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LINAC-BASED TRANSURANIC WASTE CHARACTERIZATION SYSTEM

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Abstract

Remote-handled transuranic nuclear waste poses a particular challenge for assaying due to the high neutron and γ-ray background that emanates from the non-fissile, but highly radioactive material, contained within the waste. The utilization of an RFQ linac with a neutron flux of the order of 10^{10} n/s/4π has shown that, in principle, the differential die-away technique can reliably assay this special class of nuclear waste.

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Introduction

Much work (Ref. 1 and references therein) has been accomplished and documented in the area of nondestructive characterization of contact-handled transuranic (CH-TRU) radioactive waste. The accepted neutron methods for CH-TRU waste assay are a) the differential dieaway technique (DDT)[1-3] which is based on the stimulation of fissile material with thermal neutrons and the subsequent detection of fast neutrons emitted from fission, b) the detection of delayed fission neutrons emitted after the capture of thermal neutrons by the fissile material (Cf shuffler)[4], and c) detection of coincidence fission neutrons from the spontaneous decay of specific even-numbered actinide radionuclides.[5] Fissile material fissions with absorption of thermal neutrons. CH-TRU waste, as defined by DOE Order 5820.2A, is waste possessing external dose rates, at the surface of the container, of $\leq$2 mSv/h, specific activities of TRU radionuclides >3700 Bq/g, half-lives >20 years, and Z >92. The above methods have been shown to successfully assay CH-TRU waste providing the passive neutron emission rate produced from (α,n) reactions and spontaneous fission events is $<10^4$ n/s.

Remote-handled transuranic (RH-TRU) waste, as defined by DOE Order 5820.2A, is waste possessing external dose rates, at the surface of the container, >2 mSv/h, specific activities of TRU radionuclides greater than 3700 Bq/g, half-lives >20 years, and Z

>92. In many cases, the passive neutron background is $>10^4$ n/s, incapacitating the Cf shuffler and coincidence counting techniques and severely limiting the DDT method. Because of these difficulties, less work[6] has addressed the assay of RH-TRU radioactive waste.

Present DDT systems use sealed-tube neutron generators commercially available in the U.S. and other countries and provide a neutron flux up to 10^8 n/s/4π. This neutron flux is at least a factor of 10 less than what is required to assay RH-TRU waste possessing neutron emission rates ranging from 10^6 to 10^8 n/s. Consequently, it is desirable to devise a system capable of producing a sufficient interrogating neutron flux and able to detect fission neutrons above a significant neutron (and gamma-ray) background. In the following sections, the principle of the DDT method will be described and the results of the utilization of a radiofrequency quadrupole (RFQ) linear accelerator for RH-TRU waste assay will be presented.

Principles of the DDT Assay Method

The DDT assay utilizes a pulsed-neutron source to interrogate fissile material contained in radioactive waste. The neutrons produced by a pulsed-neutron source are thermalized by various interactions including those produced by neutron moderating material located in the vicinity of the accelerator target and

moderating materials present in the waste. After cessation of the pulse, the thermalized neutron flux thus produced interacts with fissile nuclides such as ^{239}Pu and ^{235}U to produce a spectrum of fast-fission neutrons. These prompt-fission neutrons are detected by proportional counters surrounded by cadmium that absorb the interrogating thermal neutron flux (1.7 mm cadmium attenuates thermal neutrons by a factor of 10^6). A fraction of the fast-fission neutrons are slowed to thermal energies by polyethylene contained within the detector packages. ^3He proportional counters within the moderated detector packages respond sensitively to thermal neutrons.

The decay-time spectra of the interrogating and fission neutrons consist of three components (see Figure 1): the interrogating neutron pulse with a decay time characteristic of the fast-neutron energy loss (Region A), the fission neutrons with a decay time characteristic of the build up of the thermal neutron flux (Region B), and an essentially constant background resulting from passive neutron emission (Region C).

After corrections are applied for neutron moderator and absorber effects for both the interrogating thermal neutron flux and the fast-fission neutrons, and normalizing with respect to the interrogating flux, the difference in slopes of the background and fissile spectra is proportional to the quantity of fissile material present in the waste.

Experimental

The pulsed-neutron source used in this study was a RFQ linear accelerator.[7] The RFQ is a focusing-accelerating structure (Figure 2), composed of a radiofrequency cavity with a quadrupole structure running down the total length of the cavity (Ref. 8 and references therein). Each of the four vanes of the quadrupole structure has a ripple built on its edge. This ripple is cut successively deeper and longer towards the exit of the RFQ, introducing gradually an axial component of the electric field that accelerates the beam. The quadrupole structure therefore plays a dual role: via the ripple it accelerates the beam and through the quadrupole structure it bunches and focuses the beam (Figure 3). The bunching process is performed very efficiently at the RFQ. Other linacs usually discard 30-50% of the beam. In an RFQ, 80-90% of the incident beam is accelerated through it.

