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NEUTRON MONITORING BY MEANS OF NUCLEAR TRACK FILM (NTA)

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Health Physics Division

NEUTRON MONITORING BY MEANS OF NUCLEAR TRACK FILM (NTA)

Supplementary Report

J. S. Cheka

Date Issued: JAN 9 1950

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NEUTRON MONITORING BY MEANS OF NUCLEAR TRACK FILM (NTA)

Supplementary Report

J. S. Cheka

Introduction:

Preliminary studies of the use of particle or nuclear track emulsion for monitoring of plant personnel for neutron exposure have already been reported[1]. Study of emulsion characteristics and responses was continued, and calibration methods were analyzed.

Since nuclear track emulsions are a rather recent development, and have been in the process of improvement, response has lacked uniformity. The expected response has been recalculated on the basis of a recent analysis by the Eastman Laboratories.

The fading of the latent image, as it affects monitoring work, was investigated. Effects of different developers, and of different development techniques, on latent image stability were also tested.

The flux of fast neutrons in the "fast cart" was mapped by means of both Victoreen r-meter measurements and films. Some films were cross-calibrated with neutrons from a Po - Be source against a flux of fission neutrons in the "fast cart".

Emulsion Sensitivity:

During the period of several years that nuclear track emulsions have been used at this site, sensitivities have been found to vary over a wide range. The variation ranged from $(3.01 \pm 0.09) \times 10^5$ n_{th}/track in batch #N4x158384453 to $(4.4 \pm 0.44) \times 10^4$ n_{th}/track in batch #N4x18110724 3,

a factor of 6.8, for thermal neutrons; and from $(1.92 \pm 0.31) \times 10^4$ n_f/track in batch #N4x158284453 to $(2.61 \pm 0.25) \times 10^3$ n_f/track in batch #N4x181107243, a factor of 7.4, for fast neutrons.

While some of this change was obviously due to differences in emulsion sensitivity, and some to the errors involved in measurement, it is possible that a part of it could be accounted for by a change in the constituents (especially N and H content) of the emulsion itself. The more recent NTA type emulsions have been more uniform, and approximate the most sensitive of the emulsion batches tested. In order to estimate efficiency, the theoretical expectancy was recalculated on the basis of the most recent emulsion analysis available. This was obtained from J. Spence[2] of Eastman Kodak Company. This analysis shows a silver halide content of 82 + % which is probably as high as it is practical to go. Consequently, any further modifications will probably be minor ones. The analysis appears in Table I below.

Table I

Composition of Eastman NTA and NTB Emulsions

	Dry Emulsion		2.7% Moisture (50% RH - 21°C)	
Element	% Composition	Atomic Ratio Element/Silver	% Composition	Atomic Ratio Element/Silver
Ag	47.1	1.00	45.8	1.00
I	1.49	0.027	1.45	0.027
Br	33.9	0.97	33.0	0.97
C	9.47	1.62	8.26	1.62
H	1.17	2.68	1.43	3.37
N	3.06	0.50	2.98	0.50
O	4.80	0.69	7.02	1.03

Specific gravity of the emulsion is given as 3.8 @ 24°C.

In calculating the expectancy of tracks due to thermal neutrons by the $N^{14}(n,p)C^{14}$ reaction, only the nitrogen content of the emulsion is significant. It is, therefore, simpler to use the portion of the analysis given for dry emulsion. Nitrogen atom density is then given by:

$$D_N = \frac{\text{Sp. gr. x N ratio x Avogadro's No. x thickness x area}}{\text{Atomic weight of nitrogen}}$$

or, numerically:

$$D_N = \frac{3.8 \times 3.06 \times 10^{-2} \times 6.02 \times 10^{23} \times 4 \times 10^{-3} \times 1}{14}$$

$$= 2.00 \times 10^{19} \text{ N atoms/cm}^2$$

Proton density due to $N^{14}(n,p)C^{14}$ is given by:

$$P_N = D_N \times \sigma_{a_N} \times n,$$

where P_N is the proton density sought,

D_N is the nitrogen atom density found above,

σ_{a_N} is the capture cross-section of nitrogen for thermal neutrons, and

n is the thermal neutron exposure for one week's tolerance[3].

Numerically:

$$P_N = 2.00 \times 10^{19} \times 1.7 \times 10^{-24} \times 6.5 \times 10^{8}$$

$$= 2.21 \times 10^{4} \text{ protons (of 0.62 Mev)/cm}^2.$$

There is a correction required to compensate for protons which form near each surface and escape from the emulsion without having been recorded. This is not the same for all emulsions. Range in nuclear particle

emulsions of a 0.62 Mev proton is approximately 8 μ which comprises 6 to 8 silver grains. In the least sensitive emulsions over 90% of tracks noted are of but 3 to 4 grains and have a length of approximately 4 μ. It is reasonable to assume that these grains are sensitized at the end of the range where the ionization density is higher. Where the complete range gives a 6 to 8 grain track the first half of the path produces a recognizable track of 3 grains. Consequently, the layer of probable loss is reduced. Losses range from 5 to 10%. Making these corrections, expected track densities range from 2.10 to 1.99 x 10^4 track/cm^2.

From the sensitivity figures given above, 3.01 x 10^5 n_{th}/track give 2.16 x 10^3 tr/cm^2 for a week's tolerance exposure, an efficiency of 10.5% and 4.4 x 10^4 n_{th}/track give 1.48 x 10^4 tr/cm^2 for the same exposure, an efficiency of 72%.

The expectancy of H-recoil proton tracks from fast neutrons requires a similar but more intricate calculation. The flux comprising a week's tolerance of 0.10 rep[3] is estimated from a Victoreen r-meter reading, the fission neutron energy spectrum[4], and conversion factors of numbers of neutrons per rep at various energies given by C. C. Gamertsfelder[5]. H-atom density can be taken from Table I, but the variable scatter cross-section of hydrogen for different energies of fast neutrons must be considered.

The measurement of fast neutron fluxes in the "fast cart"[1] was done by means of Victoreen r-meters with 100 r chambers, the discharge of the meters being measured in arbitrary n-units. The n-unit was estimated by Aebersold and Anslow[6] to be greater than 2.0 rep, and members of the Biology Division have been using the conversion factor of 1 n-u = 2.5 rep. A previous estimate of the number of fast neutrons per rep was made[1] on the basis of the assumption that one rep of one Mev neutrons would be fairly representative of 1 rep of

fast neutrons of fission energies. The conversion was made on the basis of calculations of Gamertsfelder[5] on the loss of energy in tissue of fast neutrons. The new numerical value of the rep was derived by taking an arbitrary number of neutrons whose energies are distributed according to the fission spectrum[4], dividing the fraction of the total in each energy increment, say 0.1 Mev, by the number of neutrons per rep corresponding to each increment, and calculating the number of rep represented by this flux. This calculation gives 1 rep $\approx 3.4 \times 10^8$ n_f/cm^2 or 1 n-unit $\approx 8.52 \times 10^8$ n_f/cm^2.

Conditions of exposure necessitate further corrections. R. E. Zirkle and J. R. Raper have estimated[7] that the residual gamma ray contamination accounts for (12 ± 6) % of the r-meter readings, so that only (88 ± 6) % of each reading measures n-units of neutron flux. Making this adjustment, a reading of 1 division indicates $(7.50 \pm 0.51) \times 10^8$ n_f/cm^2.

Since the detection of fast neutrons by means of emulsion depends on track formation by recoil protons from hydrogen, the calculation of expected track density requires an analysis of this reaction. A track, to be recognizable, must have at least three silver grains. The length of such a track is about 3 μ. This would require a proton of energy greater than 0.25 Mev, since the stopping power of NTA emulsion is about 1700 times that of air[8].

The number of protons of any energy formed in the emulsion depends on the H-atom density of the emulsion, and on the scatter cross-section of hydrogen for neutrons. The H-atom density can be calculated as follows:

$$D_H = \frac{\text{Sp. gr.} \times \text{H-atom ratio} \times \text{Avogadro's No.} \times \text{thickness} \times \text{area}}{\text{atomic weight of H}}$$

Numerically:

$$D_H = \frac{3.8 \times 1.17 \times 10^{-2} \times 6.02 \times 10^{23} \times 4 \times 10^{-3} \times 1}{1}$$

$$= 1.07 \times 10^{20} \text{ H-atom/cm}^2$$

for dry emulsion. The value for 2.7% H_2O content (Table I) would be more representative of average conditions for personnel exposure at this location. H-atom density is 26% greater than for dry emulsion, or 1.35×10^{20} H-atoms/cm^2. The proton density is then determined by the following relation:

$$P_H = D_H \times \sigma_{sH} \times n$$

where P_H is the proton density due to H recoils,

D_H is the H-atom density found above,

σ_{sH} is the scatter cross-section for hydrogen, and

n is the fast neutron flux.

As σ_{sH} is a function of neutron energy, a good approximation can be reached by taking increments of energy, as above, and calculating H-atom collisions for the fraction of total flux in each energy increment, and totalling. These operations give a total of 1.67×10^4 collisions/cm^2 for one week's tolerance exposure[3] of 3.41×10^7 neutrons of fission energy distribution discounting those of energy less than 0.25 Mev. However, since only those protons are counted which produce recognizable tracks; i.e., those having energy greater than 0.25 Mev, the probability of a proton formed by any collision being above this minimum energy was calculated by the formula:

$$P = C\left(1 - \frac{1}{4E_n}\right)$$

where P represents protons of energy greater than 0.25 Mev,

C is the number of collisions, and

E_n is the energy (in Mev) of the incident neutrons.

