Nuclear Science and Technology Division

Monte Carlo Modeling for the Characterization of Nuclear Materials

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Monte Carlo Modeling for the Characterization of Nuclear Materials

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Abstract

The present paper describes the use of the Monte Carlo code MCNP-PoliMi, in conjunction with a post-processing code, to model the response of organic scintillators to neutrons and gamma rays. The need for a code to accurately describe correlation measurements is described. The main features of the code are discussed, and comparisons between simulations and measurements are presented. Emphasis is placed on the simulation of the response of organic scintillators to neutrons and gamma rays. A few possible post-processing options are presented: the simulation of source-detector covariance and bi-covariance functions and the simulation of detector pulse height responses. In each case, a comparison of simulations and measurements is presented.

1. Introduction

Recent efforts have been aimed at developing new technologies for the characterization of nuclear materials for a wide range of applications in nuclear nonproliferation, international safeguards, nuclear material control and accountability, national security, and counter-terrorism programs. The development of measurement techniques based on the detection of radiation emitted by nuclear materials often relies on the use of Monte Carlo codes. The codes are used to simulate the interaction of the radiation with the material under investigation, the environment, and the radiation detectors.

One such measurement technique is based on the detection of correlated particles emitted by fission events in active or passive measurements conducted on fissile materials [1]. In active measurements, an external interrogation source is used to induce fission in the material. The measurement is performed with plastic or liquid scintillation detectors, sensitive to fast neutrons and gamma rays. Typically, the fissile material is not moderated by external moderating material, and so the correlations are performed with timing of the order of a few tens of nanoseconds. This technique shows promise in the measurement of uranium and plutonium, in metal and oxide form. Current efforts are aimed at developing a novel neutron multiplicity counter that is based on the use of this methodology and will have a much shorter die-away time than conventional multiplicity counters.

This paper is organized as follows: Section 2 describes the main features of the MCNP-PoliMi code. Section 3 presents three possible post-processing options and the comparison of simulations and measurements. In Section 4, some conclusions are drawn.

2. Main Features of MCNP-PoliMi

Previous studies have focused on the design and validation of the Monte Carlo code MCNP-PoliMi, which was developed to simulate measurements based on neutron and gamma ray correlations from fission [2-4]. MCNP-PoliMi is a modification of MCNP-4c [5].

MCNP-PoliMi provides an improved representation of the physics of the processes involved in correlation measurements on fissile materials. The standard MCNP code was not designed for the simulation of higher-order signatures from the signals of detectors. In fact, the simulation of higher-order signatures cannot be based on average values, and average values are used in the standard MCNP code in the production of neutrons from fission and in the generation of gamma rays that originate in neutron interactions. Consequently, modifications to the MCNP code were necessary to simulate these phenomena more correctly.

The main features of the MCNP-PoliMi code can be summarized as follows. Simulation of fission: implementation of the correct multiplicity of prompt neutrons and gamma rays from spontaneous and induced fission. Generation of secondary gamma rays: implementation of the correlation between neutron interaction and secondary gamma ray production. Multi-particle sources: addition of a number of spontaneous fission and alpha/n neutron and gamma ray sources. Output format: collision output table. Further details on the modifications are given in reference 2.

An example of the collision output table is given in Table 1. In the table, single neutron and photon interactions are reported and information about each is given. Interaction type, target nucleus, energy deposited in the collision, time at which the collision occurred, and position of the collision are among the information given in the output table. The last column of Table 1 gives the energy of the incident neutron before the collision. For example, the first row of the table refers to a 1.6 MeV neutron that interacts via elastic scattering with a carbon nucleus, depositing 339 keV in the collision. The interaction occurred 9.4 ns after the originating source event, in cell number 9.

The information reported in the output file can be post-processed to obtain information on the detector response. The data output post-processing is described in Section 3.

Table 1. Excerpt of MCNP-PoliMi output file.

History number	Particle number	Projectile type*	Interaction type$^{\Delta}$	Target nucleus•	Cell number of collision event	Energy deposited in collision (MeV)	Time (shakes)	Collision position (x, y, z)			Generation number	Number scatterings	Code	Energy (MeV)
...														
151	4	1	-99	6000	9	0.33893	0.944	-16.71	-1.73	3.92	0	0	0	1.61E+00
151	4	1	-99	1001	9	0.71283	1.158	-15.55	-4.66	2.84	0	1	0	1.27E+00
151	20	1	-99	1001	8	0.10313	1.464	13.4	3.95	-4.48	3	0	0	2.61E+00
151	20	1	-99	1001	8	1.20735	1.488	13.91	3.96	-4.51	3	1	0	2.50E+00
151	20	1	-99	1001	8	0.05711	1.493	13.98	3.9	-4.49	3	2	0	1.30E+00
151	20	1	-99	1001	8	0.6134	1.64	15.9	2.76	-4.28	3	3	0	1.24E+00
151	20	1	-99	1001	8	0.42279	1.756	16.49	2.15	-3.34	3	4	0	6.24E-01
151	20	1	-99	1001	8	0.07376	1.974	17.29	2.74	-2.42	3	5	0	2.01E-01
151	20	1	-99	1001	8	0.08552	2.297	17.55	2.64	-0.89	3	7	0	1.14E-01
164	23	1	-99	1001	9	0.22836	4.21	-18.07	-4.43	-3.21	1	3	10	2.84E-01
164	4	2	1	1	8	0.11409	0.193	14.87	4.73	-1.2	0	1	0	6.49E-01

* 1 = neutron; 2 = photon
Δ -99 = elastic scattering; 1 = Compton scattering
• 1001 and 1 = hydrogen; 6000 = carbon

3. Monte Carlo Post-processing

Fig. 1 shows a sketch that illustrates the detection model implemented in the Monte Carlo simulation of plastic and liquid scintillators. As was discussed in Section 2, the MCNP-PoliMi code provides detail on the single particle interactions with the detector material. In the detection model, each interaction occurring in the scintillator generates a contribution to the pulse. This contribution depends on how much energy was deposited in the interaction, on what particle interacted (neutron or photon), and, in the case of neutrons, on what nucleus was hit (proton or carbon nucleus). On the basis of this information, the light generated in the scintillator can be determined for each interaction. Then, the light pulses occurring within a given time are added together linearly to form a total pulse. This time is referred to as the 'pulse generation time', and is typically set to 10 ns for the scintillation detectors.
This simple detection model was proven to be effective for liquid and plastic scintillators having dimensions up to 6 inches, approximately. In the case of larger scintillators, it would be possible to take into account phenomena such as light attenuation and light reflection.

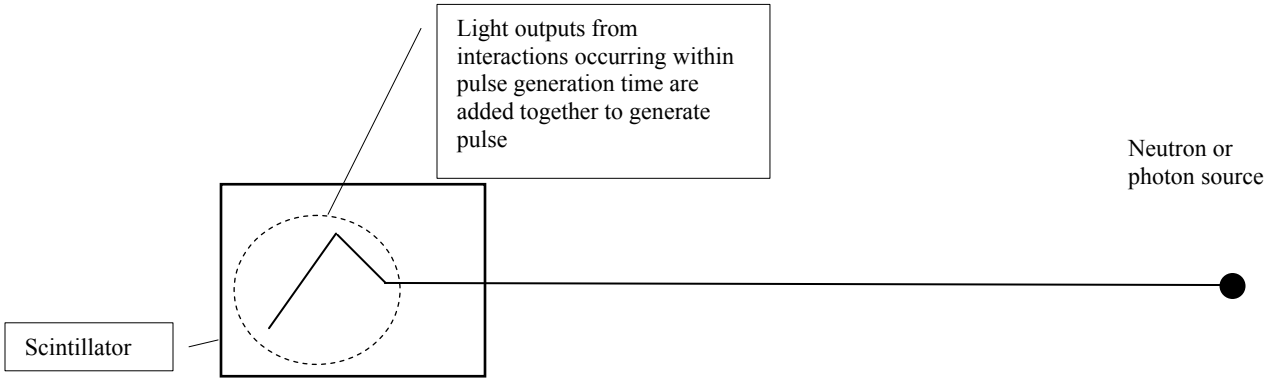


Fig. 1. Illustration of the detection model for liquid and plastic scintillators.

3.1 Simulation of Covariance Functions

The proposed detection model has been used to simulate neutron and photon detection by liquid and plastic scintillators [3]. Fig. 2 shows an example of the comparison of an MCNP-PoliMi simulation with a measurement performed with a Cf-252 source and a plastic scintillator, having dimensions 7 by 7 by 10 cm, approximately. In the experiment, the Cf-252 source was placed at a distance of 1 m from the plastic scintillator, and the source to detector time of flight was measured. The acquired signature consists of two peaks; the first, sharp peak is given by the gamma rays and the second, broad peak is given by the neutrons.