The RFQ utilized for this experiment accelerates a deuteron particle beam to a maximum energy of 900 keV. A beryllium target was used, producing via the $^{9}Be(d,n)^{10}B$ reaction approximately 10^{10} n/s with an average neutron energy of 3.5 MeV. The ion source was pulsed at frequencies ranging from 100 Hz to 400 Hz with neutron beam pulse widths ranging from 10 μs to 40 μs.

The machine was made available to ORNL by the Federal Aviation Administration for use in these experiments. It was installed at

the Lawrence Livermore National Laboratory's (LLNL) Center for Atomic Mass Spectrometry where the experiments were conducted.

The neutron detector package consisted of three ^{3}He proportional counters of dimensions 2.5 cm diameter by 61 cm length each filled with 4-atm of ^{3}He (see Figure 4). The ^{3}He tubes were encased inside 2.5 cm polyethylene to slow a fraction of the fast-fission neutrons to thermal energies. In addition, the package was surrounded by 1.7 mm of cadmium to absorb thermal neutrons from the interrogating neutron pulses. The detector package was enclosed in a 2.5-cm polyethylene box to initiate fast-neutron thermalization. Thirty six cm of lithiated paraffin were used to shield the neutron detector package from the RFQ neutron beam. The signals from the proportional counter package were properly pre-amplified and amplified with a pulse-shaping time of 0.5 μs. Because the ^{3}He proportional counters are also sensitive to gamma rays appearing as pulses of amplitude smaller than the neutron amplitude, the amplified signals were passed through a discriminator set to cut off the gamma-ray signals from the amplified spectrum. The conditioned signals were sent to a PC-based Canberra AccuSpec™ multi-channel scaler (MCS) board. The data acquisition was triggered to acquire at the end of the interrogating neutron pulse. Data were acquired with a dwell time of 10μs in 500 channels, corresponding to 5 ms of data acquisition. Data were accumulated for 50,000 sweeps to ensure adequate counting statistics.

The materials to be assayed were placed in the center of a 208-liter drum (see Figure 4) lined with 2.5-cm thick polyethylene slabs. The detector package was placed in a fixed position against the drum, while the distance between the drum and the RFQ target was varied. The neutron flux produced by the RFQ was monitored using a combination lithium-glass/liquid scintillator[9] detector. The lithium glass was used to monitor the thermal neutron flux and the liquid scintillator the fast-neutron flux.

To increase the thermal neutron flux in the middle of the drum, a polyethylene moderator was placed around the RFQ target. The arrangement used created a two-fold increase in the thermal neutron flux in the middle of the drum as compared to no moderator assembly.

The materials assayed were a) two pieces of uranium (containing 0.3% ^{235}U), total U mass of 6.3 kg, b) PuBe containing 17.7 g of ^{239}Pu yielding 2.3×10^6 n/s, and c) 18.4 μg of ^{252}Cf yielding 4.8×10^7 n/s and 10^3 γ/s. The U does not produce any appreciable neutron background and was used to confirm that the RFQ could duplicate previous DDT measurements using standard, sealed-tube neutron generators. The PuBe material served a two-fold purpose: it provided a source of fissile material and also produced a significant background neutron flux of approximately 10^6 n/s. The ^{252}Cf source simulated a high neutron and gamma-ray background not associated with fissile material.

Results

In order to duplicate previous DDT measurements using sealed tube neutron generators, the U samples were assayed. The drum containing the U samples was placed 188 cm from the RFQ target. Background spectra were acquired without any assay material in the drum followed with assay spectra of 3.1 kg U, and 6.3 kg U. Figure 5 shows the results of the measurements. In the region between 400 μs and 1.4 ms, one can observe substantial differences between the 3.1 kg and 6.3 kg U spectra (solid and dashed lines, respectively) and the background spectrum (dotted line). These differences are due to the fast neutrons emitted from the induced fissions of the fissile material. The RFQ pulse width was varied between 10 μs and 35 μs and it was found that the optimum neutron pulse width was 25 μs yielding the largest difference between the background and fissile material spectra. To ascertain that the difference observed in the region of interest was indeed due to the presence of fissile material in the drum, the U in the drum was replaced with a 7 kg piece of stainless steel. Figure 6 shows that the Fe spectrum is identical to the background spectrum clearly identifying the neutrons in the 400 μs to 1.4 ms range as due to the presence of fissile material in the drum. Although in this series of experiments we did not attempt to quantitatively assay the fissile material, one can observe in Figure 5 that there exists a substantial yield difference between the 3.1 kg U and 6.3 kg U

spectra.

Having established that the RFQ duplicated the DDT measurements obtained using sealed tube neutron generators, the U was replaced with the PuBe material which provided both the fissile material and the high neutron background simulating RH-TRU waste assay conditions. Measurements taken with the drum at 188 cm and 125 cm from the RFQ target, indicated that the interrogating neutron flux of the order of 10^8 n/s was not sufficient to differentiate the fissile material in the presence of the 10^6 n/s neutron background. Subsequent measurements obtained at a distance of 22 cm, where the interrogating neutron flux is of the order of 10^9 n/s, indicate that one can observe the fissile material in the presence of a high neutron background. As seen in Figure 7, looking again in the region between 400 μs and 1.4 ms there exists a substantial difference between the background and the Pu spectra.

