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Title: Active Interrogation of Highly Enriched Uranium

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Active interrogation of highly enriched uranium

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Abstract:
Active interrogation techniques provide reliable detection of highly enriched uranium (HEU) even when passive detection is difficult. We use 50-Hz pulsed beams of bremsstrahlung photons from a 10-MeV linac or 14-MeV neutrons from a neutron generator for interrogation, thus activating the HEU. Detection of neutrons between pulses is a positive indicator of the presence of fissionable material. We detect the neutrons with three neutron detector designs based on ^{3}He tubes. This report shows examples of the responses in these three detectors, for unshielded and shielded kilogram quantities of HEU, in containers as large as cargo containers.

Introduction

Passive detection of highly enriched uranium (HEU) containing approximately 90% ^{235}U is often difficult. The strongest gamma rays from ^{235}U at 144, 186, and 205 keV, as well as the x rays, are easily attenuated by a few millimeters of lead or its equivalent. The neutron emission rate of ^{235}U of 3×10^{-4} n·s^{-1}·g^{-1} is too low to be useful. The next most abundant isotope in HEU, ^{238}U, has a very weak gamma ray at 111 keV, which is also easily attenuated. However, ^{238}U decays to ^{234}Th (24.1 days half-life), which subsequently decays to ^{234m}Pa (1.17 m half-life). If processing of the HEU was not recent and sufficient ^{234}Th has built up, the 766- and 1001-keV gamma rays from ^{234m}Pa may be detectable. However, these gamma rays are also present in background. HEU often contains ^{232}U, which produces a 2.6-MeV gamma ray following the decay to ^{208}Tl. However, this signature is not reliable because ^{232}U is produced in a reactor and not all HEU has been in a reactor.

Active interrogation techniques are used to activate HEU and thus allow its detection. General discussions of active interrogation techniques are available in the literature.[1-3] Our technique uses 50-Hz pulsed beams of photons or neutrons for the interrogation to produce fissions in the HEU. A small fraction of the resulting fission products emit delayed neutrons. Some of these neutrons can cause additional fissions and produce more neutrons. We detect the neutrons between the interrogating pulses. If the neutron detection probability is adequate, or if the HEU system has a high neutron multiplication, two or more neutrons can be detected from a single fission event. The observation of neutrons between beam pulses is a positive indicator of the presence of fissionable material. Besides HEU, other material such as depleted uranium, natural uranium, thorium, and neptunium can also produce a positive neutron signal between pulses. The half-lives of the fission products range from less than a second to more than several minutes. When interrogation is initiated, the number of fission products begins to build up and approaches saturation after approximately one minute. Operating at 50 Hz allows us to detect the neutrons from the short-lived fission products as well as from the longer half-live fission products as they build up.

This report presents examples of data obtained with our technique. We generated the interrogating photons with an electron linac and the interrogating neutrons with a small neutron DT generator. We detected the neutrons with three neutron detector designs, all based on polyethylene-moderated ^{3}He tubes: a large area detector for maximum efficiency, an array of modular detectors, and a portable detector. The examples show responses in these three detectors, for unshielded and shielded kilogram quantities of HEU, in containers as large as cargo containers.

Experimental

The interrogating photons pulses were produced by a Linatron 2000 electron linac that had been repackaged into transportable boxes for field deployment.[4] The linac can operate at 6, 8, or 10 MeV and a pulse repetition rate of 20-120 Hz. For the measurement reported here, we operated at 10 MeV with a pulse rate of 50 Hz. The pulse width was approximately 6 μs. With a half radiation length tungsten bremsstrahlung target (1.75 mm thick) at the exit of the linac, the dose rate at 1 meter on axis was approximately 160 R/min. The photon flux was forward peaked with a FWHM < 20° for the high energy portion of the bremsstrahlung photon beam.

The interrogating neutrons were produced by an MF Physics model CC A-210 neutron generator also operated at 50 Hz. The generator uses the reaction T(d,n)^{4}He to produce 14-MeV neutrons. The pulse width was approximately 10 μs, and the output was 10^6 neutrons/pulse or 5×10^7 neutrons/s.

The large area detector was a hollow high-density polyethylene box (122 cm × 244 cm × 18 cm) containing ^{3}He tubes. The box contained four ^{3}He tubes, 5.08 cm diameter × 182.88 cm length, pressurized to 2 atm. The polyethylene was 1.27 cm thick on the front facing the incident neutrons and 5.08 cm on the back. The detector was completely covered with 0.17-cm thick cadmium to prevent low-energy background neutrons from entering the detector. The detector's efficiency for measuring neutrons from a ^{252}Cf source placed at a distance of 1 meter and centered from the detector front face was approximately 10%.

