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Title: "Nuclear and Radiochemistry Supporting National Security"

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Nuclear and Radiochemistry Supporting National Security, Carol J. Burns, Nuclear and Radiochemistry Group, Los Alamos National Laboratory, Los Alamos, NM 87545.

One of the most notable early technological ramifications of the discovery of radioactive elements (coupled with the discovery of fission) was the development of military applications. Today, those involved in nuclear and radiochemistry are involved in a wide array of applications, from environmental studies to nuclear medicine. There remains a compelling role for radiochemistry and nuclear science in national security research, however, particularly given today's emphasis on addressing nuclear security. This talk will present an examination of some of these mission areas, as well as an overview of some of the innovative research at Los Alamos National Laboratory that is being conducted in this area.

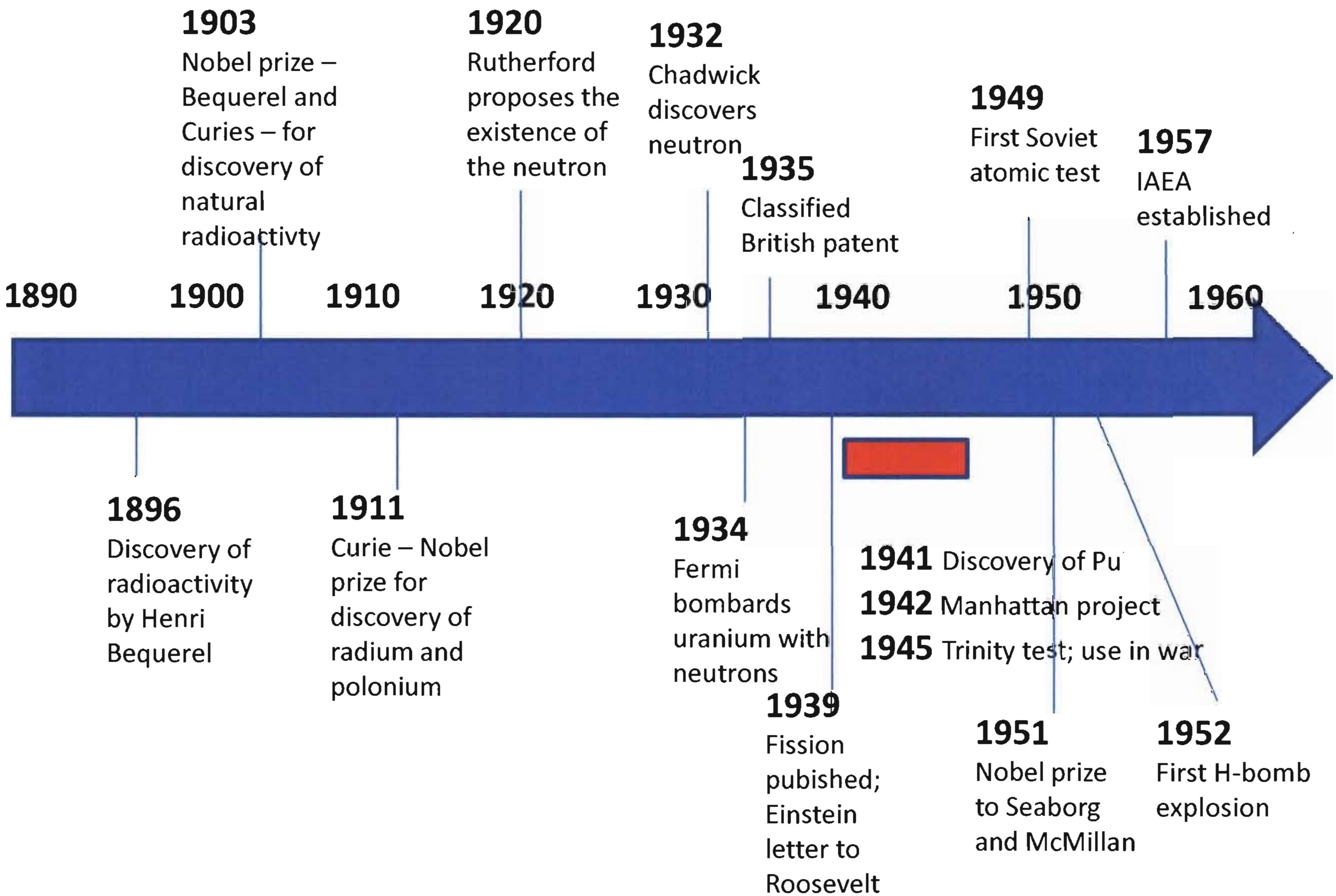
Nuclear and radiochemistry supporting national security

Carol J. Burns

August 29, 2011

LA-UR 11-XXXX

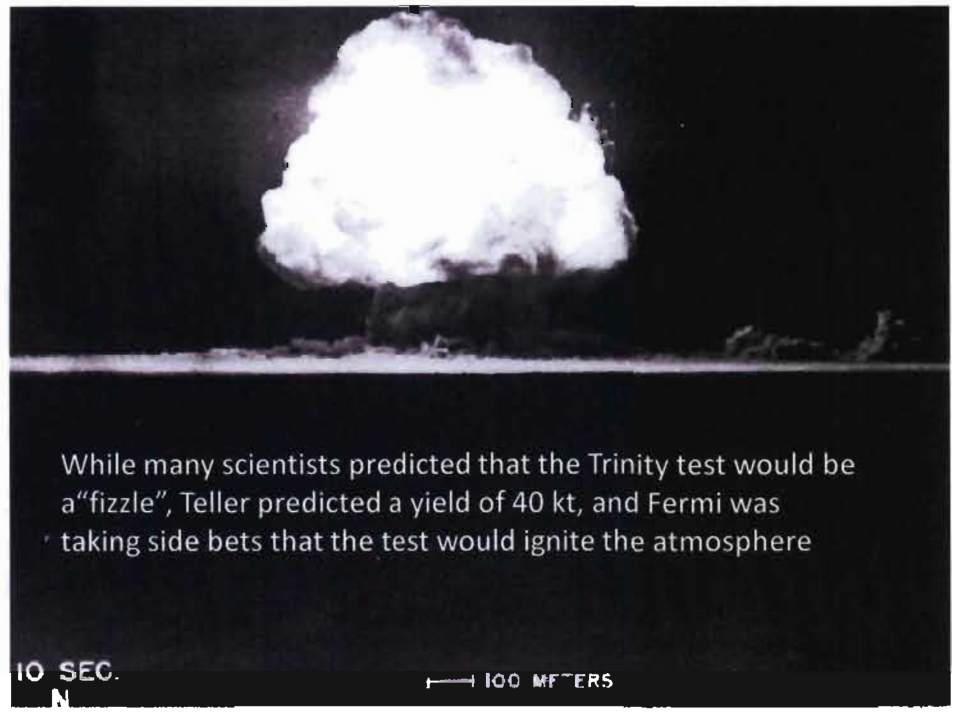
Nuclear chemistry and national security



The genesis of weapons radiochemistry

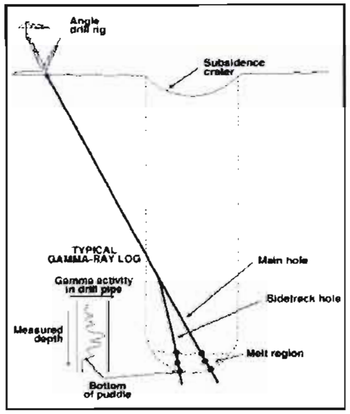
- *Some time before the Trinity event, it was proposed that radiochemistry could determine the yield of a device:*
  - Assume that the yield is solely due to fission in plutonium
  - In a sample of the debris, determine the amount of fission products relative to "unburned" plutonium to determine the efficiency
  - Given a known mass of fuel, the total number of "fissions" can be determined
  - This can be converted into energy release:

  10^{12} calories = 1 kiloton (~ ¼ mole of fissions)



Until the radiochemical data was available, the best estimate of the yield of the device was obtained by Fermi, who dropped small scraps of paper as the blast wave went by

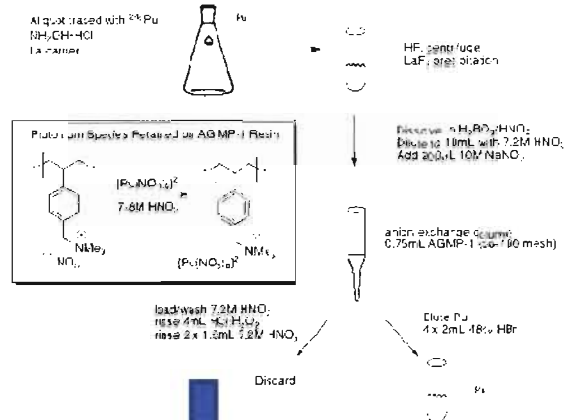
Weapons radiochemistry in the test era

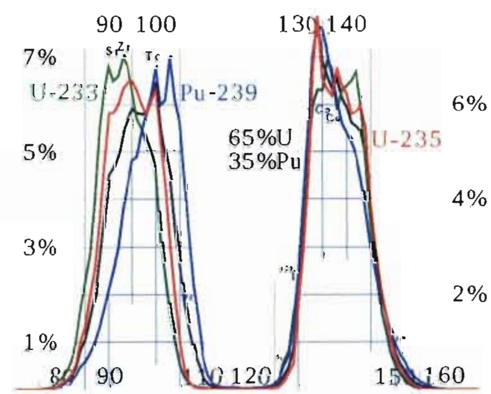


Sample recovery

Collections consist of very complex mixtures:

- **Homogenization**
- **Dissolution**
- **Separation**
- **Assay**





"*Measure fissions*" **in the sample, F**
Measure unburned fuel atoms, p

$$\varepsilon = \frac{\text{fissions}}{\text{Original atoms of fuel}}$$

$$\varepsilon = \frac{F}{F + p}$$

If total fuel known, can calculate total fissions (F_T), or energy release

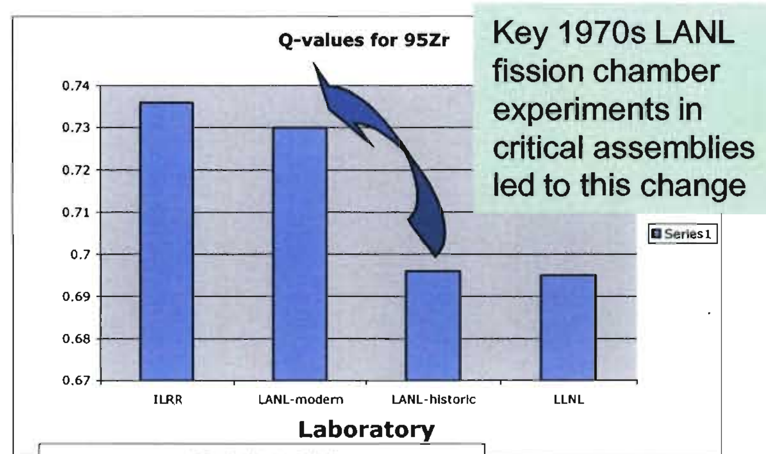
Not really this simple – other corrections apply

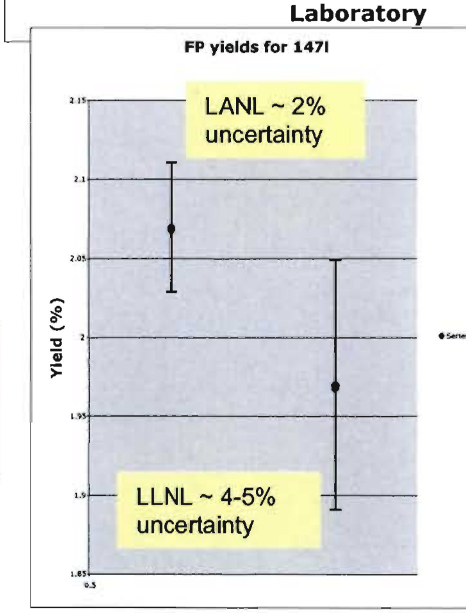
Current Interests in Weapons Radiochemistry: Resolving Interlaboratory Differences

- **Data on fission product production from fast neutrons irradiation of ^{239}Pu persisted between Los Alamos and Livermore for >10 yrs**
- **Improved precision in these values is critical to accurately determine radiochemical nuclear yields from UGTs — essential to reduce QMU uncertainties**
- **Reassessed evidence based on seminal '70s *dual fission-chamber* experiments (NBS multi-Lab consortium and LANL) significantly improves accuracy and reduces uncertainties**
- **Laboratories jointly reevaluated key nuclear data to resolve outstanding differences**
- *Nucl. Data Sheets*, 2010, *111(12)*, 2891, 2923.