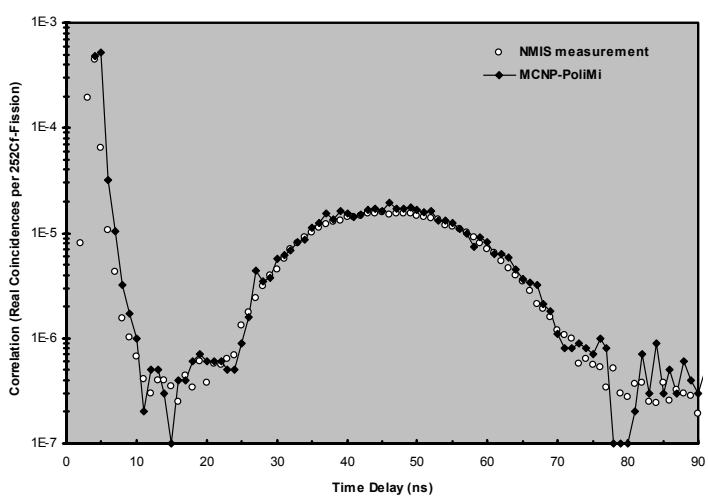


Fig. 2. Comparison of Monte Carlo simulation and measurement for a Cf-252 source and plastic scintillator.

Inspection of Fig. 2 shows that there is very good agreement between the simulation and the measurement.

3.2 Simulation of Bi-covariance Functions

The correlation between a source and two detectors is referred to as the source-detectors bi-covariance function. A contribution to the bi-covariance is made when detection occurs in two detectors as a result of the same source event.

A comparison is presented between simulated data and experimental data acquired with the Nuclear Materials Identification System (NMIS). The measurements and simulations were performed for assemblies of plutonium metal shells of varying inner and outer diameter. A sketch of the measurement configuration is given in Fig. 3. Further details on the experiments and their simulation are given elsewhere [6-7].

Fig. 4 shows the comparison of the measured source-detector bi-covariance for a plutonium metal shell (98 wt% Pu-239) having mass 4.0 kg, outer radius 6.0 cm and inner radius 5.35 cm. Comparison of Fig. 4 (a) and (b) shows that the four regions of the bi-covariance are well represented in the simulated signature. Inspection of Fig. 4 (a) reveals that there was a 1 ns time offset in the timing alignment of the two detectors used in the measurement.

Contributions to the bi-covariance can be due to neutrons or photons and to particles from the source or particles from induced fission in the fissile material. The information on what type of particle was detected is present in the collision output file and can be analyzed by a specifically designed post-processing code. This capability is illustrated in Figs. 5 and 6. Fig. 5 shows the result of Fig. 4 (b) separated into four components according to the particle type that was detected. Experimentally, a split of the bi-covariance according to particle types can be achieved by using liquid scintillators, which have pulse shape discrimination properties [8].

Fig. 6 shows a split of Fig. 4 (b) performed according to generation of the particles pairs detected by the scintillators. In our nomenclature, generation zero particles are source particles than reach the detector uncollided, and source particles that have interacted by all reactions except for nuclear fission; induced fission particles originate from fission induced inside the uranium metal casting. Integration of the surfaces given in Fig. 6 gives the total number of detected pairs per Cf-252 source fission. The number of pairs [Fig. 6 (a)] given by generation zero particles is 5.5 10^{-4}. The number of pairs given by induced fission particles [Fig. 6 (b)] is 3.18 10^{-4}. The remaining pairs are given by combinations of generation zero particles and induced fission particles [Fig. 6 (c) and (d)], for a total of 1.93 10^{-4}.

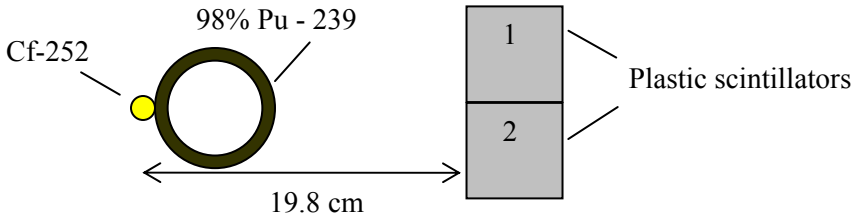
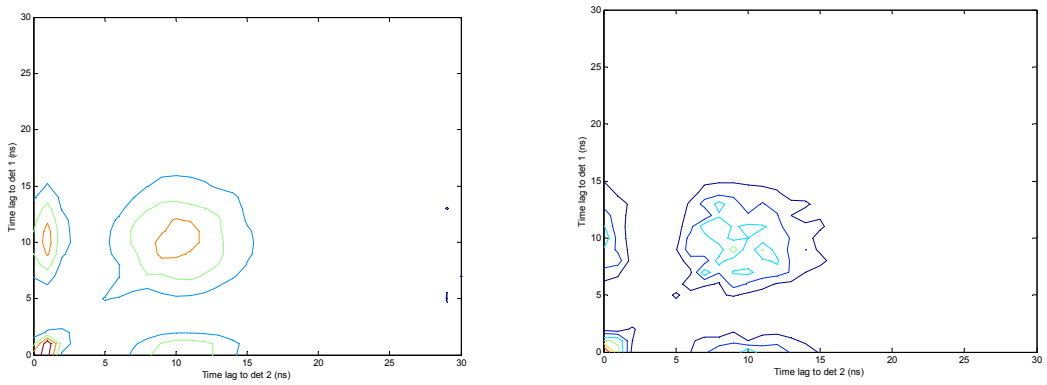


Fig. 3. Sketch of the configuration for the simulation of the source-detectors bicovariance function with plutonium sample (not to scale).



(a) (b)

Fig. 4. Source-detectors bi-covariance with plutonium sample: (a) Experimental result and (b) MCNP-PoliMi simulation.

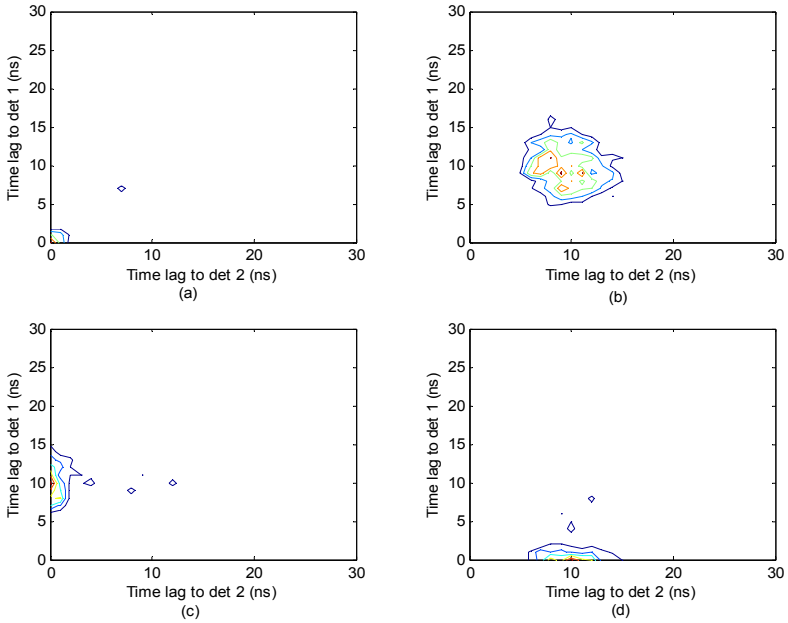


Fig. 5. Simulation of source-detectors bi-covariance with plutonium sample: (a) photon – photon pairs, (b) neutron – neutron pairs, (c) neutron – photon pairs, and (d) photon – neutron pairs.

