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NEUTRON DETECTION BY MEANS OF FILM BLACKENING AND RELATED SHIELDING PHENOMENONS

by

LCDR T. E. Shea, Jr.,* MSC, U. S. Navy

Date Issued MAR 31 1950

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NEUTRON DETECTION BY MEANS OF FILM BLACKENING AND RELATED SHIELDING PHENOMENONS

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Introduction

Blackening effect, rather than the production of film tracks, as a means for measuring fast neutron fluxes was reported by Kalmon.[1] Lapp and Andrews[2] list proton recoils in the Wilson Cloud chamber, ionization chambers, and photographic emulsions as methods for measuring fast neutron fluxes, also the displacement of normal atoms from their positions in crystal lattices with measureable changes in the electrical or thermal conductivity. Of these methods, the reduction of silver halides in photographic emulsions seems to offer a practical solution in Health Physics personnel monitoring and is reported by Yagoda[3] and Cheka.[4,5] All the reporting investigators (except Kalmon) used film track formation in specially prepared emulsions to obtain their findings. It has been suggested that blackening effect in films not specially prepared for neutron detection be studied, and their suitability for Health Physics personnel monitoring be evaluated.

Materials and Methods

Four commercially available types of Eastman Kodak films and one Ansco Company film were purchased for use in the tests. Stocks of commercial film available at the laboratory failed to give readable unexposed controls, so all film was purchased at locations removed from possible project radiation and was used before the manufacturer's recommended date for discarding the film. All films were packed for testing in light tight packages containing a single

(1) Kalmon, Ben, *The Blackening Effect of Neutron and Gamma Radiation on Dupont 552 and Eastman V-120 Film*, ORNL 394 (November 16, 1949).

(2) Lapp, R. E. and Andrews, H. L., *Nuclear Radiation Physics*, Prentice-Hall, New York, N.Y. (1948).

(3) Yagoda, Herman, *Radioactive Measurements with Nuclear Emulsions*, John Wiley and Sons, Inc., New York, N. Y., (1949).

(4) Cheka, J. S., *Neutron Monitoring by Means of Special Fine-Grain Alpha-Emulsion Film*, MDDC-890 (April 3, 1947).

(5) Cheka, J. S., *Neutron Monitoring by Means of Nuclear Track Film—Supplementary Report*, ORNL-547 (January 9, 1950).

test film in black paper and had one thickness of paper between the film and the source, although several thicknesses of paper were behind the film to complete the envelope style packet.

All films tested were packed in complete darkness, exposed to the source, opened and developed in complete darkness. The developing and fixing procedures were the same as recommended in the Eastman Kodak Company Handbook[6] for the type film studied, with time and temperature corrections made from the recommended Eastman curves. The Ansco film was developed and fixed according to the routine method of the Oak Ridge National Laboratory personnel monitoring system, using Dupont X-ray developer and fixer.

During exposures the films were held on the top of aluminum rods at fixed distances from the source. A series of positions (Fig. 6) was accurately determined on the plate, and the rods holding the test film were screwed into the plate in concentric circles at 15, 20, 25, and 30 centimeters from the source position. This permitted a maximum of eight films to be exposed at one time to the source, and since the rods holding the source and films were of standard length, a uniform geometry resulted, except that the width of the exposed film would form either the chord or the tangent of the circle, and either the edges or the center would receive a slightly larger dose. On the films tested it was not possible to measure this small difference in the blackening, so it was not corrected for in reporting this study.

The exposures were made with the face of the film perpendicular to the beam from the source, having an "open window area" (covered only with paper to keep it from being exposed to light) and a shielded area covered on the front and the back of the film by 0.07 in. thick lead. A portion of all the test films was exposed without shielding (referred to as open window), but in some tests, which will be qualified, the lead was thickened or aluminum filters plus various thicknesses of lead were added.

The fast neutron source was a Po-Be source in a metal jacket, the composition of which was not known. The flux was determined by calculation using the formula

$$\text{hours exposure} = 4\pi D^2 (A)_t / 3.6 \times 10^3 N_o (e^{-\lambda t}),$$

where D = distance from center of source to plane of exposed film,

(6) Eastman Kodak Reference Handbook (1945).

A_t = the number of neutrons to which it was desired to expose a cm^2 of test film at time t,

N_o = neutrons per second given off by Po-Be source at time $t = 0$,

λ = half-life of Po.

Results

Five commercial films (Eastman Kodak Test Films 1, 2, 3, and 4, and Ansco Test Film 1) were exposed to a fast neutron source as described under the descriptive heading of methods and materials. Tables and figures give blackening density of the film corresponding to the dose (in units of .1 rep) representing a given number of neutrons per cm^2 and the equivalent number of weeks tolerance.

Eastman Kodak Ortho Film. Table 1 and Fig. 1 show the results of exposing Eastman Kodak Ortho type film to a Po-Be source. The table shows the readings of film blackness resulting from the calculated flux. The column headed "Weeks Tolerance" is the equivalent number of weeks allowed exposure using a value of 1 rep of fast neutrons $\cong$ 4 Mev energy $\cong 2.2 \times 10^8$ N_f/cm^2 or 0.1 rep of fast neutrons $\cong 2.2 \times 10^7$ N_f/cm^2.[7,8] The figures show a plot of blackening *vs* exposure to

Table 1

NEUTRON FLUX			DENSITOMETER READING	
Film	N_f/cm^2	Weeks Tolerance	Open Window	0.07 in Pb Shield
1	0	0	zeroed	zeroed
2	2.5×10^7	1.1	0.0016	0.0023
3	5.0×10^7	2.2	0.018	0.023
4	7.5×10^7	3.4	0.025	0.059
5	1.0×10^8	4.5	film light struck	
6	2.5×10^8	11.4	0.088	0.42
7	5.0×10^8	22.7	0.27	--
8	7.5×10^8	34.1	0.40	0.56

fast neutrons in allowable weekly tolerance for an eight hour day, five day work week. All other tables and figures will represent the same treatment of the data for the film concerned.