Resolution of Radchem differences reduces UGT yield uncertainties and enable significantly improved common baselines for all US nuclear tests

Q-value = FP-yield(^{239}Pu fast) /(^{235}U thermal)





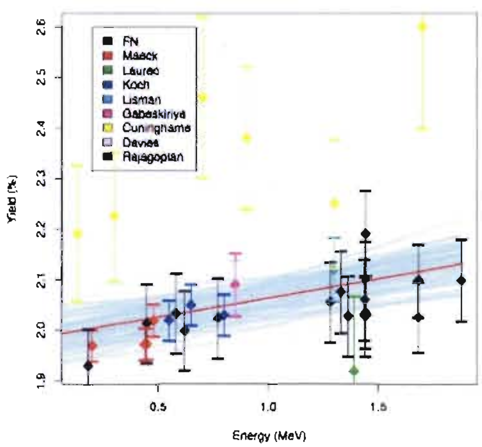
Evaluation of independently measured benchmark-quality data confirm LANL result, provide new insights into fission

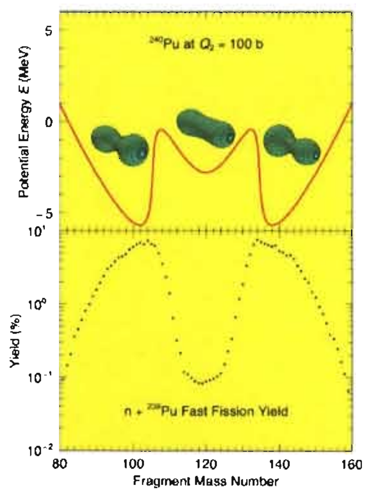
Evaluation of independently-measured benchmark fission product production data confirmed LANL experimental results

Additional efforts provided refinement of uncertainties, and revealed previously unappreciated energy dependence of production of key fission product data

Reinvigorated interest in improving predictive fidelity of fission theory:

^{147}Nd Fission Product Yield, showing energy dependence:





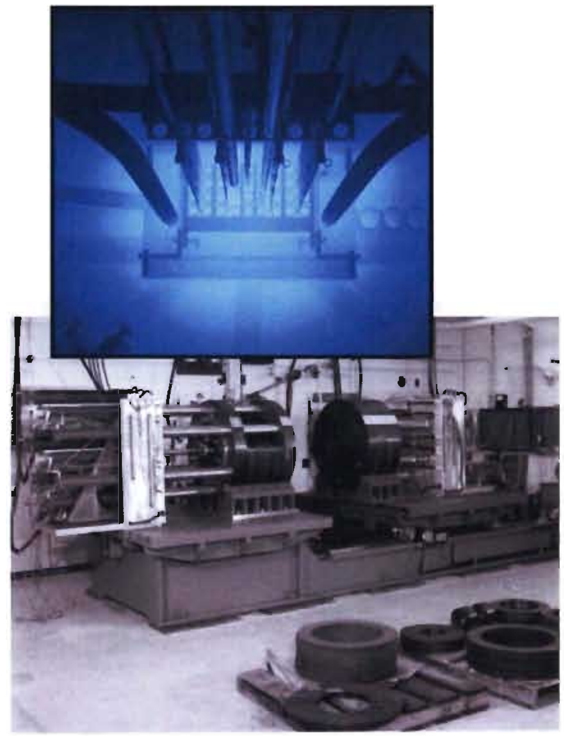
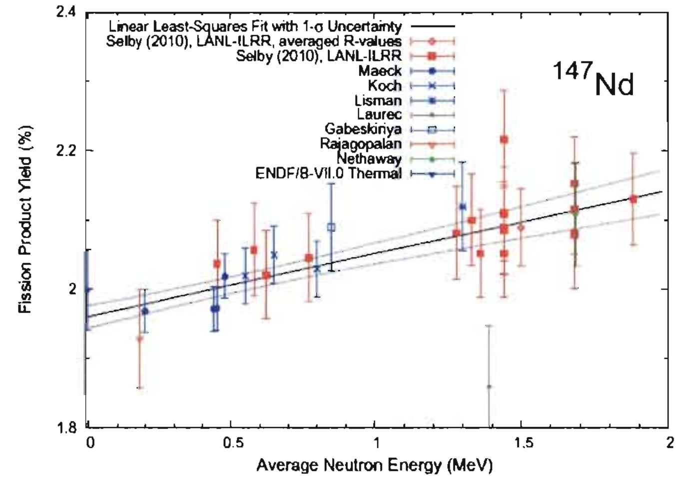


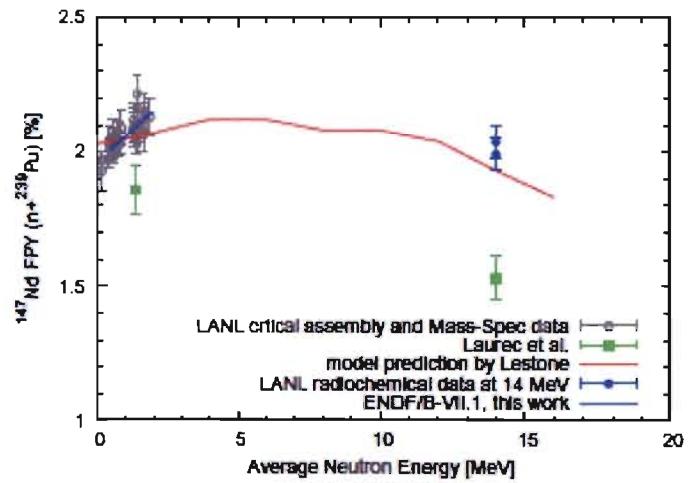
Figure 1. The Big Ten Assembly during Construction (1968).

LANL capabilities in nuclear science, radiochemistry, & nuclear theory are essential for solving such problems

Fission Fragment/Product Yields

- **There is increasing demand for high-precision measurement of, and subsequent theoretical understanding of, the yields**
  - Implications for inference of weapons yields
  - Starting point for new theoretical fission evaluation process
- **Recent work places a 2% uncertainty on Pu239 Nd-147 yield from 0.1 to 2 MeV**
  - Previous global Wahl systematics had wrong slope
  - Predictions by Lestone show 2x difference in slope
- **New experimental data are essential!!**

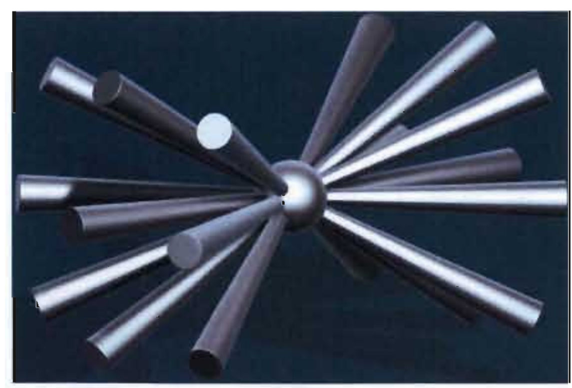




Fission Fragment/Product Measurements

- Irradiation measurements for fission product yields
  - TUNL mono-energetic source and/or NTS CEF critical assembly spectra
  - Gamma spectroscopy and/or radiochemical separation with beta counting
    - Builds on LANL's well-established R-value methodology
  - Preliminary work has begun (tests at TUNL, IER submitted to NCSP)
- SPIDER multi-arm 2E,2V measurements at LUJAN/WNR
  - Absolute measurements of fission fragment yields
    - Additional fragment state measurements of high-value to theory
  - Successful LANL LDRD proposal

FLATTOP



SPIDER

Improving nuclear data used in models – prompt fission neutron spectra

Knowledge of neutron and gamma outputs from fission as a function of incident energy important