After making this correction, 1.15×10^4 protons/cm^2 of sufficient energy to generate recognizable tracks are left. Making a further correction of 5 to 10% for protons formed near the surface of the emulsion and directed outward, the final figure for track expectancy is from 1.04 to 1.09×10^4 tracks/cm^2.

From the previously mentioned experimental values for sensitivity, at 1.92×10^4 n_f/track, one week's tolerance exposure of 3.41×10^7 neutrons/cm^2 produces 1.78×10^3 tracks/cm^2, or 16 to 17% of the expected value, and at 2.61×10^3 n_f/track the same exposure produces 1.31×10^4 tracks/cm^2, or 120 to 126% of the expected density. The latter figures show that at least some of the tracks recorded are produced by protons formed outside of the emulsions.

The tracks are counted under a microscope at approximately 900x magnification, using oil immersion and dark field illumination. A 90x apochromatic objective is used with 10x compensating eye pieces. The field diameters of the microscopes used at present are 0.16 mm, giving areas of 2.0×10^{-4} cm^2/field or 500 fields/cm^2. Since the depth of focus is only approximately 4 μ and the thickness of the developed emulsion is approximately 20 μ, the emulsion is read in depth by racking the objective up and down.

The films are processed and dried in a rack such that they dry slightly concave on the emulsion side. If they are placed on a glass slide on the microscope stage with a drop of immersion oil interposed, manipulations can be accomplished without other means of holding. After scanning, the

films are cleaned with xylene and tissue papper, and stored. The glass slide does not require removal from the stage except when cleaning is necessary.

Latent Image Instability

In 1946 Yagoda[9] described the fading of latent images in alpha emulsions in terms of densitometer readings. An investigation was made to evaluate the consequence of this phenomenon on personnel monitoring methods. In no case is the photographic density readable for tolerance doses. The basic postulate for the use of nuclear emulsions for the evaluation of neutron exposures at tolerance levels was the assumption that a track count after exposure for a reasonable length of time would be a measure of the integrated dose for the period. Since the track densities for some of the earlier batches of emulsion were less than one track per field of vision on a microscope at 900x magnification for one week's tolerance exposure, and the statistical error with such a count rate was high, there was a tendency to increase the time a film is used. However, if the fading is rapid, there would occur a time at which equilibrium is reached, assuming a uniform rate of exposure, and track density would no longer increase. Moreover, if exposure is not uniform, a large dose during the early portion of the exposure period would show no greater track density than a small dose during the latter portion.

The tests consisted of exposing groups of films to the same flux of fast neutrons producing about 30 tracks/field, developing one specimen of the group immediately, and others after various periods of delay. The fading or "decay" of the latent image after any time t was calculated from:

$$F_t = \frac{Do - Dt}{Do}$$

where F_t is the fading at time t,

D_o is original track density, and

D_t is track density on film developed after delay of time t after exposure.

The decay rate of recognizable tracks was found not to be uniform in most cases, but increased after a week or more, finally approximating an exponential rate. It was also found that decay rates in different emulsion batches differed, and that the less sensitive films, which also had shorter tracks in terms of number of grains, showed decay which sooner approached an exponential rate.

It seemed reasonable to assume that the individual grains, originally sensitized to form a latent image, become undevelopable at random with a uniform half-life. If that condition obtains, then a longer track may lose a larger percentage of its grains before becoming unidentifiable as a track. W. S. Snyder (see Appendix A) calculated the probable rate of disappearance of tracks, assuming the above condition and that less than three consecutive developable grains remaining constitutes the loss of a track. Track disappearance rate is then expressed by:

$$A_n = q\,A_{n-1} + qpA_{n-2} + qp^2A_{n-3} \quad \text{for } n > 2$$

where n is the number of grains originally activated in a track,

A_n is the probability of the loss of a track of n grains,

p is the probability of an activated grain remaining activated, and

q is the probability of an activated grain not remaining activated.

Specific values were calculated and tabulated for A_n with $n \geqq 3$. Figure I shows values for A_n = 3, 7, 12, and 20. In this figure the time axis is divided into arbitrary units such that, at the end of the unit, 0.9 of the activated grains found at the beginning are still developable.

The validity of this theory was tested by ascertaining track length distributions in terms of number of grains for various emulsions and under various conditions of development, calculating the weighted average decay rate using values indicated by the table mentioned above, and normalizing the time units. Such curves gave good fits for several sets of experimental data.

Increase of development time over that recommended by the manufacturer, while not affecting track density on immediate development, decreased the decay rate of recognizable tracks. Longer development also tended to increase chemical fog, and to produce dichroic fog. The two latter effects, which interfere with track counting when excessive, were the limiting factors in determining the development time.

DuPont Liquid X-ray developer was tried instead of the recommended Eastman D-19. This is a more vigorous developer, and brought out 10 to 20% more tracks after no delay. The increase in chemical fog was noticeable, but not troublesome. Decay rate of the latent image was also decreased. The results of tests on emulsions of batches N4x17328016473, and N4x181107551 with D-19 and DuPont X-ray developer, expressed in terms of days of delay at which half of tracks were lost, appear in Table II.

Table II

"Half-Life" of Latent Images in NTA Emulsions

Development Time (min)	Type of Developer	Half-life in days for Film	
		N4x1732806473	N4x181107551
4	D-19	12	14
5	D-19	16	17
6	D-19	23	20
3	DuPont	15	15
4	DuPont	23	20
5	DuPont	38	23

Figures II - VII show rates of loss of tracks in emuslion #N4x181107551 after exposure to approximately 0.5 n-units of fast neutrons.

Study of the experimental data revealed several interesting details. First, the sensitivity of emulsion #N4x181107551 is almost double that of emulsion #N4x1733806473. Then the shapes of the loss curves indicate the greater preponderance of longer tracks in emulsion N4x181107551. Also, the disappearance rate of the grains is greater in the more sensitive emulsion. Finally, it is very apparent that the percent probable error increases enormously as the track count decreases. This latter characteristic is one of the greatest disadvantages of this method of monitoring.

From these data it is obvious that the loss of developability of the grains of the latent image is not absolute, but a matter of degree. A longer development time, or a stronger developing agent brings out a greater number of grains. This effect is greater than apparent from the track count; for example, for emulsion N4x1732801 6473 developed in D-19 for four and six minutes, the number of tracks greater than seven grains in length was 16% and 35% respectively, at no delay. It was also noted that the proportion of long tracks dropped considerably in the case of delayed development.

The exact mechanism of the photographic effect is not too well known. It is known that development of the silver halide grains begins at discrete centers, which have been designated as sensitivity specks. These seem to contain aggregations of reduced silver atoms. It is thought that electrons, freed by the photoelectric effect of light, migrate to these centers where they reduce silver atoms. The aggregates grow according to the light exposure, and developability depends on the size of the aggregate. It is also not clear why a true photographic image is very stable compared to one formed by a highly ionizing particle, such as an alpha particle or proton. Neblette mentions[10] that smaller grained emulsions show regression of the latent image more rapidly than those of coarser grain. Nuclear track emulsions have a grain size of 0.2 to 0.4 μ, while standard photographic emulsions have a grain distribution ranging from 0.1 to 1 or 2 μ. Further than this, Demers suggests[11] that with the dense ionization due to the passage of the ionizing particle through a silver halide grain, although a large number of electrons is released along with positive ions, the strong positive charge present in the path prevents migration of many of the electrons to the sensitivity centers before recombination. The positively charged core, consisting of large ions, would disperse less quickly than electrons, and would act in competition with the sensitivity centers in capturing electrons. Yagoda[8] suggests further that hydrogen peroxide, formed in the moisture of the emulsion by particles having high specific ionization, could diffuse into the silver halide grains and, in time, resolve the silver aggregates thought to constitute the latent image.

Calibrations

Calibration of films by the method outlined above has an appreciable uncertainty. The Victoreen r-meters used to ascertain n_f flux did not give reproducible results. This condition was found both in the lack of agreement among several meters used simultaneously, and in the variation of the relation between the measured flux and the power level of the reactor. A precaution in exposure technique was also necessary to accommodate the geometry of the secondary source (uranium slugs) relative to the exposure compartment of the cart.

When it was found that Victoreen r-meter readings, taken at the same point at the same time, varied, one r-meter was sent by the Biology Division to Berkeley to be calibrated against the meter originally used by Aebersold. It was recognized that this did not give an absolute measure of neutron flux, but it was hoped that even this arbitrary unit would supply a basis of comparability for biological experiments at the various sites. A "correction factor" was applied, and some half dozen 100 roentgen r-meters were cross-calibrated with this "standard" and individual correction factors were determined. These ranged from -3 to +19%.

It was usually found that, after a period of disuse, the meters read high on the first run. Subsequent runs gave results in agreement with each other, but sometimes as much as 40% lower than the first exposure. It was further noted that in the course of time the meter readings relative to each other changed; that is, the correction factors were not constant. Since at least some of the meters change in their response to fast neutrons, it appears that any r-meter, designated as a "standard", is unreliable.