(7) Gamertsfelder, C. C., *Calculations for Use With the Fast Neutron Meter*, AECD-2173 (July 21, 1948).

(8) Cheka, J. S., *Op. Cit.* (ORNL-547).

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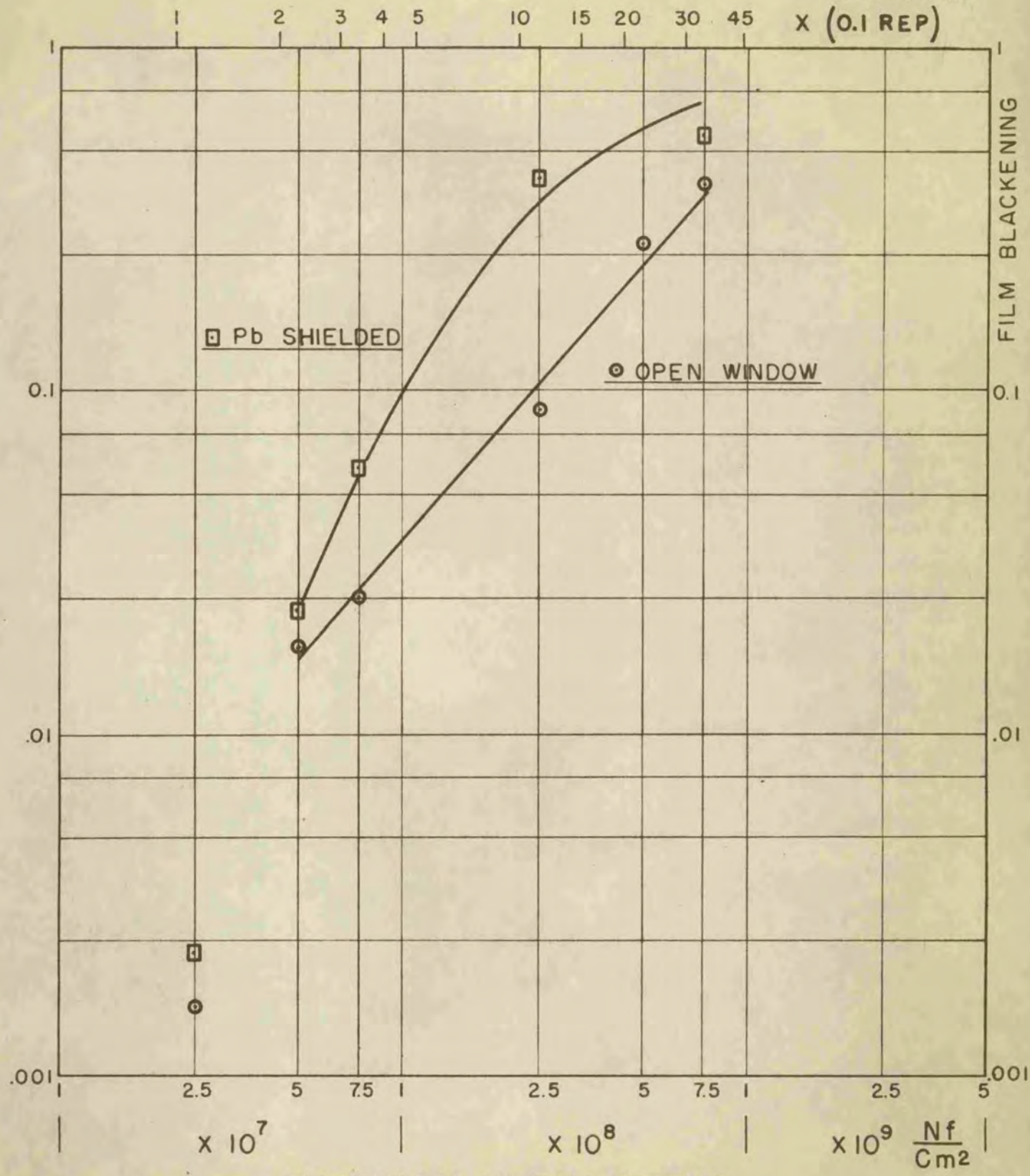

EASTMAN KODAK ORTHO

FIG. 1

Eastman Kodak Verichrome Film. Table 2 and Fig. 2 result from exposing Eastman Verichrome film to calculated numbers of neutrons from a Po-Be source.

Table 2

NEUTRON FLUX			DENSITOMETER READING	
Film	N_f/cm^2	Weeks Tolerance	Open Window	0.07 in Pb Shield
10	0	0	zeroed	zeroed
11	2.5×10^7	1.1	0.0018	0.0036
12	5.0×10^7	2.2	0.0073	0.015
13	7.5×10^7	3.4	0.020	0.042
14	1.0×10^8	4.5	0.028	0.044
15	2.5×10^8	11.3	--	--
16	6.0×10^8	27.3	0.12	0.80
17	7.5×10^8	34.1	--	--

Eastman Kodak Plus X Film. Table 3 and Fig. 3 show the results of exposing this type of film to calculated amounts of fast neutrons from a Po-Be source.

