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An Active Neutron Technique for Detecting Attempted Special Nuclear Material Diversion*

by

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ABSTRACT

The identification of special nuclear material (SNM) diversion is necessary if SNM inventory control is to be maintained at nuclear facilities. (Special nuclear materials are defined for this purpose as either ^{235}U or ^{239}Pu.) Direct SNM identification by the detection of natural decay or fission radiation is inadequate if the SNM is concealed by appropriate shielding. The active neutron interrogation technique described combines direct SNM identification by delayed fission neutron (DFN) detection with implied SNM detection by the identification of materials capable of shielding SNM from direct detection.

This technique is being developed for application in an unattended material/equipment portal through which items such as electronic instruments, packages, tool boxes, etc., will pass. The volume of this portal will be 41-cm wide, 53-cm high and 76-cm deep. The objective of this technique is to identify an attempted diversion of at least 20 grams of SNM with a measurement time of 30 seconds.

INTRODUCTION

Control of a special nuclear material (SNM) inventory implies the capability to identify attempted SNM diversion. (SNM is defined for this purpose as ^{235}U and ^{239}Pu, fissile isotopes of Uranium and Plutonium.) This control is presently based on the detection of radiation from either natural radioactive decay or fission. The threat of SNM concealment by radiation shielding materials seriously limits all methods currently used for this purpose. For this reason, efforts, under the auspices of DOE, are underway to develop a method to detect sophisticated attempts to conceal and divert SNM.

The development of an unattended material or equipment pass-through is directed at the inventory control requirements envisioned for fast-critical-reactor facilities. The pass-through or portal is designed to accommodate electronic instruments, tool boxes, packages, etc., and will be nominally 41-cm wide, 53-cm high and 76-cm deep. A threshold sensitivity of 20 grams of SNM, which is expected to be the smallest individual quantity, has been selected as the design goal. A measurement time of approximately 30 seconds has been chosen as consistent with personnel monitoring in progress concurrently with the instrument pass-through measurement.

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BACKGROUND

Several considerations must be made about the problem of identifying an attempt to divert SNM by concealment. The first deals with the methods of identification.

Radioactive decay of SNM results in relatively soft gamma radiation that is easily attenuated by several centimeters of high Z material such as iron or lead. Fission produces energetic, penetrating (neutron and gamma ray), prompt radiation that is difficult to attenuate. Delayed, penetrating radiation from fission products is both less abundant and less energetic, increasing the attenuation effect of shielding material. Fission can be induced in SNM by neutrons, with a probability that varies as a function of the energy of the neutron, and the unique property of fissile material is its affinity for slow neutron fission.

Fission occurs spontaneously at significant rates in a few non-fissile isotopes, one of which (^{240}Pu) is found in concentrations of at least a few percent with ^{239}Pu. This substantial radiation forms the basis for some passive methods of assaying and/or detecting fissile plutonium (^{239}Pu). Unfortunately, no such spontaneously fissioning uranium isotope is found with ^{235}U. For detection or assay purposes using fission radiation, ^{235}U must be induced by external means to fission and produce radiation that will lead to its identification. Some existing assay and detection methods for ^{235}U are based on active neutron interrogation.

Of the two radiations, gamma rays from radioactive decay or gamma rays and neutrons from fission, the more energetic radiations from fission provides the greater possibility of SNM detection in the presence of shielding. Several techniques have been developed for non-destructive assay of SNM based on identifying fission radiation. Four such assay methods were selected for evaluation as techniques for direct detection of concealed SNM. Of these four techniques, coincidence detection[1] and correlation counting[2] both imply the occurrence of a fission event based on timing and multiplicity of prompt fission radiation detection. Two gross count methods identify fission neutrons by time discrimination (DFN[3]-Delayed Fission Neutron) or time and energy discrimination (PFN[4]-Prompt Fission Neutron).

All of these methods were examined either theoretically or experimentally or both and in each case found to be prohibitively vulnerable to shielding.[5] Given the internal volume of the instrument portal and the shield material volume that can be accommodated, each of the direct detection methods examined must be disqualified as a stand alone method of identifying SNM if the SNM is substantially shielded.

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[1] T. J. Atwell, J. E. Foley, and L. V. East, "NDA of HTGR Fuel Using the Random Driver", Nuclear Materials Management, Fall 1974, pg. 171.

[2] Edward J. Dowdy, Carl N. Henry, Arnold A. Robba, John C. Pratt, "New Neutron Correlation Measurement Techniques for Special Nuclear Material Assay and Accountability", Los Alamos Scientific Laboratory-Report LA-UR-78-690, Submitted to: IAEA (IAEA-SM-231/69).

[3] Howard O. Menlove and Thomas W. Crane, "A ^{252}Cf Based Nondestructive Assay System for Fissile Material", Nuclear Instruments and Methods 152 (1978), pg. 549.

[4] Gerald W. Smith, "Status Report on the Development of a Prompt Fission Neutron Uranium Borehole Logging Technique", Sandia Laboratory - Report SAND77-0336, 1977.

[5] Gerald W. Smith and Lucien G. Rice, "Status Report on the Development of an Active Neutron Technique for Detecting Special Nuclear Material Diversion", Sandia Laboratory - Report SAND79-0897, 1979.

PROPOSED METHOD

The prospect of a single detection technique directly identifying SNM is not practical given the large volume of the portal available for shielding. However, a direct SNM detection measurement for unshielded or lightly shielded SNM combined with a measurement to identify shielding material capable of shielding SNM from direct detection will be described. The direct SNM measurement is intended to force the use of shield material that can be identified and distinguished from legitimate materials.