To find whether one can detect fissile material in the presence of a high gamma-ray field, the ^{252}Cf source was placed in the drum along with the U samples. Figure 8 shows the results of the investigation with the drum placed 22 cm from the RFQ target. The two U spectra show again a discernable difference from the background spectrum taken with the ^{252}Cf source in place.

Conclusions

From the results of the present investigation, one may conclude that by using the DDT method, it is possible to ascertain the existence of fissile material in the presence of a high background (>10^4n/s and >10 mSv/h gamma) if the interrogating neutron flux is > 10^9 n/s/4π. These preliminary measurements did not examine any matrix effects which can affect fissile assay measurements. The presence of various materials in the waste drum can adversely influence both the interrogating thermal neutron and the fast-fission neutron fluxes. Neither did it attempt to establish lower limits of detection of fissile materials.

Acknowledgements

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Figure Captions

Fig.1 DDT decay-time spectra: Region A corresponds to the decay time spectrum of the fast neutrons, Region B to the decay time spectrum of the fission neutrons, and Region C is the passive neutron emission background.

Fig.2 RFQ accelerator's four-vane structure.

Fig.3 RFQ beam characteristics: the ripple cut on each quadrupole vane accelerates the beam, and the quadrupole structure bunches and focuses the beam.

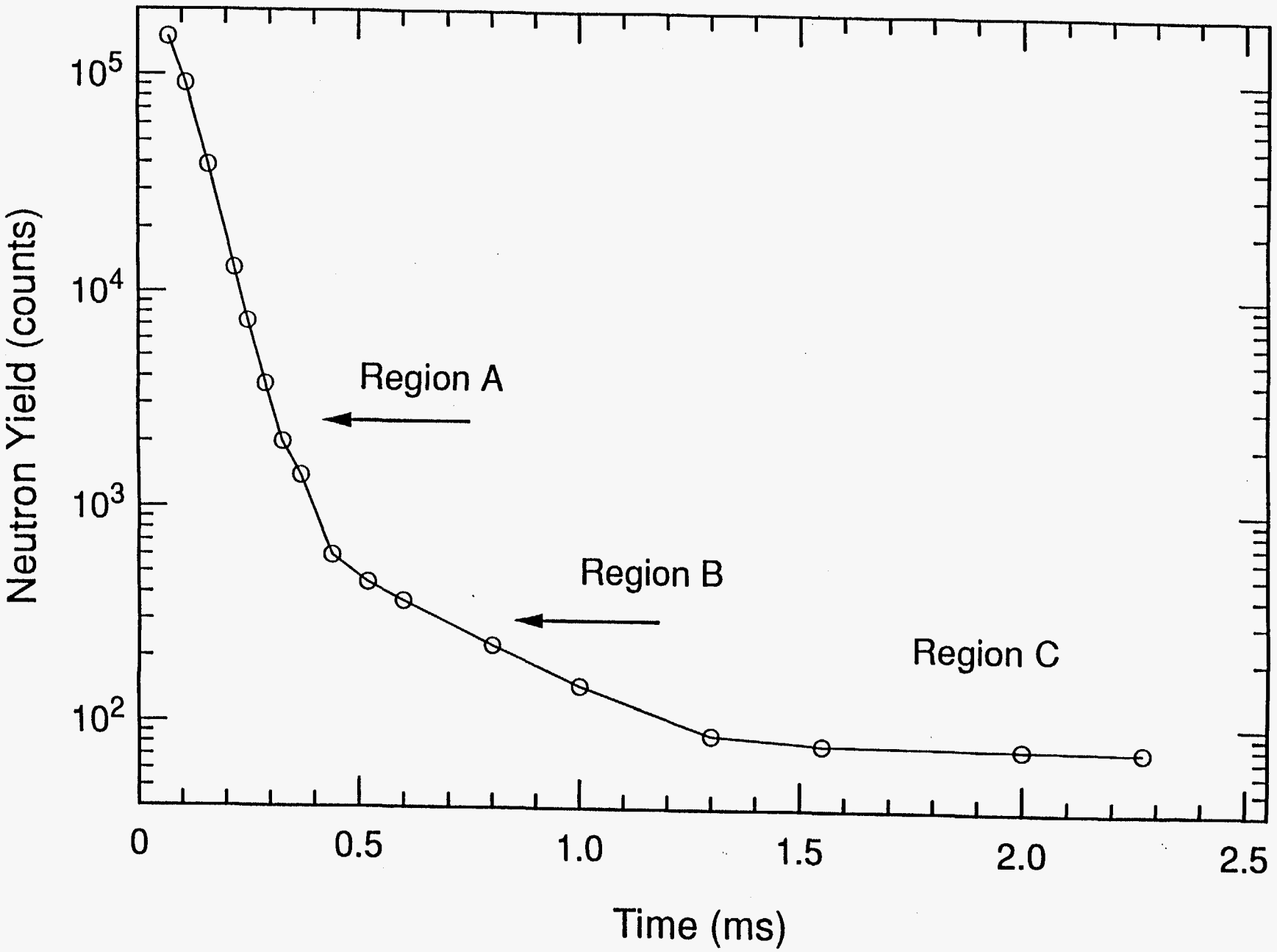
Fig.4 Experimental layout (not to scale).

