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Technical Note 29

PHOTOGRAPHIC DOSIMETRY AT TOTAL EXPOSURE LEVELS BELOW 20 mr

DEPARTMENT OF COMMERCE
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PHOTOGRAPHIC DOSIMETRY AT TOTAL EXPOSURE LEVELS BELOW 20 mr

Margarete Ehrlich and William L. McLaughlin

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FOREWORD

This Note covers the material presented by the authors at the 1959 annual meeting of the Health Physics Society. Work on some of the phases covered in the report will be continued; for this reason, it is not contemplated to publish the present results in a more formal way.

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ABSTRACT

Assemblies of commercial photographic material sandwiched between two plastic scintillators can be used to measure high-energy X- or gamma-ray exposures down to 1 mr and less. The energy dependence of the assemblies' response is much less than that of a conventional photographic dosimeter, in some instances allowing an exposure interpretation with an accuracy of ± 25 percent over the energy range from about 0.1 Mev to 1.25 Mev. However, low-intensity reciprocity failure limits the range of applicability of the system.

Conventional photographic dosimeters, not incorporating scintillators, are usually preferable for routine personnel dosimetry. By extending the monitoring period, it may be possible to avoid personnel dosimetry below 20 mr entirely. However, by doing this, one introduces additional difficulties because of instabilities in the photographic image. In some instances, an increase in effective emulsion thickness, achieved by using stacks of identical films, may lead to an increase in emulsion sensitivity sufficient to extend the useful range of a film badge well below 20 mr, without the use of a scintillator.

PHOTOGRAPHIC DOSIMETRY AT TOTAL EXPOSURE LEVELS BELOW 20 mr.

Margarete Ehrlich and William L. McLaughlin

1. INTRODUCTION

Photographic dosimetry at exposure levels below 20 mr is difficult because of insufficient sensitivity of commercial photographic emulsions, instabilities in the latent photographic image, and inaccuracies in sensitometric procedure. Some of the endeavors to cover the range below 20 mr more adequately, both with film-scintillator combinations and with conventional photographic dosimeters, will be briefly discussed.

2. FILM-SCINTILLATOR COMBINATIONS

2.1 Discussion of Method; History

One way to intensify the photographic effect of X-radiation, is to use the photographic material in contact with a scintillator. In such a system, a considerable fraction of the film response stems from the effect on the film of the visible light emitted by the scintillator, rather than from the direct effect of the primary radiation. In fact, if film and scintillator are suitably chosen, the response of the film to the primary radiation is so small in comparison with that to the visible light, that--essentially--the scintillator becomes the radiation detector, while the film merely takes the place of the recording apparatus (similar to a photomultiplier and associated electronic circuitry in the more elaborate scintillation-type survey instruments).

Since the intrinsic efficiency of the luminescence process depends only very little on the energy of the incident photons, the total amount of light produced in the scintillator is roughly proportional to the radiation energy absorbed in the scintillator. Therefore, for air-equivalent scintillators of the order of equilibrium thickness, one can expect that--as a function of radiation energy--the scintillator response is proportional to the ionization in air, as measured in roentgens. Unfortunately, the scintillators used in practice are not completely air equivalent, and, in addition, the fraction of the light produced that is incident on the film varies with photon energy.[1] As a consequence, the response of the film-scintillator system is energy dependent, although much less so than the response of a photographic detector used without a scintillator. Also, with proper choice of scintillator shape, the film-scintillator system may be made independent of the direction of the incident radiation--which is usually impossible with film alone. The only severe handicap of the film-scintillator detector is its rate dependence, a phenomenon that will be dicussed later.

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[1] In the photon energy range under consideration, the fraction of the visible light produced close to the film increases with increasing photon energy, and, as a consequence, the fraction of the light registered by the film also increases. On the other hand, at any given photon energy, increasing the scintillator size beyond a certain optimum, decreases the amount of light registered, because of the attenuation of the primary radiation and of the visible light on its travel to the film. This decrease is especially pronounced for photons of low energy, which are strongly absorbed in the surface layers of the scintillator.

Table 1 shows the important features of some of the published work on dosimeters using film-scintillator combinations. While the Ansco dosimeter was specifically designed as a personnel dosimeter and is useful for total exposures down to about 20 mr, and for rates down to about 5 mr/hr, the dosimeter of the British Atomic Weapons Research Establishment apparently was developed specifically for use with low total exposures in radiation fields of high intensities. Its response is essentially independent of the direction of incidence of the primary radiation. The Russian device is the most sensitive; no information is available on its rate dependence.