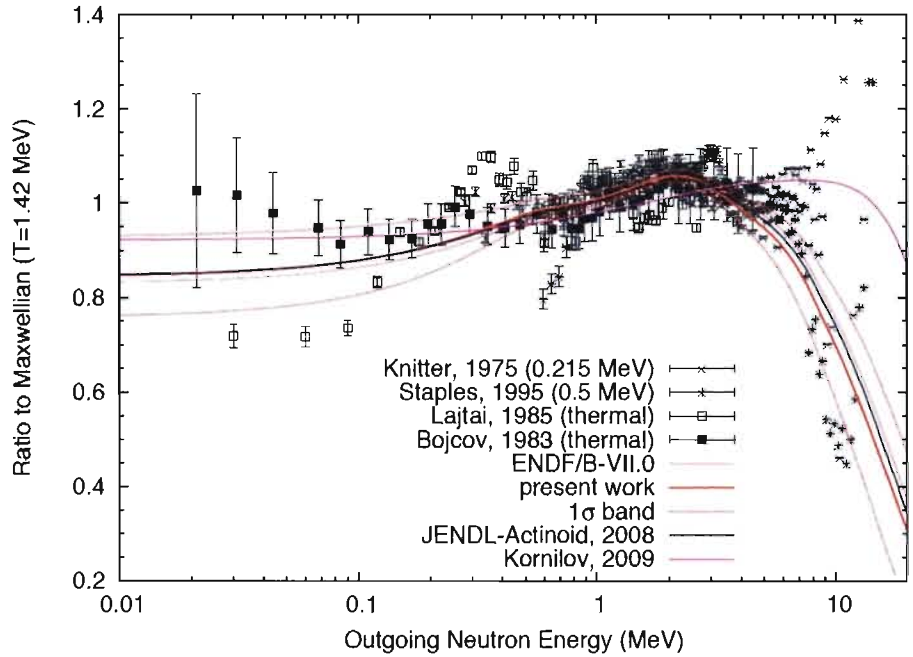
Criticality

> 30% of all neutrons are emitted with E < 1 MeV

10% uncertainty is a 0.1-0.2% effect on multiplicity

Both theory and experiments show at least 10% discrepancies

There is a paucity of experimental data in some parts of the energy spectrum



Radiochemistry

Magnitude of high-energy neutron emission is dropping exponentially with E

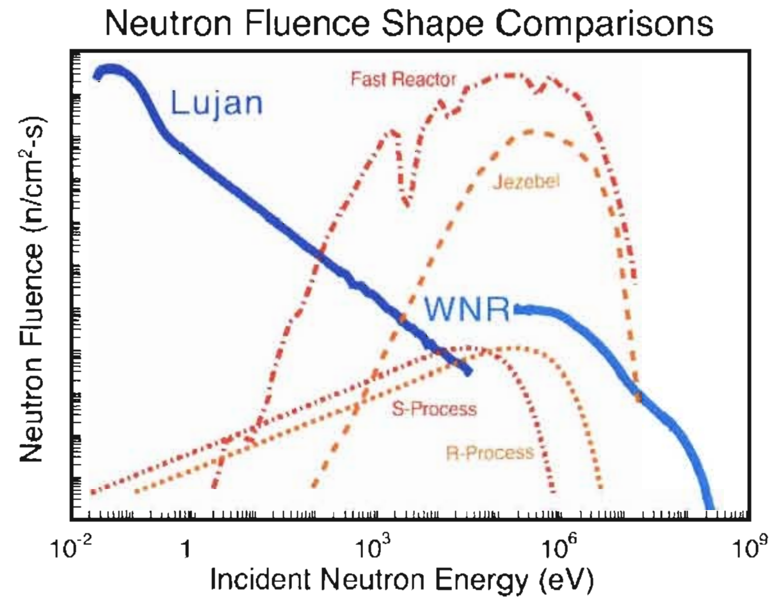
These neutrons have significant effects on diagnostics of the high-energy flux in weapons and transmutation products in reactors

IAEA CRP on this topic

Improved (n,f) cross sections

- The right-hand side of the Boltzmann Equation defines the fission neutron source term – neutron flux * chi emission distribution * nubar neutrons per fission * sigma-fission
  - To reduce the uncertainty on the fission source term, uncertainties in both sigma-fission and chi must be addressed

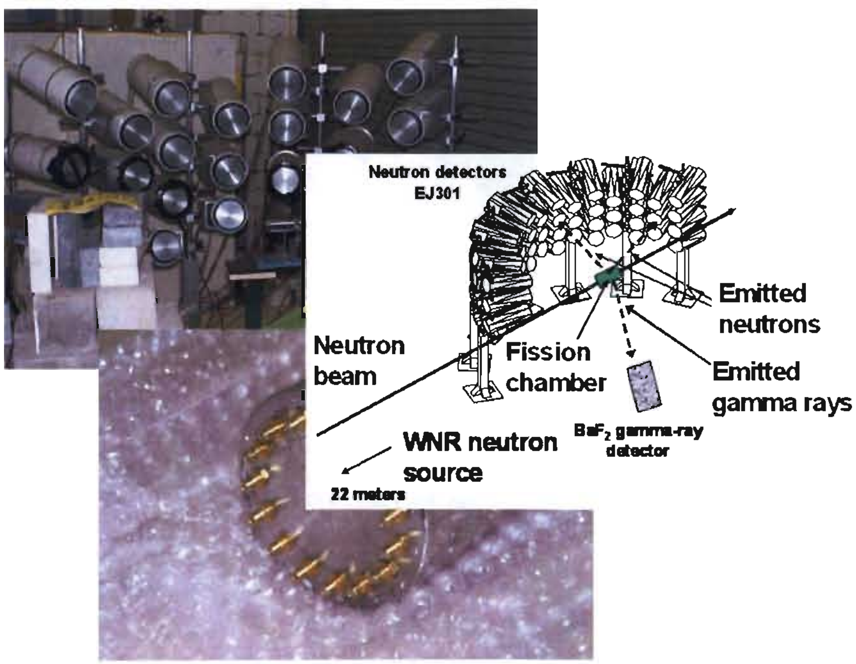
- Fission cross-section uncertainties are a significant question for both national security and energy applications
  - For weapons, the fission cross section affects the time evolution of neutron flux
  - For reactors, it is a key player in reducing safety margins
  - Our shared goal is to reduce fission cross-section uncertainties to below 1%