Table 3

NEUTRON FLUX			DENSITOMETER READING	
Film	N_f/cm^2	Weeks Tolerance	Open Window	0.07 in Pb Shield
19	0	0	zeroed	zeroed
20	2.5×10^7	1.1	0.0032	0.004
21	5.0×10^7	2.2	0.038	0.042
22	7.5×10^7	3.4	0.044	0.064
23	1.0×10^8	4.5	0.056	0.068
24	2.5×10^8	11.3	light struck	
25	5.0×10^8	22.7	0.12	0.80
26	7.5×10^8	34.1	0.13	0.82

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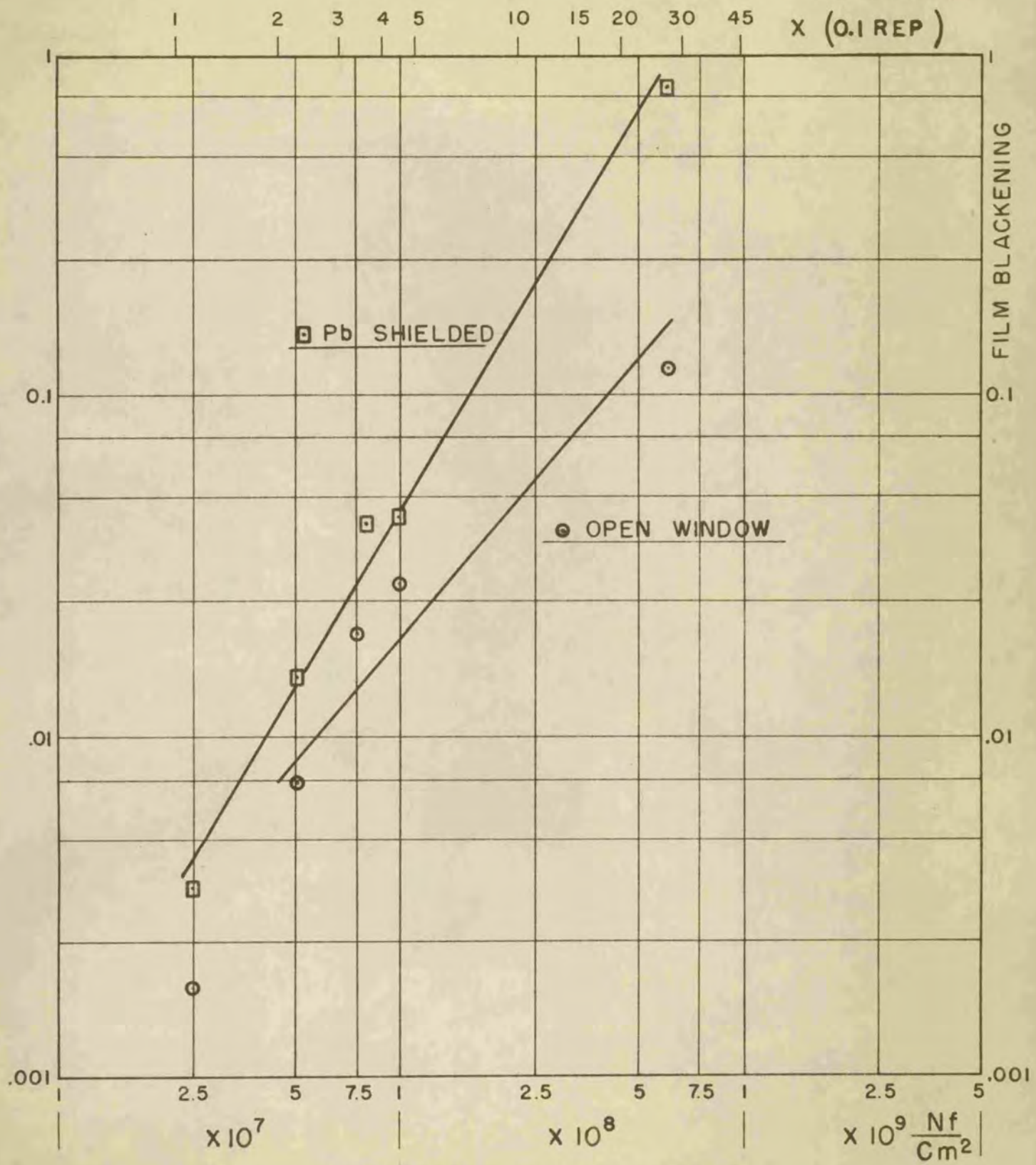
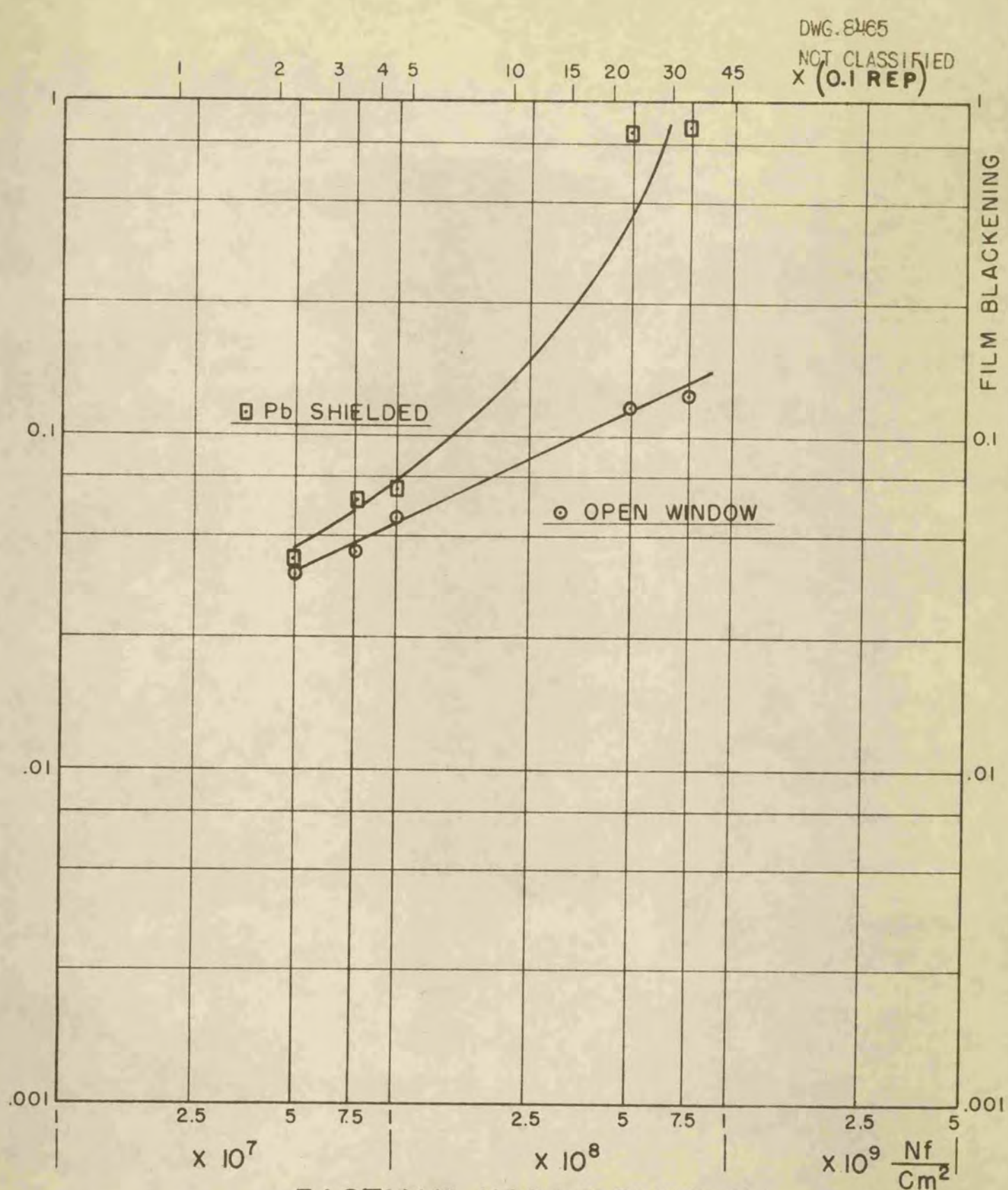


