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NEUTRON DOSIMETRY WITH ACTIVATION FILTERS AND TLD^(a)

by

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NEUTRON DOSIMETRY WITH ACTIVATION FILTERS AND TLDABSTRACT

A TLD dosimeter technique has been developed to quantitatively determine fast and thermal neutron dosages from mixed radiation exposures. For fast neutron dosimetry, this method takes advantage of the scattering of thermalized neutrons from a hydrogenous moderator.

The dosimeter housing consists of a standard Hanford beta-gamma film badge dosimeter. Three filters, consisting of combinations of lead, stainless steel, tin, cadmium, and rhodium are matched so as to produce equal TLD response from photon radiations. The fast and thermal neutron exposure is determined from the TLD response produced by the capture of moderated neutrons in the filter material. Where heterogeneous radiation fields are encountered, a portion of the thermal neutrons is absorbed in one of the rhodium filter sandwiches. This differential forms the basis of a system to distinguish between fast and thermal neutrons. Rhodium was picked as the prime element of the filter system because it has an unusually large resonance absorption for neutrons of 1.26 eV. The capture cross section at 1.26 eV is 4500 barns. Any neutron with a energy near the 1.26 eV resonance will be preferentially absorbed causing the rhodium to become activated. For a particular incident neutron energy, the activation of the rhodium should be directly proportional to the exposure or total flux.

During design and exposure, a large piece of polyethylene was used as a phantom. Dosimeters were attached to the polyethylene and exposed to radiation from various isotopic neutron sources (PuF_4 and PuBe) and monoenergetic neutrons generated by a positive ion accelerator. Thermal neutron exposures were conducted in a calibrated paraffin tank using the PuF_4 source to supply the neutrons. The dosimeter can also serve for photon dosimetry.

Introduction

The search for a personnel neutron dosimeter for adequate sensitivity and satisfactory energy response has continued for at least the last ten years. Work conducted at the old Hanford Labs, Battelle-Northwest, and such other AEC installations as Brookhaven, Oak Ridge, and Argonne has led to a number of reasonably accurate active dosimetry systems. But as yet, no passive device is available that is suitably accurate over the required neutron energy spectrum without an undesirable sensitivity to gamma radiation.

The use of NTA nuclear emulsion continues today at many universities and atomic energy sites, where the properties of the emulsion are not detrimental to the accurate measurement of high energy neutron and charged particle radiation. At installations where radiation fields are a mixture of neutron and other types of radiation, the interpretation of nuclear track emulsion is at best difficult and in many instances impossible due to the interference reaction in the emulsion. In many cases near reactors where a degraded fission neutron spectrum is mixed with gamma radiation, no logical results can be obtained from the NTA system since the low energy cutoff of the emulsion may be as high as 0.8 MeV, which is approximately the most probable energy of the fission neutron spectrum.

The shortcomings of emulsions have led to numerous investigations of various luminescent materials, fissionable materials, TLDs, and elements which may be activated by neutrons of various energies. A particular element (rhodium) has been investigated to determine its characteristics when exposed to mixed radiation fields and is of universal interest. This report includes what is known at this time about the characteristics of a rhodium-TLD combination dosimeter developed with the intention of replacing nuclear emulsion film.

History

In early 1963, investigations of the use of various combinations of rhodium cadmium, tin, and iron as filters in front of ordinary beta-gamma dosimetry film started, Figure 1. Rhodium was picked as the prime element of the filter system because it has an unusually large absorption resonance for neutrons of 1.26 eV. The cross section of 1.26 eV is 4500 barns compared to the 2200 meter/sec cross section of 150 barns. The basic idea of the rhodium system is then to cause high energy neutrons incident on the dosimeter and human body to be moderated and backscattered into the rhodium. Any neutron with energy near the 1.26 eV resonance will be preferentially absorbed causing the rhodium to become activated. For a particular incident neutron energy, the activation of the rhodium should be directly proportional to the neutron exposure or total flux.

The path of an incident fast neutron is as follows:

- (1) Penetration through the dosimeter components into the body, or penetration in the body near the dosimeter.
- (2) Moderation in the body tissues.
- (3) Backscatter from the body to the neutron dosimeter.
- (4) Capture by an activation foil in the dosimeter.

