^4\ cm^{-2}\ s^{-1}$)

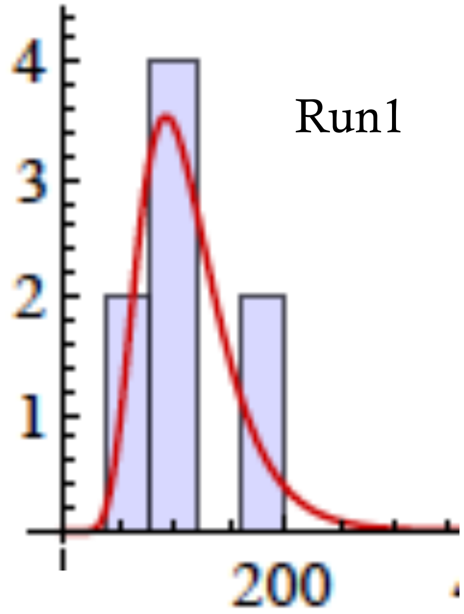


$$F = (D - D_{bkg}) / (cT)$$

Flux estimate, T being time of exposure

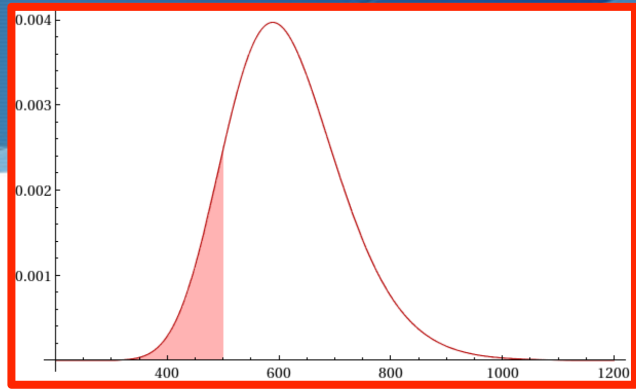
CR-39: Background

- Estimated with non-irradiated detectors
- Fits with a log-normal function (R. Bedogni, et al, Radiat. Meas. 43, 1108 (2008)):



$$L(D \mid \mu, \sigma) = \exp\left[-(\log D - \mu)^2 / (2\sigma^2)\right] / (\sqrt{2\pi} D \sigma)$$

CR-39: analysis



- Estimate Prob(D) Probability of being signal if a track density D is measured
  - Declare a signal significant if Prob(x) >99%

$$\mathcal{P}(D) = \int_0^D L\,(x \,|\, \mu, \sigma)\, dx$$

- If no significant signal, compute upper limit Dmax @95%C.L. with a bayesian approach

$$P(x < Dmax|D) = \frac{\int_0^{Dmax} L(D|\mu+\xi,\sigma)d\xi}{\int_0^{\infty} L(D|\mu+\xi,\sigma)d\xi} = 95\%$$

Cross check with a neutron source

A run was taken putting inside the electrolytic cell (switched off) a **5.5 10^4 n/s AmB neutron source.**

➔ Measurements by the detectors on the exterior of the cell (~5cm from source), **We have a partially moderated (PM spectrum):**

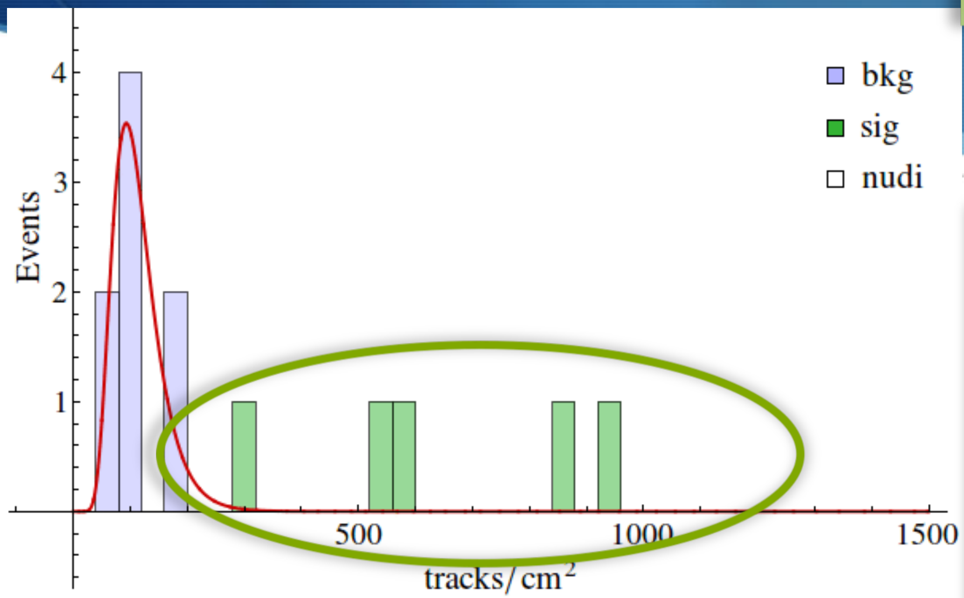
Cr-39 with ^{10}B	85±12 neutrons/cm^2/s
Indium disk	375±125 neutrons/cm^2/s