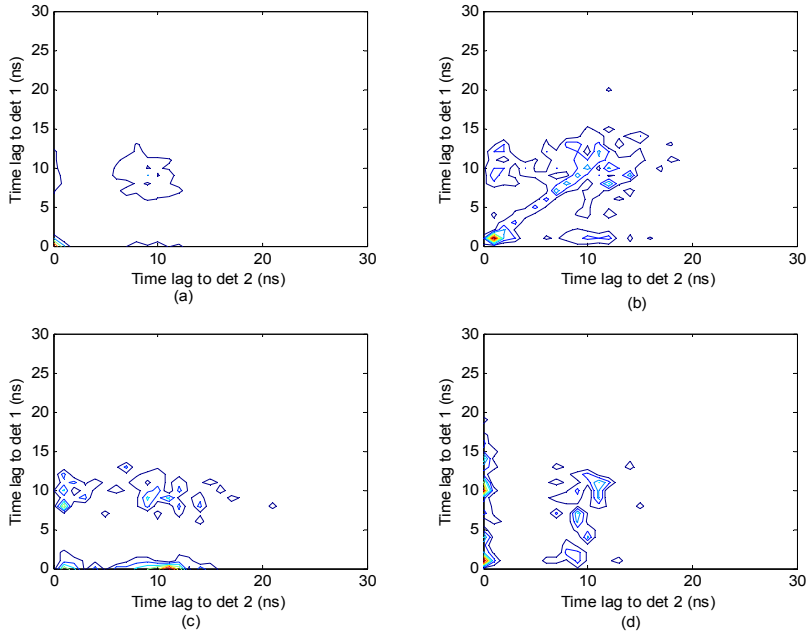
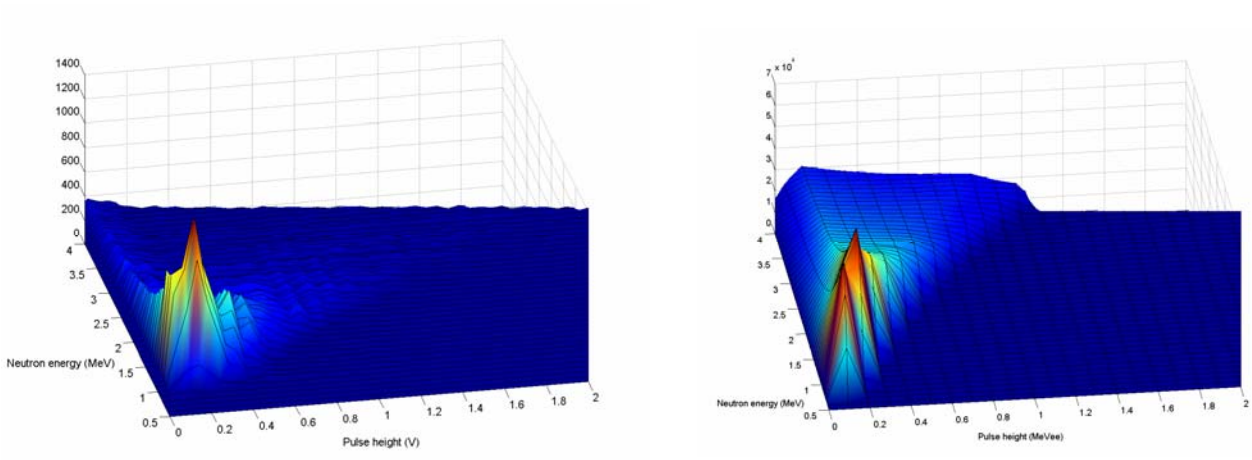


Fig. 6. Simulation of source-detectors bi-covariance with plutonium sample: (a) generation zero – generation zero pairs, (b) induced fission – induced fission pairs, (c) generation zero – – induced fission pairs, and (d) induced fission – generation zero pairs.

3.3 Simulation of Pulse Height Spectra

Previous studies were aimed at simulating and measuring the neutron response matrix for a liquid scintillator (pulse height – neutron energy) [9]. The simulation relies on the fact that the information given in the collision output data file can be post-processed to obtain the pulse height spectra from the detector. This is done by accumulating the light outputs for particle interactions occurring in the detector and storing this information together with the information on the neutron energy.

Fig. 7 shows the result of the measurement and simulation of the pulse height distribution of neutron pulses measured with a liquid scintillator [9]. The measurement was performed with a timed Cf-252 source, and the incident neutron energy was determined by time of flight.



(a) (b)

Figs. 7(a) and (b). Pulse height – incident neutron energy for liquid scintillator: (a) Experimental result and (b) MCNP-PoliMi simulation.

A qualitative comparison of the results of the Monte Carlo simulations and the measurement shows reasonable agreement in the shape of the response matrix (Figs. 7). The simulated response matrix contains more data at the higher neutron energies than the measured data. In fact, the Cf-252 neutron spectrum does not contain many high energy neutrons and therefore convergence of the response matrix at higher energies requires long measurement times. Ideally, the measurement could be performed with neutrons from a linear accelerator. As an alternative, a measurement could be performed using Am/Be or Pu/Be sources, which have a harder neutron

spectrum than Cf-252. Such a measurement could be performed using a second scintillator placed in contact with the source to provide a trigger pulse for the time of flight.

4. Conclusions

This paper described the use of the Monte Carlo code MCNP-PoliMi, in conjunction with a post-processing code, to model measurements for nuclear material identification that are based on the use of organic scintillators to detect neutrons and gamma rays. A few possible post-processing options were presented: the simulation of covariance and bi-covariance functions and the simulation of detector pulse height responses. Other post-processing options can be developed to simulate particle detection by other types of detectors.

Acknowledgments

The author gratefully acknowledges John Mihalczo of Oak Ridge National Laboratory and Vitaliy Dubinin and colleagues of the Russian Federal Nuclear Center, All-Russia Scientific Research Institute of Experimental Physics, for making available the data taken with NMIS on the plutonium assembly.

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9. S.A. Pozzi, "Determination of Liquid Scintillator Response Matrix for Neutron Spectrum Unfolding," 7th International Conference on Facilities Operations – Safeguards Interface, Charleston, South Carolina, February 29 - March 4, 2004.

UT-BATTELLE

Monte Carlo Modeling for the Characterization of Nuclear Materials

Sara Pozzi
Nuclear Science & Technology Division

International Meeting on Reactor Noise (MORN)
Budapest, Hungary
May 17-19, 2004

Outline

- Nuclear Materials Identification System (NMIS): brief overview
- Main features of MCNP-PoliMi
- Nuclear Materials Identification System measurements on plutonium metal
- Measurement of neutron response matrix of liquid scintillator
- Conclusions

Use of Organic Scintillators

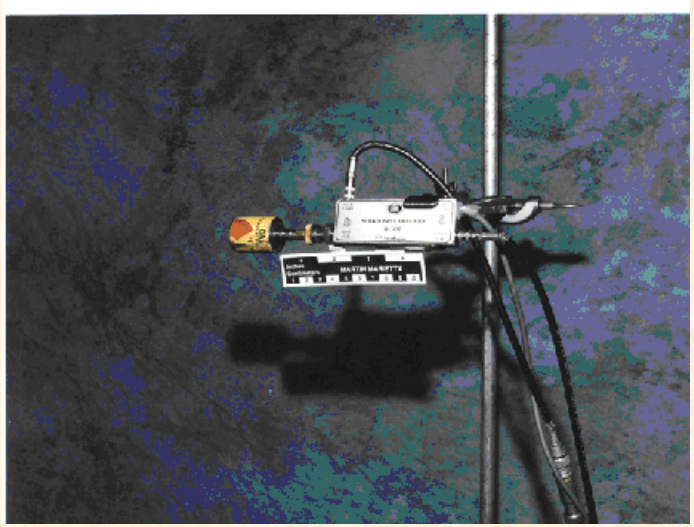
- Sensitive to fast neutrons and gamma rays
- Fast response (few tens of ns): reduces dieaway time
- Neutron spectroscopy

Nuclear Materials Identification System
JT Mihalczo et al., NIM A, 450, pp. 531 (2000).

Characterization of nuclear materials

Correlation measurements

Sources for Active Measurements



- Cf-252 sources in ionization chamber. Operated in pulsed mode, provides timing.

- DT generators produce monoenergetic neutrons in a directed cone. Neutrons are time-tagged using an alpha detector.

MCNP-PoliMi

- MCNP-PoliMi: a modification of MCNP-4C, originally developed to simulate correlation measurements

Code was developed at the Polytechnic of Milan, Italy by Enrico Padovani and Sara Pozzi
Code is distributed by RSICC – ORNL

- Also used to simulate detector response

- Monte Carlo output is in the form of a table, containing information on particle collisions in the detector cells

SA Pozzi et al., NIM A, 513/3 pp. 550-558.

Collision Output File From MCNP-PoliMi (excerpt)