Gamma rays are effectively attenuated by dense material such as iron and lead, which for the most part must be considered legitimate material and passed. Neutrons are best attenuated by a shielding material with a large concentration of hydrogen for moderation and a neutron absorber such as boron, cadmium, lithium, etc., for absorbing the moderated neutrons. A hydrogenous material such as polyethylene, paraffin, or water containing a small amount of neutron absorber could constitute effective shielding for concealing SNM against neutron interrogation and fission neutron detection. This is a material that could be administratively prohibited from routine passages through the portal, if it can be uniquely identified. In order to insure the exclusion of such material, a measurement of the perturbing effect that a thermal neutron absorber will have on a thermal neutron population can be used to identify its presence.

As seen in Figure 1, the effectiveness of a neutron absorber (increased absorption) increases as the energy of the neutron decreases. Through moderation, a neutron will reach equilibrium with its surroundings, and by convention this equilibrium condition has been established at 300°K with the resulting energy of equilibrium neutrons being 0.025 eV.

The most sensitive measurement for the perturbing effect produced by the presence of a neutron absorber would be made with a population of neutrons most affected, neutrons at equilibrium (thermal) energy.

In a steady state neutron environment, neutrons from an isotopic source, energies ranging from thermal equilibrium to generation energy would coexist. In a pulsed neutron environment, pulse timing and rate can be selected such that all energetic neutrons will be moderated to thermal equilibrium energy and survive exclusively as thermal neutrons for a time governed by the absorption and diffusion properties of the materials defining the portal test volume. By selecting materials that minimize absorption and diffusion, the mean lifetime of thermal neutrons will be maximized, increasing sensitivity to the presence of a neutron absorber.

Thermal Die-Away Perturbation

A pulsed neutron source creates mono-energetic neutrons that are moderated by the scattering properties of the materials they encounter. The time histories of fast, intermediate, and thermal neutron groups are shown in Figure 2.

The die-away of thermal neutrons is described by the exponential expression

$$n(t) = n(o)e^{-\alpha t}$$

where $n(t)$ is the instantaneous thermal neutron rate and $n(o)$ is the initial rate. The decay constant α is a function of absorption and diffusion as follows

$$\alpha = \nu\Sigma_a + \nu DB^2$$

where ν is the thermal neutron velocity, Σ_a is the macroscopic absorption coefficient, D is the diffusion coefficient and B^2 is the geometric buckling. The thermal neutron mean lifetime is

$$T = \frac{1}{\alpha} = \frac{1}{\nu\Sigma_a + \nu DB^2}$$

which illustrates the mean lifetime dependence on the two loss mechanisms of absorption and diffusion. If the materials and geometry are such that the geometric buckling is zero, the only thermal neutron losses would be to absorption and the mean lifetime would be maximized.

However, in most finite systems diffusion losses to buckling will be a factor depending on materials. For analysis, those losses can be expressed as absorption by an effective macroscopic absorption cross section for the portal which is defined as

$$\Sigma_{eff} = \Sigma_a + DB^2$$

and the thermal neutron mean lifetime of the portal will be

$$T = \frac{1}{v\Sigma_{eff}} .$$

Material that will best define the portal test cavity and reveal the presence of absorbing material in a neutron shield will be material that effectively moderates fast neutrons and, of those thermalized neutrons created, captures very few. A figure of merit for materials suitable to this application is the moderation ratio

$$MR = \xi\bar{\Sigma}_s/\Sigma_a$$

where ξ = average logarithmic energy decrement per collision

$\bar{\Sigma}_s$ = average macroscopic scattering cross section (0.025 eV $\leq$ energy $\leq$ 0.1 MeV

Σ_a = macroscopic absorption cross section for thermal neutrons.

The product $\xi\bar{\Sigma}_s$ is called the slowing down power. While large moderation ratio is a good indication of properties desirable for this application, a small absorption cross section must be considered most important. Table 1 is a list of these properties for good neutron moderating materials. Deuterated materials are by far superior because they combine very small absorption with large slowing down power. Unfortunately, they are very expensive and will not be considered at this point in the system development. Carbon has a very small absorption cross section (most desirable) but poor slowing down power. Beryllium is possibly suitable but also expensive. Polyethylene has very large slowing down power but its absorption cross section is large.

No single material, other than deuterated materials, has all of the desired properties for this application, but a reasonable set of properties can be developed by a combination of carbon and polyethylene. With carbon slabs backed by polyethylene lining the portal cavity, the high energy neutrons will be efficiently slowed by the bulk polyethylene and the thermal neutrons that find their way into the cavity will have a long die-away time due to the very small absorption cross section of carbon.

Another factor to be considered is the effective reflection coefficient (albedo) which is an indication of diffusion losses. Albedos of the carbon and polyethylene as a function of thickness are shown in Figure 3. Thermal neutrons that diffuse through the carbon are likely to be absorbed, with an undesirable increase in the effective absorption of the cavity. This diffusion loss can be reduced by a carbon slab that is sufficiently thick to maintain a large albedo within the carbon (perhaps 10 to 15 cm). A further increase in carbon slab thickness will tend to decrease the net thermal neutron population in the cavity because fast neutrons are being moderated further away from the volume and an increase in carbon albedo will tend to reflect these neutrons away from the cavity. A 15 cm thickness of carbon was selected for testing. Figure 4 shows an experimental portal designed for evaluation of this measurement technique.

The neutron source is a DT generator (14 MeV) that produces greater than 10^6 neutrons per pulse and can be operated at pulse rates up to 100 pulses per second. The thermal neutron detector is a single ^{3}He proportional counting tube, 1-cm in diameter and 25-cm active length with a ^{3}He gas pressure of 10 ATM. The delayed fission neutron detector consists of 30 ^{3}He proportional counting (PC) tubes in an array outside the carbon cavity liner on three of the side walls. Each PC tube is 5-cm in diameter and 61-cm active length with a gas pressure of 4 ATM. Figure 5, measured time histories for different portal contents, shows the thermal neutron die-away change resulting from absorption losses to two shield material blocks of 32% borated polyethylene at 7.5-cm and 12.5-cm thickness. These changes are based on thermal neutron mean lifetime change. Either of these shields would be readily distinguished from the empty cavity reference.