About two years ago, we were asked by the AEC Division of Biology and Medicine to investigate whether the use of the film-scintillator system could be successfully extended to exposure rates close to natural background, and total exposures down to 1 mr or less. Thus, the idea was to extend the usefulness of the method to rates lower than the ones limiting the response of the previous systems, and to total exposures comparable to those recorded with the Russian instrument.[2]

2.2 Rate Dependence of the Photographic Process

The reason for the rate dependence of film-scintillator dosimeters is to be found in the peculiarities of the photographic action of visible light. Photographic emulsions consist of AgBr crystallites

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[2] The results of a similar endeavor, also initiated by the AEC Division of Biology and Medicine, were reported by O'Brian, K., Solon, L.R., Lowder, W.M., in Rev. Sci. Instrum., 29, 1097, 1958.

(grains) embedded in a gelatinous matrix. Upon absorption of photons or of their corpuscular secondaries by the emulsion, some of the AgBr is reduced to free silver, which--when clustered in aggregates of several silver atoms--forms units that are fairly stable at room temperature and not too high a relative humidity. These aggregates are the smallest units of the so-called latent image, which, according to modern theory, requires for its formation at least three silver atoms.[3] During development, these units serve as centers for the further reduction of AgBr in the crystal, a process that eventually leads to the visible silver aggregates known as the photographic image.

Rate dependence of photographic material, referred to as reciprocity failure, i.e., a failure of the photographic effect to be proportional to the product of the intensity of the exposing radiation and the exposure time, occurs when more than one ionizing event is required to make one of the photographic emulsion's silver halide grains developable. A photon of visible light creates only one photoelectron upon its absorption in a silver halide crystal. This is not sufficient for the production of a latent image and for grain developability. As a consequence, if the illuminance is so low that the time between ionizing events in any one grain approaches the lifetime of the tiny, very unstable, precursors of the latent image, the number of grains actually made developable depends on the rate of incidence of the photons, and the photographic process shows what is known as low-intensity reciprocity failure. There exists also a high-intensity failure, but it is not of interest to the present discussion.

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[3] Mitchell, J.W., Photographic Sensitivity, p.433 of Report on Progress in Physics, Vol 20, The Physical Society, London.

When a photographic emulsion of sufficiently high sensitivity is exposed to X- or gamma radiation, the number of electrons produced by one single absorbed photon usually is large enough to make a grain developable. Therefore, the photographic effect of X- or gamma rays as a rule shows no rate dependence. However, if a photographic material is exposed to X- or gamma rays in conjunction with scintillators (which usually emit visible light of a wavelength in the neighborhood of 4000 Å) the assembly is rate dependent--at least at very low and very high radiation intensities. Therefore, the design of dosimeters embodying film-scintillator assemblies is actually more straightforward for the intermediate rate range than for the very low range of exposure rates. The present attempt to go to background rates was only partially successful.[4]

2.3 Experiment

The Pilot B plastic scintillator, which consists of diphenylstilbene in a 100% hydrocarbon plastic, was chosen as the intensifier. Figure 1 shows the smallest and the largest assembly used. The scintillators were painted with Tygon paint on all but the surfaces in contact with

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[4] E.T. Larson, and H.A. Levine of Ansco reported very interesting results on the reduction of reciprocity failure with mercaptoozazoline development retarders (Phot. Sci. and Eng., 1, 59, 1957). It is likely that, with the aid of such retarders, most of the rate dependence of the system could be removed. However, the associated decrease in sensitivity would probably make it impossible to use the present assembly for total exposures of less than 20 mr.

the photographic material, and then wrapped in black tape. The surface upon which the radiation was incident was covered only with a thin layer of Bakelite (in practical use, a Bakelite container, rather than tape, could be used to enclose the entire system). Black tape also shielded one-half of the photographic emulsion from the visible light emitted by the scintillator. This provided a comparison between photographic densities due to high-energy photons alone and due to light photons plus high-energy photons. In the exposure range covered in this study, the area shielded from the scintillator light showed no density above that due to base and emulsion fog.

Experiments with several different photographic materials finally led to the decision to investigate more extensively the Eastman spectroscopic plates, type 103a-O, usually employed for astronomical photography. In this material, the onset of low-intensity reciprocity failure is shifted to lower intensities than in other commercial emulsions. Emulsion chemists achieve this partially by increasing the number of silver ions on the emulsion surface, which facilitates the formation of precursors of the latent image, which then grow into latent images upon addition of only very little energy. This explains why such emulsions are less stable than other commercially available photographic emulsions. In spite of their relatively low stability, it was possible to use the Eastman 103a-O plates at room temperature and at 40 percent to 50 percent relative humidity over at least a period of one week, without any appreciable changes in background and in sensitivity. Stored in the refrigerator, the plates remained usable for more than a year. We obtained the commercial product, consisting of a single emulsion layer coated on glass, and made a plate-scintillator sandwich, whenever possible employing a plastic spacer to avoid undue pressure on the emulsion. When ordered in bulk, the same emulsion can also be obtained coated on one or both sides of a film base.