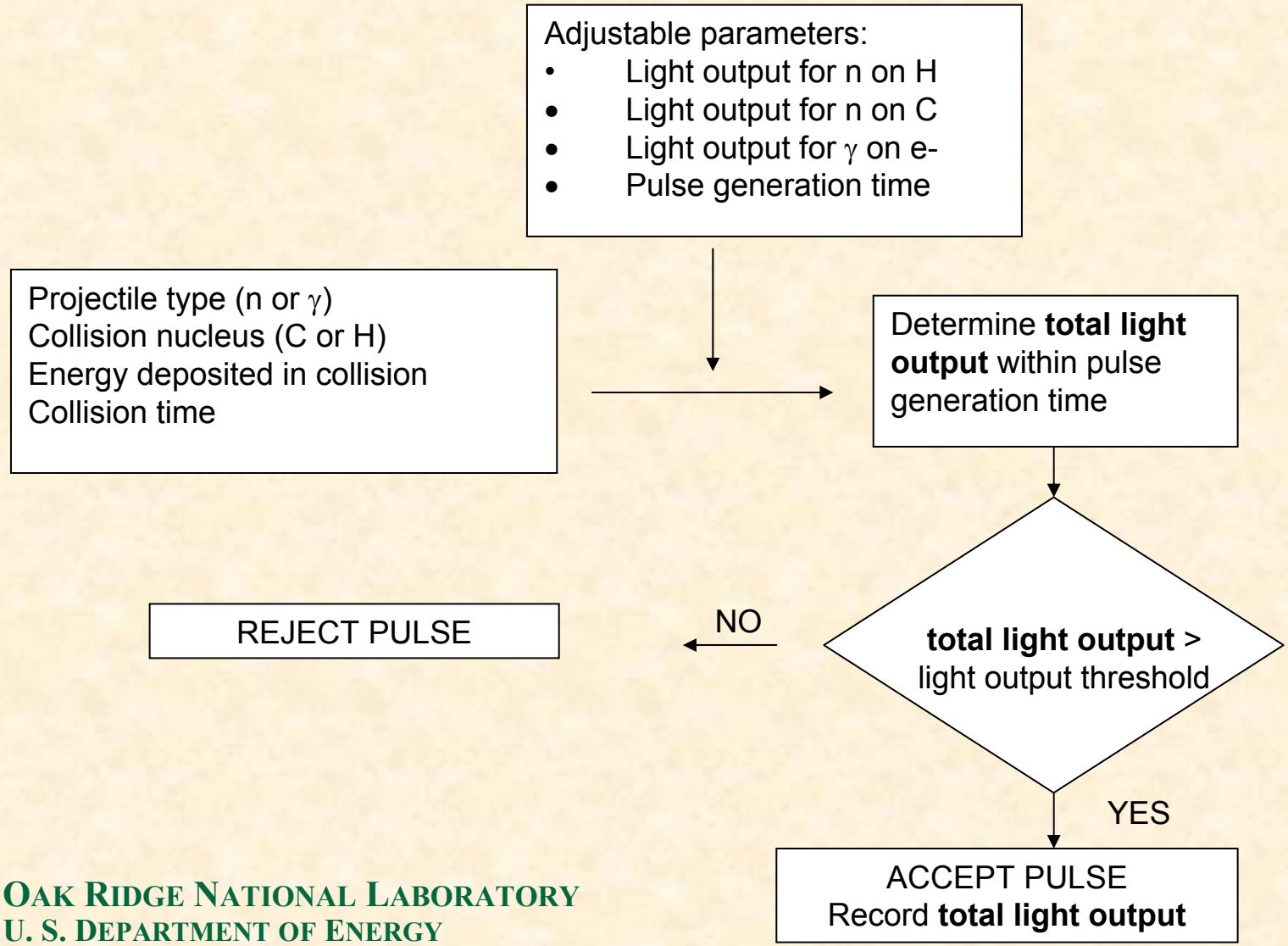
History number	Particle number	Projectile type*	Interaction type$^{\Delta}$	Target nucleus*	Cell number of collision event	Energy deposited in collision (MeV)	Time (shakes)	Collision (X)	position (Y)	(Z)	Particle weight	Generation number	Number scatterings
...													
29	20	1	-99	1001	2	0.3105	2.743	-25.64	14.37	8.7	1	2	2
29	20	1	-99	1001	2	0.1894	2.846	-26.01	-14.8	9.48	1	2	3
29	20	1	-99	1001	2	0.1617	3.045	-27.27	15.07	10.1	1	2	4
29	43	2	1	6	1	0.7734	6.514	31.83	7.07	11.2	1	2	1
29	8	1	-99	1001	1	0.3731	3.965	27.84	18.82	-0.6	1	0	1
...													

* 1 = neutron; 2 = photon

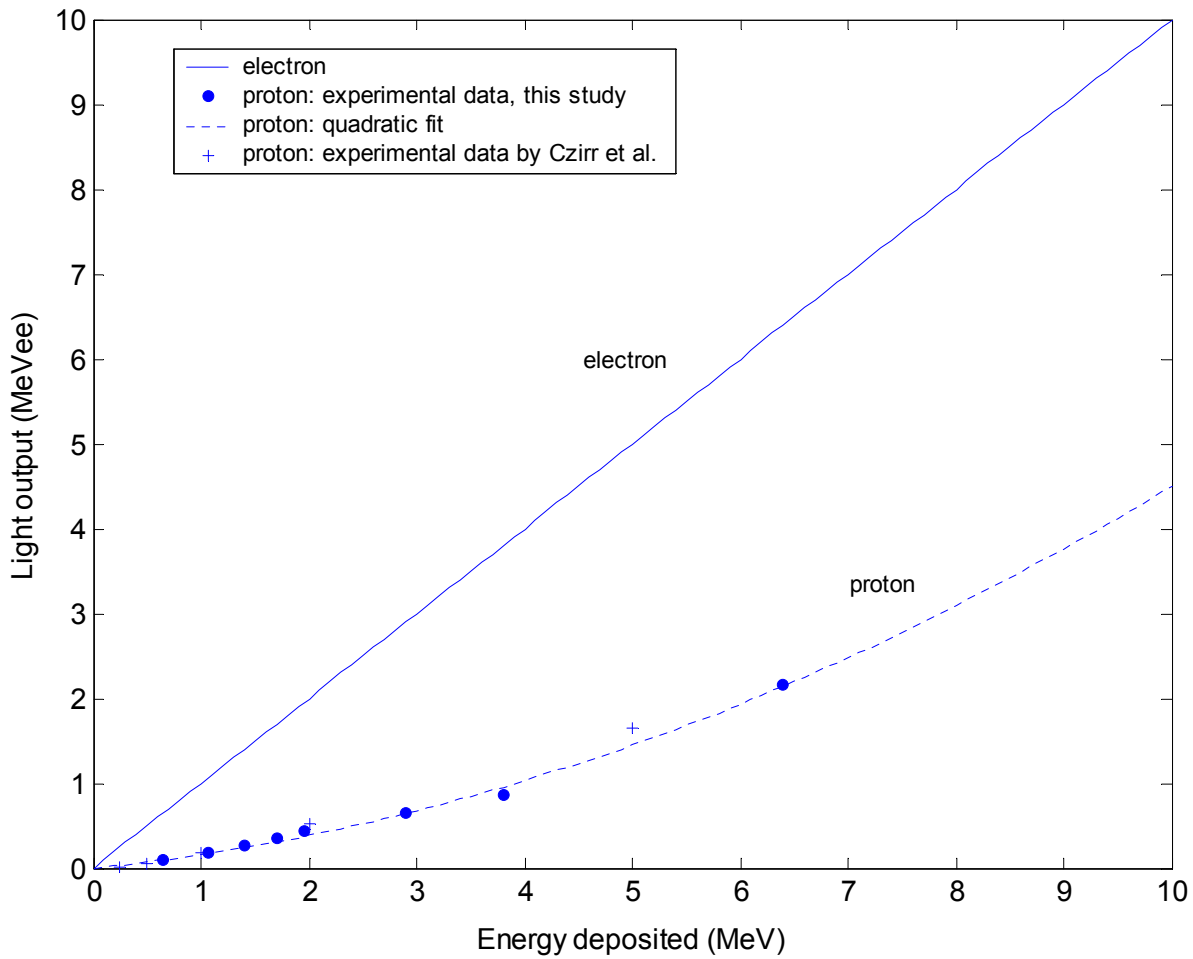
Δ -99 = elastic scattering; 1 = Compton scattering

• 1001 = hydrogen; 6 = carbon

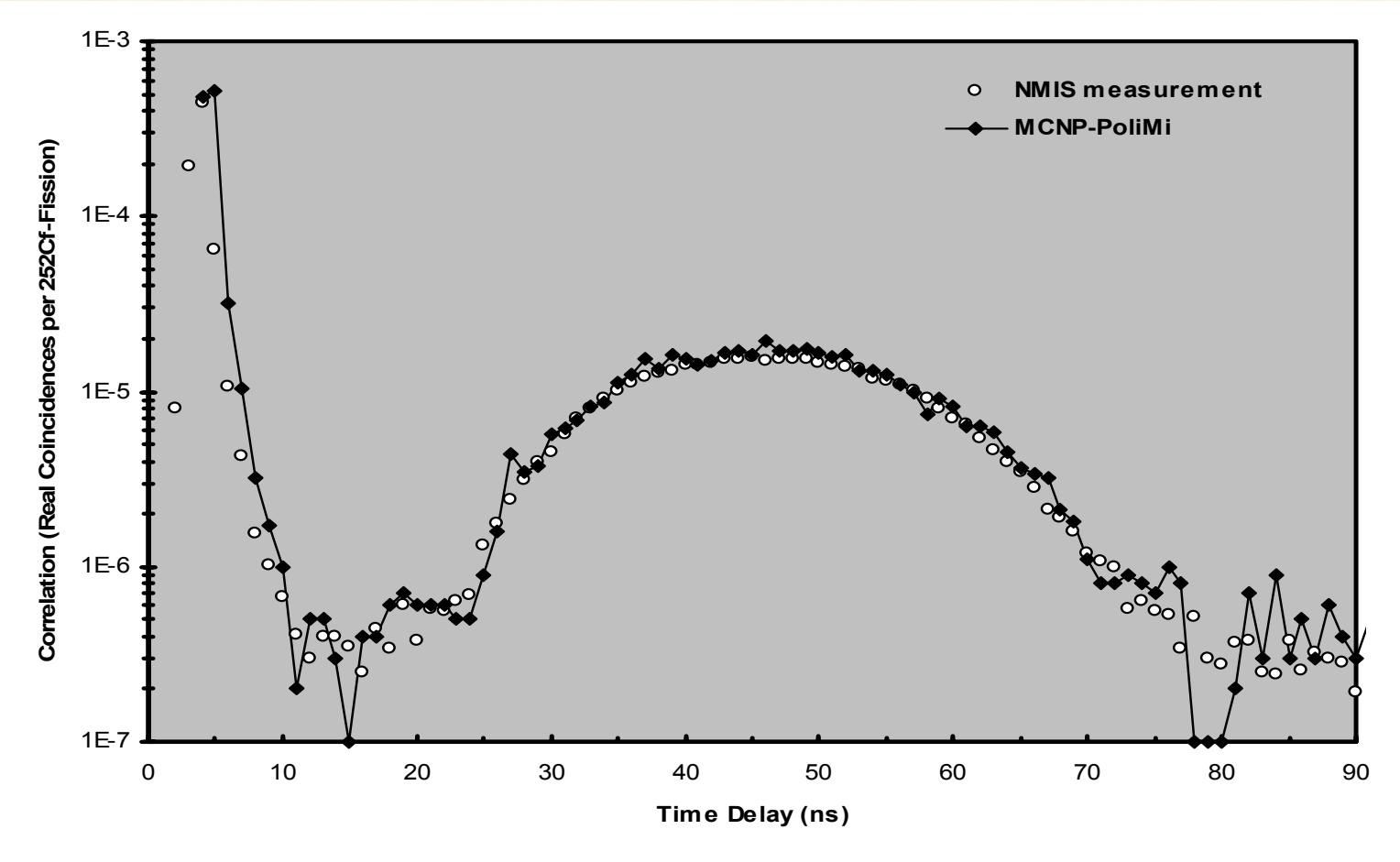
Post-Processing Code for MCNP-PoliMi Collision Output File