A record of the factor n-u/Kwh of reactor energy noted since 1946 by this writer and some members of the Biology Division reveals that the

ratio has fluctuated widely. Table III shows part of the record. This is not continuous because the tunnel was not used continuously.

Table III

N-Units/Kwh of Reactor Energy in the "Fast Cart"

Date	n-u/Kwh
February 1946	0.040
January 1947	0.0402
March 1947	0.0366
April 1947	0.036 - 0.0337
September 3, 1947	0.0474
November 20, 1947	0.0528
November 26, 1947	0.0488
December 17, 1947	0.0530
January 19, 1948	0.0558
September 16, 1949	0.047

Information from Operations indicates that changes in fuel loading of the reactor were insufficient to account for these fluctuations. It might be presumed, then, that the discrepancies are largely due to changes in r-meter response. This is further indicated by a recent experiment of members of the Biology Division[12] where a larger number of n-units was required for a certain biologic effect than in 1946. Since the ratio of n-units to reactor energy has been high in the last few years, it may be that the n_f flux per indicated n-unit has decreased.

An experiment was also run to map the neutron flux distribution

on the floor of the "fast cart". This was done both with Victoreen r-meters and with films. Figure VIII is a diagram showing the averaged corrected readings of a series of exposures with the chambers in the positions shown. Figure IX is a diagram showing track densities in tracks/field at the positions shown. It appears, from the diagrams, that only the center of the cart has a uniform flux field. The effect is accentuated with the r-meter readings, probably because of the secondary gamma radiation (at different geometry) to which the films do not respond. At any rate, this condition entails the precaution that all exposures be made in the center of the cart for reproducible results.

Since the calibration method based on r-meter readings leaves much to be desired, several irradiations of films were made with Po - Be neutrons. The source strength in n_f/curie is given by the supplier, and the Po decay rate is known so that source strength at any subsequent time can be easily calculated. The volume of the source is small enough to approximate a point source at a reasonable geometry.

The exposures were made on a light aluminun rack with a 15 cm radius. The source was placed in the center, and the whole placed atop a rod in the middle of a room to minimize backscatter. The exposure was calculated by the formula:

$$n_f = \frac{n_s \times t}{4\pi r^2}$$

where n_f is the number of neutrons/cm^2 for the exposure,

n_s is the source strength in neutrons/sec,

t is the exposure in seconds, and

r is the distance of the film from the source

The expected track density was calculated in the manner previously outlined, using the Po - Be neutron spectrum reported by Demers[13]. The measured track density was about four times as high as was expected. This was due to the effect of recoil protons formed in the paper wrapper of the film packet being recorded in the emulsion and was much greater than that from the exposure to fission neutrons. The average energy of the fission spectrum is about one-third that of the Po - Be spectrum. Moreover, approximately 77% of Po - Be neutrons are above 4 Mev, and only approximately 5% of fission neutrons fall in this category. If one assumes the stopping power of paper for protons to be equivalent to that of air, the 23 mg/cm^2 of paper covering the film is equivalent to the range of a 3.8 Mev proton. Consequently, all protons $\gtrsim$ 4 Mev normal to the surface of the film will penetrate to the emulsion even if produced at the outer surface.

An experiment to measure this effect was performed. Four films were exposed at 15 cm from the Po - Be source with surfaces normal to the flux. A was in the standard wrapper (23 mg/cm^2) with the emulsion facing the source, B with the standard wrapper and 1 mm of cadmium interposed between the wrapper and the emulsion, C bare with emulsion toward the source, and D bare with emulsion away from the source. The exposure was 3.15 x 10^8 n_f/cm^2. The expected track density, calculated as above, was 6.27 x 10^4 tr/cm^2 or 12.5 tr/field. Enough fields were counted to include 1000 tracks. The track densities observed appear in Table IV below:

Table IV

Film Exposure to Po - Be Neutrons

Film	t/f	Remarks
A	39.4 ± 3.0	standard packet
B	13.1 ± 1.6	1 mm cadmium shield
C	10.3 ± 1.7	bare emulsion
D	43.6 ± 2.4	n flux through cellulose acetate

The number of tracks per field observed in the emulsion of films B and C are in the range of the expected value which, themselves, have uncertainty depending on water content of the emulsion. The track density resulting from exposure of film D, through approximately 35 mg/cm^2 of cellulose acetate film base, is in the same range as that in film A through 23 mg/cm^2 of paper.

A later experiment gave comparison between the above batch of film (30 μ emulsion) and a new batch (40 μ emulsion). The 40 μ emulsion was 22% more sensitive than the 30 μ, rather than 33% as might have been expected. However, the differences in track densities between wrapped and unwrapped emulsions were identical, implying that the variation in emulsion sensitivity may be a matter of proton production rather than recording efficiency.

Track densities, equivalent to that produced by one week's tolerance exposure to fission neutrons, as determined by calibration in the "fast cart", were obtained by exposure to 1.52×10^7 n/cm^2 of Po - Be neutrons of packets of both batch #42245475666 (30 μ) and #4230104406 (40 μ). If this equivalence still obtains on specimens of other batches, calibrations can be both simplified and made more reliable by the use of a Po - Be source instead of the reactor. It should be noted that 2.2×10^7 n/cm^2, rather than 1.52×10^7 n/cm^2, is the tolerance value for neutrons of the Po -Be spectrum.

Collateral Studies:

Comparisons were made between neutron response of Eastman's NTA emulsion, batch #N4x202733491, and that of several other nuclear emulsions. The latter group comprised Eastman's NTB coating #353,448, and Ilford's

nuclear research emulsions B_2 and C_2. All of these test emulsions were on glass plates. Each of the test emulsions showed greater sensitivity and latent image stability than this batch of NTA films. Results of response tests appear in Table V.

Table V

Comparative Neutron Response of Nuclear Emulsions

Emulsion	Tr/cm^2-nu ($\times 10^{-5}$)	Emulsion Thickness (μ)	Tr/cm^2-nu -μ ($\times 10^{-3}$)
Ilford B_2	4.62 ± 0.26	52	8.89 ± 0.50
Ilford C_2	3.53 ± 0.18	24	10.56 ± 0.75
Eastman NTA	1.98 ± 0.15	40	4.94 ± 0.38
Eastman NTB*	2.79 ± 0.51	15	18.60 ± 3.40

*Results for the NTB emulsion are not strictly comparable because it was exposed at a different time. The response is subject to fluctuation with moisture content, and there is an uncertainty in the stability of the "n-unit".

The NTB emulsion was the most sensitive of those tested, besides which, the tracks were long and there was not much more fog than in NTA. Both of the Ilford emulsions showed good sensitivity, but were coarser grained than the Eastman emulsions. Development took longer and there was considerable fog even with unexposed plates. The coarseness of the silver grains and the prevalence of fog interfered somewhat with dark-field microscopy. Latent image stability was good in all three of the experimental emulsions, only about 20% of tracks being lost in 35 days

in each. The NTA films, meanwhile, showed 50% loss in 18 days.

A batch of NTB emulsion on film backing instead of glass was ordered for further testing. These tests showed that this emulsion on film did not reproduce the results obtained with glass plates, either with respect to sensitivity or to latent image stability. The results were very much in line with those shown by NTA films.

Because of this difference in response of the same emulsion on glass and on plastic backing, and also because of greater facility in manipulation of glass than flexible films during microscopic scrutiny, a change to glass backing was considered. Because the neutron monitoring film is placed in the badges with the x-ray film used to monitor personnel for gamma exposure, the effect of the presence of such glass on the gamma response of x-ray film was measured. The glass in the badge increased open window readings by approximately 4% and decreased shield readings by about 8%, where percent gain or loss was determined by the difference in roentgens of exposure in a given instance, and the roentgens required to produce the same densitometer reading in a standard badge. Since relatively few of the personnel on the plant area require neutron monitoring, it was not thought that two complete gamma film calibrations were justified.

In the course of several exposures to Po - Be neutrons, it was noted that the response in neutrons/track varied by as much as 15% in specimens from the same batch exposed at different times. The best response occurred during exposure periods of highest average absolute humidity. Mees[14] states that all gelatins absorb or give up water until an equilibrium condition with the circumambient atmospheric moisture is attained. The equilibrium value is not the same for all gelatin, but depends on the pH of the sol just before drying. Consequently, a reliable correction factor

could not be ascertained even if atmospheric humidity were not very variable.

These emulsions are more sensitive to the condition of the developer solution than are x-ray or photographic emulsions. Depletion of the solution by use, or oxidation by exposure to air causes a much greater loss of efficency of development than would be expected from observation of these effects on x-ray films. The irregularities in apparent fading, seen in figures II-VII, are probably due to these effects. Test developer solution batches were mixed and stored in half full bottles for periods of 4 to 16 days. A group of NTA films was then exposed uniformly and specimens developed in the variously aged solutions. The results were very erratic, showing losses up to 42% but evidencing no direct correlation between efficiency of development and age of the solution. It is evident, however, that for consistency, freshly mixed developer must be used.

Occasionally it is necessary to expose several films to the same number of neutrons for purposes of comparison, since absolute exposures are difficult to evaluate. The flux distribution on the floor of the fast cart having been mapped, it was desired to determine whether greater uniformity could be achieved by piling up the films or by making a horizontal distribution. Fifteen films of batch #N4x202733491 were piled up and taped in position for exposure. They were irradiated with about 0.5 n-u. Track density readings showed a slight rise on the second film and then a gradual fall off. The sixth film had approximately 90% of the density of the first, the 9th approximately 79%, and the 15th approximately 70%. It appears, then, that the 110 mg/cm^2 of each film causes sufficient scatter when piled up to disturb results significantly, and exposure can be made more uniform by spreading the films over the uniform central section of the exposure cart.