EASTMAN VERICHROME

FIG. 2



EASTMAN KODAK PLUS X

FIG. 3

Eastman Kodak Super XX Film. Table 4 and Fig. 4 represent the results obtained from exposing Eastman Super XX film to calculated numbers of fast neutrons using a Po-Be source.

Table 4

NEUTRON FLUX			DENSITOMETER READING	
Film	N_f/cm^2	Weeks Tolerance	Open Window	0.07 in Pb Shield
28	0	0	zeroed	zeroed
30	4.0×10^7	1.8	0.008	0.036
31	7.5×10^7	3.4	0.040	0.062
32	1.0×10^8	4.5	0.070	0.072
33	2.5×10^8	11.3	0.096	0.14
34	5.0×10^8	22.7	0.13	0.22
35	7.5×10^8	34.1	film light struck	
36	1.0×10^9	45.5	0.140	0.40

Ansco Process Film. Table 5 and Fig. 5 represent the results of exposing Ansco Company Process film to calculated numbers of fast neutrons from a Po-Be source.

Table 5

NEUTRON FLUX			DENSITOMETER READING	
Film	N_f/cm^2	Weeks Tolerance	Open Window	0.07 in Pb Shield
37	0	0	zeroed	zeroed
39	5.0×10^7	2.3	0.0056	0.01
40	7.5×10^7	3.4	0.007	--
41	1.0×10^8	4.6	0.011	0.02
42	2.5×10^8	11.4	0.02	0.04
43	5.0×10^8	22.8	0.058	0.10
44	7.5×10^8	34.1	0.070	0.120
45	1.0×10^9	45.5	0.10	0.150
46	1.4×10^9	51.8	0.165	0.244

Ansco Process film, having shown a response to neutron flux plotted as a straight line, was further tested to determine, if possible, the blackening effect of the fast neutrons in the mixed field of radiation of a Po-Be source.

The above tables represent the blackening by neutrons and all other radiation present. While, theoretically, Po-Be should not originally contain any large amount of radiation other than neutrons, it is unavoidable that

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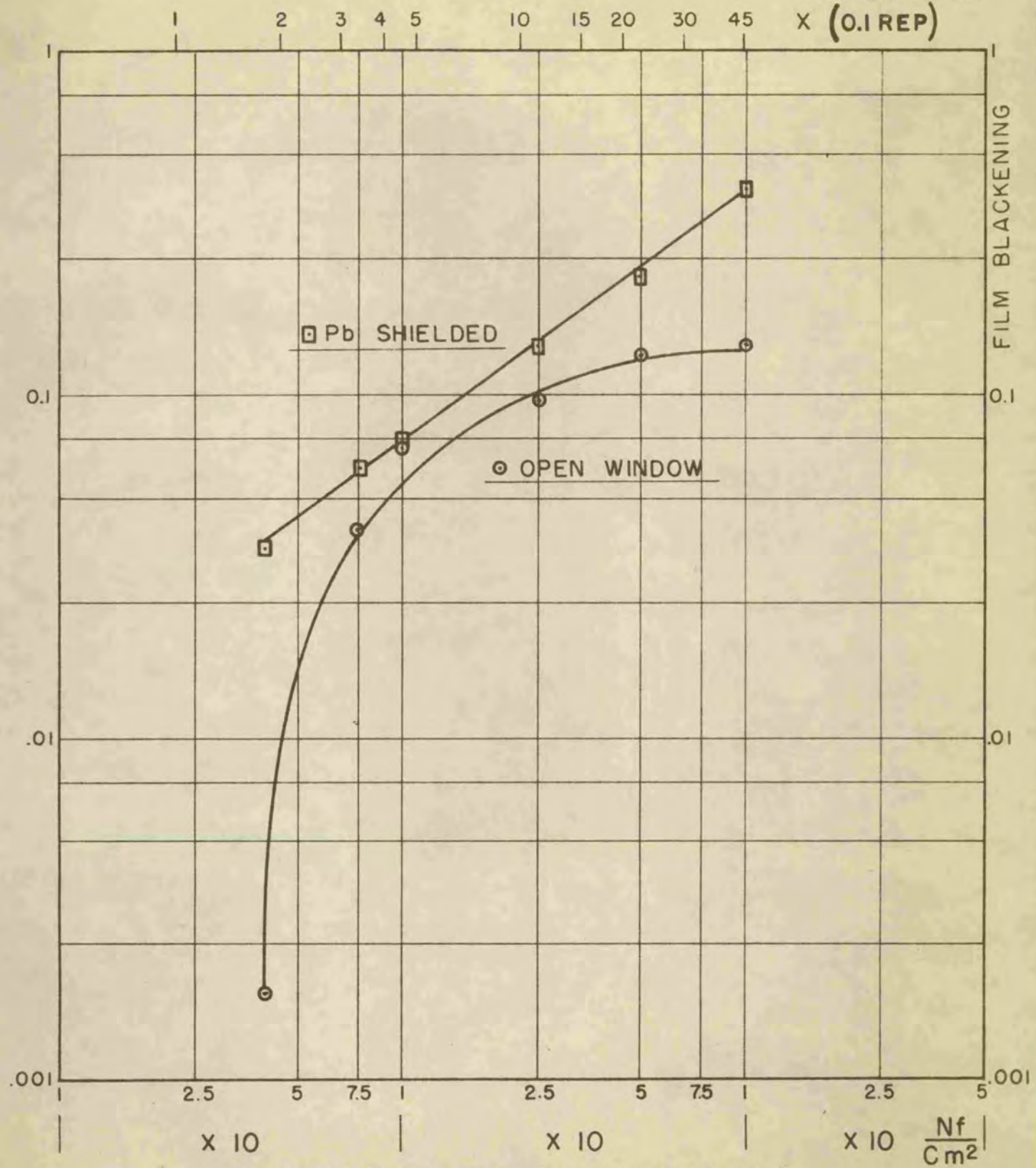