The response of the plate-scintillator assemblies was studied as a function of total exposure, exposure rate, and photon energy, with the radiation incident perpendicularly to the surface of the photographic plate. The response was not studied as a function of the direction of the incident radiation. Because of the similarity in shape, it is likely that the response of the assemblies as a function of the direction of radiation incidence is quite similar to that of the AWRE assemblies (see table 1 for reference).

2.4 Results

a. Characteristic curves

Figure 2 shows the response of the large scintillator assembly to gamma radiation at several different rates from about 12 mr/hr down to 0.08 mr/hr, and to the laboratory background rate of 0.023 mr/hr. The assembly records exposures as low as about 0.5 mr and less; however, the characteristic curves shift and change shape with exposure rate. The change in shape is most pronounced for the background rate.

Figure 3 shows the same phenomenon for the small scintillator. This assembly makes possible an exposure interpretation down to about 2 mr, but the rate dependence is seen to be even more pronounced;

this is to be expected, since the smaller scintillator emits less light per incident gamma photon than the larger scintillator. Note that while rates down to about 0.02 mr/hr were included in figure 2, the lowest rate shown here is 1 mr/hr.

b. Limitations due to rate dependence

Figure 4 shows--for both scintillators--the exposures required for three different densities as a function of exposure rate. With the small scintillator, the useful rate range only extends down to about 15 to 20 mr/hr. With the large scintillator, at a density of 0.5, corresponding to about 0.5 mr, there is little rate dependence all the way down to laboratory background rates. For higher densities, the exposure interpretation is accurate to within ± 25 percent from about 0.1 mr/hr up. It is to be expected that, with scintillators larger than our 2" x 2" specimens, and with double-coated photographic material, it will be possible to obtain an adequate interpretation of exposures as low as 1 mr for rates all the way down to natural background.

c. Energy dependence

There is, however, one disadvantage in employing large scintillators. As the scintillator size is increased, the energy dependence of the assembly's response increases due to the change in spatial distribution of the attenuation of the primary radiation and of the luminescence light, as discussed in footnote 1. Figure 5 shows that, over the energy range from about 100 kev to Co^{60} energies and higher, the sensitivity of the assembly employing the 1" x 1/2" scintillator varies by only ± 25 percent, while for the large scintillator, the variation over the same range is nearly ± 50 percent.

It may be noted that, wherever possible, the same exposure rate was used to obtain all energy-dependence data. Because of the change in curve shape with exposure rate, the shape of the energy dependence curve changes slightly with exposure rate--and also with the density level for which the sensitivity is calculated. However, the rates here employed are high enough to make these variations unimportant for the present study.

d. Summary

The results of this study indicate that it is quite feasible to use large film-scintillator assemblies to measure very low total exposures at very low rates, but only over a limited range of rather low film densities. This requirement can be met by limiting the total time over which the assembly is exposed. Smaller film-scintillator assemblies can be used only to interpret exposures at rates above a certain minimum value. Such a complicated procedure cannot be recommended for routine personnel dosimetry.

3. LIMITATIONS IN CONVENTIONAL PHOTOGRAPHIC DOSIMETRY DUE TO ENVIRONMENTAL EFFECTS

In personnel dosimetry, it is still advisable to use conventional film packets in suitable holders, and to extend their useful range by extending the period over which the holders are carried. This procedure is gaining in popularity, particularly in view of the change in thought on how to specify maximum permissible exposure in a biologically meaningful way.[5] As a consequence, we have been asked on several occasions for advice on the stability of the latent image in the time interval between its formation and processing.

Results on studies of changes in photographic response with storage time and with temperature during and after exposure have been obtained earlier at the National Bureau of Standards, but only over periods of one week or less.[6] Data over periods up to thirteen weeks were more recently obtained by Ziegler and Chleck[7] under controlled laboratory conditions. A more realistic study over a period of thirteen weeks is now being performed at the National Bureau of Standards. In general, it may be said that the interplay of a variation in temperature and in the relative humidity of the atmosphere produces a complicated pattern of behavior of the latent image. According to the particular experimental conditions employed, the result may be either an increase or a decrease in the photographic response.

In personnel dosimetry, there is little one can do to protect the monitoring films from large changes in ambient temperature. However, the influence of humidity and of changes in the chemical composition of the atmosphere can be readily eliminated. A simple hermetically sealed plastic container provides adequate protection.

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[5] Handbook 59, Addendum, U.S. Department of Commerce, National Bureau of Standards, 1958.

[6] Handbook 57, U.S. Department of Commerce, National Bureau of Standards, 1954.

[7] Ziegler, C.A., Chleck, D.J., Latent-image fading in film badge dosimeters, to be published.