Conclusion from the test: both detectors are sensitive to a source 1-2 order of magnitudes smaller than Cirillo et al.
Note: Cr-39 calibration constants, obtained with a thermal neutron spectrum, was used, there might be corrections to apply

Results

Run1

CR-39 detectors with thin Boron showed a small but significant excess !!



Run	Duration [']	Voltage Range [V]	n-flux (In) [n cm^{-2} s^{-1}]	n-flux (CR-39) [n cm^{-2} s^{-1}]
1A	8	150-200		280 ± 32 (ins.)
				259 ± 30 (ins.)
1B	5	250	< 1.5	275 ± 35
1C	12	150-200	< 0.7	109 ± 14 (ins.)
1D	4	150-300		166 ± 32 (ins.)

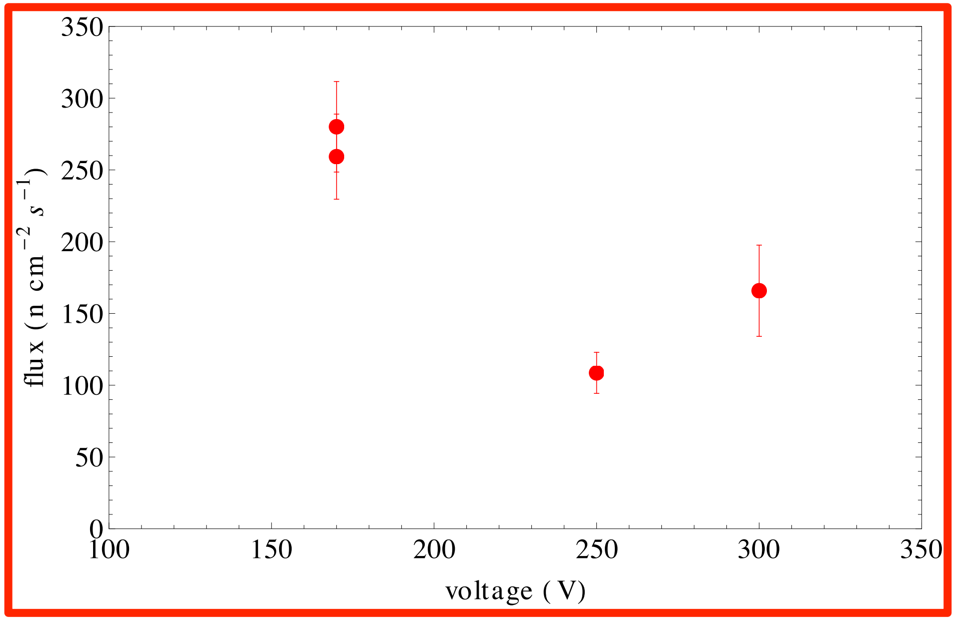
Indium disks and CR-39 with thick boron showed no signal

→ upper limit 10^4 times smaller than Cirillo et al. (72000 n/cm^2/s)

If we believe Cr-39 the could be a neutron flux, but it is ~300 times smaller than Cirillo et al

Characteristics of CR-39 excess

- Does not depend on position (inside/outside cell)
- The only configuration that does not show it is the closest to Cirillo et al.: inside and with thick boron
- Does not depend on Voltage