EASTMAN KODAK SUPER XX

FIG. 4

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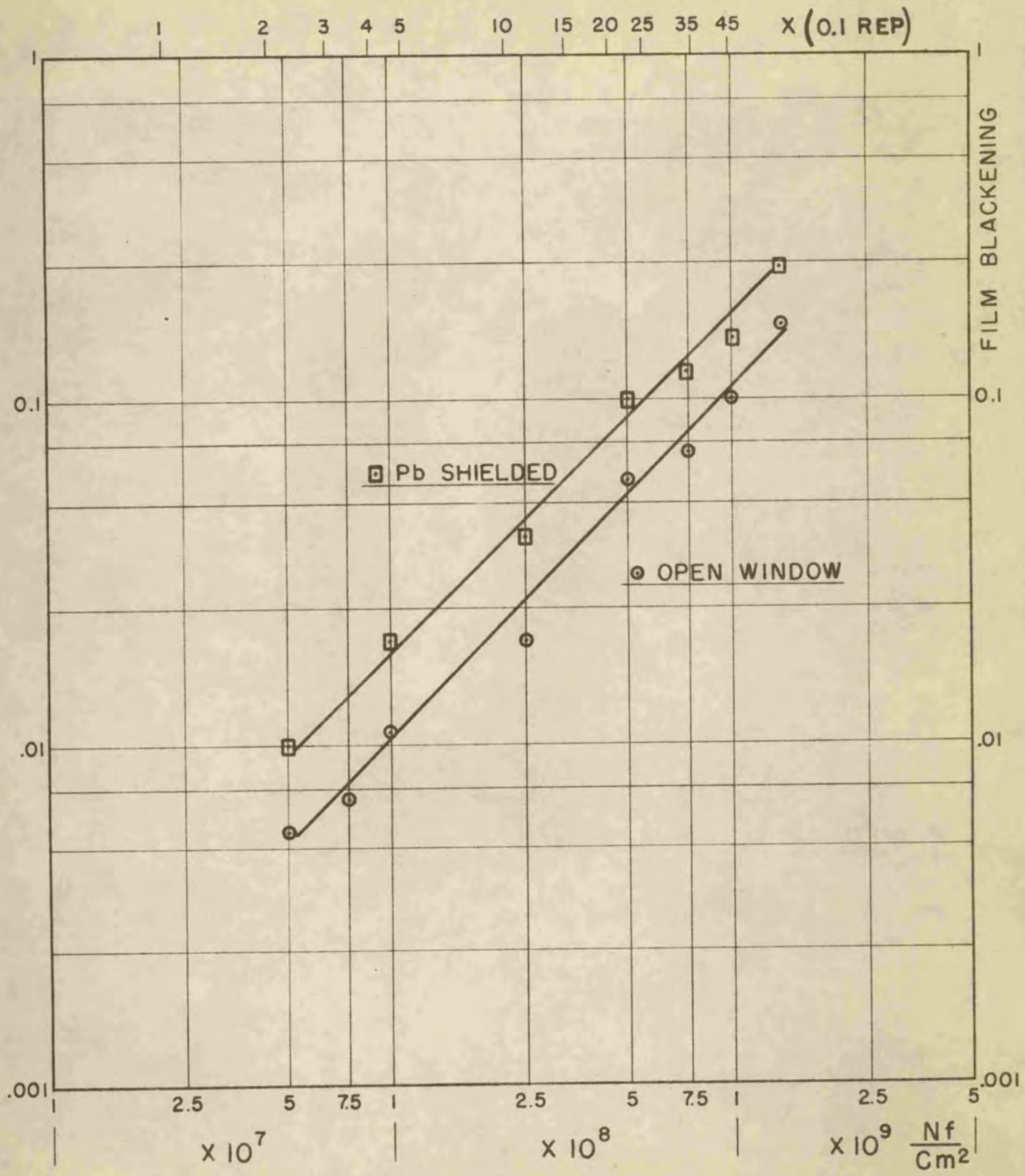