The array of detectors consists of modules for maximum flexibility in configuring a system. Each module is 13 cm × 25 cm × 117 cm and has a mass of 33 kg (Fig. 1). Each module contains three ^{3}He tubes pressurized to 2 atm, and the intrinsic efficiency is 27%. Each module is completely shielded, including the ends, by 0.81 cm of cadmium. We have 16 modules, but for the results reported here we used a stack of five modules.

The portable detector system was designed for maximum efficiency in a small system that could be easily deployed [5]. The detector measures 10 cm × 43 cm × 51 cm and has a mass of 25 kg. It contains fifteen ^{3}He tubes, 2.54 cm (diameter) × 38.1 cm (length), pressurized to 10 atm and inserted into holes in a polyethylene block (Fig. 2). The polyethylene block, including the top except for the connectors, is completely covered by 0.81 mm of cadmium. The intrinsic efficiency is 20%. The absolute efficiency is lower than the efficiency of the other two systems because of its limited size. A compact electronics package (not shown), originally developed for passive measurements, is mounted on top of the polyethylene block for data acquisition and analysis. We used external electronics for the active measurements, but intend to use the internal electronics after some modifications are made.

The systems used custom electronics for processing the signals. For the large area detector and the array of modular detectors, the combined signal from the ^{3}He tubes was fed into the preamplifier amplifier discriminator emitter coupled logic module (PADEM) and pulse arrival-time recording module (PATRM).[6] The PADEM produces a logic pulse for all signals above a threshold. The PATRM provides a time stamp for each event. The data from the detectors were gated off for ~2 ms during and after the interrogating pulse to allow the detector to recover from saturation and for all neutrons produced during the pulse to exit the detector. The data were stored event by event in the PATRM internal memory of one million 32-bit data words. At the end of each measurement the date were transferred to an IBM-compatible computer and saved to a hard disk.

The portable detector contained internal electronics that included a preamplifier and discriminator on each ^{3}He tube to accommodate high counting rates. For the portable detector measurements reported here, which used the neutron generator at the interrogating source, the logic pulses from the fifteen tubes were combined into a single output. This output was fed to an ORTEC MCS-pci™ multichannel scaler controlled by a LabView program. The start input pulse for each pass of the multichannel scaler was provided by the neutron generator at the beginning of each interrogating pulse.

The data from the large area detector and the array of modular detectors were analyzed with the Feynman variance method[7] as implemented in a C++ program. This program determines the number of correlated neutrons ("singles," "doubles," triples," etc.) as a function of time after the beam pulse. The number of "singles" is typically two or three orders of magnitude greater the number of "doubles." For the portable detector data, no analysis was performed to determine the number of correlated neutrons and only the sum of all types of events is displayed.

Results

The response of a system depends on the mass of the HEU, the distance between the detector and HEU, the intervening materials, and the measuring time. We have investigated the effect of varying these parameters. For interrogation of large objects, such as cargo containers and semi-trailers, with the cone of radiation from the linac, the object must be placed far enough from the linac so that the whole object is within the cone or the object must be scanned, which is slower. Figure 3 shows an example in which 22 kg of HEU was placed 7 m from the linac and two large area neutron detectors were placed side by side downstream of the HEU at a distance of 2 meters. The neutron counting rate is approximately a factor of five times the active background, which is a measurement with beam on but no HEU present.

High-Z materials are expected to strongly attenuate photon beams. Figure 4 shows the effects of inserting various thicknesses of lead between the linac and the HEU. A 22-kg mass of HEU was on the beam axis and 2.34 m from the linac. Two large area neutron detectors were at 90° to the beam and on opposite sides of the HEU at 1.37 m and 2.23 m from the beam axis. Even with 15.24 cm of lead, the response is still distinguishable from the active background, also measured with 15.24 cm of lead. The response is not reduced as much as expected because of photoneutron production in the lead, which causes neutron interrogation of the HEU.

Similarly, materials containing hydrogen, such as polyethylene, are expected to attenuate the neutrons, both the interrogating neutrons and the fission neutrons emitted by the HEU. Figure 5 shows the effects of shielding 5 kg of HEU with various thicknesses of polyethylene. The 5-kg mass of HEU was on the beam axis and 3.7 m from the linac. Only one detector, located at 90° and 2.23 m from the beam axis, was used for these measurements. With 15.24 cm of polyethylene the response is still distinguishable from background.

One possible application involves adding modular neutron detectors to a scanning radiograph system that uses a fan beam. If the photon energy exceeds the photofission threshold of approximately 5.8 MeV for ^{235}U, then HEU can be detected simultaneously with forming an x-ray image of an object. Figure 6 shows the response with a vertical slit 0.7 cm wide, 30.5 cm high, and 40.6 cm long using lead bricks placed in front of the linac to produce a fanned beam similar to the one expected in a scanning radiography system. The 22-kg mass of HEU was on axis at approximately 244 cm from the accelerator. An array of five modular detectors was out of the beam at approximately 152 cm from the beam axis.