Direct SNM Detection

For unshielded or lightly shielded SNM, direct identification of the SNM is accomplished by detecting delayed fission neutrons resulting from fissions produced by the source neutrons. Prompt fission neutron (PFN) detection as described in reference 5 was initially considered for direct SNM detection but found to be less suitable than DFN because of shielding vulnerability. The total number of delayed neutrons (N_T) from a ^{235}U sample under interrogation is the product of the number of fissions produced (F) and the total number of delayed neutrons per fission ($\frac{N}{F}$).

$$N_T = F \times \frac{N}{F}$$

For the portal configuration described in Figure 4, the fission production efficiency (f), fissions per gram of SNM and per source neutron as a function of shielding is given in Figure 6. The fission production efficiency is derived from Monte Carlo analysis of the portal. The Monte Carlo code (MORSE) is a three dimension, time dependent, neutron transport code. The total number of fissions produced is

$$F = f \times m \times N_s$$

where (m) is the mass of ^{235}U and (N_s) is the total source neutrons. The number of delayed neutrons that are detected by the neutron detector is directly proportional to the delayed neutron fraction transmitted through the shield (ϵ_s) shown in Figure 7. The absolute efficiency of the delayed neutron detector array is (ϵ_{PC}). The number of delayed neutrons detected also depends on the time window available for counting (T), the time available for counting normalized to the total time required to sample all delayed neutrons. The number of delayed neutrons counted is

$$N_D = \epsilon_{PC} \times \epsilon_s \times T \times N_T .$$

If all source neutrons were generated in a single pulse and sufficient time after the pulse was used for counting, essentially all delayed neutrons generated are available for detection and counting and the time fraction (T) approaches unity. However, for the case of a rapid pulse generator, the optimum time window for sampling delayed neutrons is a function of the pulse rate.

The time required for a population of source neutrons to moderate and be captured or diffuse from the system is approximately 10 milliseconds. Therefore, the generator cannot be operated at a pulse rate greater than 100 pulses per second. Based on estimates of the total source neutron requirement, not less than 10^3 pulses of the neutron source will be required for each measurement which must be made in 30 seconds or less. The minimum rate at which the source can operate and generate 10^3 pulses in 30 seconds is 33 pulses per second. Figure 8 shows the dependence of the time fraction (T) on neutron source pulse rate. From Figure 8, the optimum source rate for maximum sampling interval is 40 pulses per second which permits the detector to look for 45% of the delayed neutrons generated.

The final expression for the number of delayed neutrons detected is

$$N_D = \epsilon_{PC} \times T \times \frac{N}{F} \times \epsilon_s \times f \times N_s \times m$$

where ϵ = 0.036 (absolute delayed neutron efficiency)
T = 0.45 DFN measurement time fraction
$\frac{N}{F}$ = 0.0165 $\frac{\text{delayed neutrons}}{\text{fission}}$ (^{235}U)
ϵ_s = delayed neutron fraction transmitted through shielding
f = fission production efficiency
m = ^{235}U mass
N_s = Total source neutrons.

From this model, threshold mass for direct detection of ^{235}U are shown in Figure 9 as a function of background for various shielding conditions.

A critical parameter for threshold estimates is the background and the acceptable false alarm criteria. Laboratory background for these measurements was 160 counts. For unattended applications such as this portal is intended, a very low false alarm rate is desirable. For the purpose of this evaluation, the false alarm probability has been chosen to be 0.001, one false alarm every 1000 measurements. Each shielding configuration in Figure 9 shows two detection thresholds, one representing a detection probability of 0.999 and the other a detection probability of 0.9.

The SNM thresholds shown in Figures 9 and 10 indicate how good the thermal neutron die-away perturbation measurements for shielding must be in order to prevent 20 grams of ^{235}U from being successfully concealed. For the laboratory background of 160 counts unshielded ^{235}U in quantities of a few grams could be reliably detected. If the SNM were concealed in a thin cadmium envelope essentially all thermal fissions are eliminated and the threshold is degraded nearly an order of magnitude. However, even with a cadmium shield 20 grams is detectable.

For more substantial shielding in the form of borated polyethylene, Figure 9, the detection threshold is seriously compromised. Borated polyethylene in the amounts shown must be identified by thermal neutron die-away perturbation if detection is to succeed.

Figure 10 shows effects on detection threshold produced by pure polyethylene. The pure poluethylene shields moderate neutrons sufficiently so that penetration of the carbon cavity liner is effectively reduced and fewer neutrons reach the neutron detector array. The fact that pure polyethylene contains no significant thermal neutron absorber severely limits the effectiveness that can be expected of the thermal die-away perturbation measurement.

Measured mean lifetimes of the thermal neutron die-away for these various shields are shown in Figure 11 on the left of the time axis. On the right are die-away times for the empty cavity and legitimate items for passage through the portal.

CONCLUSION

From Figures 9, 10 and 11, it is clear that the direct DFN detection of 20 grams of ^{235}U is not possible for some shielding conditions. Furthermore, not all of the successful shield configurations are distinguishable by thermal neutron die-away perturbation from legitimate items requiring passage.

A number of options that will improve the detection threshold are available for consideration. Generate more source neutrons, reduce measurement background, and improve DFN detector efficiency, are possibilities obvious in the delayed neutron production equation. For shield identification, there is the possibility of further material identification by spectral signatures of thermal neutron capture gamma radiation. This possibility is currently being examined.