ANSCO PROCESS

FIG. 5

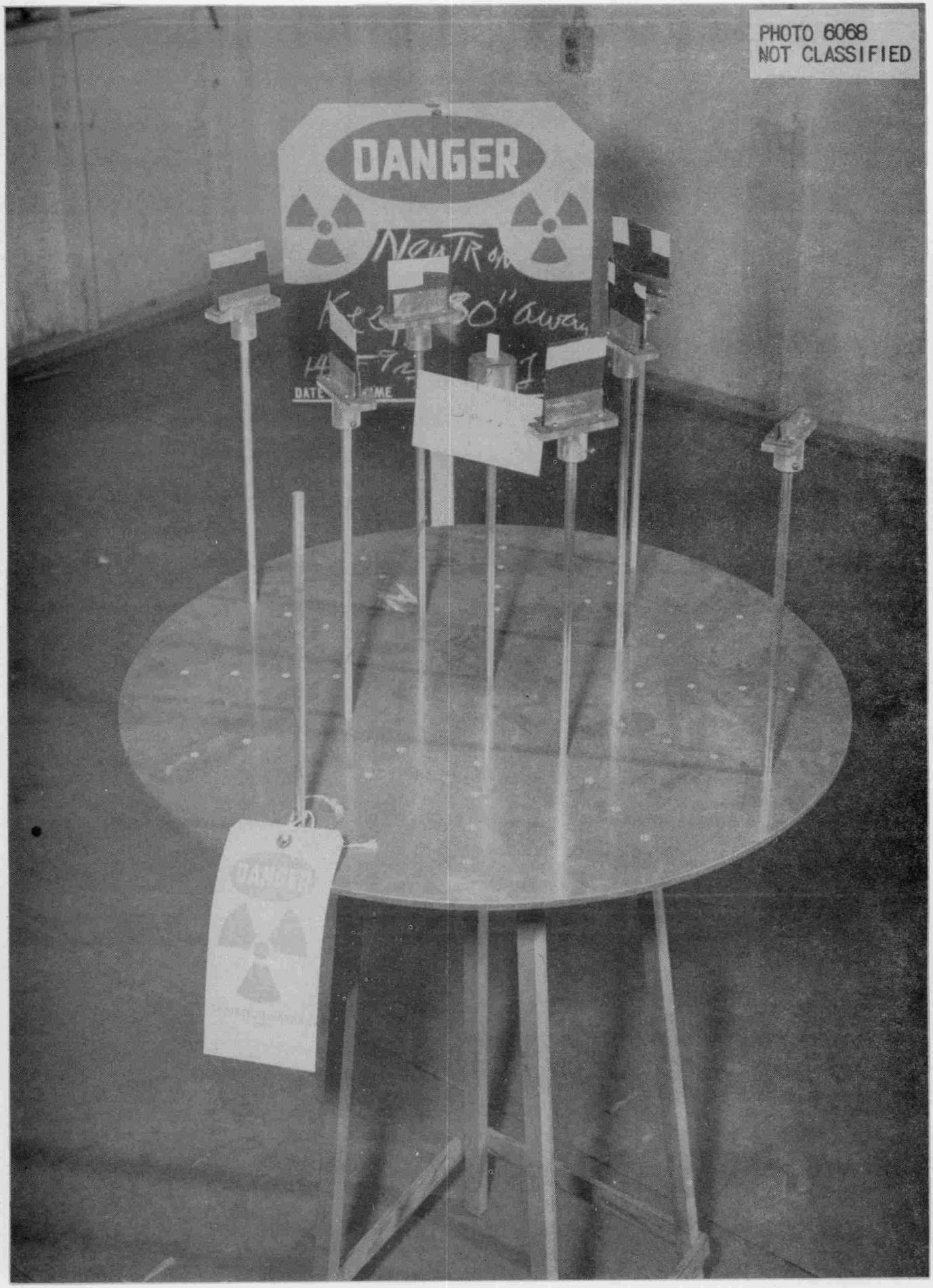


FIG. 6

gamma and photoelectrons be produced in the container, in the lead shield, and in the beryllium where the neutrons are produced. It is important, therefore, to investigate what portion of the blackening resulted from the neutrons, and what amount of blackening resulted from other radiation. Under practical conditions X rays and electrons will always be present.

Primary Gamma Source (Radium). Four films were exposed to primary gamma from a radium source, whose average energy of 0.8 Mev approximates the 0.77 Mev[9] single energy gamma of polonium. The radium source was shielded by a platinum capsule which permitted only the gamma rays to escape; the films had open windows and were exposed as follows:

1. 0.07 inches Pb exposed to 0.55 r of gamma.
2. 0.07 inches Pb + aluminum thickness varying from 0.5 to 1.5 mm, exposed to 0.55 r of gamma.
3. 0.07 inches of Pb only exposed to 1.1 r of gamma.
4. 0.07 inches of Pb only + aluminum thickness varying from 0.5 to 1.5 mm, exposed to 1.1 r of gamma.

After being brought to zero with the unexposed film, the densitometer readings on the various films were as follows:

	Open Window	Shielded Portion
1.	0.070	0.065
2.	0.054	0.040
3.	0.122	0.120
4.	0.118	0.078

The readings showed a reduction in blackening behind the shield in all cases as compared to the blackening in the open window portion exposed to equal doses; a reduction in blackening behind the shielded portion, when aluminum was interposed between the Pb shield and the film, was likewise evident.

Primary Gamma Sonrce (Unshielded Cobalt). Four films were exposed to an *unshielded* cobalt source of primary gammas. The gamma from cobalt averages 1.24 Mev (50 percent 1.16 Mev, 50 percent 1.32 Mev);[10] it is therefore, for all practical test purposes, monoenergetic, and not the average of a wide gamma spectrum as is that from radium. The unshielded exposure at 15 cm distance was 1.1 r for the four films exposed, but the shields were varied in front of the films in the following manner:

(9) Siegbahn, K. and Statis, H., "Gamma Radiation from Polonium and from Lithium Bombarded with Alpha Particles", *Nature*, V. 159, 471 (1947).