Film was first chosen for a number of reasons as the sensitivity material to detect the decay products of the activated foils.⁽¹⁾ Film is extremely sensitive to the beta and gamma radiation emitted by rhodium. The energies of this radiation occur at the peak in the unshielded response of the film. The film is identical to film used in the routine beta-gamma dosimeter used for personnel monitoring; thus, no unusual processing was required for the neutron dosimeter film. When TLD materials became readily available, it was chosen as the sensitivity material because of its characteristics.

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Several combinations of the above metals were used in various thicknesses to achieve the proper response to beta and gamma radiation. These radiations must be taken into account to achieve uniform response at each filter location. The early work from 1963-1965 consisted largely of working out the proper combinations.

Dosimeter and Experimental Design

After many preliminary designs using metal foils of varying thicknesses, a final design was determined by calculation and experimentation in mid 1965. The configuration of the dosimeter is given in Figure 1. The filter materials and thicknesses are as follows:

	<u>γFilter</u>	<u>Fast Neutron</u>	<u>Thermal Neutron</u>
Badge Front	Sn-0.040"	Cd-0.030"	Sn-0.030"
	Stn Steel-0.003"	Rh-0.010"	Rh-0.010"
Badge Back	Pb-0.010"	Pb-0.010"	Pb-0.010"
	Sn-0.010"	Rh-0.010"	Rh-0.010"

With the above filter combinations, the TLD response behind the three filters due to gamma radiation is essentially equal for energies lying in the range 0.020 MeV to 3 MeV.

The response behind the tin-iron filter is produced almost entirely by photon radiation. The response behind the cadmium-rhodium filter is produced by gamma radiation and "body moderated" fast neutrons. Some response is, also, due to prompt gamma radiation after thermal neutron capture in the cadmium. The response at the tin-rhodium filter is produced by gamma radiation, fast neutrons and thermal neutrons.

With this system, three quantities are measured and related to three types of radiation (photon, thermal neutron, and intermediate to fast neutron) through a set of simultaneous equations. The filters are of sufficient density thickness to yield essentially zero response behind all three filters for beta particles of energies <3 MeV. The response to beta radiation is assumed to be zero in the following derivations. Generally, in a mixed radiation field, the following equations describe mathematically the TLD response behind each filter region: (1)

$$D_{\gamma A} + D_{N_f A} + D_{N_t A} = D_A \quad (1)$$

$$D_{\gamma B} + D_{N_f B} + D_{N_t B} = D_B \quad (2)$$

$$D_{\gamma C} + D_{N_f C} + D_{N_t C} = D_C \quad (3)$$

where

$D_{\gamma A}$ = Response behind filter A due to photon radiation.

$D_{N_f A}$ = Response behind filter A due to intermediate and fast neutron radiation

$D_{N_t A}$ = Response behind filter A due to thermal neutron radiation

D_A = Total response behind filter A.

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The symbols in equations 2 and 3 are defined in a similar way for filters B and C.

The photon absorption properties of all three filters is such that the response behind each filter is equal when the system is exposed to photon radiation only.

$$D_{YA} = D_{YB} = D_{YC} \quad (4)$$

Filters B and C are chosen so the response is equal when the system is exposed to intermediate and fast neutrons.

$$D_{NF B} = D_{NF C} \quad (5)$$

All three filters provide unequal responses to thermal neutron.

$$D_{Nt A} \neq D_{Nt B} \neq D_{Nt C} \quad (6)$$

Using the condition given in equation 4 and subtracting equations 2 and 3, the result is a relation between densities due to thermal neutrons.

$$(D_{Nt C} - D_{Nt B}) = D_C - D_B \quad (7)$$

As long as the assumptions hold, this relationship can be used to measure thermal neutron exposure, since the difference ($D_C - D_B$) is due only to thermal neutrons.