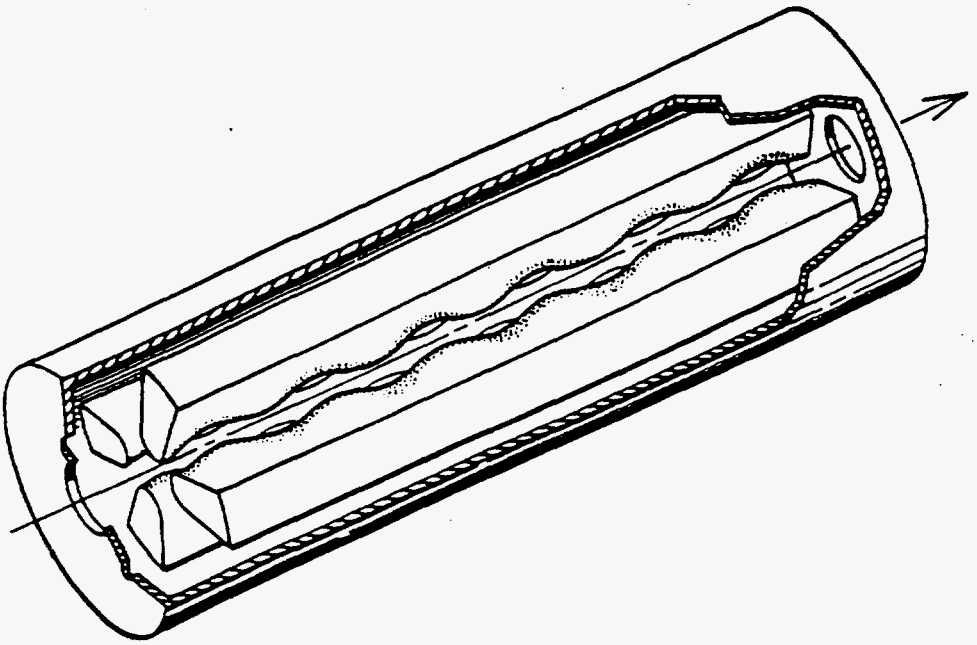
Fig.5 Neutron time spectra of 3.1 kg U (solid line), 6.3 kg U (dashed line) and background (dotted line).

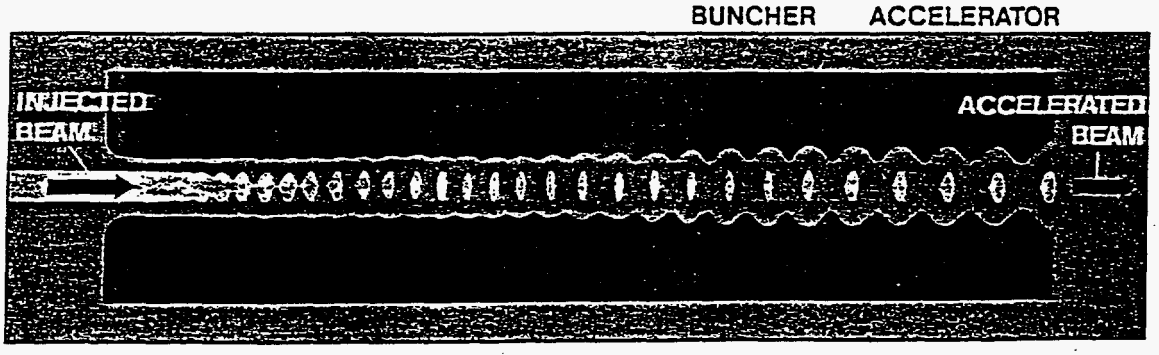
Fig.6 Neutron time spectra of 7 kg Fe (solid line) and of background (dotted line).