For field applications we intend to use the neutron generator and the portable detector because these items are much more easily deployed than the linac and the large area detector or the modular detectors. For most field interrogations the neutron generator and the neutron detector can be placed in contact with the object. This tight geometry will partially compensate for the lower interrogating flux from the neutron generator and the lower absolute efficiency of the detector. Figure 7 shows the response to 5 kg of HEU using the neutron generator and the portable detector. The neutron generator and the detector were side by side and the HEU was 50 cm from both.

Discussion

For the measurements presented in this report, the HEU masses ranged from 5 to 22 kg. Smaller masses can be detected with the response approximately scaling with the mass. Additional shielding and collimation around the detector and source can reduce the background and thus improve the signal-to-noise ratio. The sensitivity can also be improved by placing the object as close as possible to the source and the detector because the response as a function of distance between the linac and the HEU and the response as a function of distance between the HEU and the detector both scale as $\sim 1/(\text{distance})^2$. Higher intensity sources can be used as long as safety requirements are still met. Longer measurement time will increase the sensitivity, but operational requirements will limit the time in most applications.

One major concern in detecting HEU, either actively or passively, is the possible present of thick shielding around the HEU. The shielding might be incidental, as in a large cargo container loaded with a variety of items, or deliberate to hide the HEU. We believe that radiography is the most practical method to check for this possibility. Radiography systems are already widely used. If the energy of a radiography system exceeds the 5.8-MeV ^{235}U threshold for fission, then it should be possible to add neutron detectors to the system to simultaneously make an x-ray image and detect HEU.

We have presented results for interrogation with pulsed beams of bremsstrahlung photons or neutrons from a DT neutron generator and detection of neutrons between pulses. Other techniques are possible. We tried inserting a beryllium block in the bremsstrahlung photon beam to produce a more intensity beam of neutrons than the one provided by the DT neutron generator and obtained results similar to the results presented here for photons.[2] Detection of the prompt fission neutrons, which have a higher yield than the delayed neutrons, is also possible, but prompt neutrons can also be produced by photoneutron reactions on many other elements. Detection of prompt or delayed gamma rays, which also have a higher yield than the delayed neutrons, has been investigated.[8-11]

Conclusions

We have shown that kilogram quantities of HEU can be detected using active interrogation with photons or neutrons to activate the HEU. Even if the HEU is shielded by lead or polyethylene and cannot be detected passively, active interrogation can detect it. We have described a photon source based on an electron linac and a neutron source based on DT generator. We also described three detector systems based on ^3He tubes that can be used in a wide range of active interrogation applications. Interrogation times are the order of a few minutes with these systems. We believe that active interrogation of HEU is ready for more routine applications.

Acknowledgements

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Figure Captions

Fig. 1. Cross section of a neutron detector module.

Fig. 2. Portable detector polyethylene block and ^{3}He tubes.

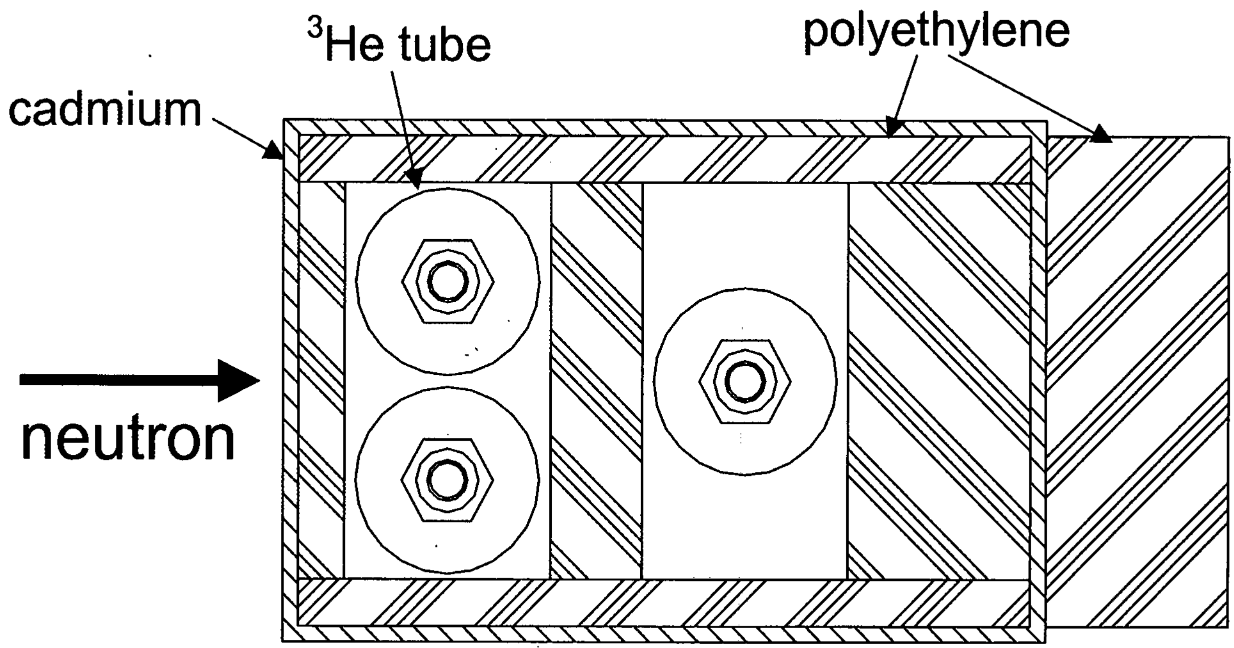
Fig. 3. Neutron counting rate as a function of time after the beam pulse from the linac for 22 kg of HEU at 7 m. The measurement used 36,000 pulses from the linac.