After subtracting equations 1 and 2 and applying condition 4, the resulting equation gives the relationship of the response due to fast and thermal neutrons.

$$(D_{NF B} - D_{NF A}) = D_B - D_A - (D_{Nt B} - D_{Nt A}) \quad (8)$$

$$\text{but } (D_{Nt B} - D_{Nt A}) = k(D_{Nt C} - D_{Nt B}) \quad (9)$$

The quantity ($D_{Nt B} - D_{Nt A}$) must be determined by exposures in a "pure" thermal neutron field. The constant K can be determined and equation 8 modified to:

$$(D_{NF B} - D_{NF A}) = D_B - D_A - k(D_{Nt C} - D_{Nt B}) \quad (10)$$

Thus, once k is determined, it is possible to separate the difference due to fast and intermediate neutrons from the difference due to thermal neutrons. These differences are then related to the neutron exposure. For gamma and low energy photon radiation, response reading at the tin-iron filter is calibrated to exposure.

Three quantities of importance can be determined: exposure to electromagnetic radiation, to thermal neutrons, and to fast and intermediate neutrons. The basic problem with this type of neutron dosimeter is its non-linear energy response. The non-linearity makes necessary a calibration of the dosimeter in a fast neutron field closely duplicating the neutron spectrum to be encountered in the dosimetry application.

To take the place of the human body during design and exposure, a large piece of polyethylene was used. Dosimeters were attached to the piece of polyethylene and exposed to various radiations from isotopic neutron sources

(PuF_4 and PuBe) and a positive ion accelerator. The neutron energy spectrum from PuF_4 is continuous from low energies to approximately 3.0 MeV. The average energy of the PuF_4 neutron spectrum is between 0.8 MeV and 1.0 MeV. The PuBe neutron source produces neutrons in several energy groups from fission neutron energies (~0.8 MeV) to approximately 10 MeV.⁽²⁾ Neutrons produced at the positive ion accelerator are very nearly monoenergetic when a thin target is used. The range of neutron energies available is from ~10 keV to as high as 16 MeV, though all these possible energies were not used. Dosimeters were exposed to monoenergetic neutrons from about 0.027 MeV to 5.0 MeV to determine the energy response of the dosimeter system.

Thermal neutron exposures were conducted in a calibrated paraffin tank using the PuF_4 and PuBe sources to supply the neutrons. The paraffin tank was calibrated by exposing gold foils in the tank and in a standard graphite pile where the thermal neutron flux is known.

Gamma radiation from radium and cobalt-60 sources and X-rays from 16 keV to 170 keV were used in developing the filter system and measuring the response of the final dosimeter design. A number of K-fluorescent X-ray sources up to 100 keV are available, while above 43 keV, filtered X-rays were used.

Exposures and Results

The first step in developing the dosimetry system is to achieve gamma equivalence for all three metal filters. This is a process of calculation and trial until a satisfactory compromise is reached. For experimental purposes, the X-ray sources and radium and cobalt-60 sources were used. A large number of exposures were made to various energies of gamma and X-ray radiation. The gamma energy response was then determined by measuring the relative sensitivity as a function of energy while keeping the exposure constant. The result is shown in Figure 2. The TLD response readings for this study are shown in Table I.

To determine the thermal neutron response and obtain a value for k to use in evaluating fast neutron exposures, a set of thermal neutron exposures was made using the PuF_4 source in a large paraffin tank. The amount of paraffin between the source and the badge is adequate to moderate most of the fast neutrons emitted from the source, but a few fast neutrons are still present. Thus, the determination of the constant k is somewhat inaccurate. The results for thermal neutron exposures are shown in Figure 3. For the measurements made, the average value of k was found to be 1.6.

Exposure response for fast neutrons was measured using a PuF_4 source with varying lengths of exposure time. The response is linear with exposure. The minimum detectable exposure is about 10 mrad for the PuF_4 spectrum. These data are shown on Figure 4. The fast neutron energy response was measured using a positive ion accelerator. Exposures were made to fast and intermediate energy neutrons from 0.027 MeV to 5.0 MeV and the difference in the cadmium-rhodium and the tin-iron filters determined. This difference is related to the fast neutron exposure as shown by the equations in the design section of this report. While this response is related to fast neutron exposure, the magnitude of the difference for a given exposure of 100 mrad is a function of the incident neutron energy. The observed neutron energy response for the experimental exposures between 0.027 MeV to 5.0 MeV varies by a factor of about 100. In all fast neutron exposures, there was no significant thermal neutron contribution. Neutron doses were calculated using the curve in NBS handbook 63. Total neutron fluence needed to accumulate the desired dose was calculated by using known long counter and BF-3 tube responses available from the Radiological Physics group. Errors in this information are not (under usual conditions) greater than $\pm 20\%$.