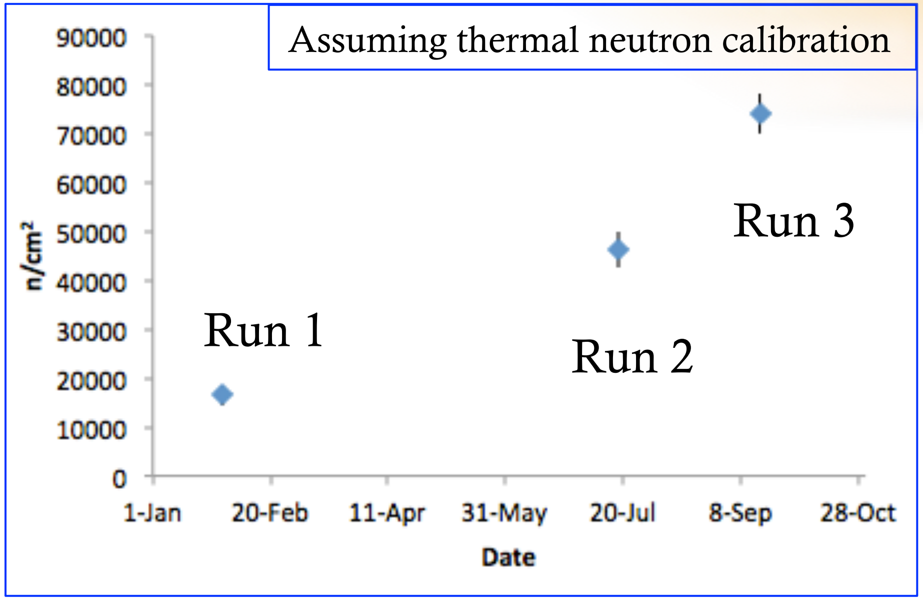


Investigating dicrepancies

NOTE: Run1 and Run2/3 CR-39 came from two different sets, but they were produced on the same day

- We realized background CR-39 had an inconsistent treatment:
  - they were **not wrapped with Boron**
  - They were **analyzed** when calibration took place**: 40 days before** the run with the cell