An exposure of a stack of 9 films to 1.8×10^8 n_f/cm^2 of Po - Be

neutrons showed similar effects, but not as accentuated. The maximum value occurred in the third film and amounted to approximately +3%, and the 9th film had 93% of the track density of the first. It was noted, however, that all of these values were 8.7 to 18% greater than track density on a single film of the same type exposed to the same total flux.

Discussion:

The use of nuclear track emulsions as a method of monitoring personnel for neutron exposure is not as satisfactory as methods available for monitoring for other types of radiation exposure. The problem could be simplified for thermal neutrons by the loading of the emulsion with lithium or boron. Both of these elements have high capture cross-section for thermal neutrons, and the captures result in an alpha and a triton and in an alpha particle, respectively. The tracks of each are more easily discernible than those of protons, such as are formed by capture by nitrogen. However, since the use of the (n,p) reaction of the nitrogen in the gelatin permits the indication of thermal neutron tolerance with the same range of track density as is produced by hydrogen recoils by a fast neutron tolerance dose, and since a heavier concentration of alpha tracks would tend to obscure the less prominent proton tracks due to H-recoils, loaded emulsions would not be advantageous in a location such as this where there is as much likelihood of exposure to fast neutrons as to thermals, or some combination of the two.

The lack of reproducibility of emulsion sensitivity, as shown by the approximate factor of 7 between least and most sensitive specimens tested is a major drawback. This is somewhat mitigated by the fact that recent fluctuations have been much less, and have tended to the more sensitive range.

A greater cause for concern is the low sensitivity of these emulsions. Since the method consists of counting of random events, there is always a statistical uncertainty which tends to be large for a small total number of events.

Table VI shows some representative experimental data. The probable error shown is calculated according to Bessel's formula:

$$P.E. = 0.6745 \sqrt{\frac{\Sigma(\delta^2)}{n-1}}$$

where δ is the deviation of any reading from the mean, and n is the number of readings (fields) taken.

Table VI

Track Counting Data with Probable Errors

Film No.	Fields Counted	Tracks/field	P. E.	% P. E.
1	25	46.4	±2.6	±5.6
2	40	24.0	±2.1	±8.8
3	50	11.6	±1.4	±12.1
4	24	1.00	±0.67	±67.
5	24	0.38	±0.43	±113.

The first three sets of figures above represent the magnitudes encountered in calibrations. The latter figures are in the range of monitoring readings. Let us assume, for purposes of demonstration, that #1 represents a calibration reading at, say 1.2 divisions, and #3 represents the background correction for irradiation during the run into and out of the exposure end of the tunnel. Then, $(46.4 \pm 2.6) - (11.6 \pm 1.4) = 34.8 \pm 2.95$

represents the difference, due to the measured exposure. Assuming the r-meter readings to have an uncertainty of 10% (fluctuation in n-u/Kwh values indicate 11%), and applying the correction factor 0.88 ± 0.06 n-u = 1 division, the exposure is $(1.2 \pm 0.12) \times (0.88 \pm 0.06)$ or 1.06 ± 0.13 n-u. Then one n-u is indicated by $\frac{34.8 \pm 2.95}{1.06 \pm 0.13} = 32.8 \pm 4.89$ t/f, or

1 rep = 13.1 ± 1.96 t/f or 0.1 rep = 1 week's tolerance = 1.31 ± 0.196 t/f. If one further assumes that #4 represents a field reading and wishes to evaluate it in terms of week's tolerance, one gets $\frac{1.00 \pm 0.67}{1.31 \pm 0.196} = 0.76 \pm 0.51$ week's tolerance. This is a much larger uncertainty than is desirable.

The above values were calculated on tolerance values published in 1948[3]. The recent trend is to revise these downward (unpublished). Such a move will cause smaller numbers of tracks to represent tolerance values, with the resultant increase in uncertainty. The use of an insensitive batch of films has the same result even at the present tolerance values.

A slight improvement can be attained by using a thicker emulsion. There is a practical limit to this since an emulsion thicker than 50 μ requires special techniques in processing, and so does not lend itself well to routine monitoring operations.

Conclusion:

Nuclear track emulsions are a fair method of monitoring personnel for neutron exposure, both in the thermal and "fast" ranges of energies. They do not supply an accurate measure of exposure at tolerance ranges, but become more accurate as exposure increases. They do, however, indicate exposures in this range within a factor of two. The greatest virtue of the method is that nothing better has yet been suggested.

FIG. 1

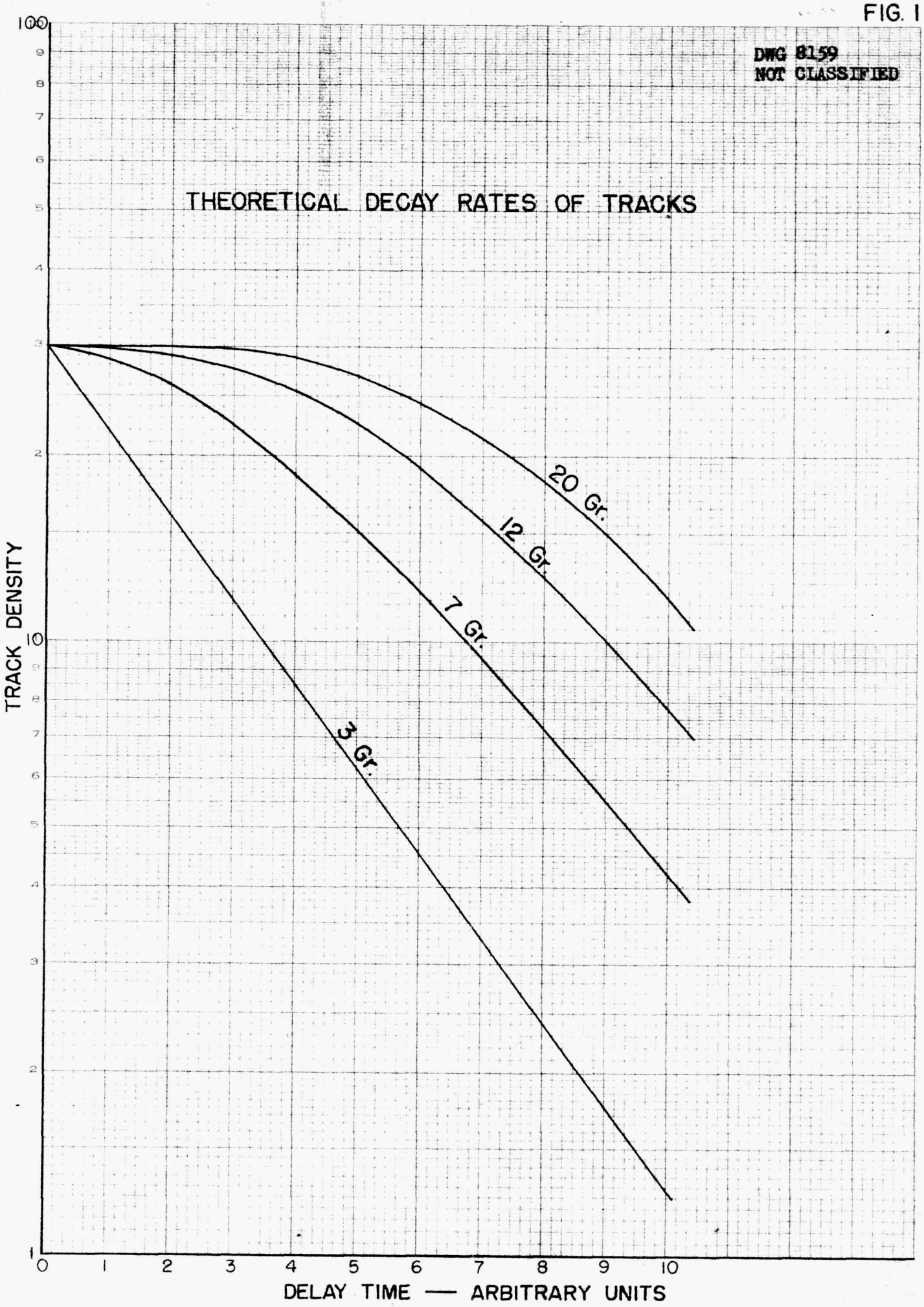

DWG 8159
NOT CLASSIFIED
THEORETICAL DECAY RATES OF TRACKS
20 Gr.
12 Gr.
7 Gr.
3 Gr.
TRACK DENSITY
DELAY TIME — ARBITRARY UNITS
100
10
1
0
1
2
3
4
5
6
7
8
9
10