Collaborative experiments are addressing these data needs for Pu-239

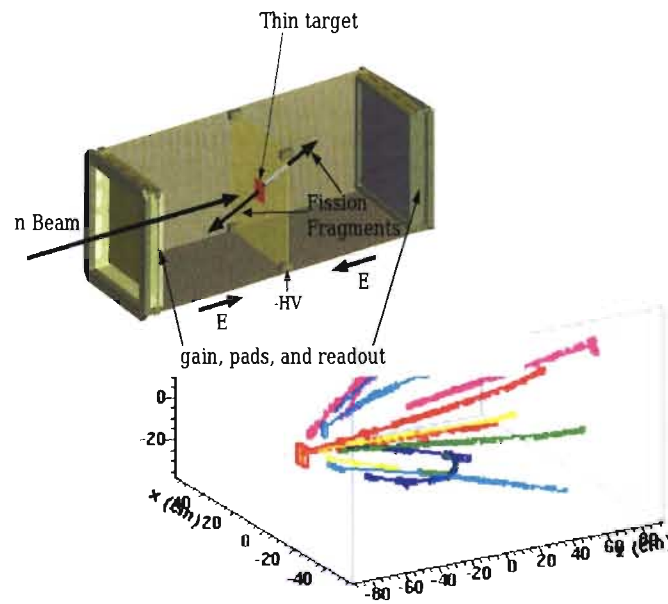
Fission neutron spectrum

- *Fission neutron output spectrum (chi-matrix) over the entire energy range of neutron input energies and neutron output energies*