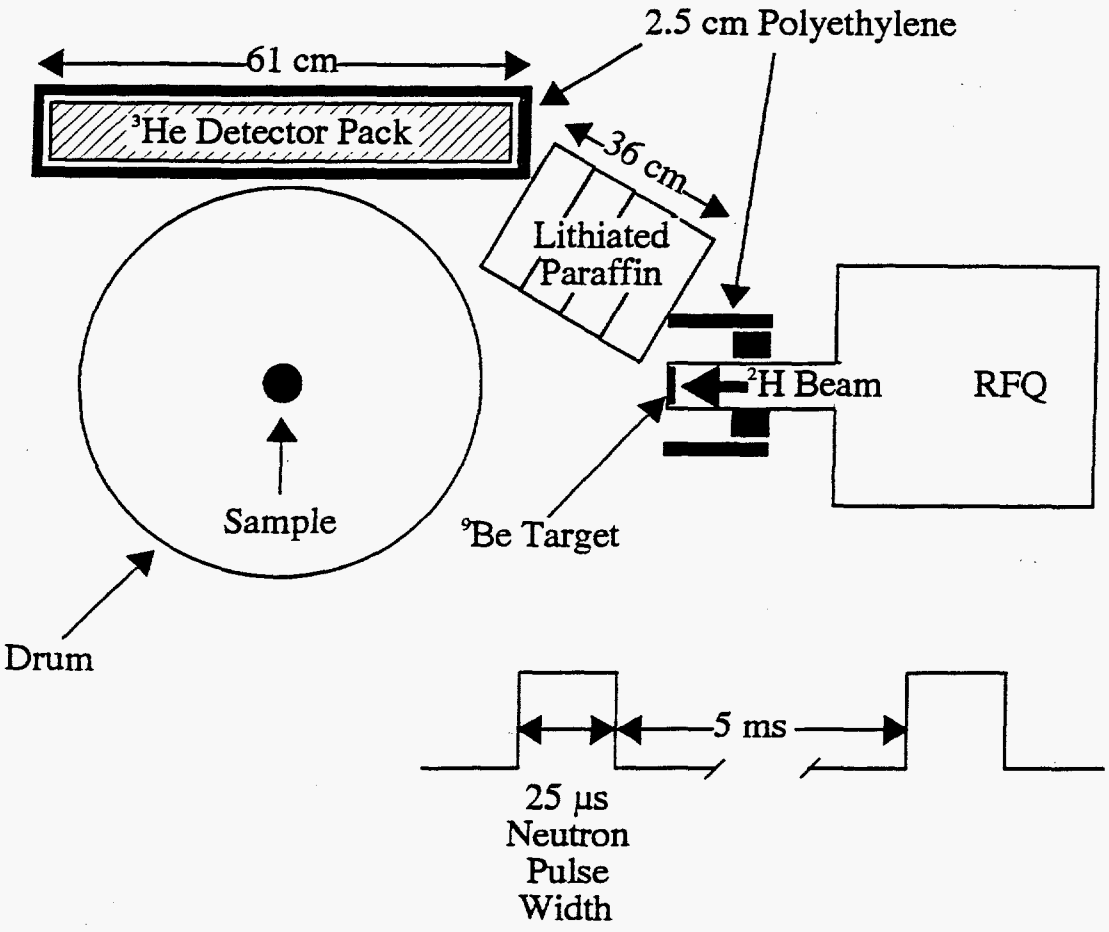
Fig.7 Neutron time spectra of 17.7 g PuBe (solid line) and of background (dotted line). Background was offset to aid comparison of the spectra in the 0.4 ms to 1.5 ms region.

Fig.8 Neutron time spectra of 3.1 kg U (solid line), 6.3 kg U (dashed line), and of background (dotted line) in the presence of 18.4 μg ^{252}Cf.


Neutron Yield (counts)
10^5
10^4
10^3
10^2
Region A
Region B
Region C
0
0.5
1.0
1.5
2.0
2.5
Time (ms)