As can be seen from Figure 5, the energy response varies across the entire energy spectrum. Thus, the dosimeter, to be useful, would have to be calibrated in a neutron radiation field quite similar to the field where personnel dosimetry is required. In situations where it is convenient to make such a calibration and where no large amounts of moderating material exist close by, the rhodium filter system can perform adequately. This dosimeter could be used to advantage where plutonium is being processed through the PuF₄ stage, but the dosimeters would need not be limited to use at those locations. If an exposure to a different spectrum were incurred, say about 0.4 MeV to 2.0 MeV an error of no greater than 2.0x would be realized.

Conclusion

In the preceding discussion, the various formulae and experimental measurements leading to a personnel neutron badge have been presented. The particular filter system described has been developed and checked to a logical termination. The rhodium filter badge does provide an excellent improvement over the WTA film badge, in that it offers good sensitivity for thermal and intermediate neutrons (whereas WTA offers no sensitivity at these energies) and improved sensitivity at high energies. As has been stated before, the accuracy of this system is good if the neutron spectrum is known. The fact that TLD is used, permits convenient handling of the badge, and does not present the normal problems associated with film--those of heat fogging and gamma-ray background.

References

- (1) BNWL-339, Dosimetry Technology Studies, Dosimetry Technology Staff, November, 1966.
- (2) NBS Handbook 72, page 64.

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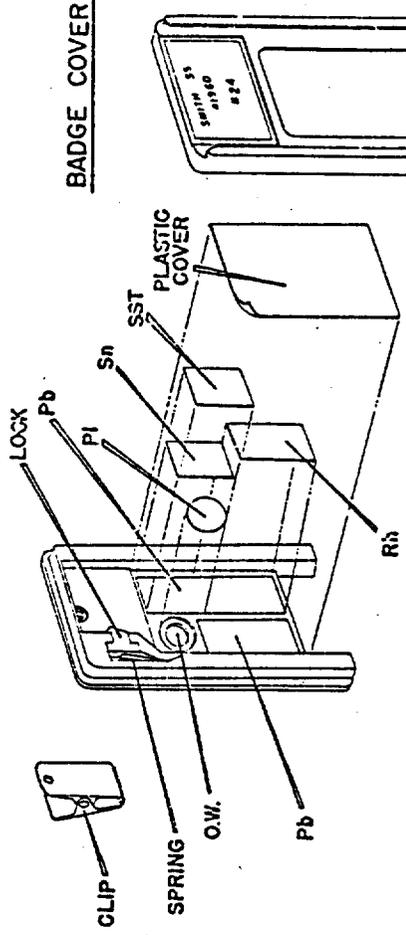
TABLE I

RESPONSE

<u>Energy</u> <u>(keV)</u>	<u>Y</u> <u>Filter</u>	<u>N_f</u> <u>Filter</u>	<u>N_t</u> <u>Filter</u>
16	130	130	140
23	380	370	380
34	480	480	510
43	520	540	540
62	510	490	510
82	420	400	430
100	430	450	420
120	450	440	410
170	470	460	470
Ra-γ	470	480	470
⁶⁰ Co	460	470	470