FIG. 2

DWG 8160
NOT CLASSIFIED

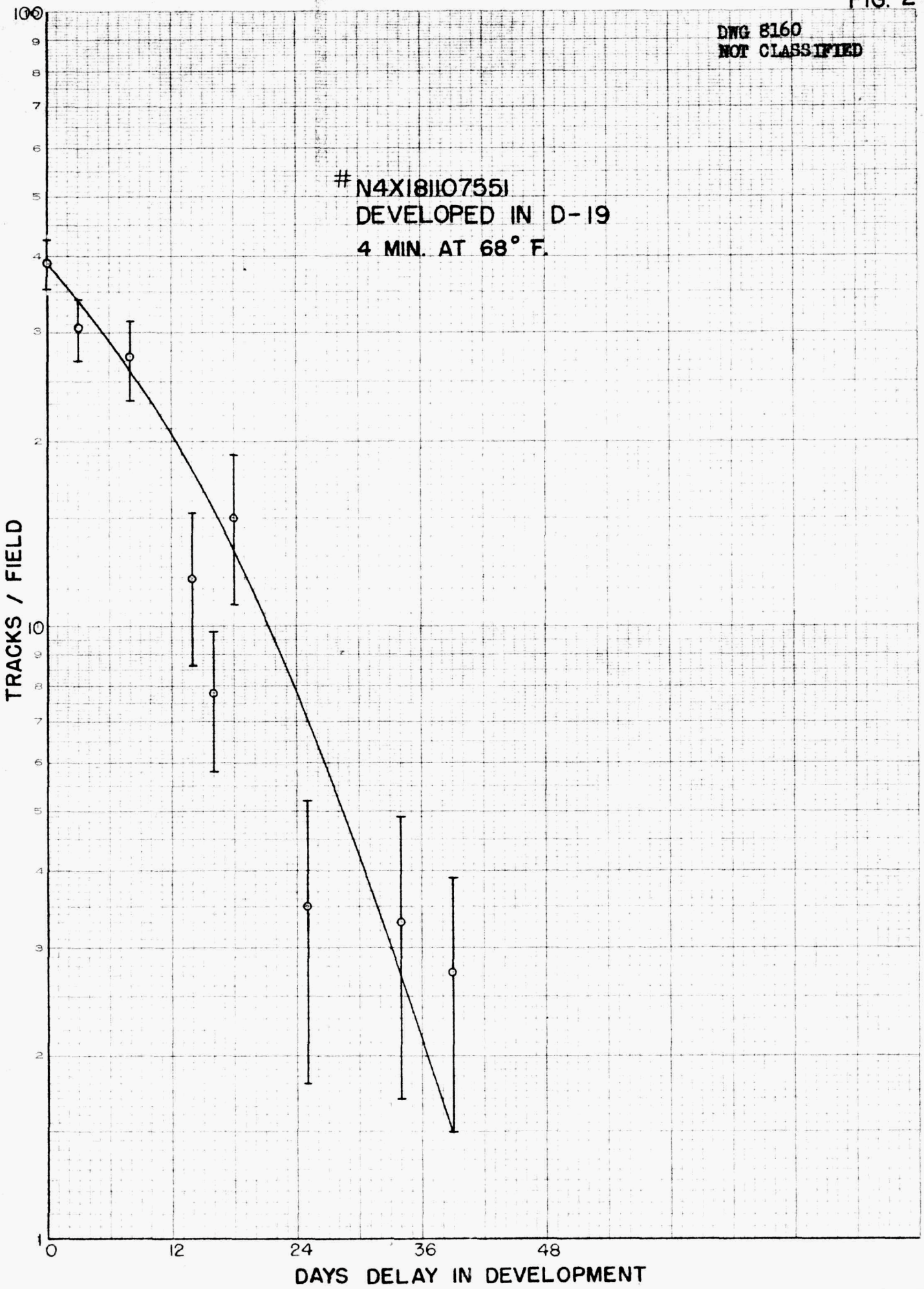


FIG. 3

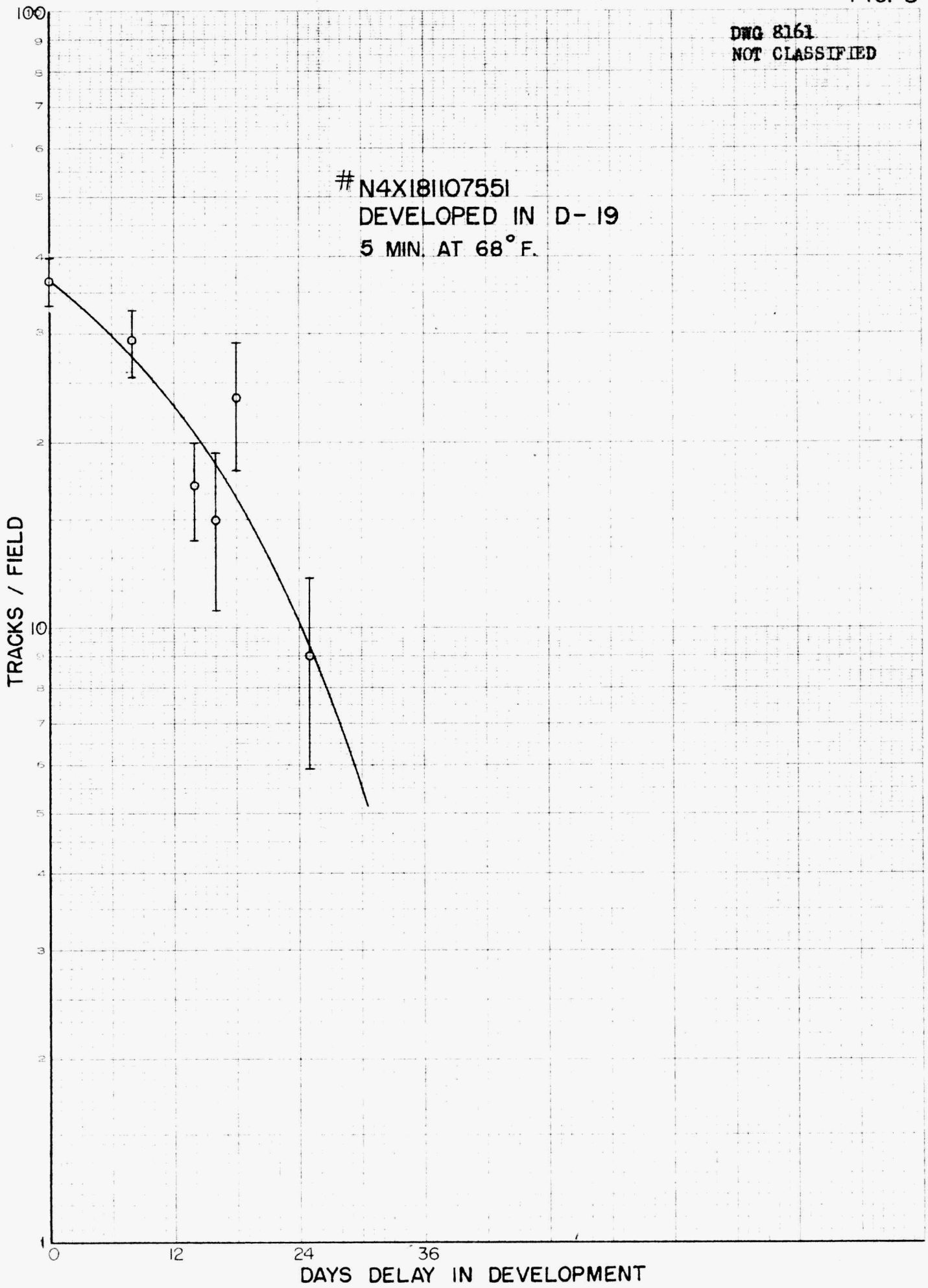

DWG 8161
NOT CLASSIFIED
#N4X18110755I
DEVELOPED IN D-19
5 MIN. AT 68° F.
TRACKS / FIELD
100
9
8
7
6
5
4
3
2
10
9
8
7
6
5
4
3
2
1
0
12
24
36
DAYS DELAY IN DEVELOPMENT