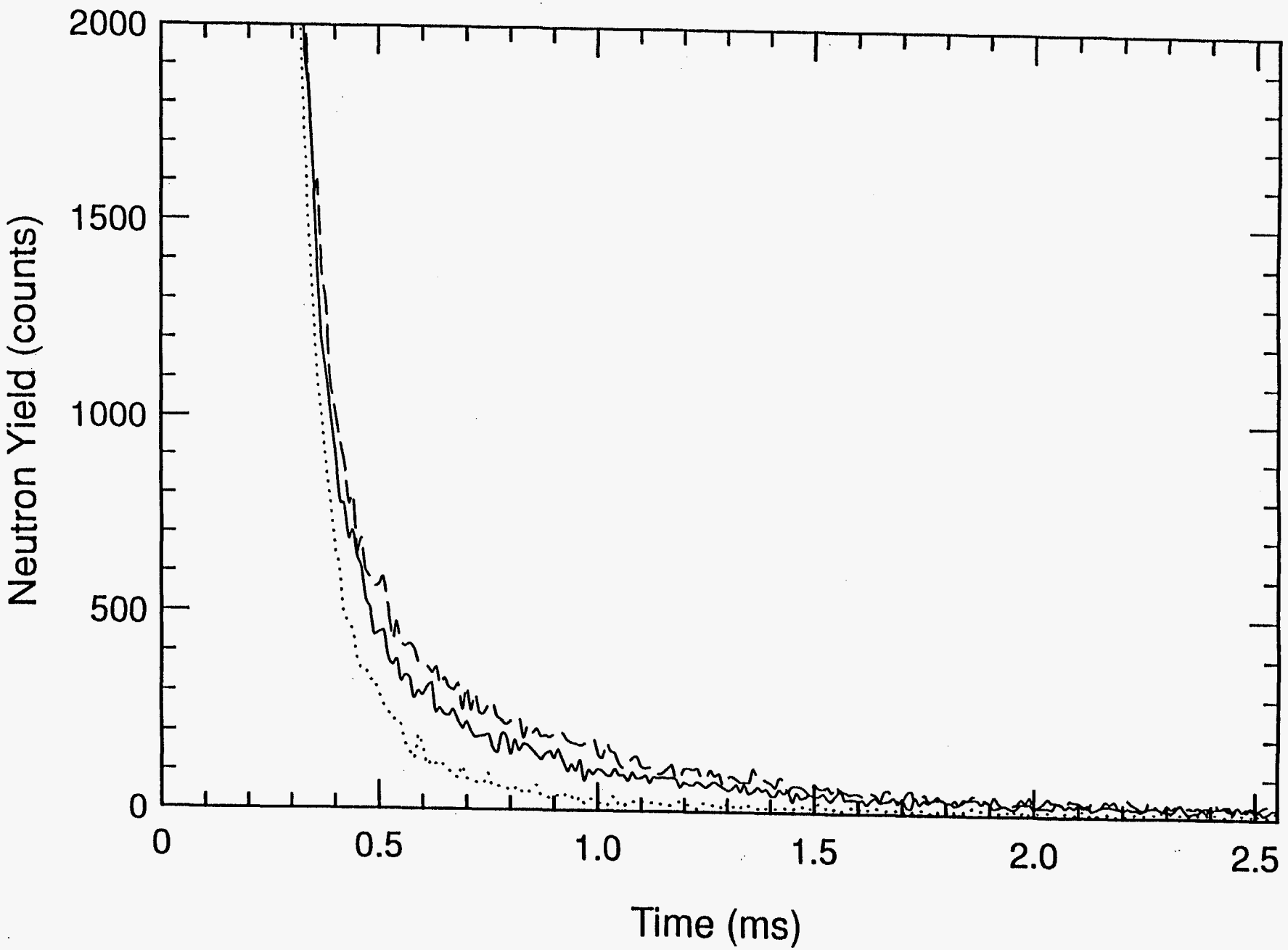



BUNCHER
ACCELERATOR
INJECTED
BEAM
ACCELERATED
BEAM



Train of Neutron Pulses

Fig. 11


2000
1500
1000
500
0
Neutron Yield (counts)
0
0.5
1.0
1.5
2.0
2.5
Time (ms)