➔ study time dependence of background

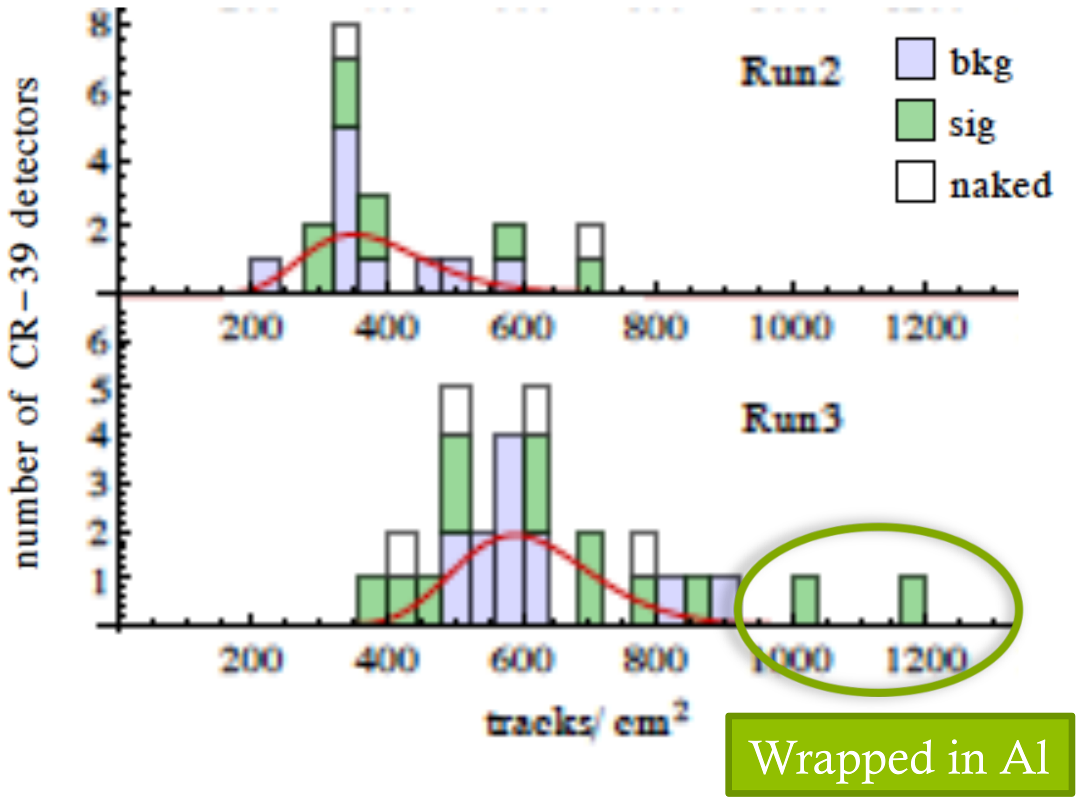


Slope between Run2/3 ≈ 460 n/cm²/day
Naked CR-39 detectors show same behavior

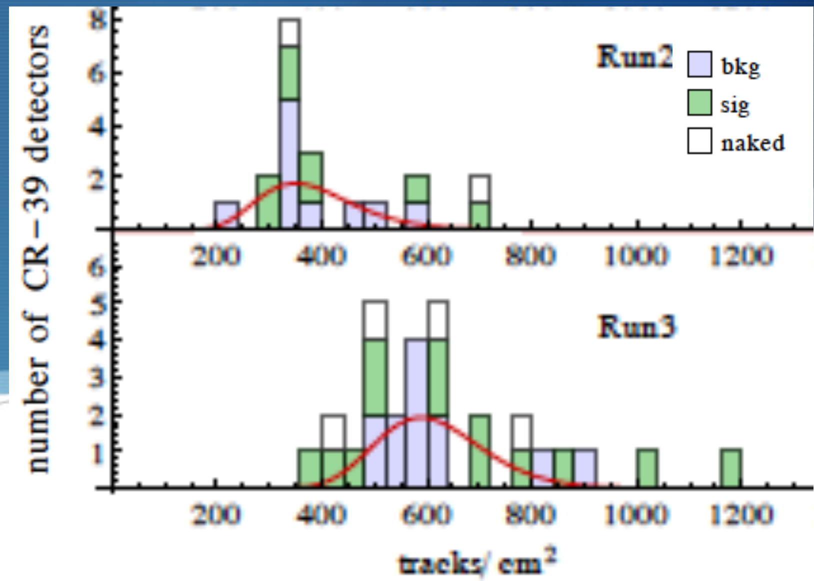
High energy background
Cfr cosmic rays≈ 850 particles/cm²/day

Run2 and Run3: results

- Repeat experiment with uniform background treatment
- All detectors where outside the cell (5cm from cathode)
- Wrap some detectors in Al (screen e.m. background) or Cadmium (screen neutrons)



Run2 and Run3: Comments



- No significant signal observed neither in In disks nor CR-39 detectors
  - Only outliers CR-39 detectors with Al (photo-electrons?!?)
- Limits set at less than 1 (In disks) and tens (CR-39) neutrons/cm^2/s (cfr. 72000 n/cm^2/s from Cirillo et al)

Run	Duration [']	Voltage Range [V]	n-flux (In) [n cm^{-2} s^{-1}]	n-flux (CR-39) [n cm^{-2} s^{-1}]
2	13	220-300	< 0.6	< 49, < 14 < 23, < 18 (Al) < 16, < 21 (Cd)
3A	20	150	< 0.4	< 26, < 27 89 ± 15 (Al) < 19, < 5 (Cd)
3B	21	200-300	< 0.4	< 11, 39 ± 12 < 39, < 8 (Cd) 56 ± 13 (Al)

Comparison with the "Cirillo et al experiment"

- **Differences between the experiments**: neutron detectors and their location.
- The comparison between the experiments depends on the hypothesis on the **production energy spectrum**
  - Since the detectors used in the "Cirillo et al Experiment" (CE) are shielded from thermal neutrons, the spectrum of the allegedly produced neutrons would be faster ➔ will assume AmB
- **Detectors placed inside the cell**: during Run1, with incorrect background suppression, some CR-39 detectors were placed inside the cell
  - Those with thick Boron (same as CE) showed no signal
  - Those with thin Boron showed an excess 100 times smaller than the CE
- **Detectors placed outside the cell:** for an AmB like spectrum we set a limit (with In) $\Phi<64$ $n/cm^2/s$ @95%C.L. @5cm from cathode.
  - Correcting for geometry and spectrum (with MC) we estimate $\Phi<900$ $n/cm^2/s$ @95%C.L @2cm from cathod (cfr 72000 n/cm2/s measured in CE)