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BADGE BACK



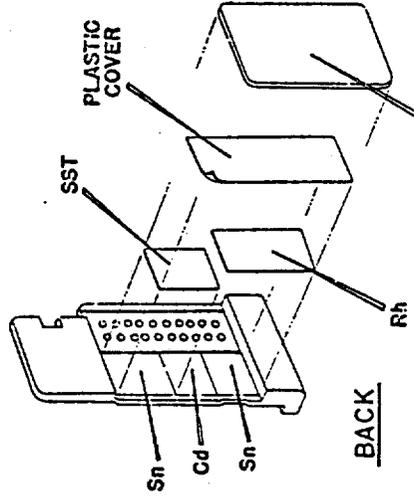
BADGE COVER



SECURITY CREDENTIAL

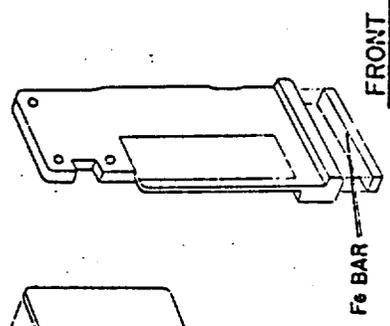


ASSEMBLY



BACK

BADGE SLIDE



FRONT

DOSIMETER TECHNOLOGY
JANUARY 10, 1965

FIGURE 1
Activation Neutron Dosimeter

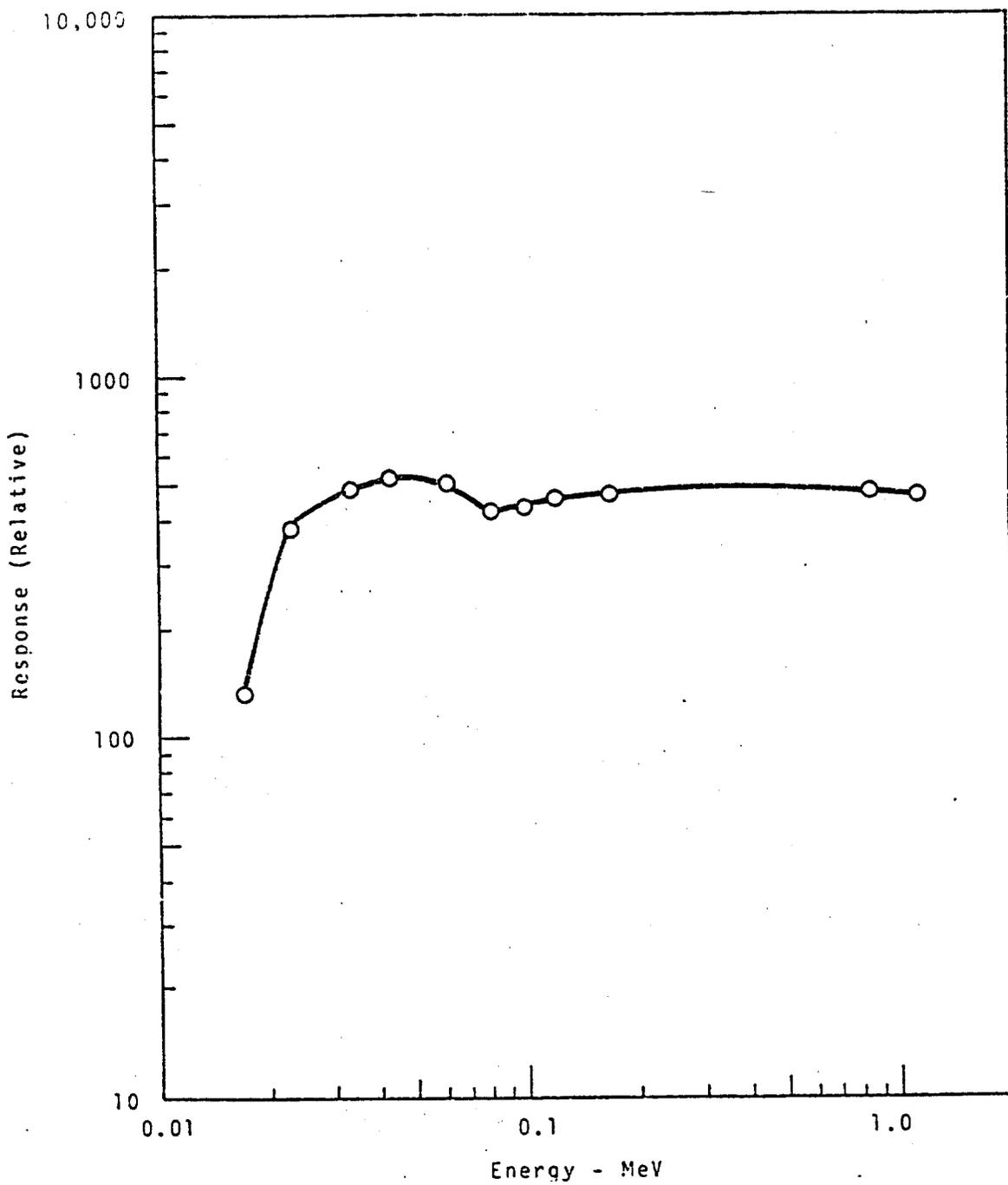
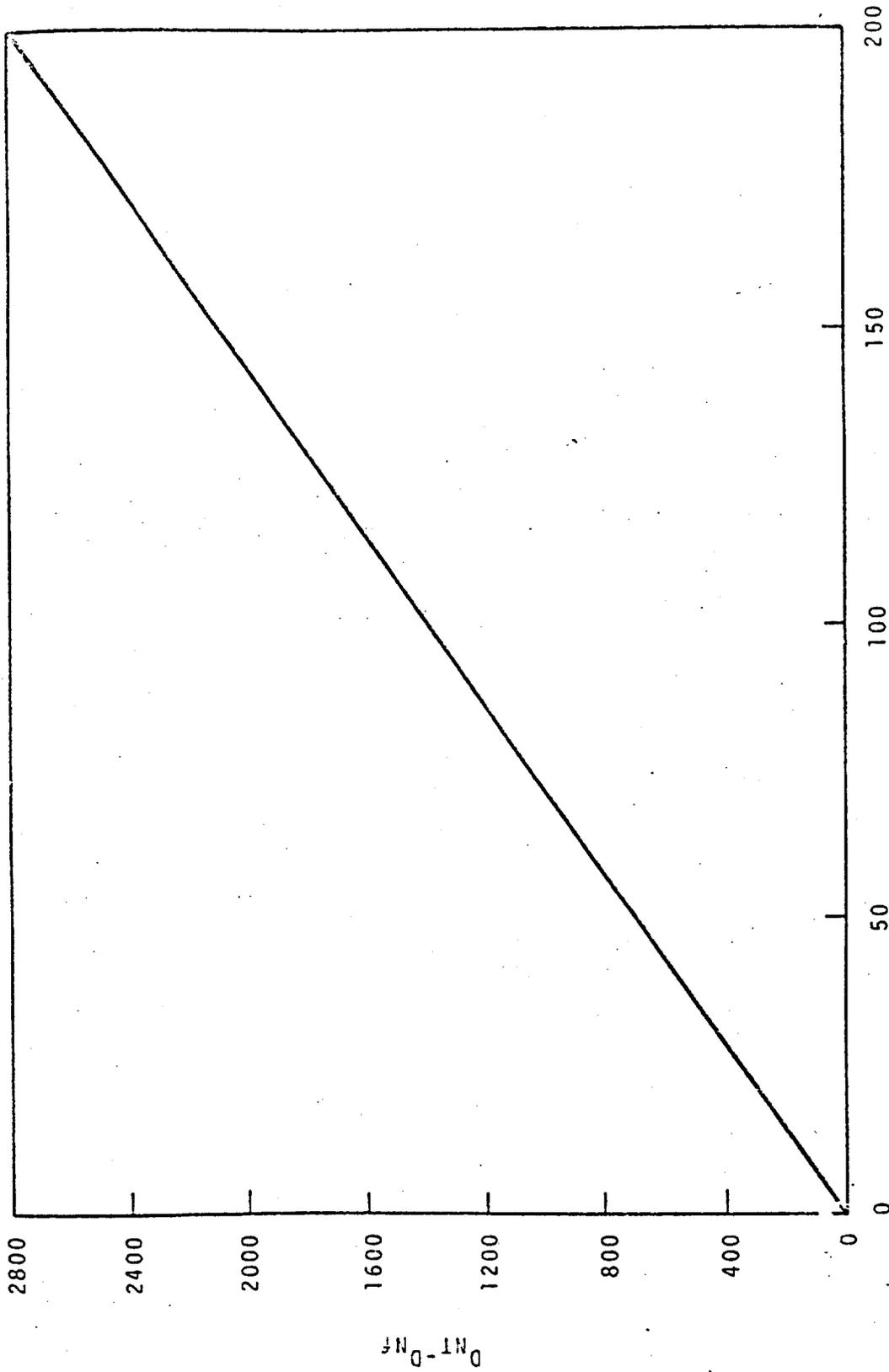