(10) "Nuclear Data Committee, Summary of Nuclear Data, 1947," *Nucleonics*, Vol. 2, No. 5, Part 2, 171 (May 1948).

1. 0.07 inches Pb only.
2. 0.07 inches of Pb + 0.5 to 1.5 mm Al thickness.
3. 0.21 inches of Pb only.
4. 0.21 inches of Pb + 0.5 to 1.5 mm Al thickness.

The densitometer readings above the transmission through an unexposed film were

Film	Open Window	Shielded Portion
1	0.239	0.218
2	0.328	0.220
3	0.219	0.182
4	0.300	0.172

which in all cases showed true shielding effects when the open window readings of the shielded portion of the film are compared to the densitometer readings of the shielded portion of the film. While all distances were originally thought to be at 15 cm (for this particular test), remeasurement showed (2) and (4) to be at 14.25 cm and the other two at 15. While this would account for some of the increased density of 328 and 300 open window readings, individual film response will have to account for the high open window reading of (2).

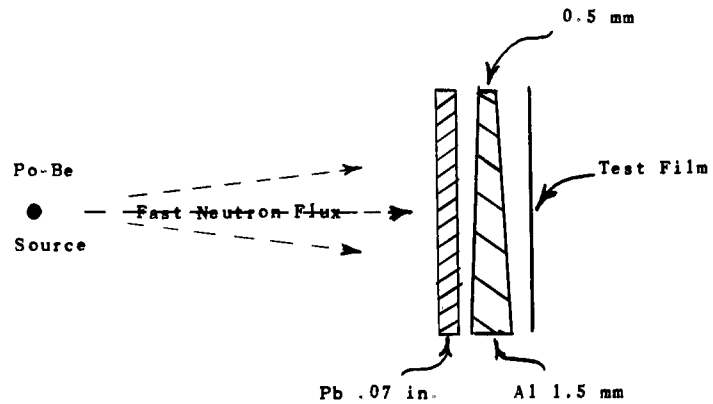
A Primary Gamma Shielded Source. The cobalt source was enclosed in a cylinder of Pb closed at top and bottom and having 1.5 inch lead walls. Two films were exposed to the source, both films having an open window portion plus a shielded portion in front of the film.

Film	Distance from Source	Shield	Density Open Window	Density Shield
1	15 cm	0.21 in. Pb	0.316	0.238
2	14.25 cm	0.21 in. Pb + Al (.5 to 1.5 mm thick)	0.282	0.24

The aluminum filters were placed between the lead and the front of the film, so that the radiation from the original source had to pass through 1.5 inches of lead, then 15 cm (less the space for shielding) of air, and then in the case of film (2) the radiation emerging from the back of the 0.21 inches of lead was filtered through the aluminum.

An Unfiltered Fast Neutron Source, Film Shielded with Lead and Aluminum. Two film packs were exposed to 1×10^9 N_f/cm^2 from an unfiltered Po-Be source. An aluminum wedge was placed between the 0.07 inches of lead and the film (as

illustrated), intending to cut off blackening effect from soft X rays and secondary electrons, provided it was the direct cause of the additional blackening behind the shield.



Open window blackening varied only from 0.144 to 0.145 after setting the densitometer to zero with a blank film. Blackening at the thin end (0.5 mm) of the aluminum wedge was 0.159 while that near the thick edge (1.5 mm) of the aluminum gave a reading of 0.098.

Open Window	Shield .07 inches Pb + mm Al	Shielded Density	True Shielding (Percent)
0.145	0.5	0.159	-10
0.144	1.5	0.098	+32

Tracings made on a micro-densitometer showed that the true shielding effect of the thick end blended into the intensification phenomenon at approximately .7 mm of aluminum. This thickness of aluminum could only take out soft X rays and electrons. If this is calculated as beta equivalent, the beta or equivalent radiation would be 2.7 grams/cm^3 of Al × 0.07 cm Al = 0.189 grams/cm^2 which would suppress beta radiation of less than 0.6 Mev.[11]

A Primary Fast Neutron Source With Aluminum Between Source and Lead Shield. In order to demonstrate that the presence of aluminum behind the shield acted to screen radiation produced in the lead shield, and was not merely attenuating the primary radiation, a film pack was tested with the aluminum in front of the lead. The open window showed a value of 0.241 and the 0.5 to 1.5 mm of

(11) Lapp, *et al*, *op. cit.*

aluminum + lead shield showed a blackening value of 0.354 or an intensification average of 14.7 percent.

A Primary Fast Neutron Source Through Increased Thickness of Lead Shield. A film pack was exposed to fast neutrons, with a portion of the film being shielded by three times the usually reported thickness of lead. The density behind 0.21 inches lead shield was 0.288, while the value for the open window was 0.21, which is an intensification of 37 percent by the shield.

Proton Recoil Tracks. Process films exposed to a fast neutron flux from a Po-Be source were examined according to the method described by Cheka,[12] and tracks were found in the open window and shielded portion. To demonstrate presence of long chains some of the films were exposed so that the plane of the film was parallel to the beam of neutrons, thus providing means by which many successive silver grains could be blackened. The presence of long chains (more than three successive grains) of proton recoil tracks were readily identified, although this work on tracks did not lend itself to a determination of the relative numbers.

Nuclear Track Films. Nuclear track films prepared especially for maximum sensitivity to neutrons in the presence of gamma[13] were exposed in an attempt to determine the number of neutrons being scattered by the shielding. Where the Po-Be source was *not shielded,* the presence of various shields in front of the films never showed a reduction in proton recoil chains of more than the probable error of the open window reading.

Shield Before Film	Fields Counted	No. of N_f Chains
open window	14	42.3 ± 2.0
0.07 inches Pb	14	39.8 ± 3.5
open window	15	43.8 ± 3.3
0.21 inches Pb	15	40.6 ± 2.1
open window	10	45.1 ± 2.5
0.07 inches Pb	10	41.0 ± 2.8
0.03 inches Al		
open window	10	42.3 ± 3.5
0.07 inches Pb	10	40.0 ± 2.6
0.05 inches Al		

(12) Cheka, J. S., *op. cit.* (MDDC-890).