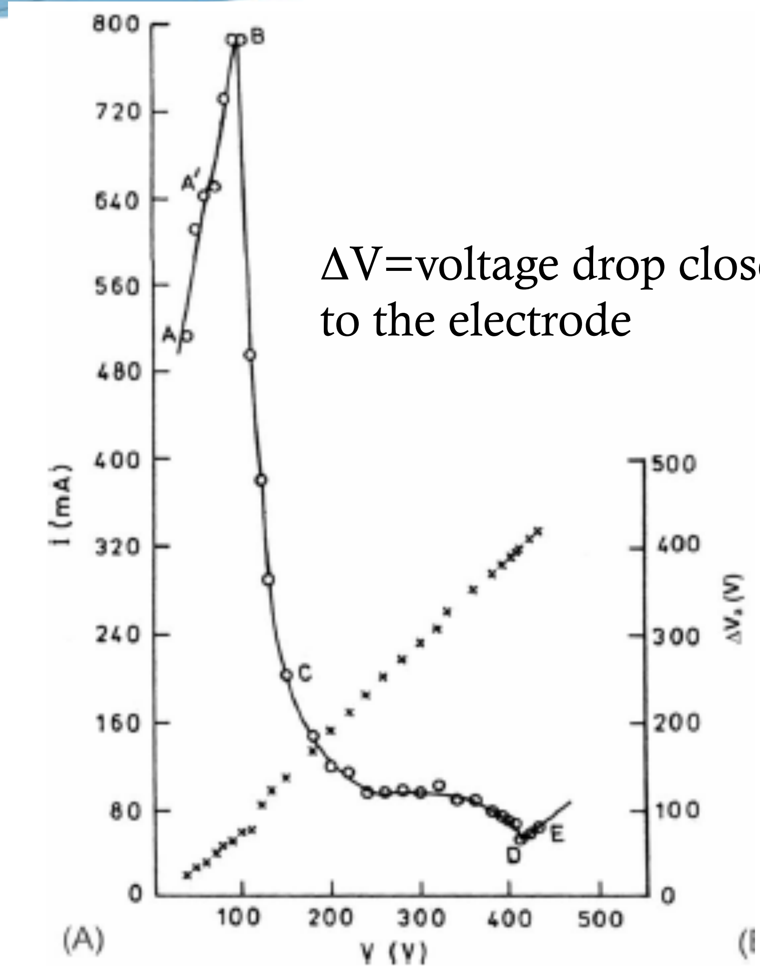
Conclusions

- We built a group of experts from ENEA, INFN, and "Sapienza" to verify the evidence of neutron production in electrolytic cells with Tungsten cathode and K_2CO_3 solution
- We could not reproduce the existing experiment
- We evidenced limits in the use of CR-39 for measurements of neutron fluxes:
  - Background from ambient (cosmic) sources
  - Instrumental effects from wrapping (Boron/Al/…)

BACKUP

From electrolytic to Mizuno cell

- Electrolytic cell powered with Voltage V ➔ Several regions
  - Normal electrolysis A—B
  - Break-down: B
  - Transition: B—C
  - Contact glow discharge (aka plasma) electrolysis: C—D



ΔV=voltage drop close to the electrode

<u>Hypothesis</u>: accelerated H (or D) neuclei have a fusion in the shielding of the cathode metal

Cella elettrolitica

- Elettrodi in soluzione salina
- Catodo (-) ➔ riduzione; anodo (+) ➔ ossidazione
- Leggi Faraday:
  - la quantità di elementi prodotti da un processo di elettrolisi è direttamente proporzionale alla quantità di corrente che ha attraversato la cella elettrolitica;
  - a parità di quantità di corrente, le quantità dei diversi elementi ottenuti è proporzionale ai pesi equivalenti delle specie chimiche.
- Cella Mizuno:
  - catodo di materiale in grado di assorbire H o D
  - Anodo di materiale resistente a corrosione elettrochimica (e.g. platino)