FIGURE 2
Gamma Energy Response Characteristic

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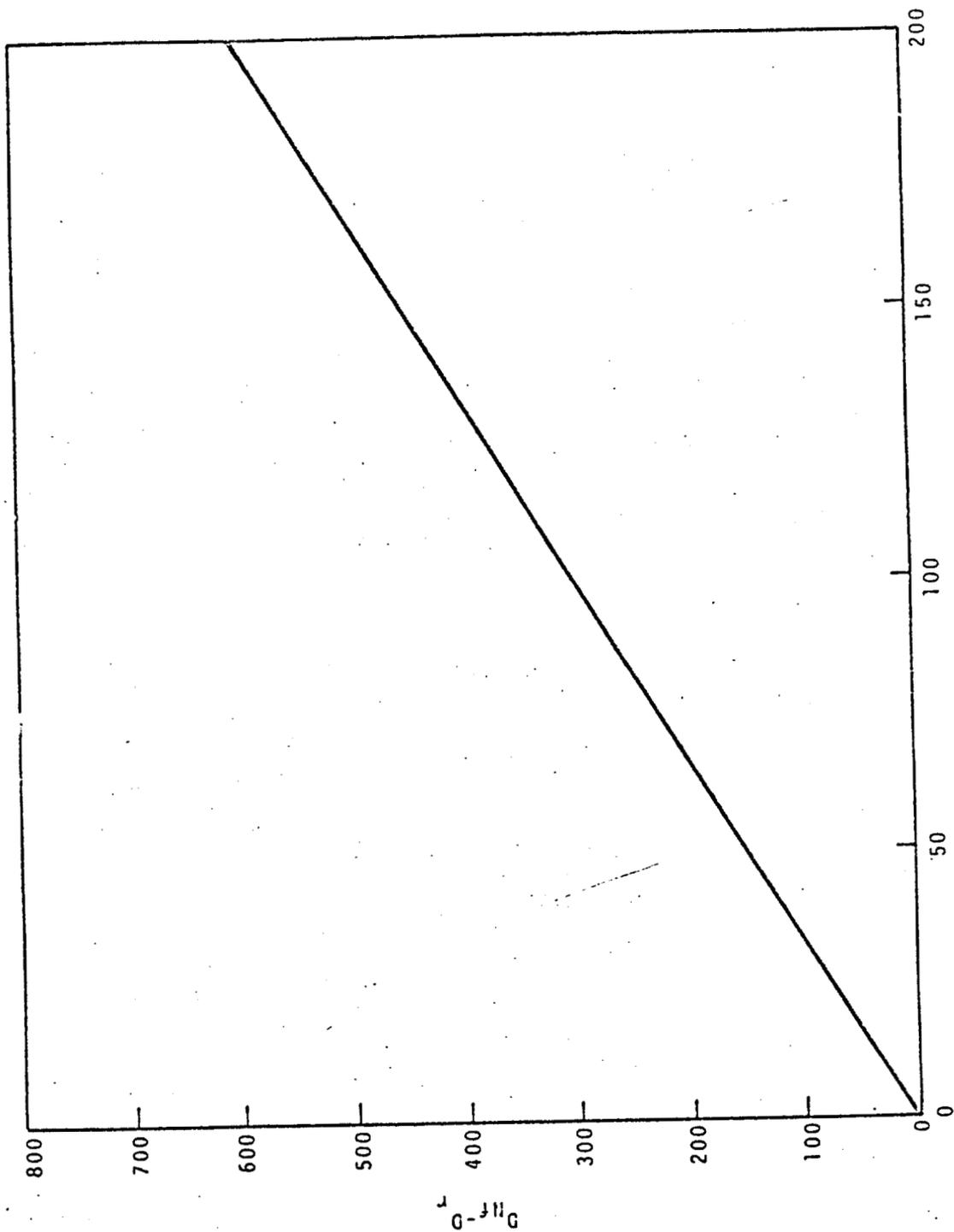