FIG. 4

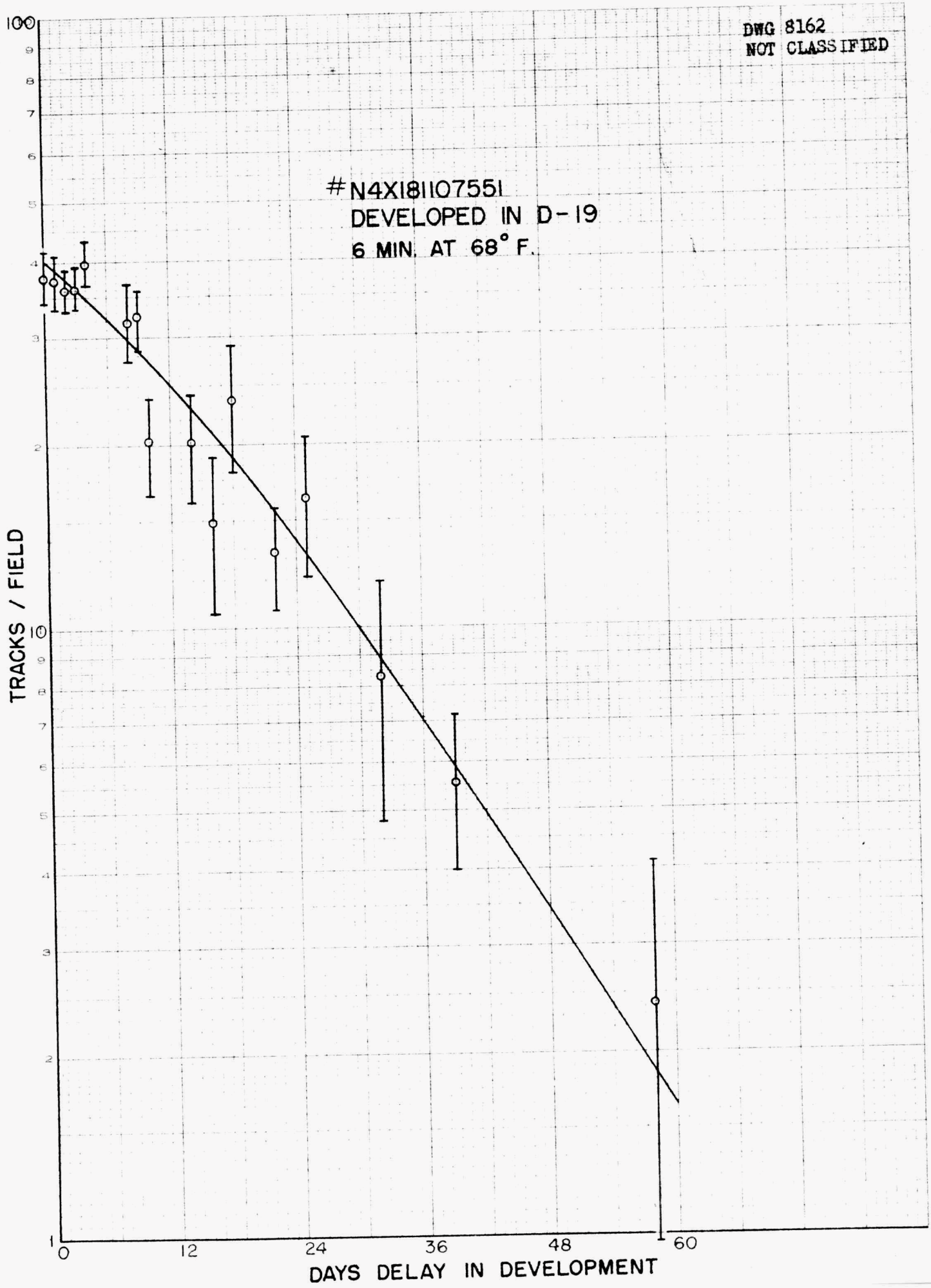

DWG 8162
NOT CLASSIFIED
#N4X18110755I
DEVELOPED IN D-19
6 MIN. AT 68° F.
TRACKS / FIELD
100
9
8
7
6
5
4
3
2
10
9
8
7
6
5
4
3
2
1
0
12
24
36
48
60
DAYS DELAY IN DEVELOPMENT

FIG. 5

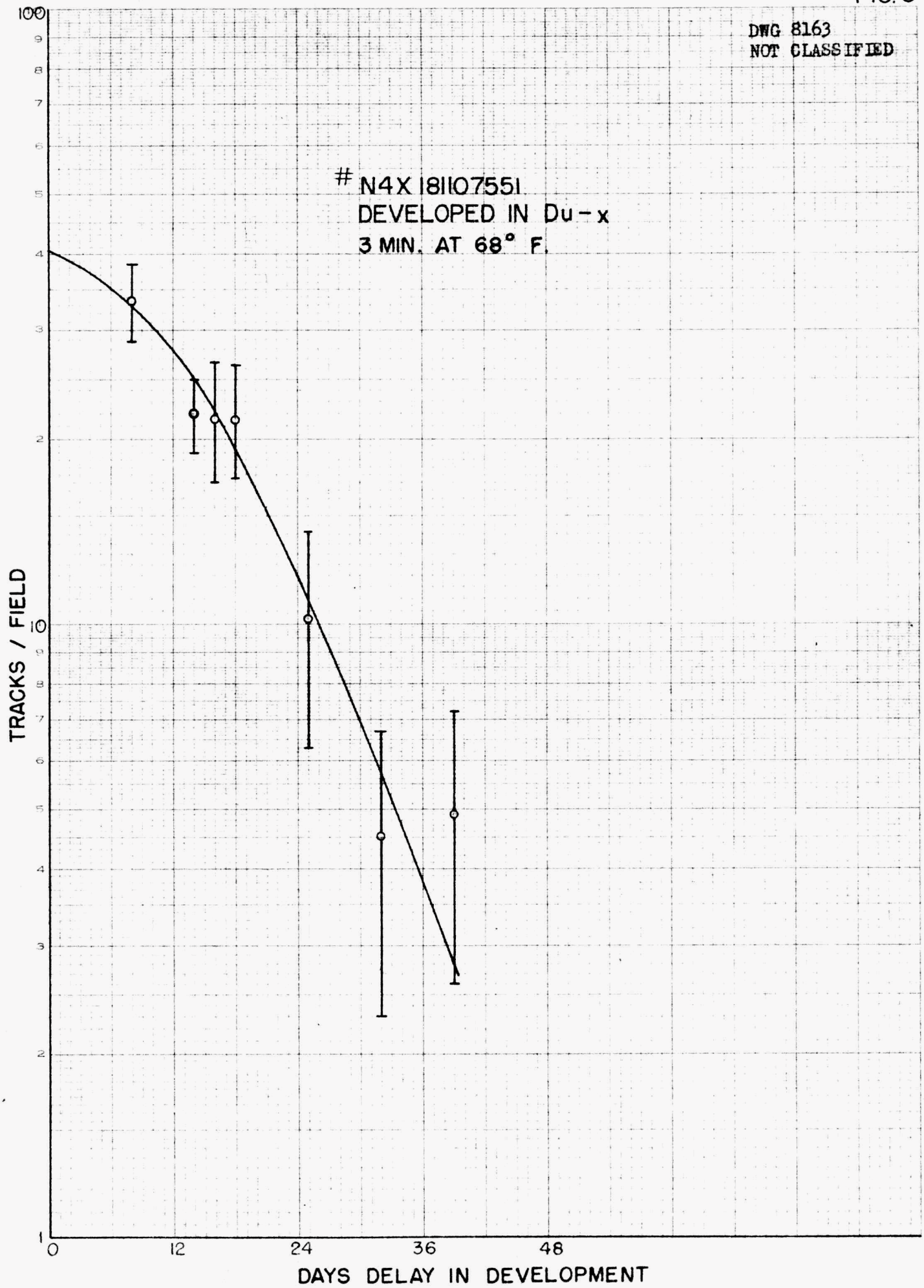

DWG 8163
NOT CLASSIFIED
N4X 18110755 1
DEVELOPED IN Du-x
3 MIN. AT 68° F.
TRACKS / FIELD
100
9
8
7
6
5
4
3
2
10
9
8
7
6
5
4
3
2
1
0
12
24
36
48
DAYS DELAY IN DEVELOPMENT