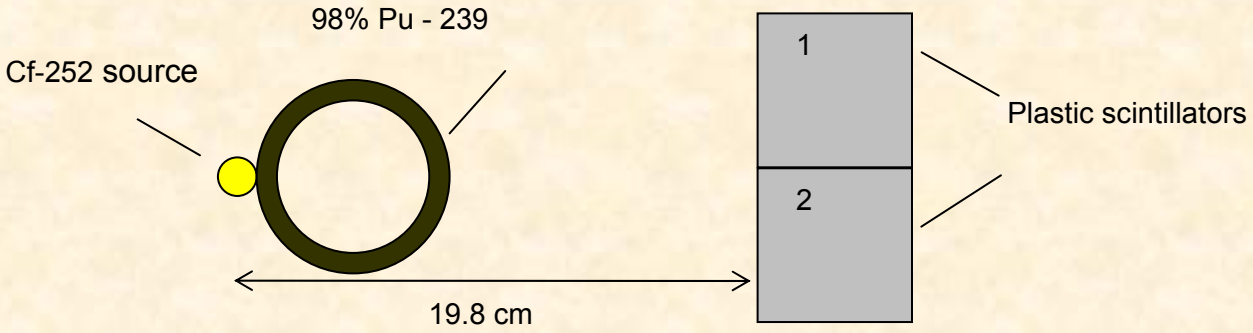
Measured Light Output for Liquid Scintillator



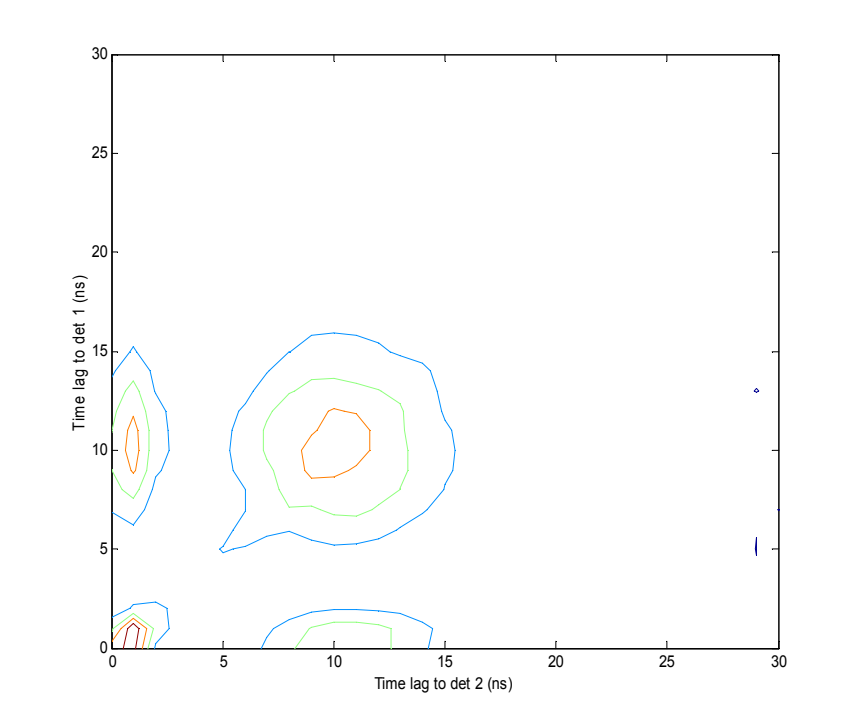
Collision Output File From MCNP-PoliMi (excerpt)



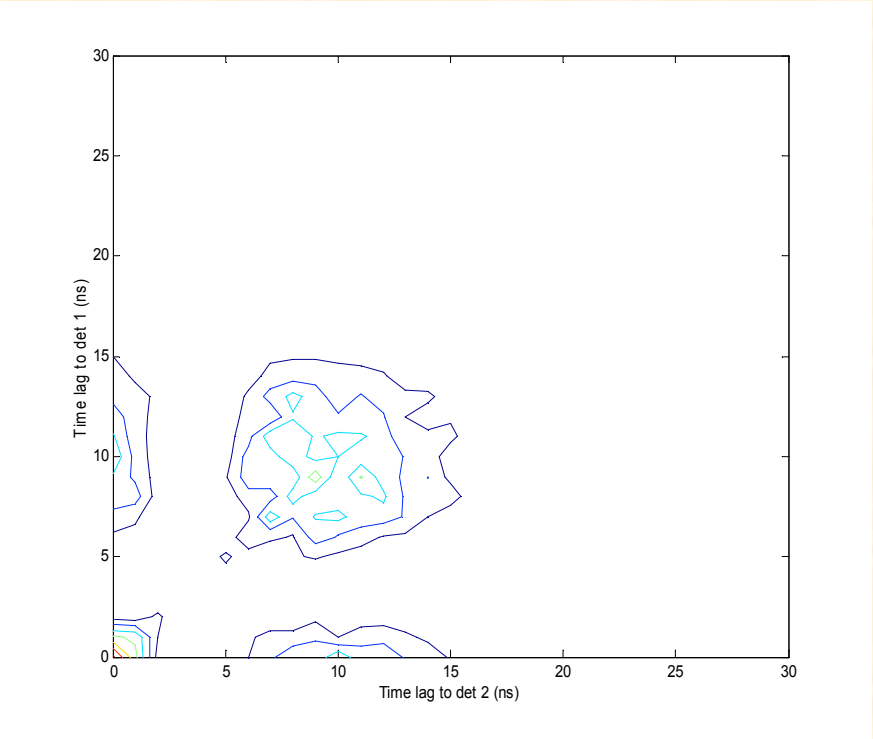
NMIS Experiment With Plutonium Metal Shell (4.0 kg)