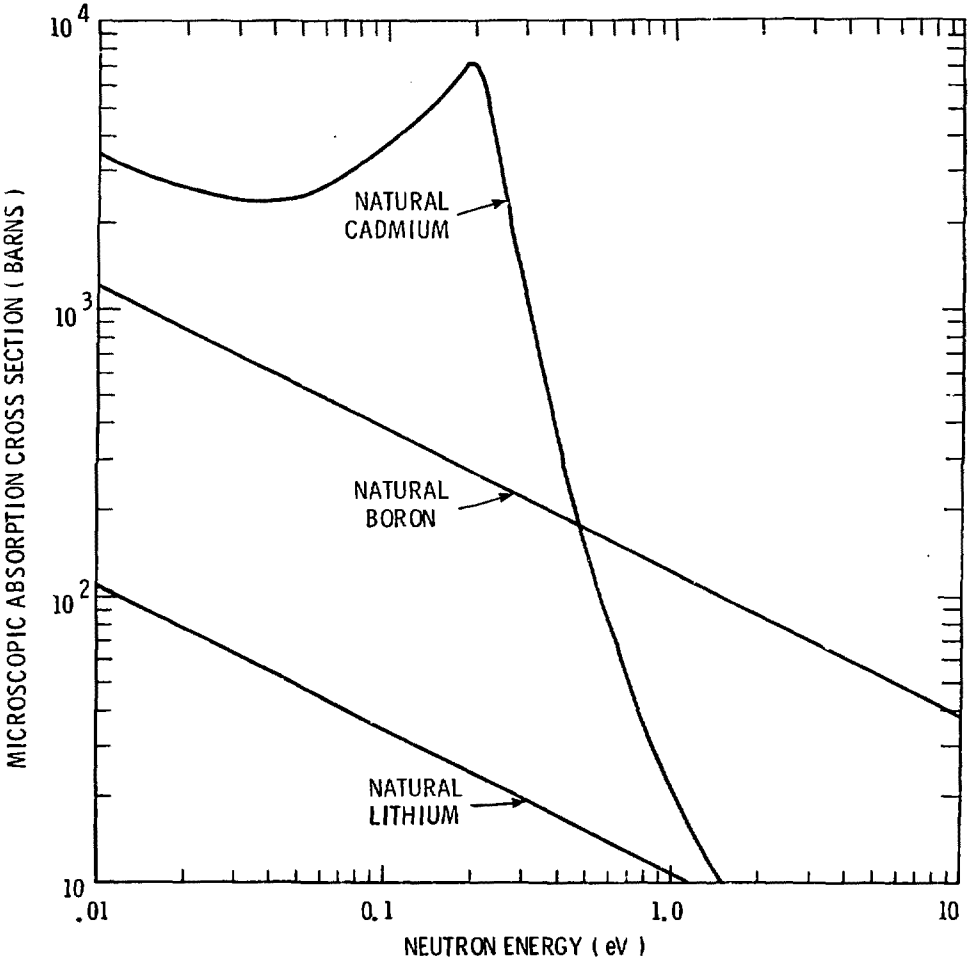


FIGURE 1. NEUTRON ABSORBER CROSS SECTIONS

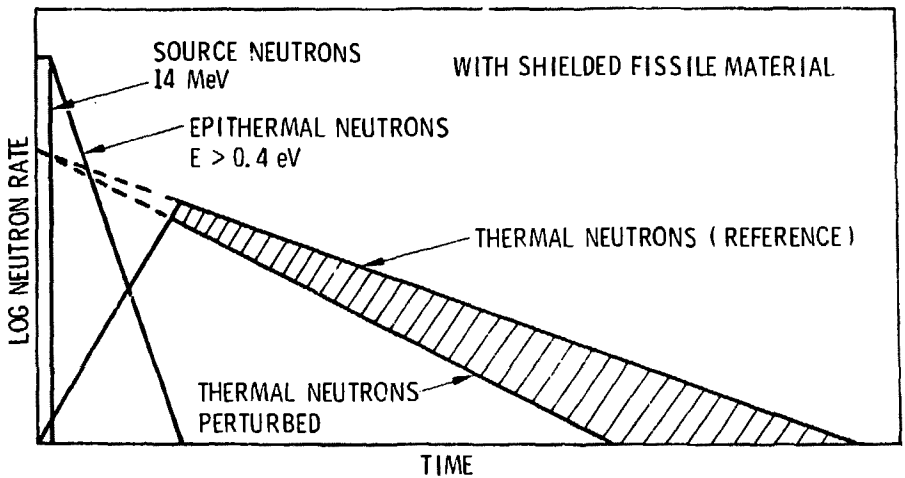


FIGURE 2. NEUTRON TIME HISTORIES

TABLE 1

NEUTRON MODERATION PROPERTIES OF VARIOUS MATERIALS

MODERATOR	SLOWING DOWN POWER (CM^{-1}) $0.025eV \leq ENERGY \leq 0.1$ MeV	ABSORPTION CROSS SECTION Σ_a (CM^{-1}) E = 0.025 eV	MODERATION RATIO
HEAVY WATER * D_2O	0.18	0.000031	5670
DEUTERATED POLYETHYLENE' CD_2	.21	0.00018	1130
CARBON* C	0.060	0.00031	192
BERYLLIUM * Be	0.158	0.0011	143
WATER * H_2O	1.35	0.019	71
POLYETHYLENE' CH_2	1.65	0.026	64