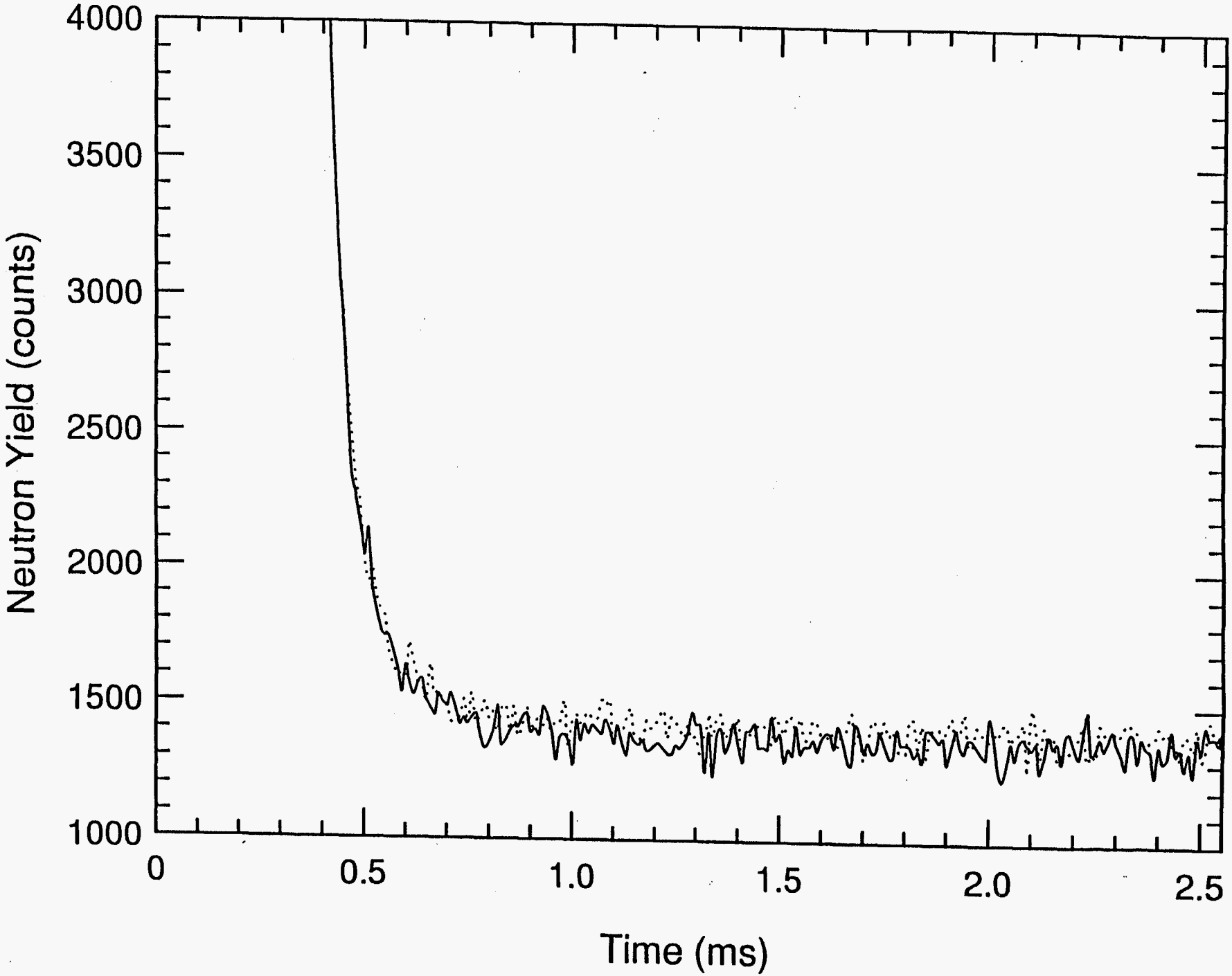

4000
3500
3000
2500
2000
1500
1000
Neutron Yield (counts)
0
0.5
1.0
1.5
2.0
2.5
Time (ms)