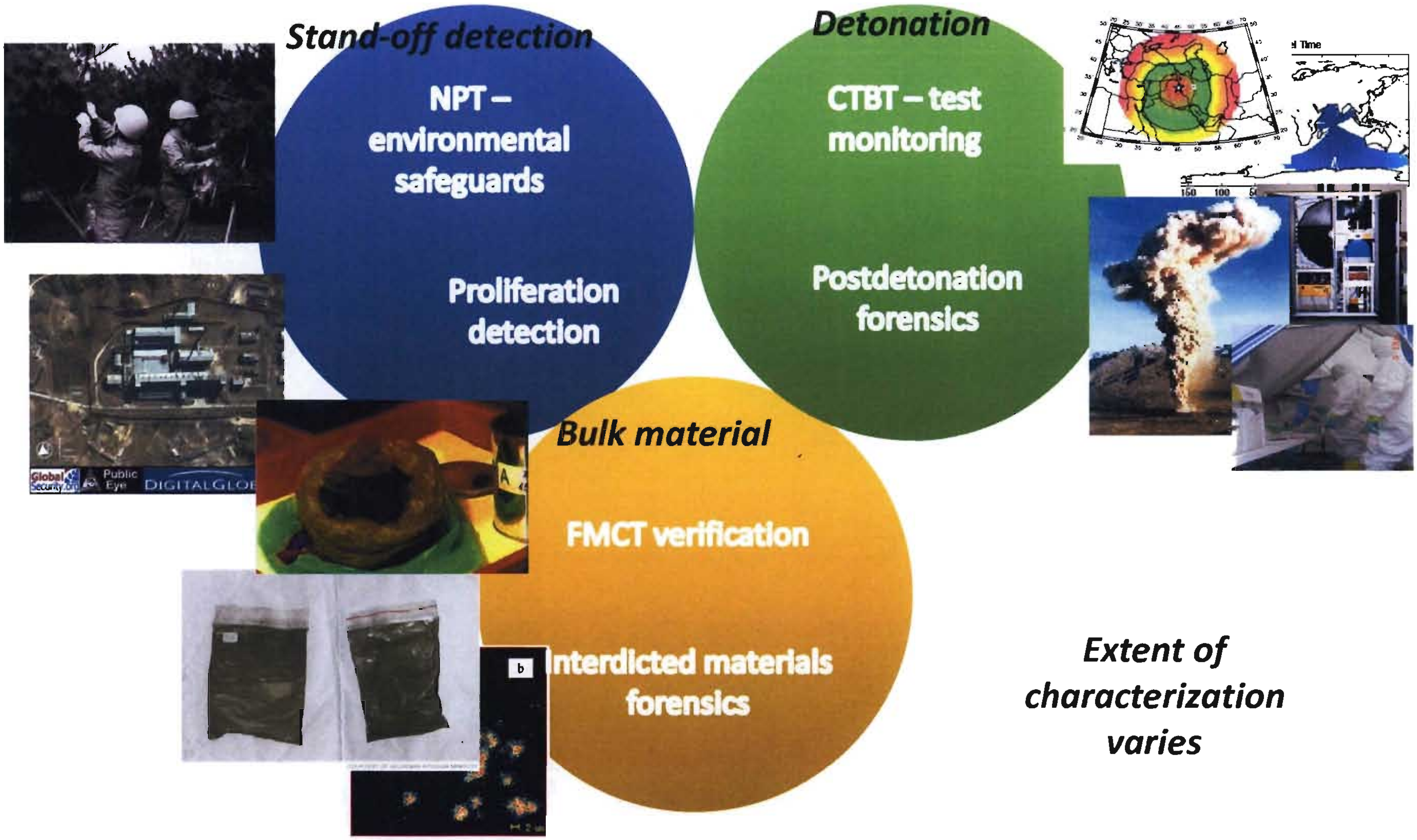


High precision (n,f) cross section

- *1% precision measurement proposed using a time-projection chamber (TPC)*



The range of requirements is expanding

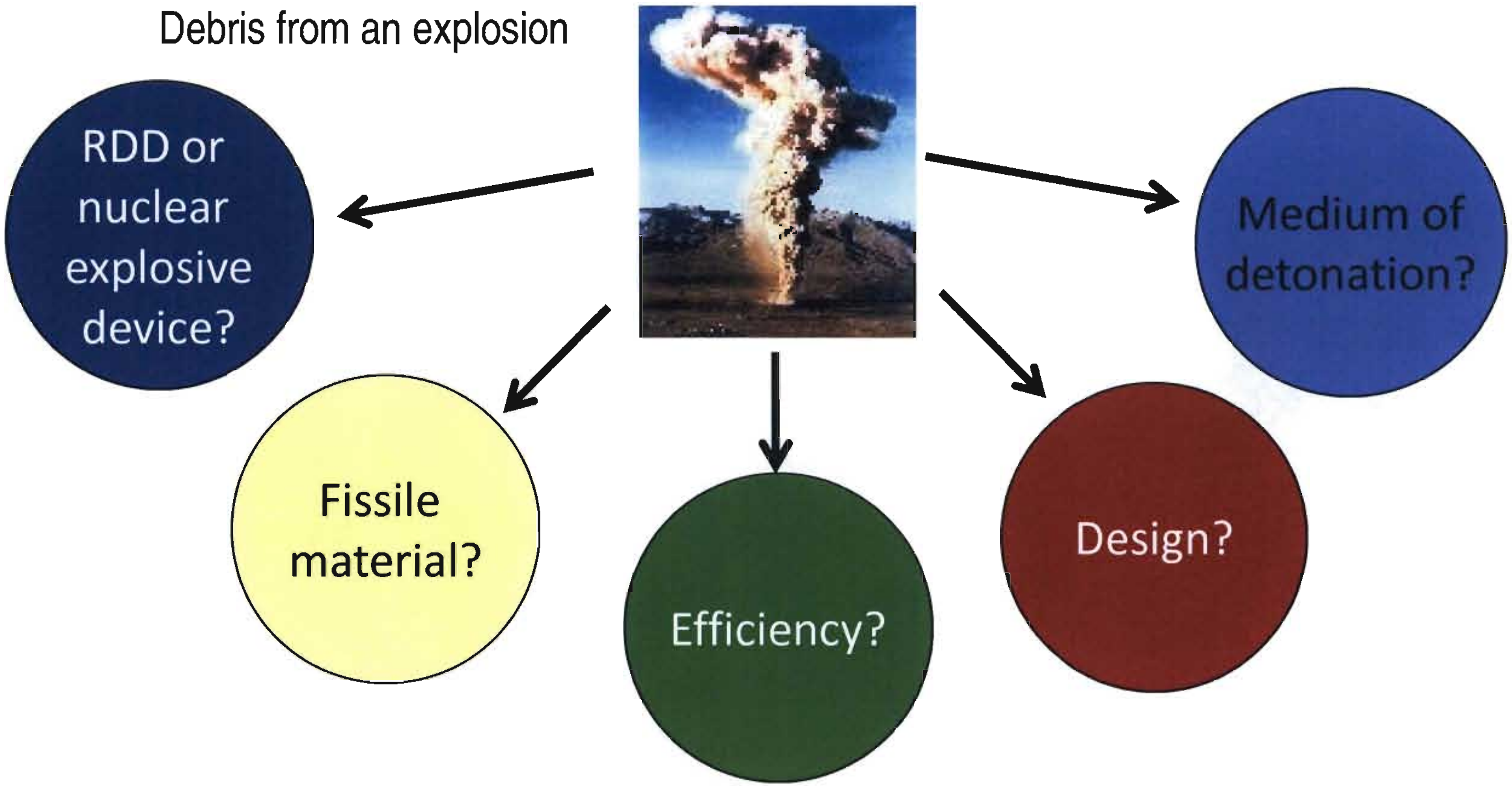


Extent of characterization varies

Forensic Investigation of Debris

To perform forensic analysis on debris, using developed data interpretation techniques to learn about the device

Debris from an explosion



The difference between this application and weapons radiochemistry: we can still measure the efficiency, but we don't know the ingoing mass/type of fuel - now we need to get yield some other way

New technical challenges – forensic analysis of debris

Collections:
- Models/tools for site characterization
- Communications
- Tools for sampling in differing environments
- New signatures

Was the detonation nuclear?

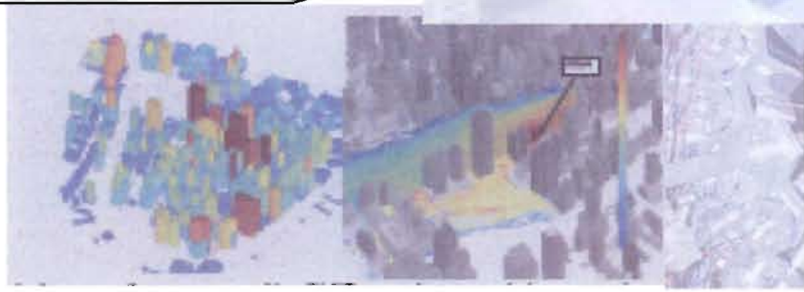
What was the fissile material?

How well did the device perform?

What can we tell about the design?

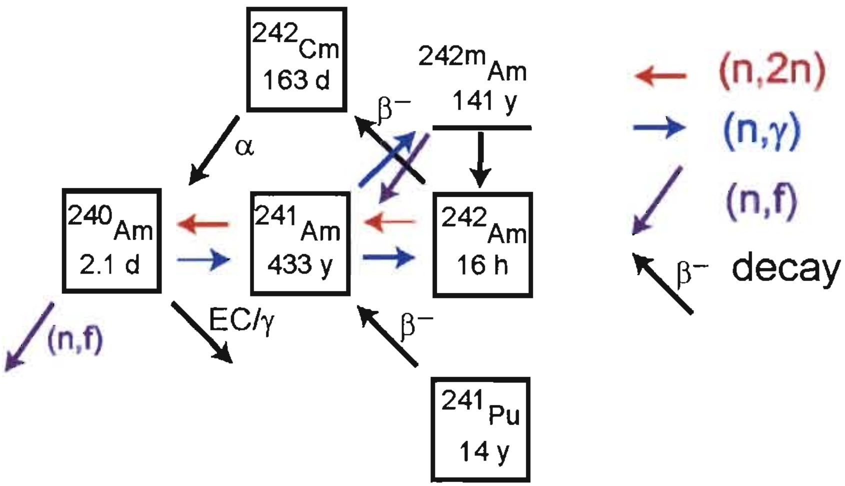
What was the medium of detonation?

Analysis:
- Rapid/automated separations
- New instrumental methods
- Fieldable analyses
- New signatures



Modeling
- Code development
- Incorporating new nuclear data
- Modeling outputs
- Addressing physics degeneracies.
- Making tools more "usable"

Isotopic ratios provide valuable information in nuclear reactions:



- Pu produced in reactors
- Always some ^{241}Pu produced
- ^{241}Pu β-decays to ^{241}Am
- Use ^{241}Am as an internal neutron fluence monitor

$\Delta A = {}^{240}Am/{}^{241}Am$

- Sensitive to high-energy neutrons ^{241}Am(n,2n) threshold = 6.7 MeV
- $^{240/241}Am \sim \frac{1}{2}\, \sigma_{n,2n}\, \Phi_n$