FIG. 6

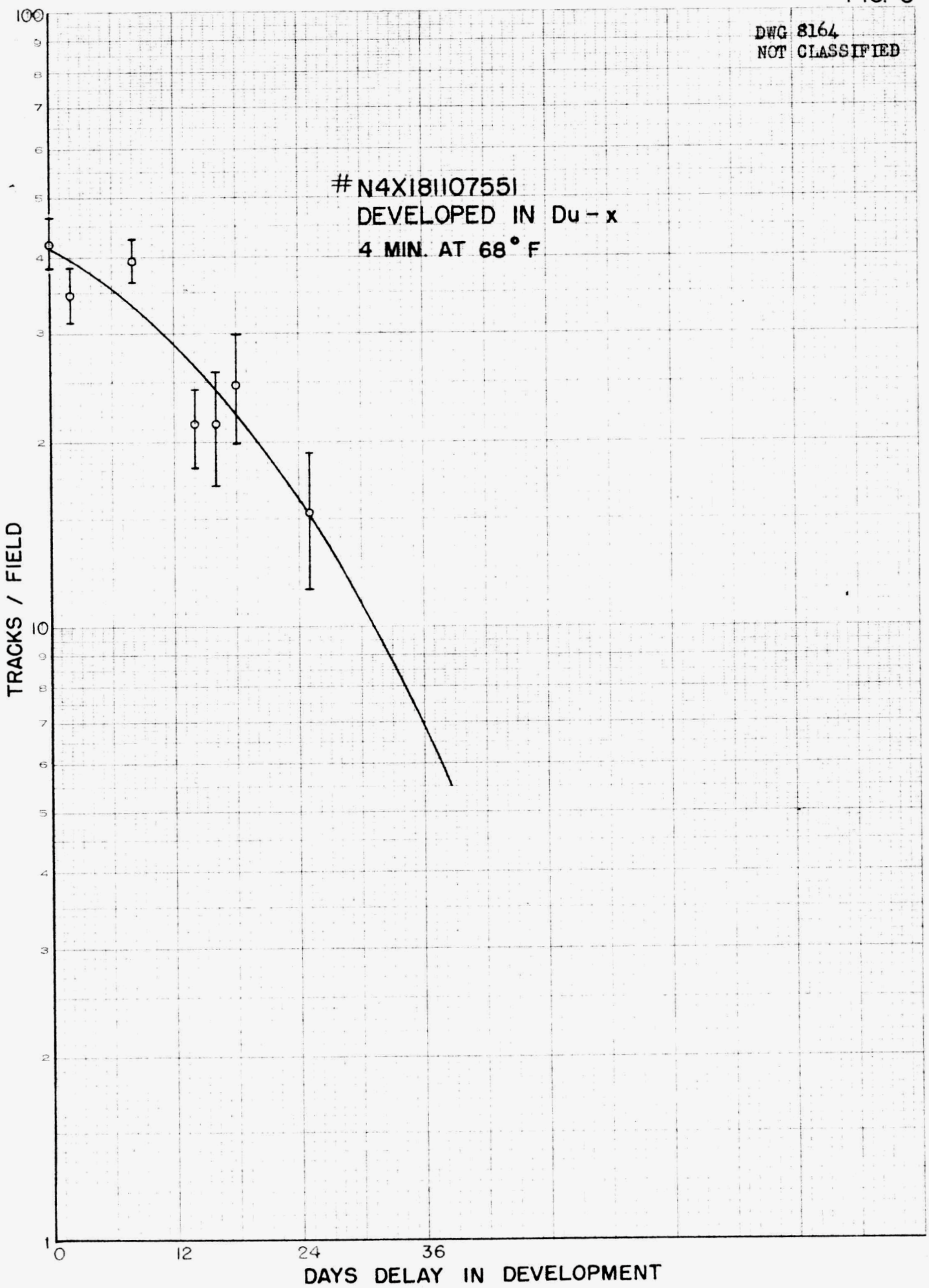
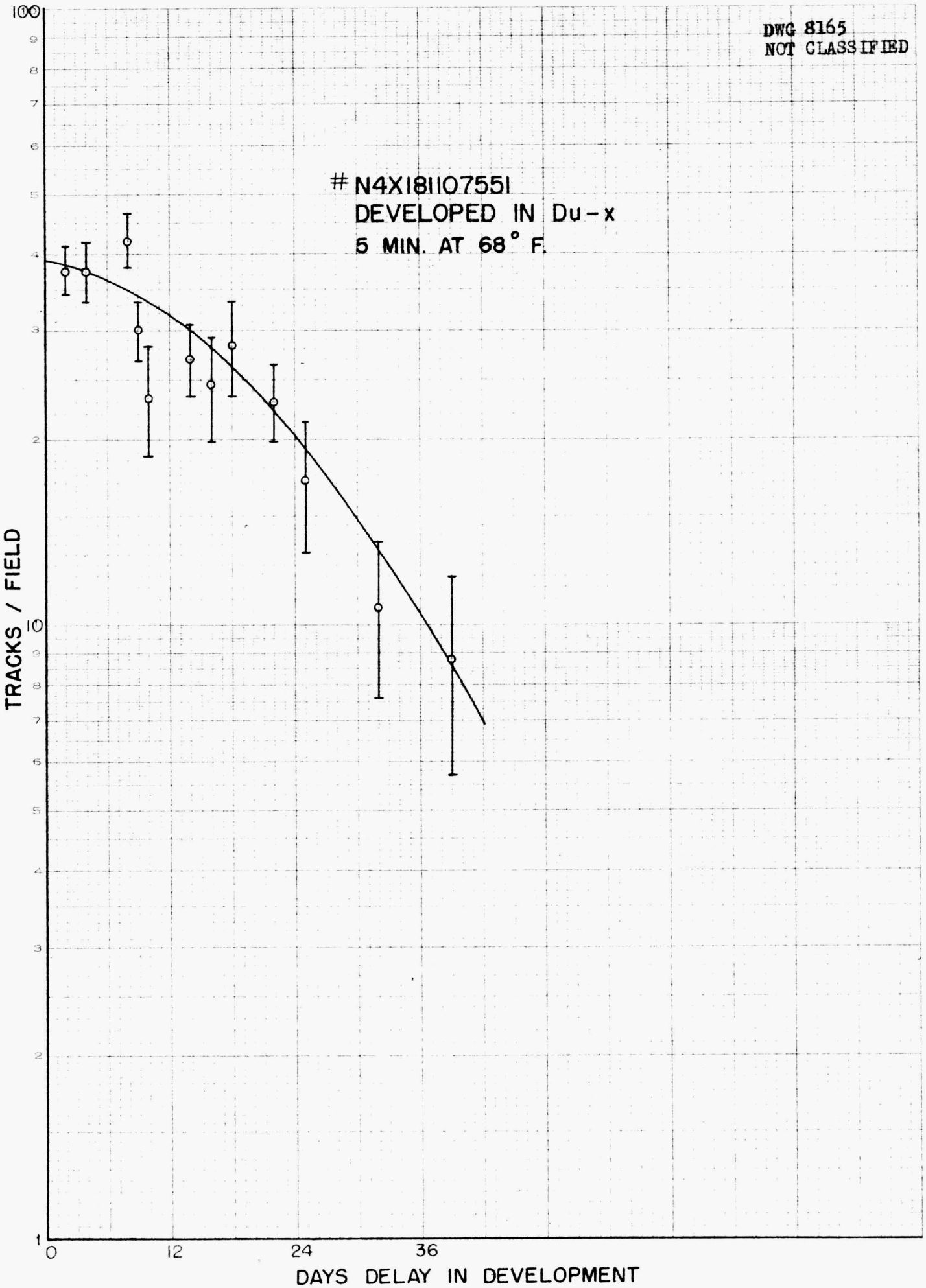

DWG 8164
NOT CLASSIFIED
#N4X18110755I
DEVELOPED IN Du-x
4 MIN. AT 68° F
TRACKS / FIELD
100
9
8
7
6
5
4
3
2
10
9
8
7
6
5
4
3
2
1
0
12
24
36
DAYS DELAY IN DEVELOPMENT

FIG. 7


DWG 8165
NOT CLASSIFIED
N4X18110755I
DEVELOPED IN Du-x
5 MIN. AT 68° F.
TRACKS / FIELD
100
9
8
7
6
5
4
3
2
10
9
8
7
6
5
4
3
2
1
0
12
24
36
DAYS DELAY IN DEVELOPMENT

DWG 8166
NOT CLASSIFIED

VICTOREEN RDGS. IN "FAST CART"