* "NEUTRON PHYSICS ", K. H. BECKURTS, KWIRTZ

' CALCULATED VALUES

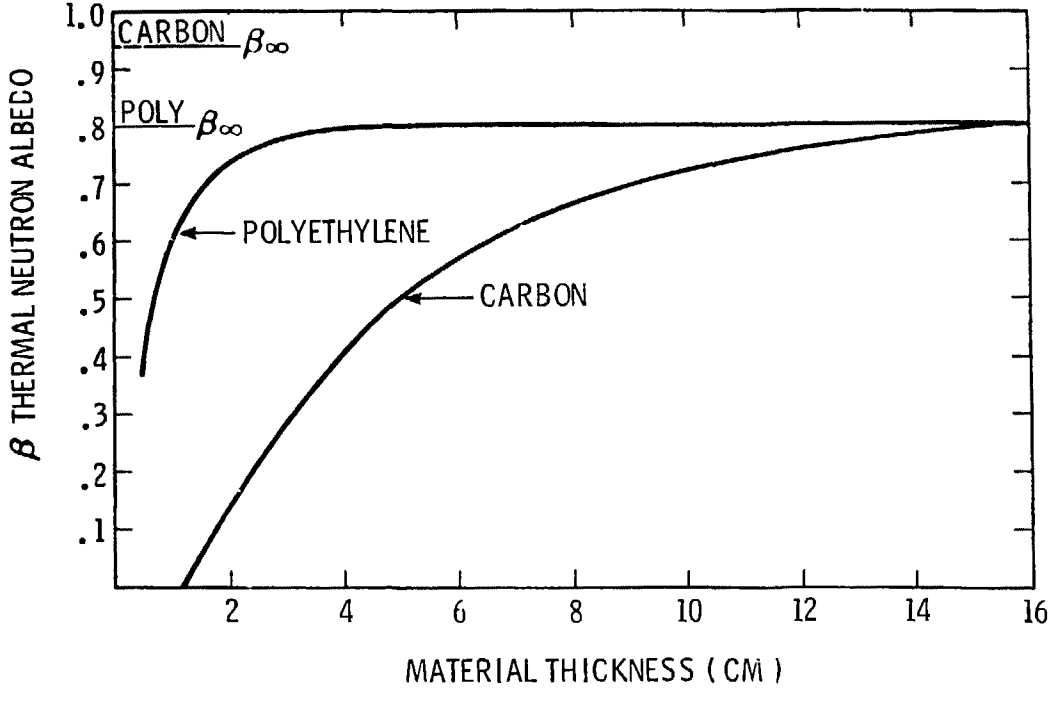


FIGURE 3. THERMAL NEUTRON ALBEDOS

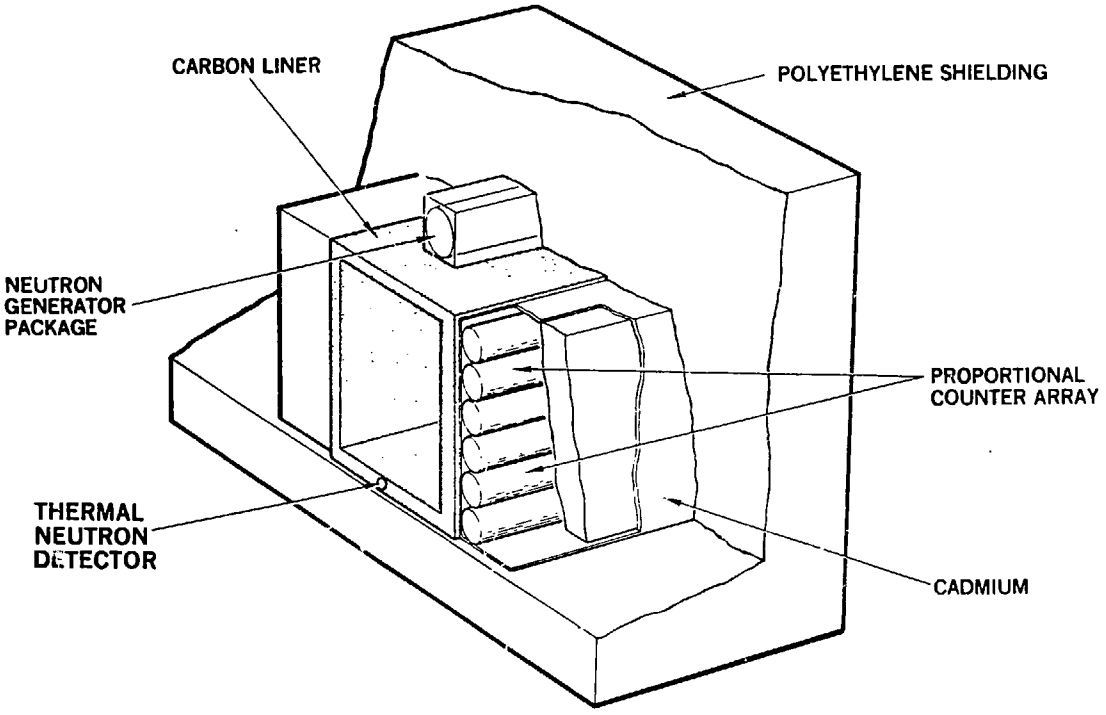