Comparison of NMIS Experiment and MCNP-PoliMi Simulation

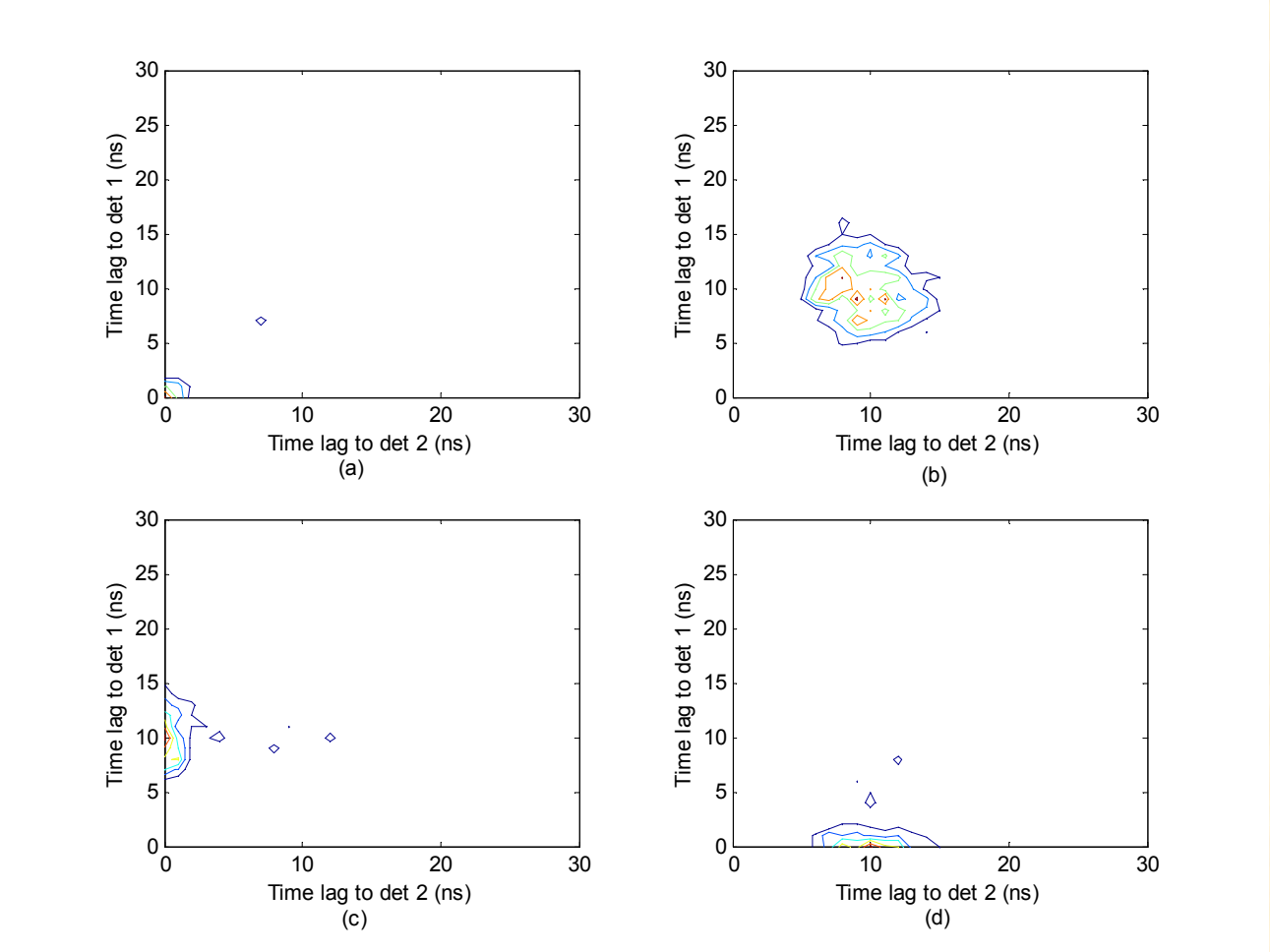


NMIS experiment – note 1 ns detector misalignment



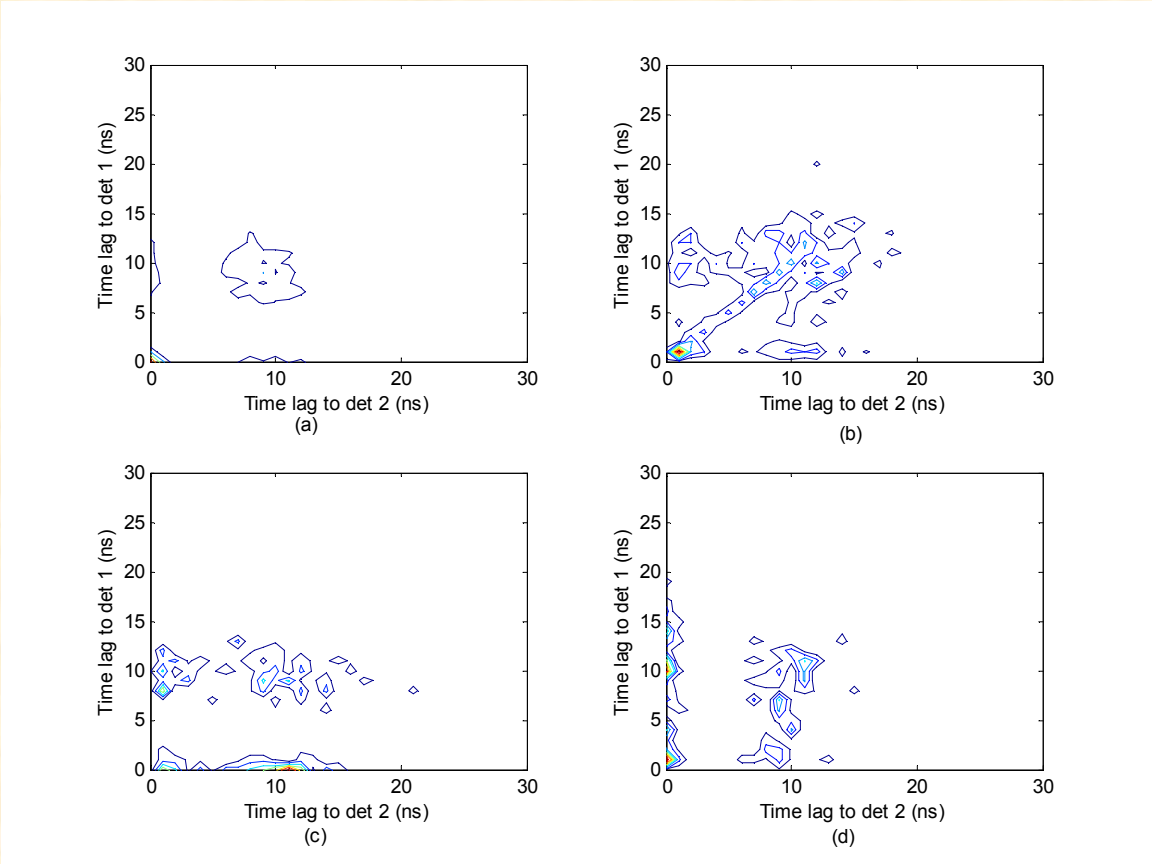
MCNP-PoliMi simulation

Simulation With Pu Metal Shell



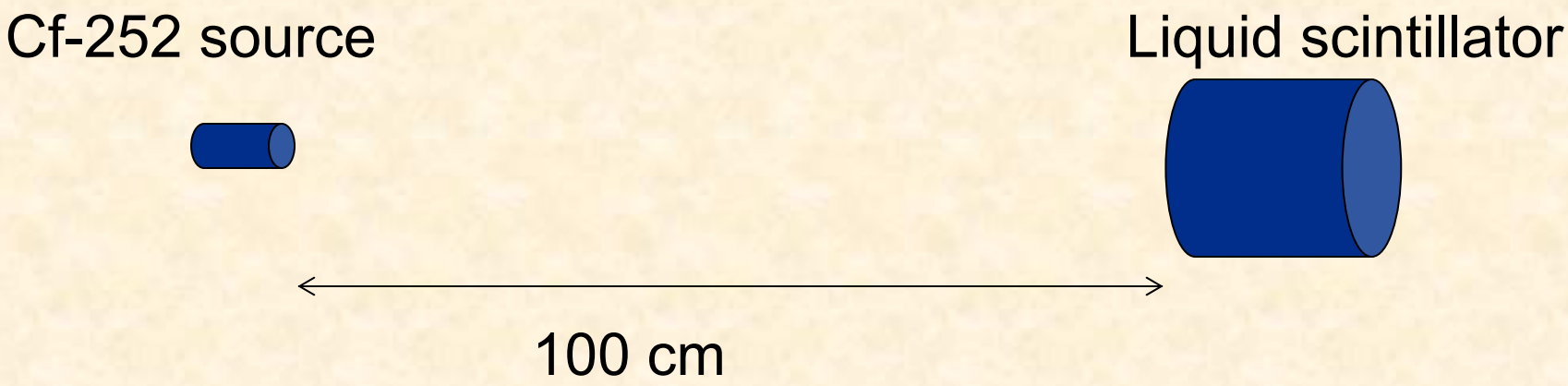
(a) photon – photon pairs, (b) neutron – neutron pairs, (c) neutron – photon pairs, and (d) photon – neutron pairs.

$R(\tau_1, \tau_2)$ Subdivided Into Generation Zero and Induced Fission Particles