Fig. 4. Neutron counting rate for several thicknesses of lead between the linac and 22 kg of HEU and for an active background measurement. Each measurement used 27,000 pulses.

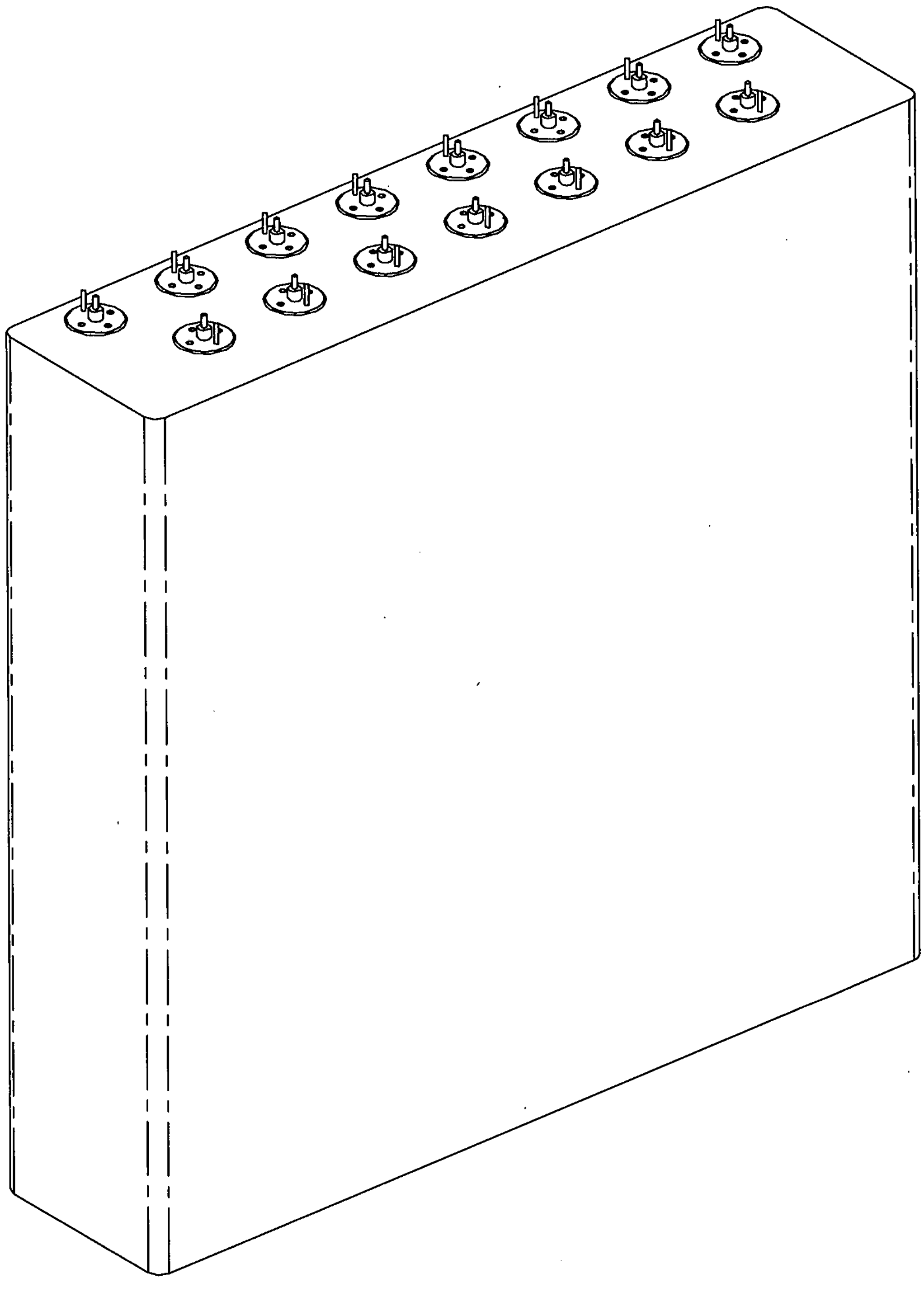
Fig. 5. Neutron counting rate for several thicknesses of polyethylene around 5 kg of HEU and for an active background measurement. Each measurement used 27,000 pulses.