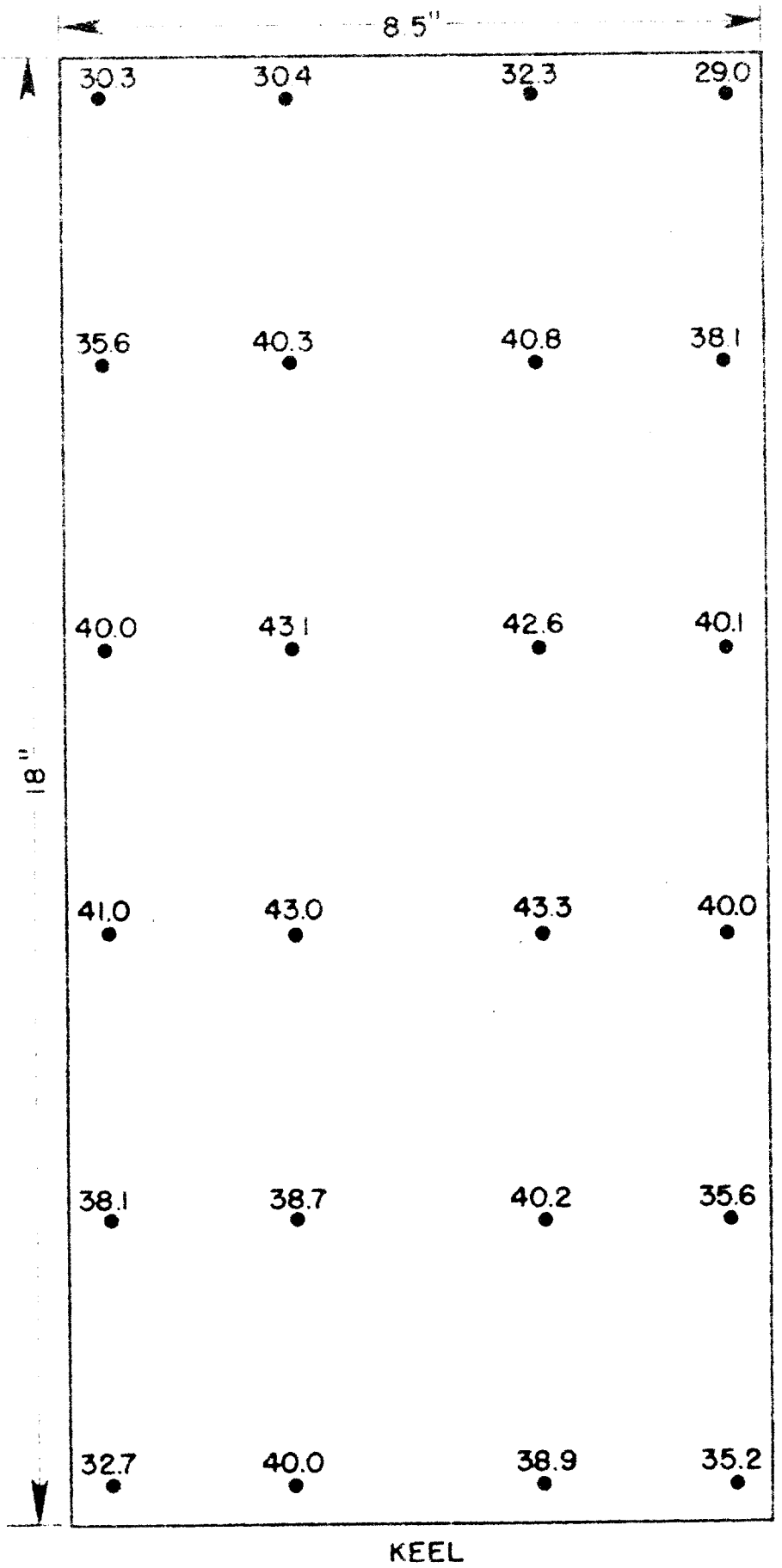
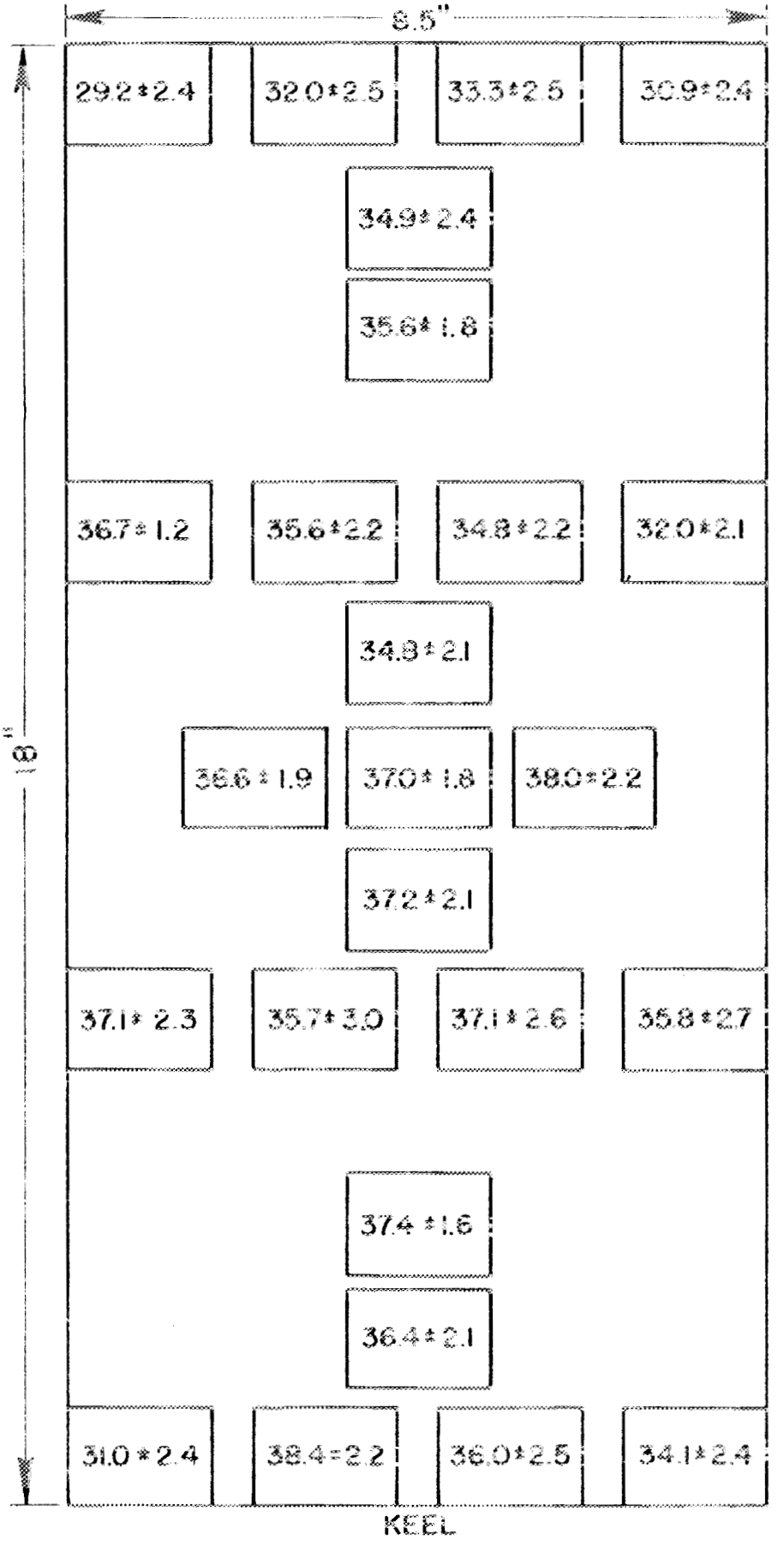


FIG. 8

FILM RESPONSE TO NEUTRONS IN "FAST CART" — (~0.73 n-u)



DWG. 8167
NOT CLASSIFIED

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APPENDIX A

By W. S. Snyder

Let p be the probability that an activated silver halide particle will remain developable after an assigned lapse of time, and let $q = 1-p$. Define a "track" to consist of n activated particles equally spaced along a straight line. Let A_n be the probability that after the assigned lapse of time no group of 3 consecutive particles of the original track remain activated. Evidently,

$$A_o = A_1 = A_2 = 1 \qquad (1)$$

For $n > 2$, we have the recursion formula,

$$A_n = qA_{n-1} \quad qpA_{n-2} \quad qp^2A_{n-3} \qquad (2)$$

To see this let $\{P_1, P_2, \ldots\ldots, P_n\}$ $n > 2$ be a track of n particles occurring in this order. The track $\{P_1, P_2, \ldots\ldots, P_n\}$ can fail to retain three consecutive activated particles after the assigned time interval in one and only one of the following three ways:

1. P_1 is deactivated and the subtrack $\{P_2, P_3, \ldots\ldots, P_n\}$ does not retain three consecutive activated particles. The probability of this occurring is qA_{n-1}.

2. P_1 remains activated, P_2 is deactivated and the subtrack $\{P_3, P_4, \ldots\ldots, P_n\}$ does not retain three consecutive activated particles. The probability for this alternative is qpA_{n-2}.

3. P_1 and P_2 remain activated, P_3 is deactivated and the subtrack $\{P_4, \ldots\ldots, P_n\}$ does not retain three consecutive activated particles. The probability of this alternative is qp^2A_{n-3}.

The probability that the chain $\left\{P_1, \ldots\ldots, P_n\right\}$ does not retain three consecutive activated particles is then (2),

$$A_n = qA_{n-1} + qpA_{n-2} + qp^2A_{n-3}$$

since (1), (2), and (3) are mutually exclusive alternatives and exhaust the possibilities for this occurrence.

The unique solution of (2) and (1) is given by,

$$A_n = A\alpha^n + B\beta^n + C\gamma^n \tag{3}$$

Where α, β, γ, are roots of the cubic,

$$y^3 - qy^2 - qpy - qp^2 = 0 \tag{4}$$

and A, B, C, are determined by,

$$\begin{aligned} A + B + C &= 1 \\ A\alpha + B\beta + C\gamma &= 1 \\ A\alpha^2 + B\beta^2 + C\gamma^2 &= 1 \end{aligned} \tag{5}$$

The cubic (4) has, for $0 < p < 1$, one real root, which we henceforth denote by α, and two complex roots, hereafter denoted by β and γ. If we put $B = Pe^{i\phi}$ and $\beta = \mu e^{i\theta}$, then (3) becomes,

$$A_n = A\alpha^n \left\{1 + 2\frac{P}{A}\left(\frac{\mu}{\alpha}\right)^n \quad \text{Cos}\,(n\theta + \phi)\right\} \tag{6}$$

The quantities occurring in (6) may be conveniently studied by making use of the transformation,

$$y = \frac{p}{W}, \frac{p}{q} = r \tag{7}$$

Then (4) becomes,

$$W^3 + W^2 + W - r = 0 \tag{8}$$

and if we use λ to denote one of the complex cube roots of 1, then

$$W = 1/3 \left\{ \lambda\pi + \lambda^2\zeta - 1 \right\} \tag{9}$$

with $$\pi = \sqrt[3]{27r + 7 + \sqrt{(27r + 7)^2 + 32}}$$

and $$\zeta = \sqrt[3]{27r + 7 - \sqrt{(27r + 7)^2 + 32}}$$

gives the three roots of (8).

We have $\left(\frac{\mu}{\alpha}\right)^2 = \frac{\beta\gamma}{\alpha^2} = \frac{p^2q}{\alpha^3} = \frac{W_1^{\,3}}{r}$ where W_1 is the real root of (8). Evidently $\frac{\mu}{\alpha} < 1$ so that for large n, $A_n \approx A\alpha^n$. But $\lim_{p \to 1} \frac{\mu}{\alpha} = 1$ so that the damping effect $\left(\frac{\mu}{\alpha}\right)^n$ is relatively small for p close to 1 unless n is very large. For example, when $p = .9$, $\frac{\mu}{\alpha} = .7$

Moreover,

$$\frac{P^2}{A^2} = \frac{3W^6 + 2W^5 + W^4}{3W^6 + 8W^5 + 16W^4 + 18W^3 + 16W^2 + 8W + 3} \tag{10}$$

and thus, $0 < \frac{P}{A} < 1$ with $\lim_{p \to 1} \frac{P}{A} = 1$.

When $\mu = 3/4$, $W = 1$ and $\frac{P}{A} = .3$

Thus $A_n \approx A\alpha^n$ with a high degree of approximation except when p

is very close to 1 and n is small. The error will be less than 1% if $\mu < .15$ for all n, and the values of n when the error becomes 1% or less are tabulated below.

p	n
.2	1
.3	1
.4	2
.5	4
.6	5