FIGURE 4. INSTRUMENT PORTAL TEST FACILITY

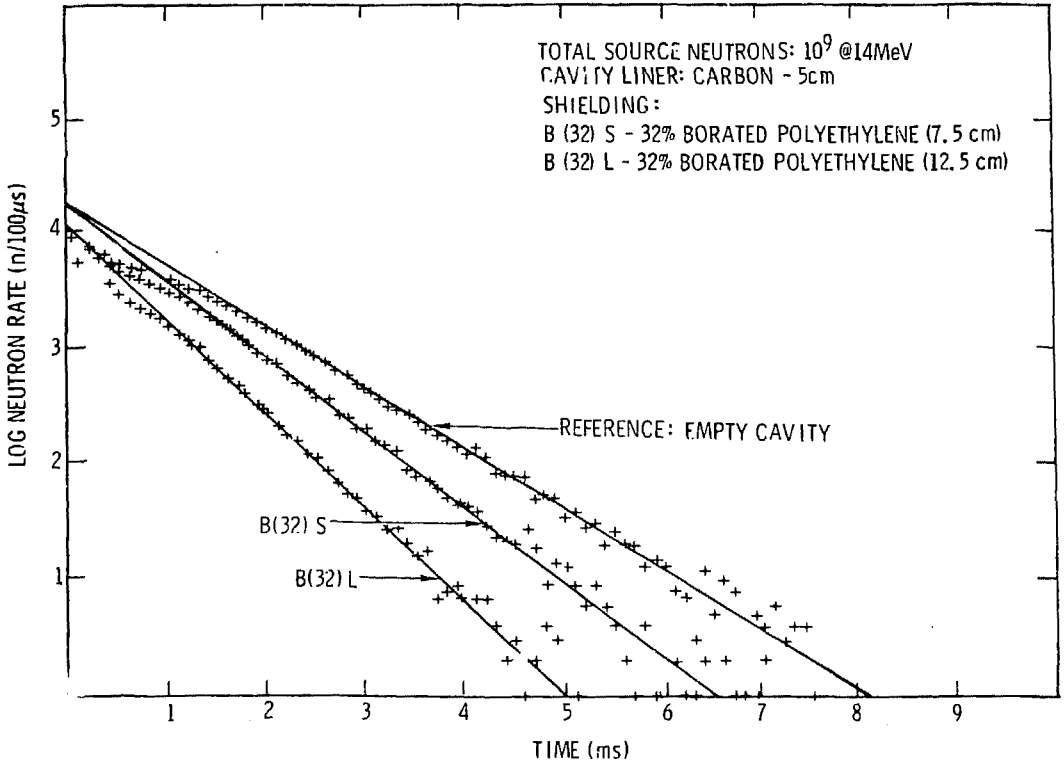


FIGURE 5. TIME HISTORIES

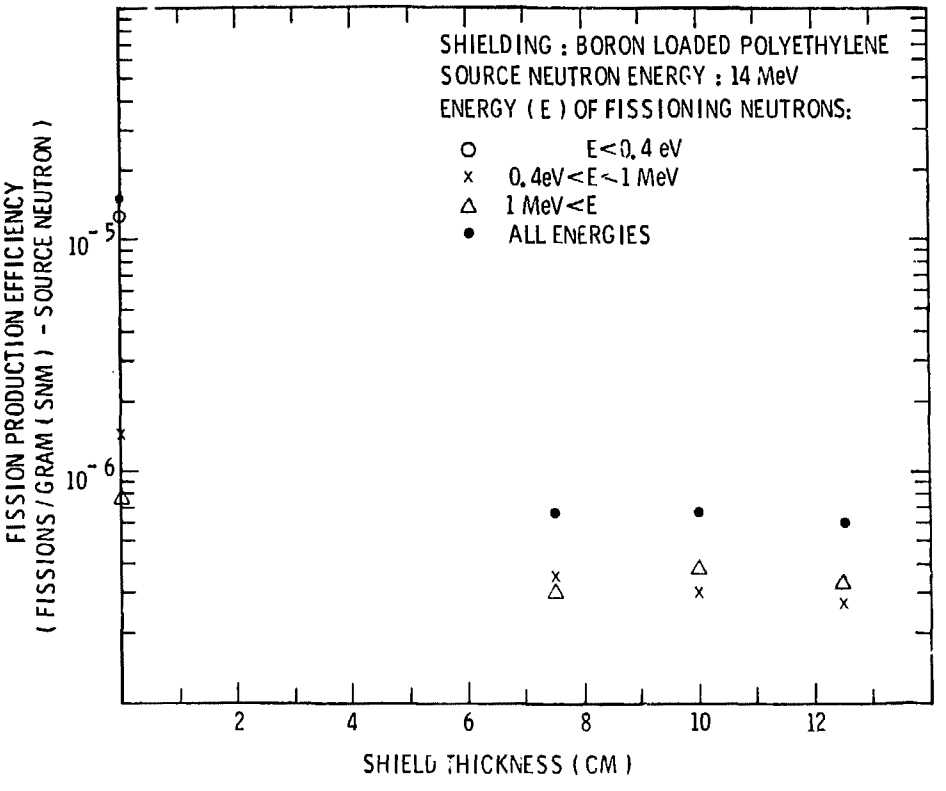


FIGURE 6. INDUCED FISSIONS IN SNM