Fig. 6. Response of five modular detectors using an interrogating fan beam from the linac. Each measurement used 8,000 pulses.

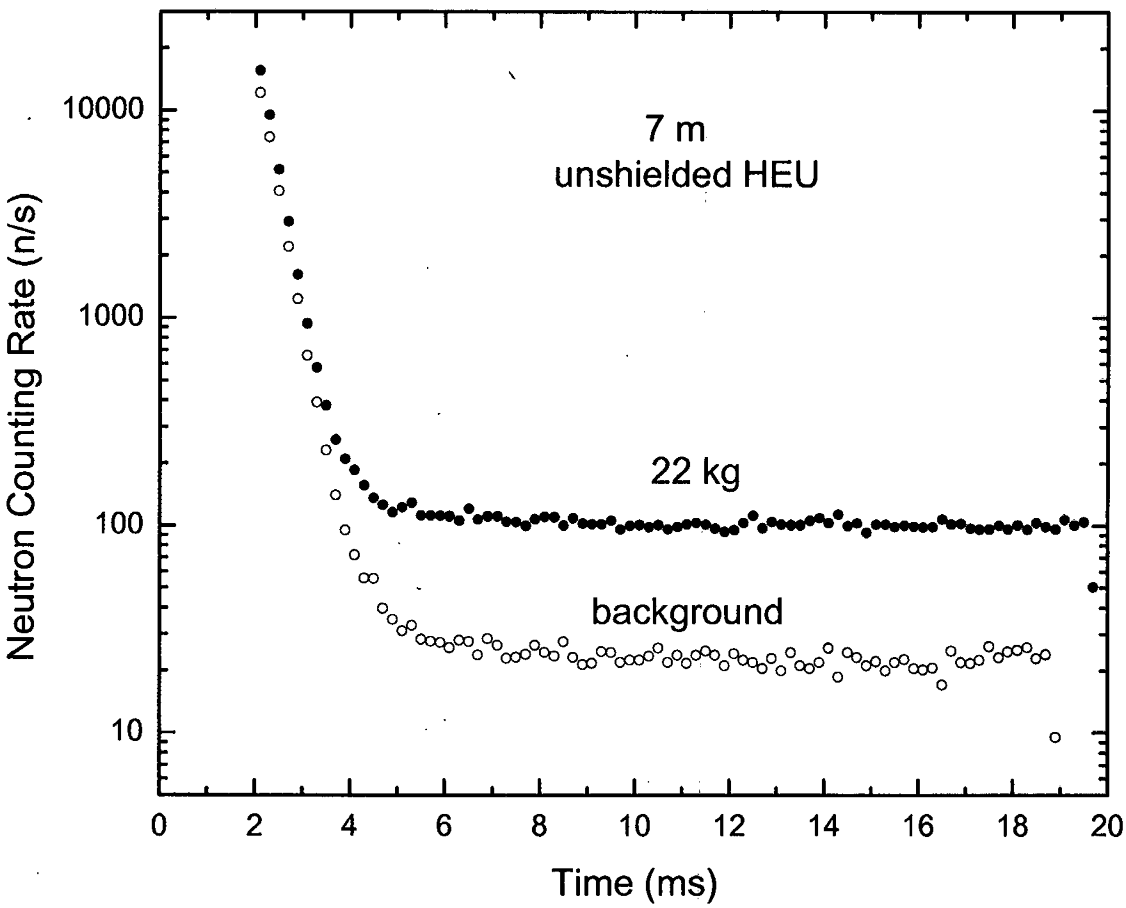
Fig. 7. Response of the portable detector using the neutron generator to 5 kg of HEU. The measurement used 8000 pulses.