Thermal Neutron Exposure (mrads)

FIGURE 3

Thermal Neutron Response

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Fast Neutron Exposure (mrads)

FIGURE 4

Fast Neutron Response

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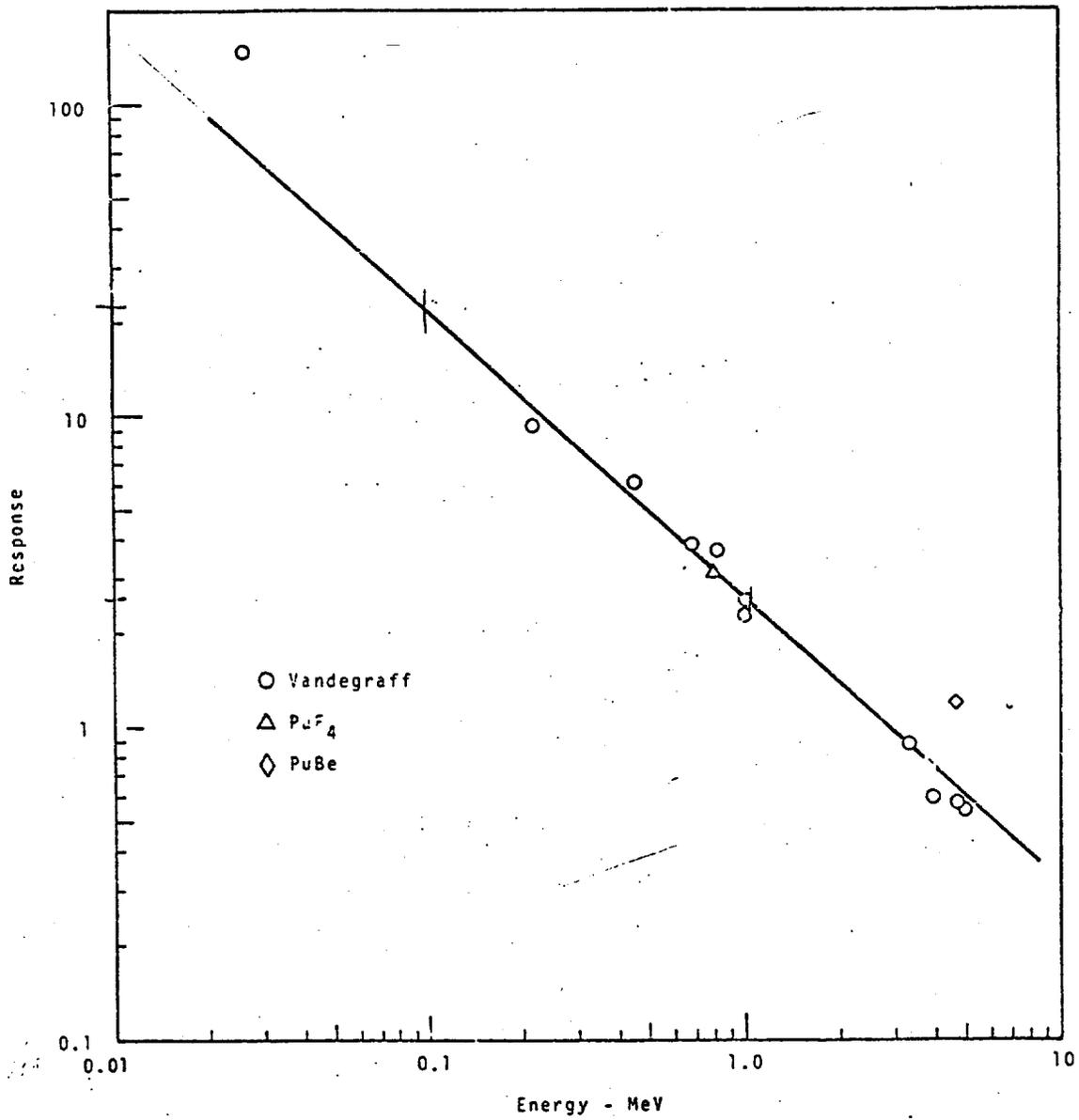


FIGURE 5
Neutron Energy Response Characteristic

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