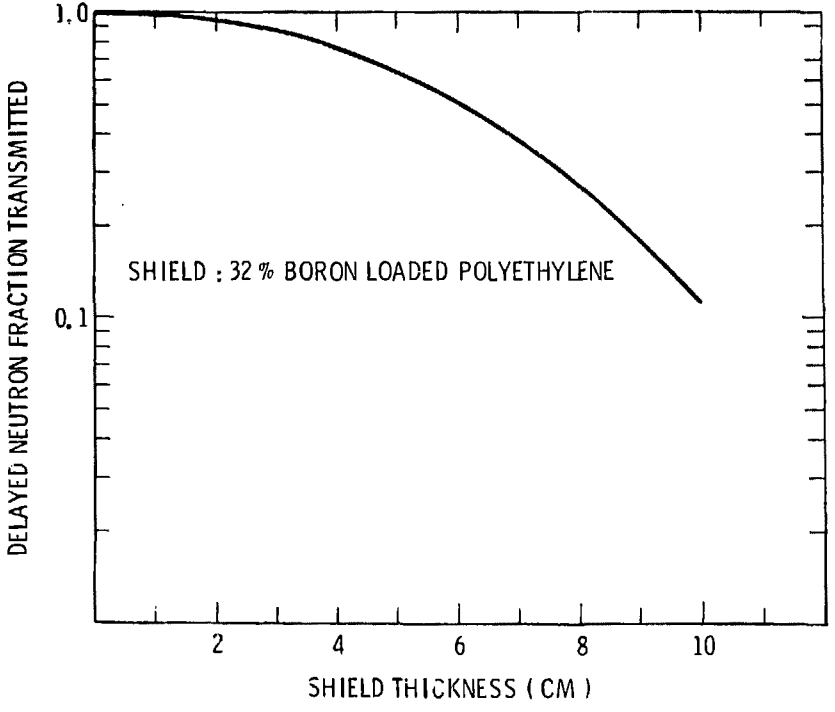


FIGURE 7. DELAYED NEUTRON SHIELD LOSSES

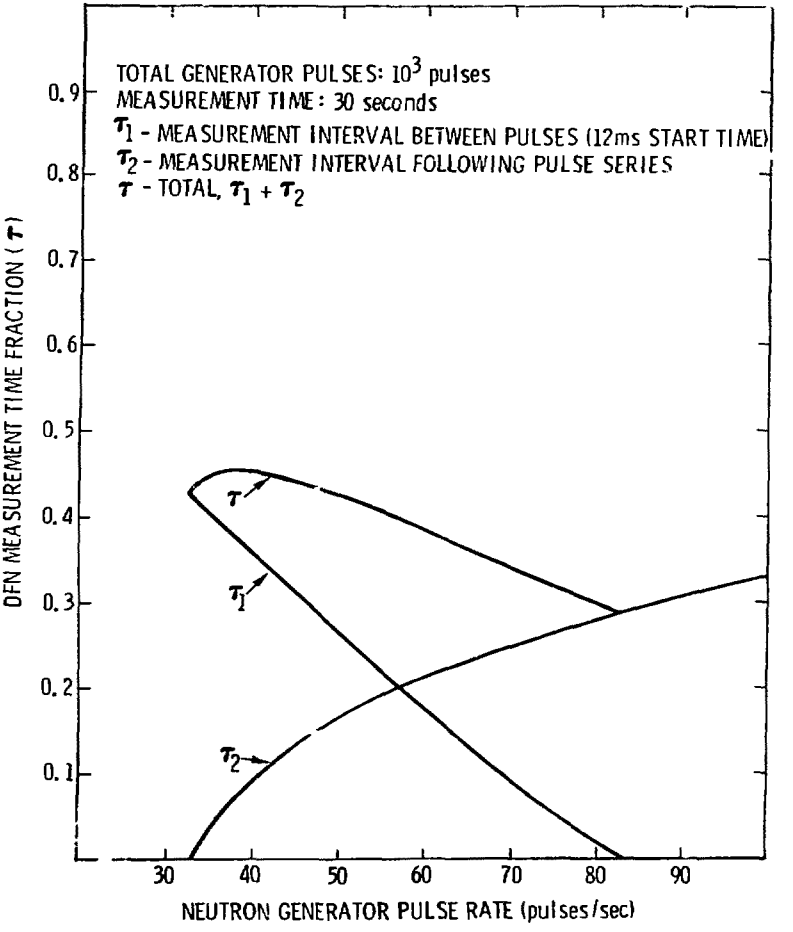


FIGURE 8. DFN MEASUREMENT TIME FRACTION

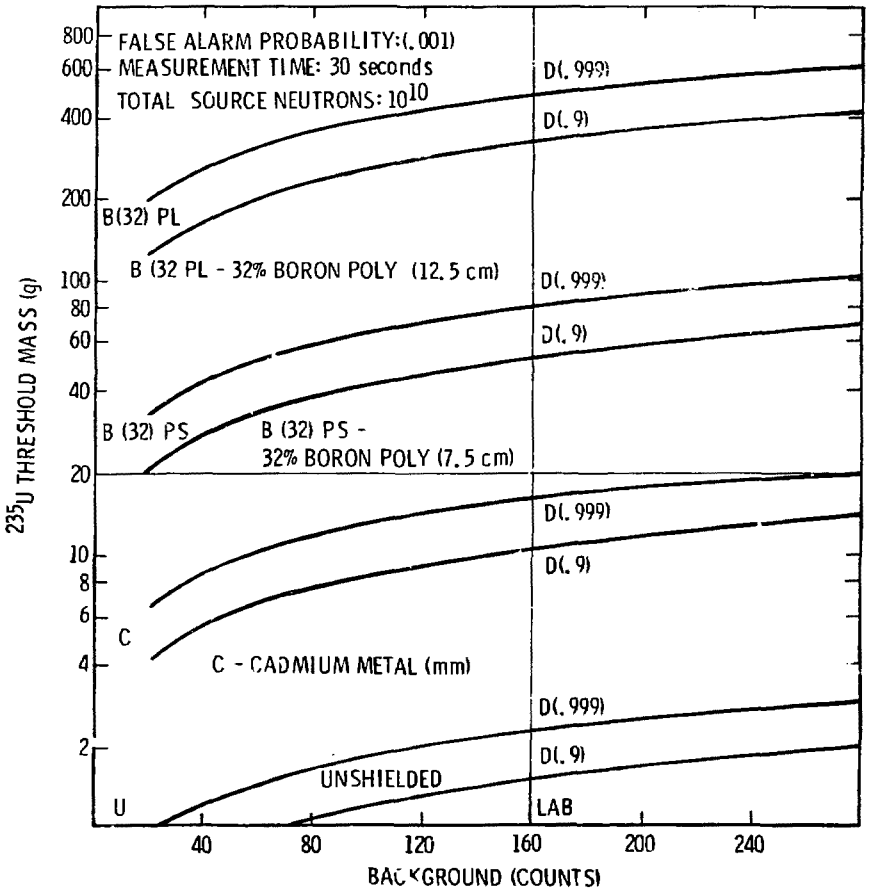