Moss, Fig. 1

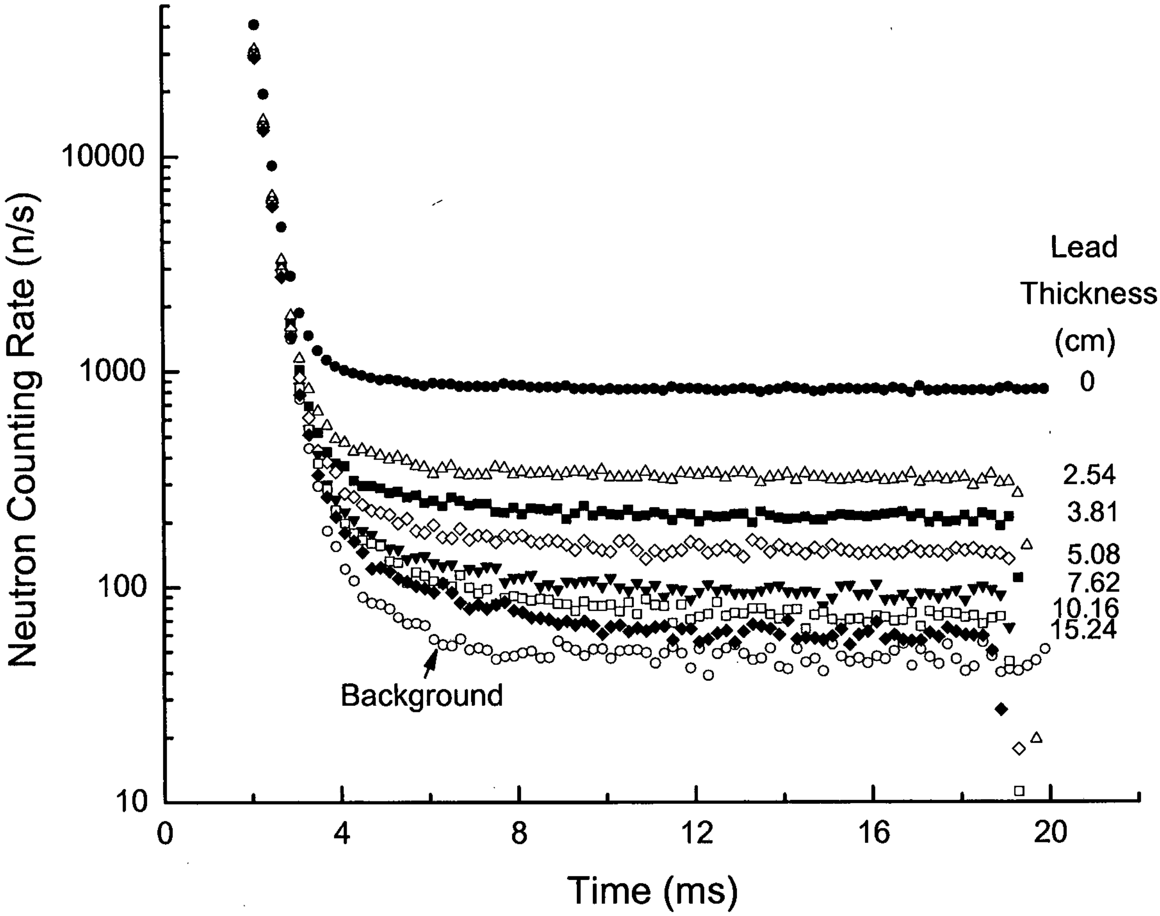


Moss, Fig. 2



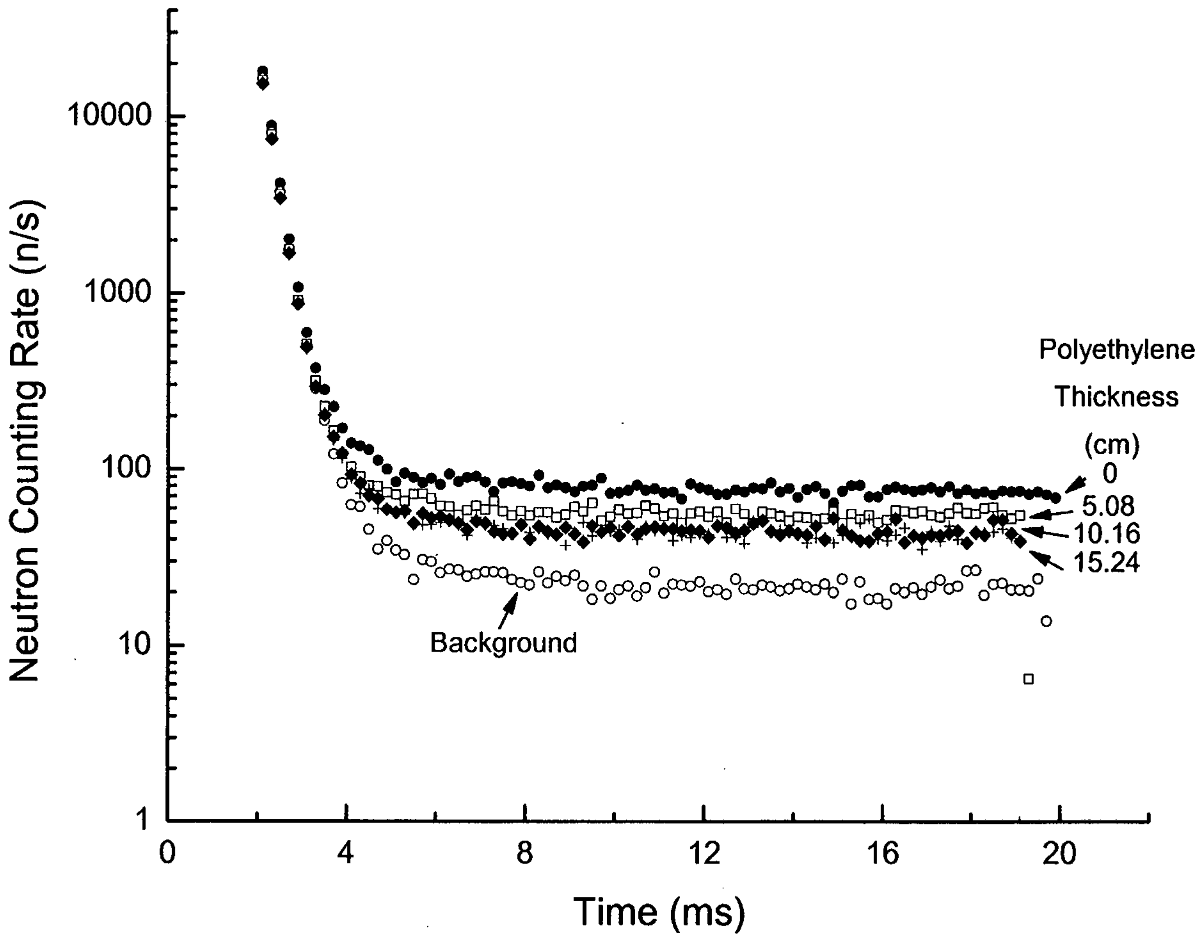
7 m
unshielded HEU
22 kg
background
Neutron Counting Rate (n/s)
10000
1000
100
10
0
2
4
6
8
10
12
14
16
18
20
Time (ms)