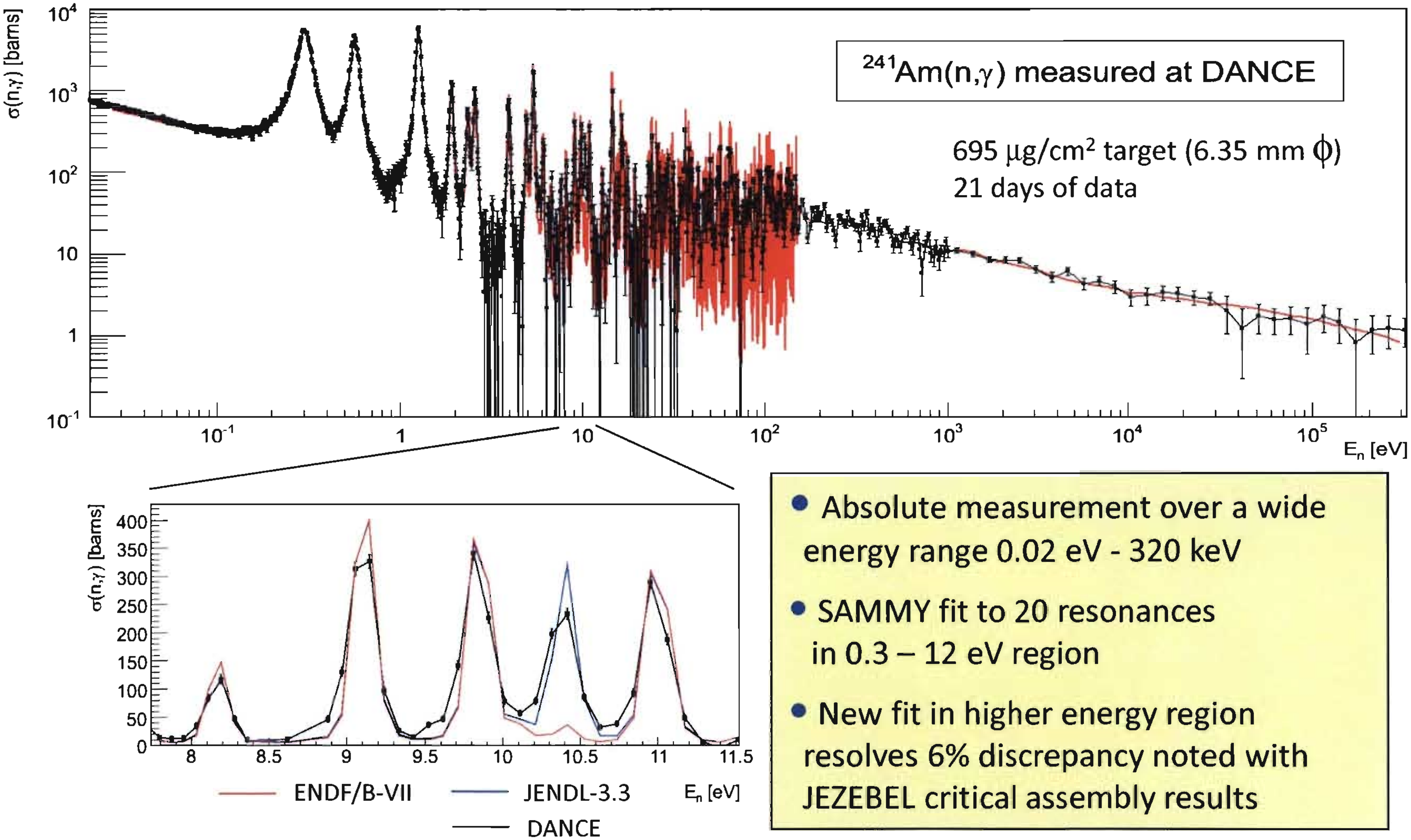
$^{242}Cm/^{241}Am$

- ^{242}Cm produced by ^{241}Am(n,γ) ^{242g}Am (16 hr) -> ^{242}Cm (163 d)
- Capture to ^{242m}Am (141 yr) also possible – ^{242m}Am/^{241}Am
- Neutron capture is sensitive to low-energy n's, $\sigma_\gamma \sim 1/v$

More precise Am(n,γ) & (n,f) measurements are needed

$^{241}Am(n,\gamma)$ Measurement at DANCE (Vieira, et al.)

M. Jandel *et al.*, Phys. Rev. C 78, 034609 (2008)

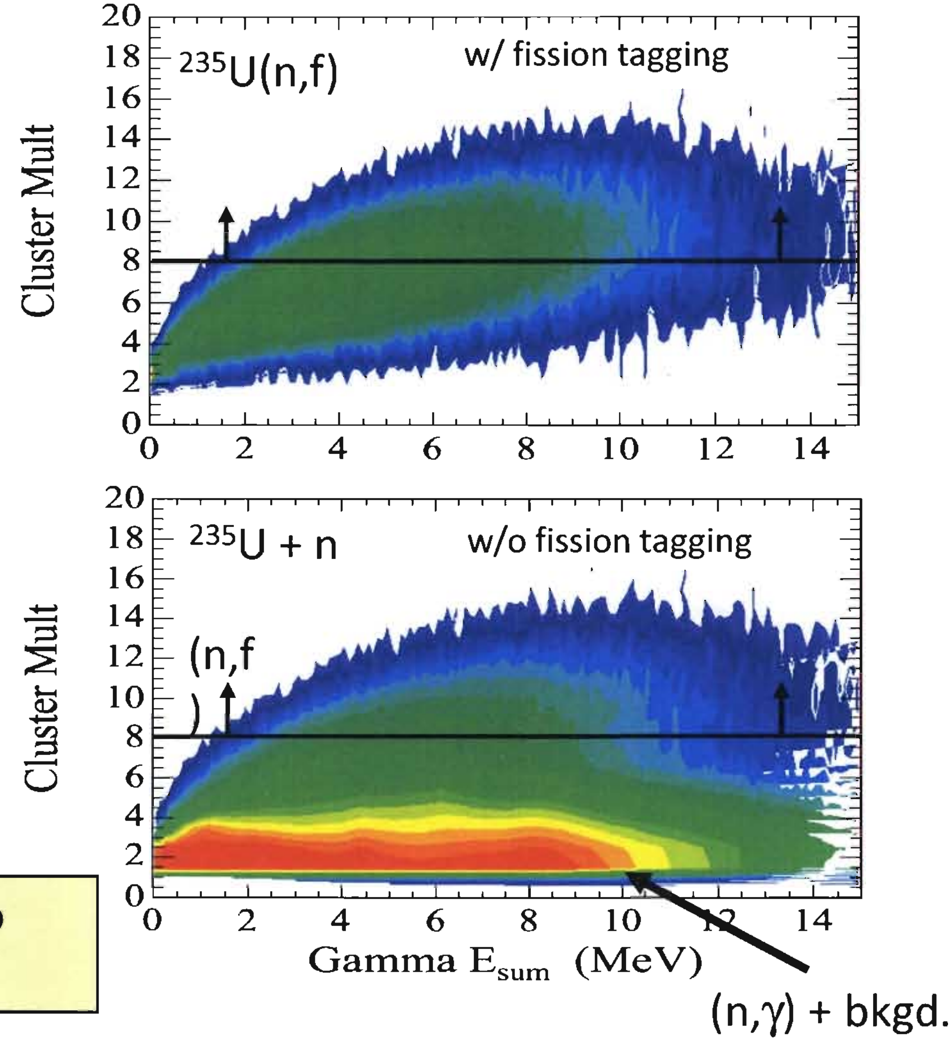


- Absolute measurement over a wide energy range 0.02 eV - 320 keV
- SAMMY fit to 20 resonances in 0.3 – 12 eV region
- New fit in higher energy region resolves 6% discrepancy noted with JEZEBEL critical assembly results

(n,fission) Reactions – also Emit γ-rays

To separate capture from fission events, we developed a 4π fission detector that surrounds the target