Fig. 6

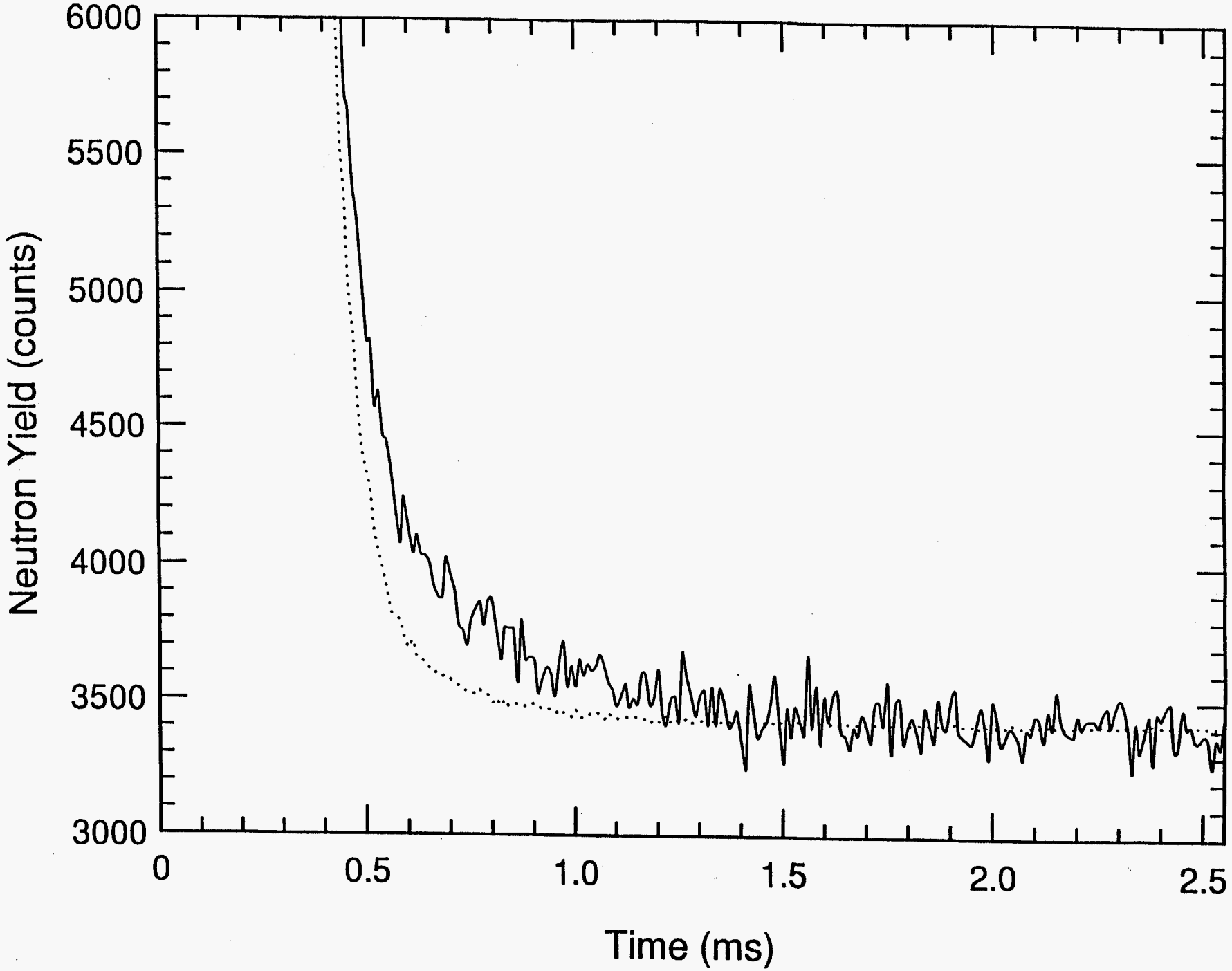

6000
5500
5000
4500
4000
3500
3000
Neutron Yield (counts)
0
0.5
1.0
1.5
2.0
2.5
Time (ms)

Fig. 7

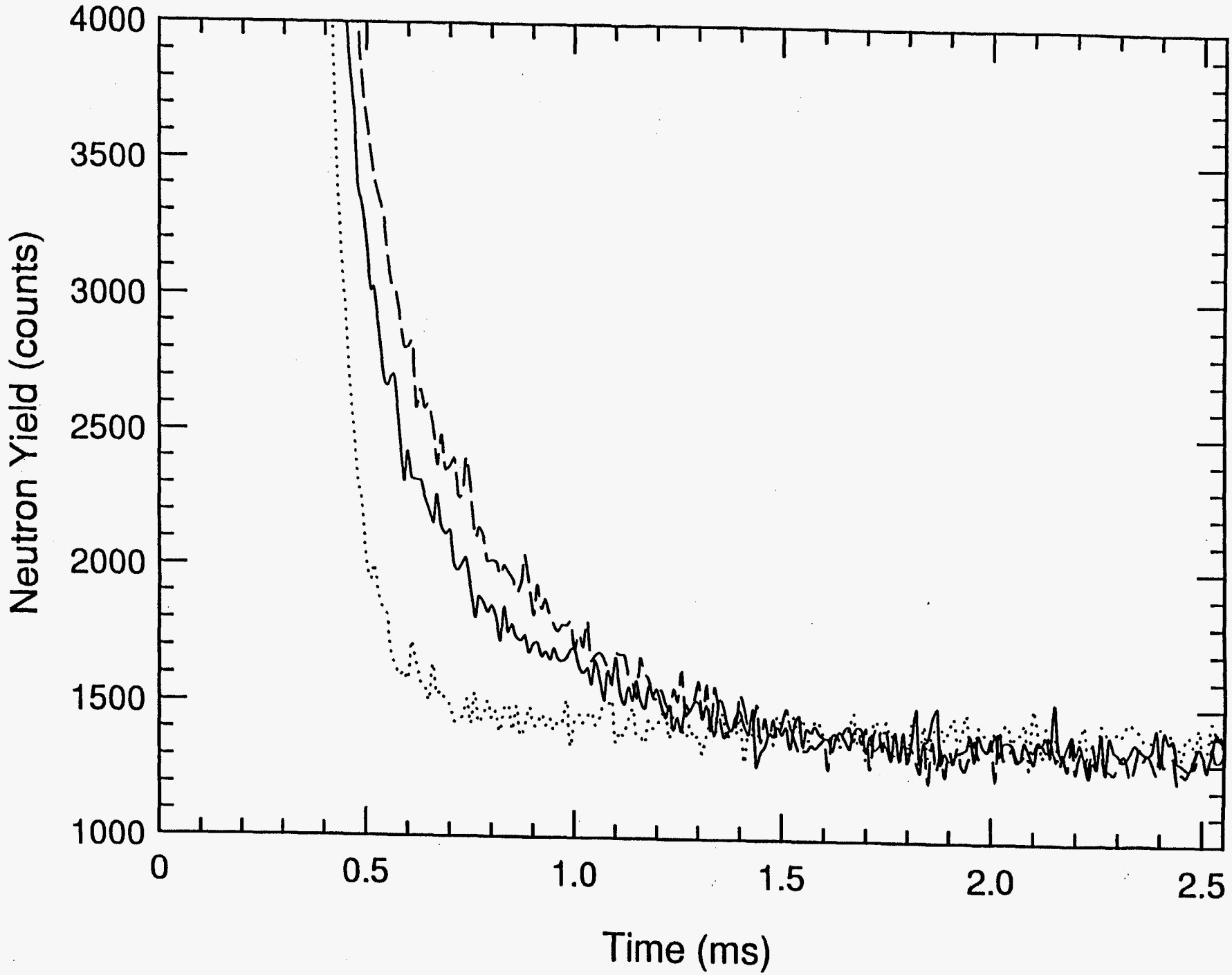

4000
3500
3000
2500
2000
1500
1000
Neutron Yield (counts)
0
0.5
1.0
1.5
2.0
2.5
Time (ms)

Fig. 8

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