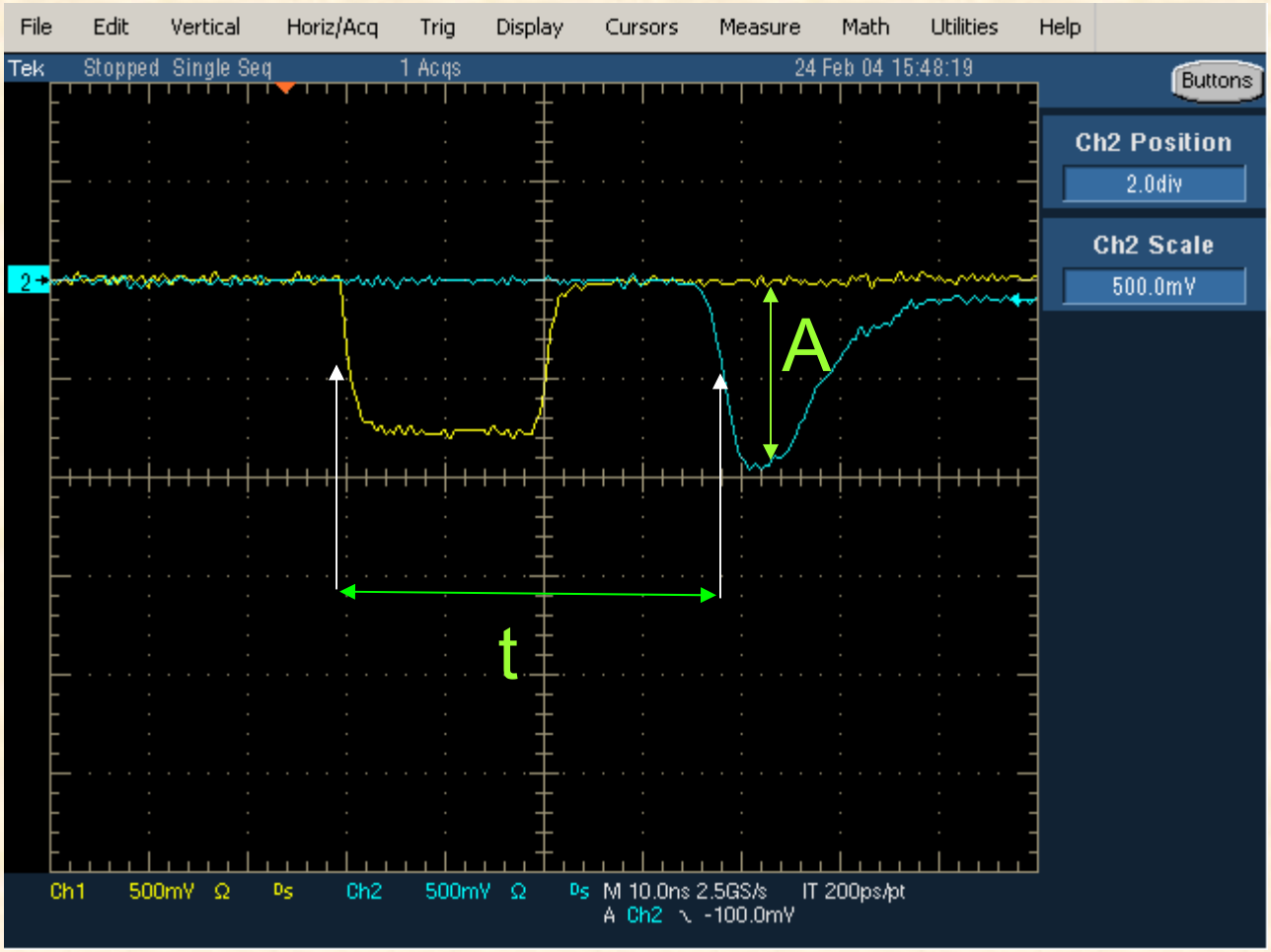


Simulation of source-detectors bicovariance with plutonium shell:
(a) generation zero – generation zero pairs, (b) induced fission – induced fission pairs, (c) generation zero – induced fission pairs, and (d) induced fission – generation zero pairs.

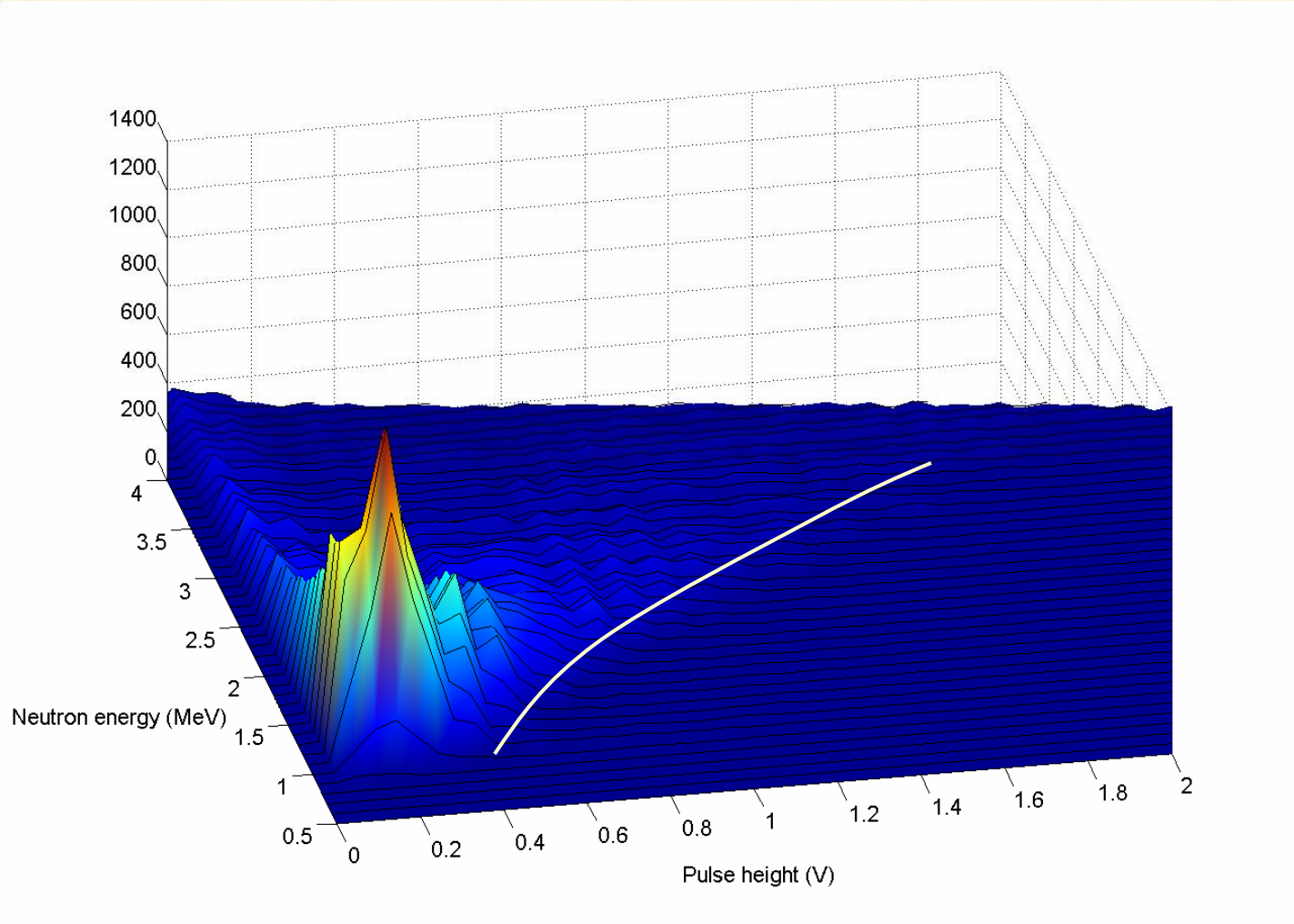
Measurement of Neutron Response Matrix of Liquid Scintillator