Moss, Fig. 3

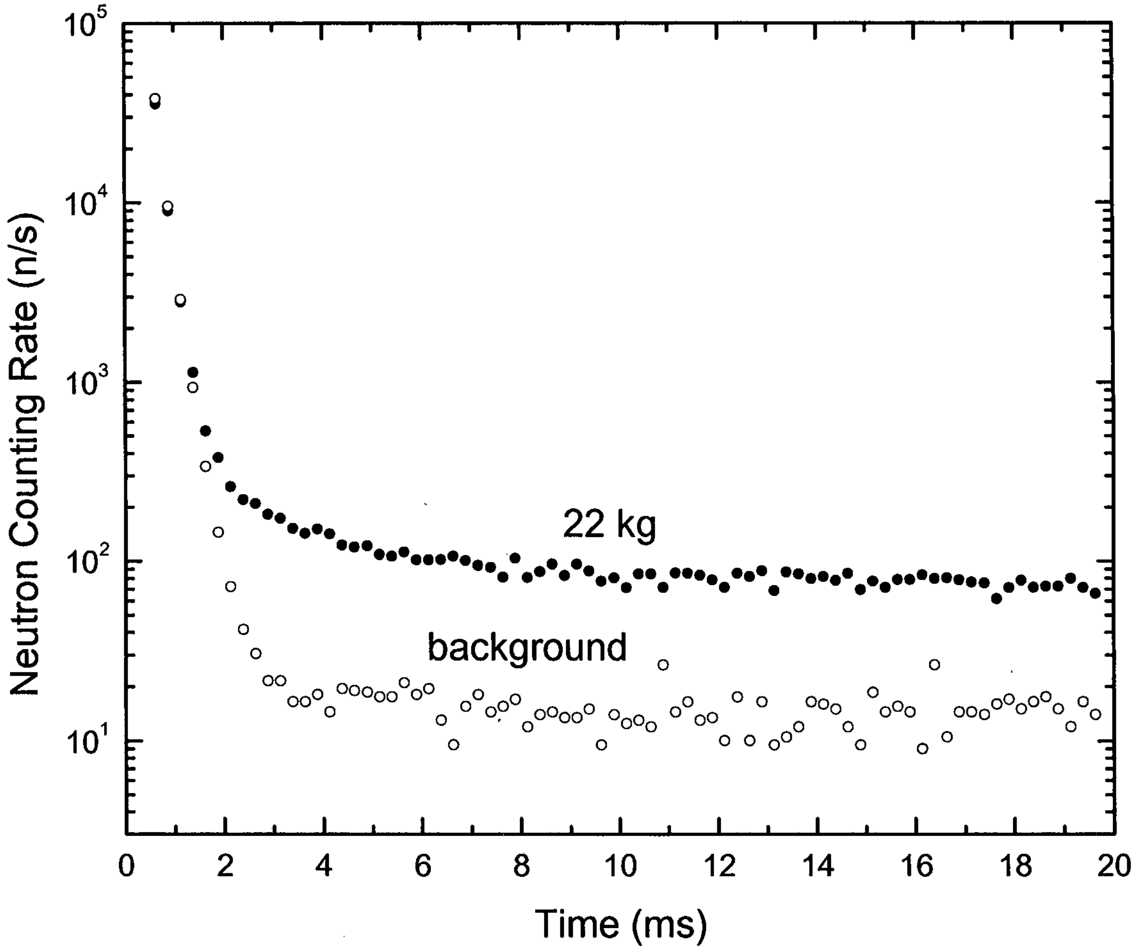


Neutron Counting Rate (n/s)
10000
1000
100
10
0
4
8
12
16
20
Time (ms)
Lead
Thickness
(cm)
0
2.54
3.81
5.08
7.62
10.16
15.24
Background

Moss, Fig. 4

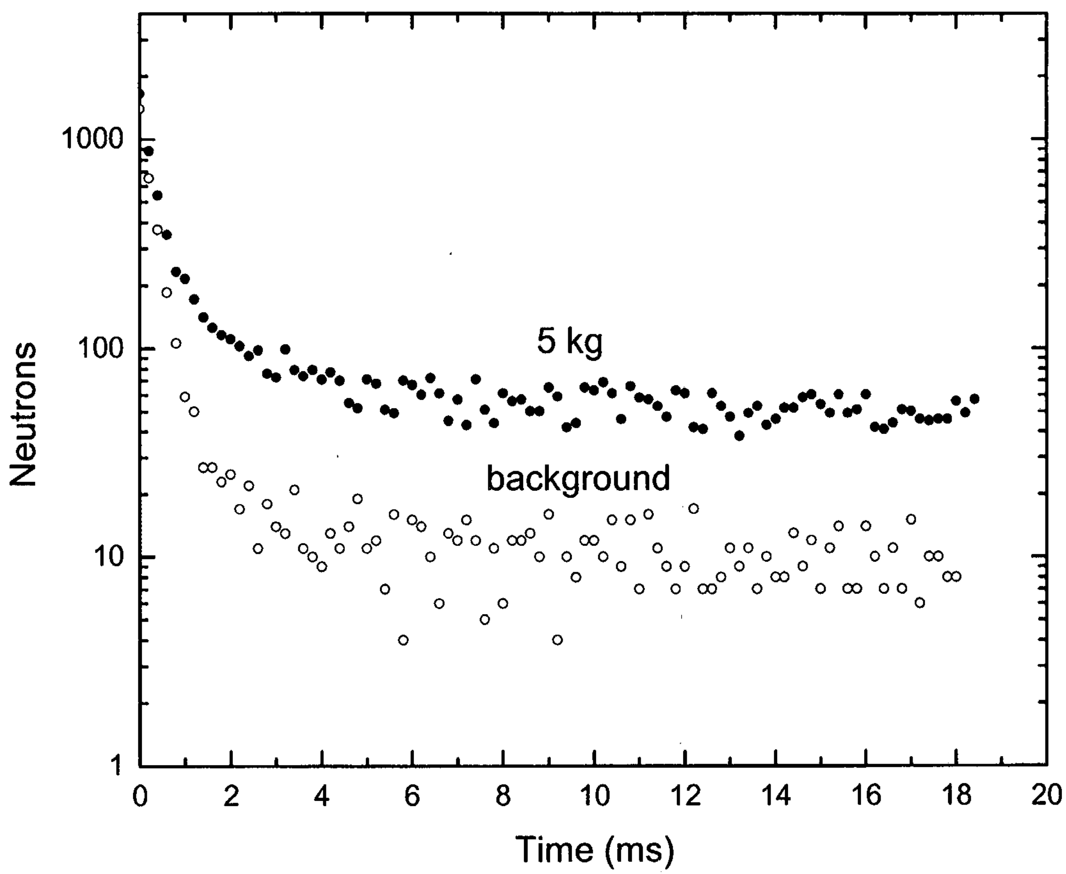

Neutron Counting Rate (n/s)
10000
1000
100
10
1
0
4
8
12
16
20
Time (ms)
Polyethylene
Thickness
(cm)
0
5.08
10.16
15.24
Background

Moss, Fig. 5



10^5
10^4
10^3
10^2
10^1
Neutron Counting Rate (n/s)
22 kg
background
0
2
4
6
8
10
12
14
16
18
20
Time (ms)

Moss, Fig. 6


1000
100
10
1
Neutrons
5 kg
background
0
2
4
6
8
10
12
14
16
18
20
Time (ms)

Moss, Fig. 7