DANCE 4π PPAC Detector

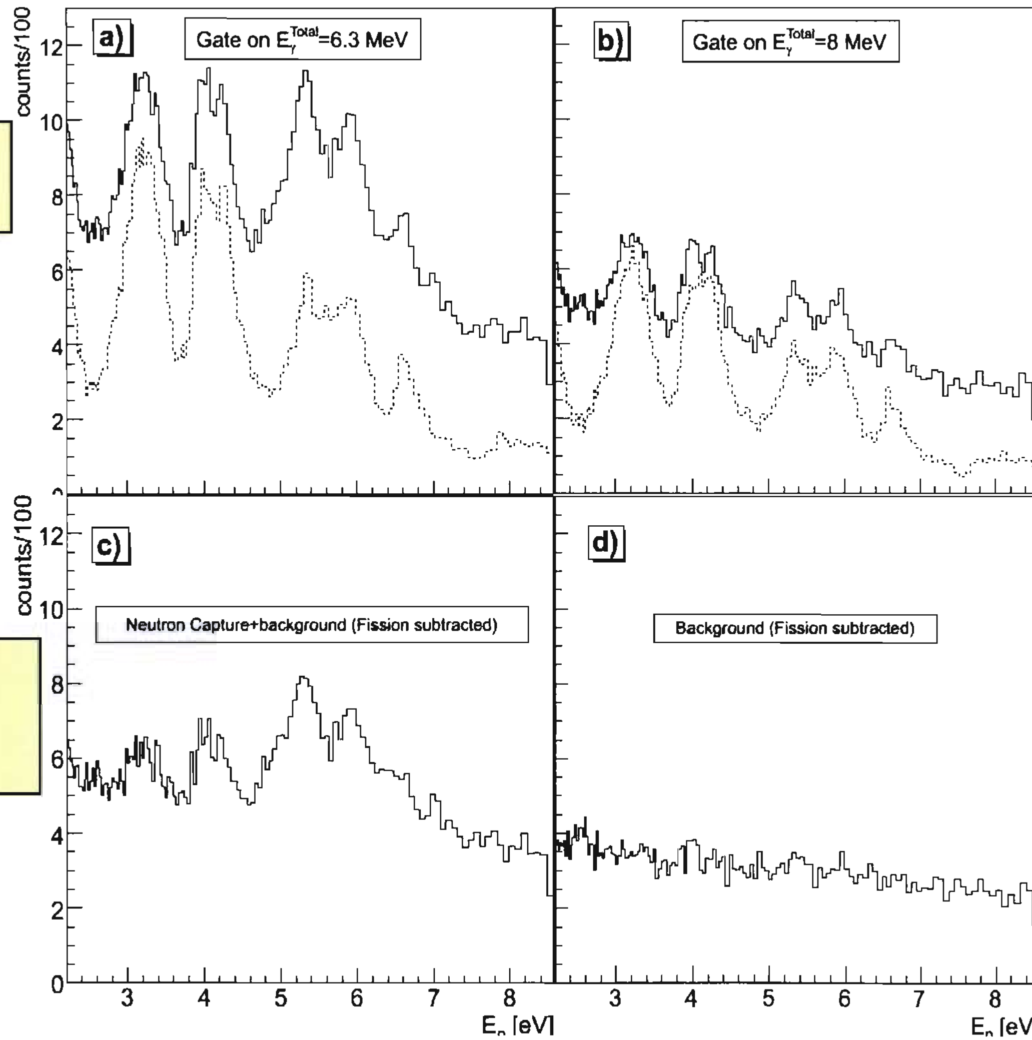


Normalize & subtract out (n,f) to measure (n,γ) and σ_γ / σ_f

Subtract out (n,f) events to extract $^{242m}Am(n,\gamma)$

a) All data w/ $M_\gamma > 4$ + $5.8<E_{sum}<6.8$ MeV

b) All data w/ $M_\gamma > 4$ + $7.5<E_{sum}<8.5$ MeV (scattered neutrons captured in BaF_2)

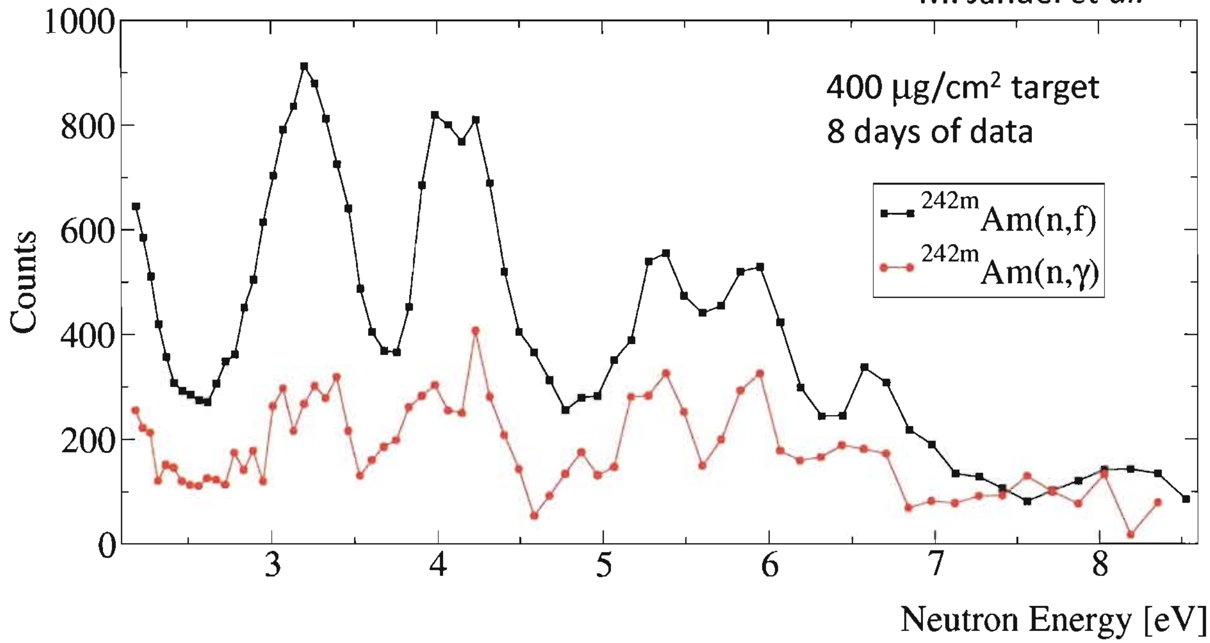


c) Untagged–tagged*eff $M_\gamma>4$ + 5.8-6.8 MeV $^{242m}Am(n,\gamma)$ + bkgd.

d) Untagged–tagged*eff $M_\gamma>4$ + 7.5-8.5 MeV bkgd.

First Resonances Measured in $^{242m}Am(n,\gamma)$

M. Jandel *et al.*



- First measurement of (n,γ) resonance in ^{242m}Am, final analysis in progress
- Theorists expected (n,γ) to be 100x small than (n,f), but we observe only 2-5 x smaller

Conclusions

- Developments in national security have closely followed those in the fields of nuclear and radiochemistry
- Radiochemistry continues to play a significant role in today's stockpile stewardship program
- These disciplines now play a role in a wider range of programs (nonproliferation, homeland security, etc.).

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