(13) Cheka, *ibid.*

When the fast neutron source *was* shielded by a 1.5 inch cylindrical lead shield, the loss of neutrons by scattering was found not to be significant behind the shielded portion of the film when compared to the open window portion. Moreover, track densities differed by very little from the exposure with the source not shielded, the two groups shown having been exposed to essentially the same number of primary neutrons.

Shield Before Film	Fields Counted	No. of N_f Chains
open window	24	39.2 ± 2.6
0.07 inches Pb	24	38.9 ± 2.9
open window	10	44.9 ± 3.1
0.21 inches Pb	10	42.1 ± 2.1
open window	10	44.6 ± 2.6
0.07 inches Pb	11	42.4 ± 2.2
0.5 mm Al		
open window	10	42.7 ± 2.5
0.07 inches Pb	10	42.1 ± 3.0
1.5 mm Al		

Discussion

Commercial type photographic film (not especially prepared for radiation detection) was tested to determine if film blackening could be plotted as a function of fast neutron flux or of neutron dosage. Of the films tested in this study, only one type displayed the desired response. The acceptable film was "slow" to light, while the other four were increasingly fast to light from test film (1) through list film (4). Film speed is dependent to a large extent upon grain size, the larger silver halide grains being found in the faster film. The acceptable film became increasingly black as the exposure time to the neutron flux was increased, and it was possible to plot a straight line relationship between flux and density. One other type film tested, of approximately the same film speed, produced greater darkening when exposed to an equivalent neutron flux; however, the response was not linear. Differences in manufacturing techniques plus the fact that the films were intended for use in "light photography," and thus were not subject to controls to insure uniform response to wave lengths outside the visible light spectrum, might explain the variation between the two films.

The small total cross section of about 2.2 barns[14] for lead made it advisable to study the radiation effect that was really causing the intensification behind the shield. Since both fast neutrons and gammas are given off from a Po-Be source, gamma radiation without neutrons present was investigated. As shown in the section of this report giving the test results of primary gamma radiation, both radium and cobalt showed less film blackening behind the film shield than resulted in the open window. This true shielding effect was the reverse of the blackening intensification resulting from the Po-Be source.

Aluminum wedges were then inserted between the lead shield and the film, and it was found that the intensification resulting from exposure to a primary neutron source was reversed by a 0.7 mm aluminum filter.. This suggested that inelastic scattering in lead nuclei caused blackening from the gamma or nuclear excitation, or the ionization in the lead from secondary gamma caused the blackening from the resulting photo-electron emission.

The lead shield was thickened from 0.07 to 0.21 inches of lead, and while there was less blackening behind the shield than with the thinner shields, there was still 13 percent greater density when the radiation from the Po-Be source went through the lead. This would indicate that more tests should be made to determine if a certain thickness of lead shield would produce a maximum intensification for a given average energy of fast neutrons. When gamma sources free of neutrons were used against even the thin lead shields, true shielding effects were noted.

The Eastman NTA films, which are relatively insensitive to gamma, showed the neutrons behind the close lead shield to have been reduced by less than the probable error when the proton recoil tracks were counted and compared to the open window. In the test films, recoil proton tracks resulting from the primary neutrons were demonstrated in films exposed at right angles to the beam of the neutron source; there were tracks in the area of the open window and also behind the shield. These tracks could be identified as long chains when the films were exposed parallel to the beam. None of these films could be used to determine the numbers of neutrons in the flux at the exposure distance by counting the chains, because of the large number of randomly developed grains.

(14) "Nuclear Data Committee ....", *op. cit.*

Summary

1. As a result of the findings at the Oak Ridge National Laboratory by Ben Kalmon, an investigation into the usefulness of commercial films (not special particle emulsion formulae) for fast neutron monitoring was carried out. The substitution of darkening rather than the counting of "proton recoil tracks" was investigated as a method of detection. Lead shields as an intensifying screen in fast neutron flux was studied.

2. Five commercial type films were exposed to Po-Be fast neutron flux representing 2.5×10^7 to 1×10^9 N_f/cm^2. The density of the open window portion of the film was in all cases less than the blackening behind the lead shield. All the films were sensitive to radiation from 5×10^7 N_f/cm^2 or greater.

3. One type of film (Ansco Process) showed straight line response when increasing flux was plotted against density.

4. Methods were used to demonstrate that the primary fast neutrons were responsible for the "behind the shield intensification". It was necessary to use indirect methods since there is always gamma present in a Po-Be source.

a. Primary gamma (no fast neutrons present) showed true shielding effects behind the lead screens.

b. Primary fast neutrons (some gamma present) showed only a slight reduction in the number of proton recoil chains present behind the lead shield and in the open window with film specially prepared to be sensitive to fast neutrons.

c. Primary fast neutrons (some gamma present) were passed through lead shields and then through aluminum between the lead and the film. 0.5 mm to 0.7 mm of aluminum permitted intensification of the darkening behind the lead, but 0.7 mm to 1.5 mm of aluminum absorbed the radiation which caused the intensification.

d. The blackening behind the shield could be caused by an (n,n) reaction of fast neutrons in lead, as the inelastic scatter produces a secondary gamma and a neutron with resulting ionization after release from the excited lead nucleus. 0.7 mm of aluminum will absorb beta equivalent radiation of less than 0.6 Mev, and very soft gamma.

Conclusions

1. Film blackening may offer a method of measuring fast neutron flux.

2. Lead shields will intensify the blackening when films are exposed to fast neutron flux.

3. No convenient method is available to separate the total effects of the mixed radiation, when film darkening is used as a measure of neutron flux.

4. All fast neutron sources (except those produced by accelerators) will contain appreciable mixed radiation.