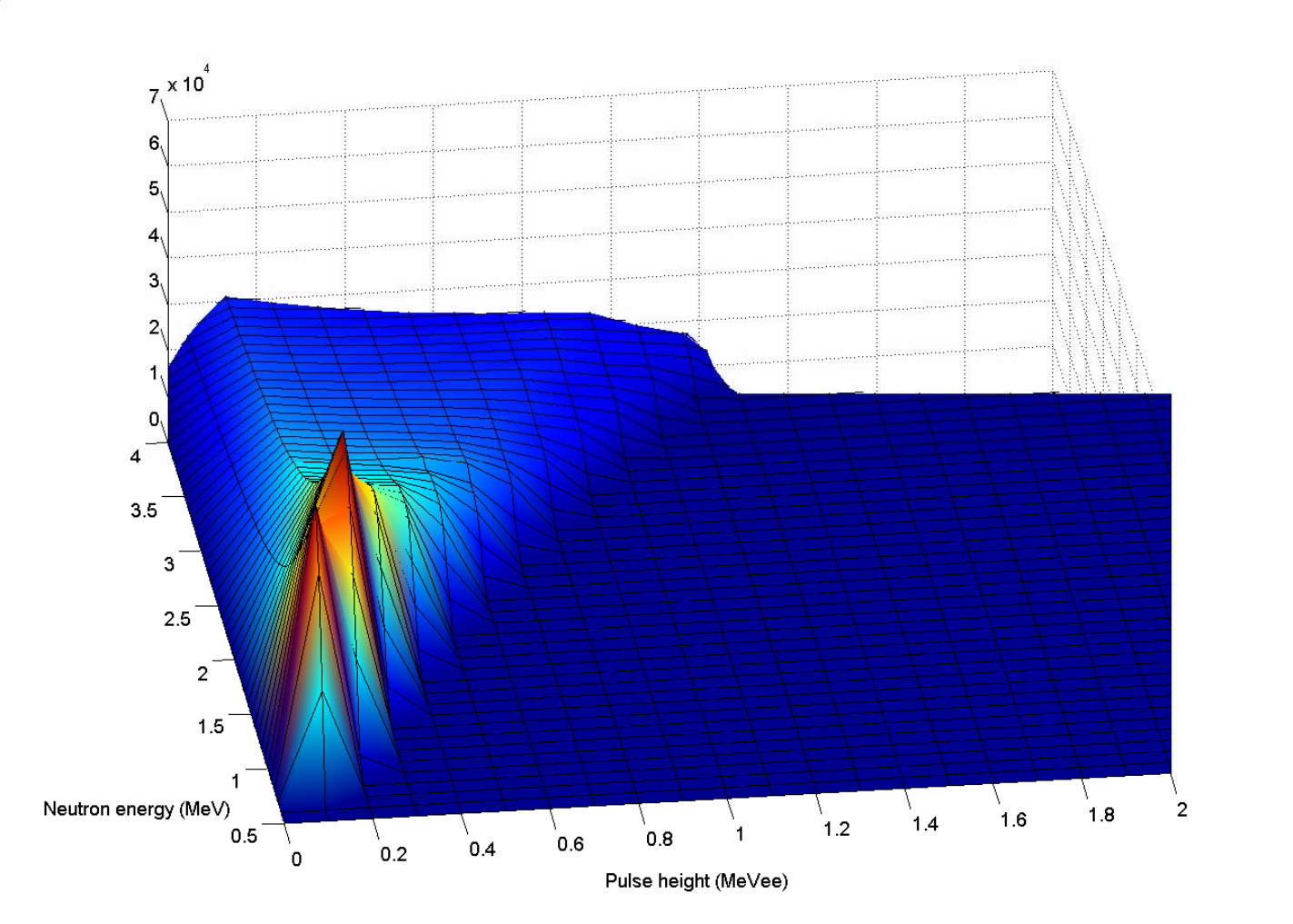
Response Matrix Measurement



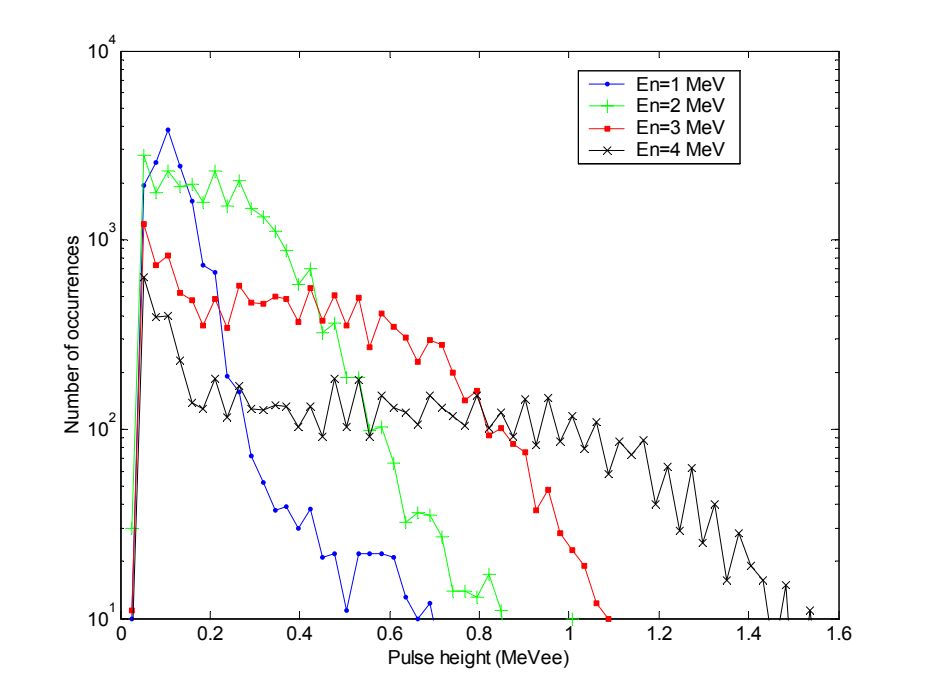
Measured Response Matrix

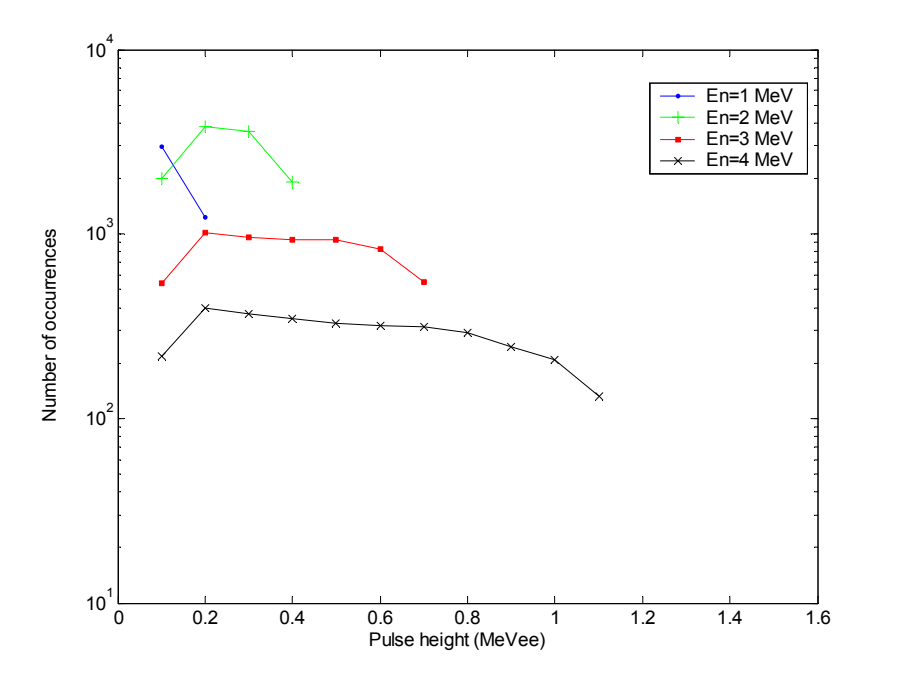


Simulated Response Matrix



Simulated and Measured Response En=1 to 4 MeV





Conclusions

- MCNP-PoliMi: a modification of MCNP-4C, originally developed to simulate correlation measurements
- Also used to simulate detector response
- Monte Carlo output is in the form of a table, containing information on particle collisions in the detector cells

Acknowledgments

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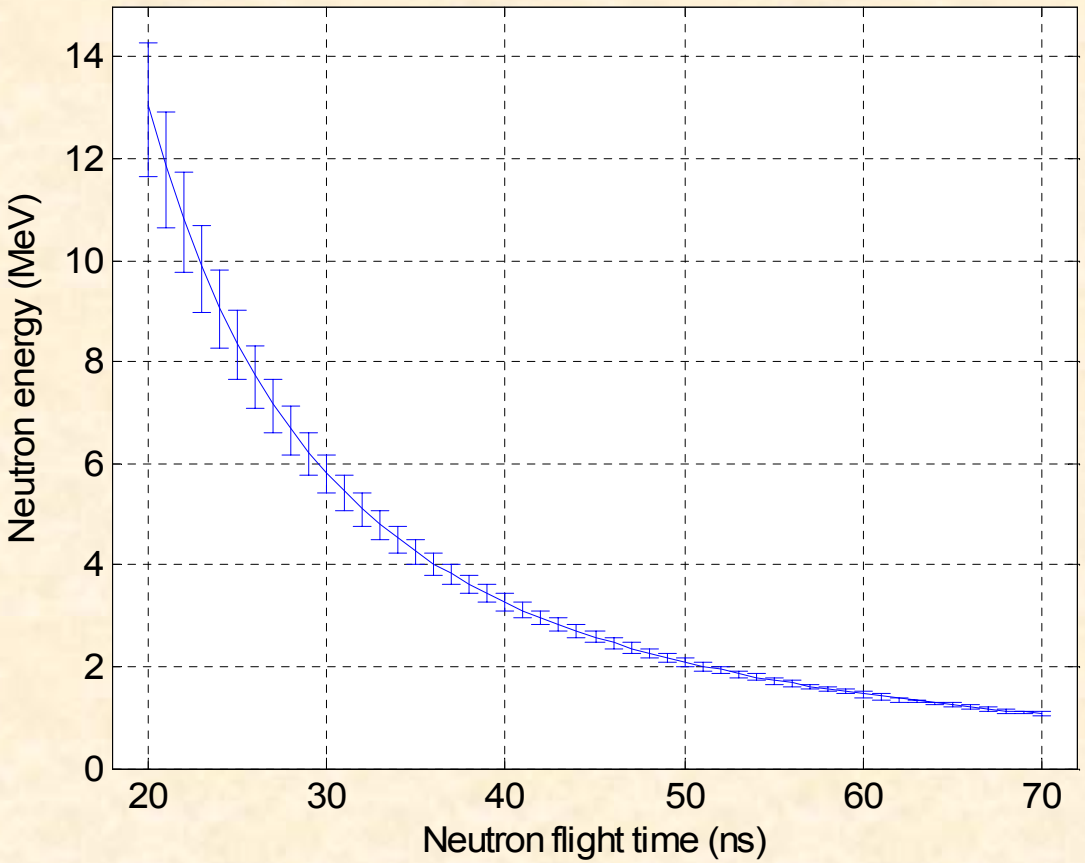
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