FIGURE 9. DFN DETECTION THRESHOLDS

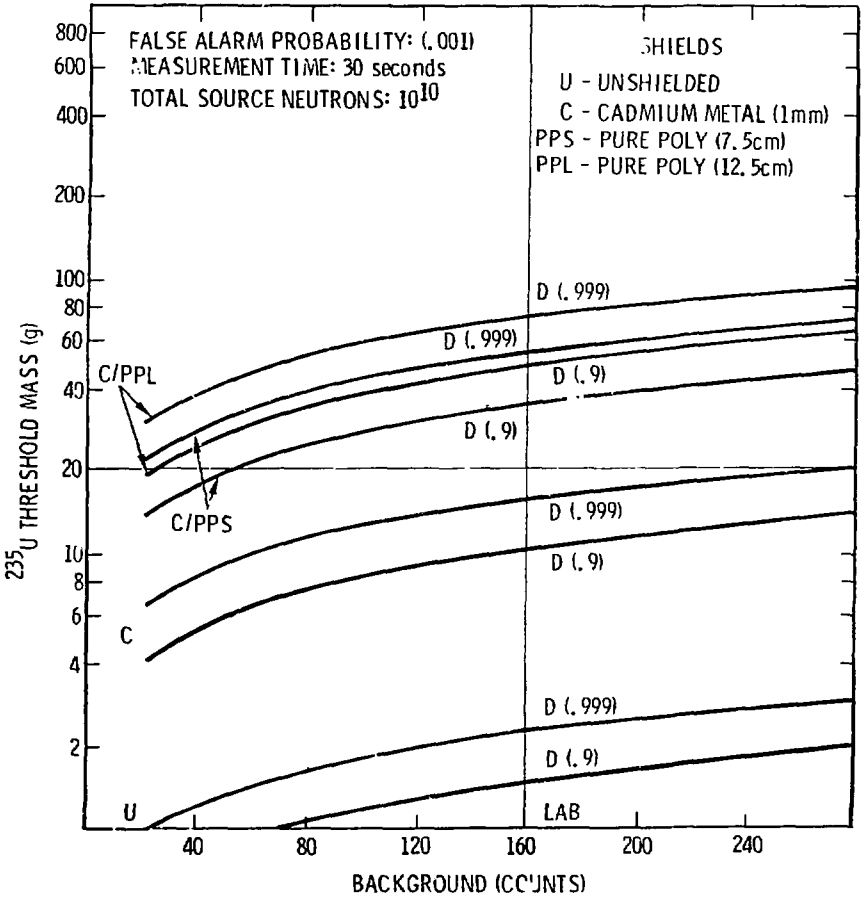
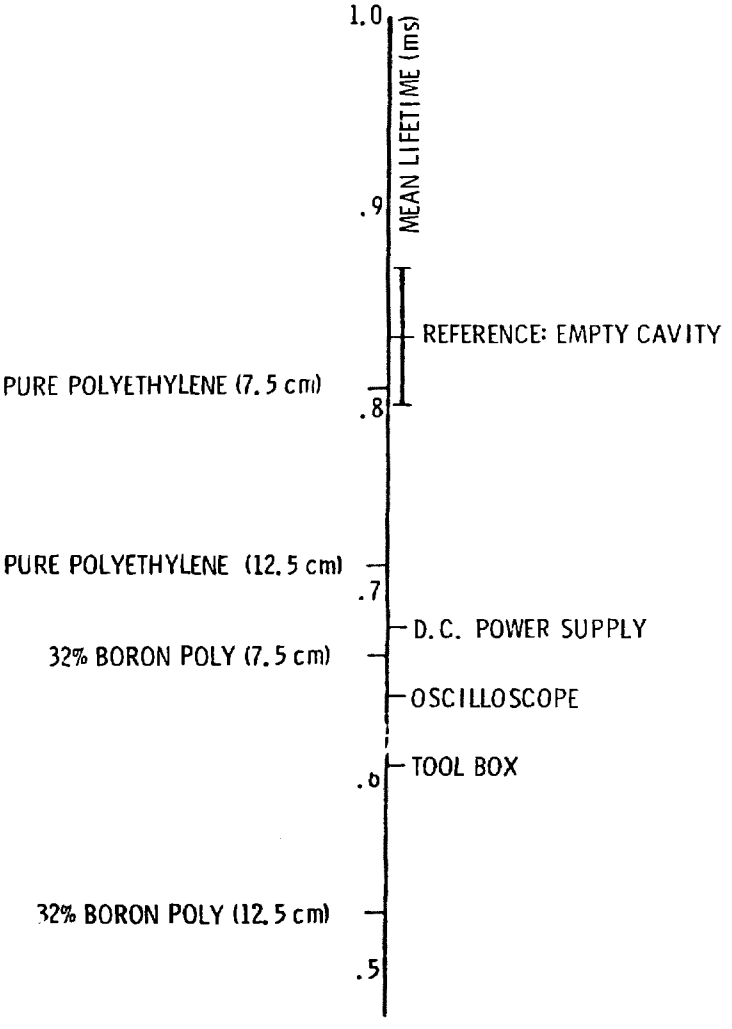


FIGURE 10. DFN DETECTION THRESHOLDS



FICURE 11. MEAN THERMAL LIFETIMES