4. INCREASE IN SENSITIVITY WITH EFFECTIVE EMULSION THICKNESS

In conclusion, it will be demonstrated that, in some instances, an increase in effective emulsion thickness, achieved by stacking a number of identical films during exposure and densitometry, may lead to an extension of the useful range of a film badge below 20 mr, without the use of a scintillator.

Figure 6 shows a characteristic curve of DuPont film type 502, obtained with narrow bands of X-radiation of about 210-kev effective energy, once with single films, and the other time with three films exposed and densitometered on top of each other. Percentagewise, the limitation in precision of the exposure interpretation resulting from variations in emulsion coating and uneven processing is the same for the two curves. However, if one uses a densitometer whose precision limit is, roughly, ±0.02 density units (such as, for instance, the Ansco Sweet Color Densitometer), tripling the density for a given exposure improves the accuracy of the exposure interpretation considerably.

Table 2 shows the extent of improvement in the precision of the exposure interpretation with DuPont film type 502, under the assumption that the density produced by a given exposure is reproducible to within ± 5 percent or ± 0.02 density units, whichever is greater. Notice that, whereas with a single film the precision in the interpretation of an exposure of, for example, about 100 mr is only ± 40 percent, it is ± 14 percent with a stack of three films.

Figure 7 shows the results of triple versus single densitometry on the more sensitive DuPont film type 555 (packaged without lead backing as personnel monitoring film type 554), exposed to Co^{60} gamma radiation. While, with a single film, the low-exposure limit for a ± 25 percent precision in exposure interpretation is 61 mr, it is 22 mr for triple stacks.

Figure 8 shows that for the even more sensitive Kodak film type KK, (packaged as personnel monitoring film type 1), the low-exposure limit for a ± 25 percent precision is 23 mr for the single film, and 8.2 mr for the triple stack. Actually, an exposure of about 4 mr can still be interpreted with an accuracy of about ± 50 percent with three films stacked, while no reasonable information at all could have been obtained for such a low exposure with one single KK film.

5. SUMMARY AND CONCLUSIONS

Because of insufficient sensitivity of available photographic materials, photographic X- and gamma-ray dosimetry at total exposures in the range from 1 to 20 mr requires special procedures, which, in turn, may introduce new problems.

If one chooses to intensify the high-energy photon image through the action of visible light from a scintillator, one reduces the energy dependence of the film device without introducing metallic filters, but, at the same time, one restricts its applicability because of the presence of low-intensity reciprocity failure. In some instances, an increase in effective emulsion thickness, achieved by using stacks of identical films, may lead to an increase in emulsion sensitivity sufficient to extend the useful range of a film badge well below 20 mr, without the use of a scintillator.

In applications to personnel dosimetry, it may be possible to avoid dosimetry below 20 mr entirely, by extending the monitoring period. However, by doing this, one introduces additional difficulties because of instabilities in the photographic latent image. Realistic long-term studies of the effect on the latent image of variations in relative humidity and temperature are scarce. More work in this direction is under way at the National Bureau of Standards.

Table 1. Photographic film - scintillator dosimeters

Authors, References	Type of		Range of			Accuracy of exposure interpretation
	film	scintillator	exposure dose	exposure rate	photon energy	
Hoerlin, H., Kaszuba, F. Bull.A.P.S., 27,23,1952	Ansco, special, with little reciprocity failure	intensifying screens P-terphenyl + $PbCl_2$ in Lucite	20 mr to 10 r	down to ~ 5 mr/hr	from ~ 20 kev up	± 25%
Barnaby, C.F., AWRE O-28/56	Ilford dosimeter film type PM-1	cylinders, ~ 5 cm diam. x 3 cm height P-terphenyl in polystyrene	5 mr to 10 r	down to ~ 300 mr/hr	from ~ 50 kev up	± 25%
Kogan, K.M., Pereyaslova, N.K., Prib. i Tekhn.Eksp., 4, 25, 1957	Photofilm ϕC-3	cylinders, 3 cm diam. x 1 cm height NaI(Tl)	0.5 mr to 50 r	not discussed		

Table 2. Improvement of precision in exposure interpretation through stack densitometry.

Exposure (mr)	Precision Limit			
	stack		single	
	(mr)	(percent)	(mr)	(percent)
24.6	± 14	± 57	± 40	± 160
26.8	± 14	± 52	± 40	± 150
49.2	± 14	± 28	± 40	± 81
55.4	± 14	± 25	± 40	± 72
98.4	± 14	± 14	± 40	± 41
110	± 14	± 13	± 40	± 36
277	± 15	± 5.4	± 40	± 14
554	± 29	± 5.2	± 40	± 7.2
1100	± 65	± 5.9	± 56	± 5.1

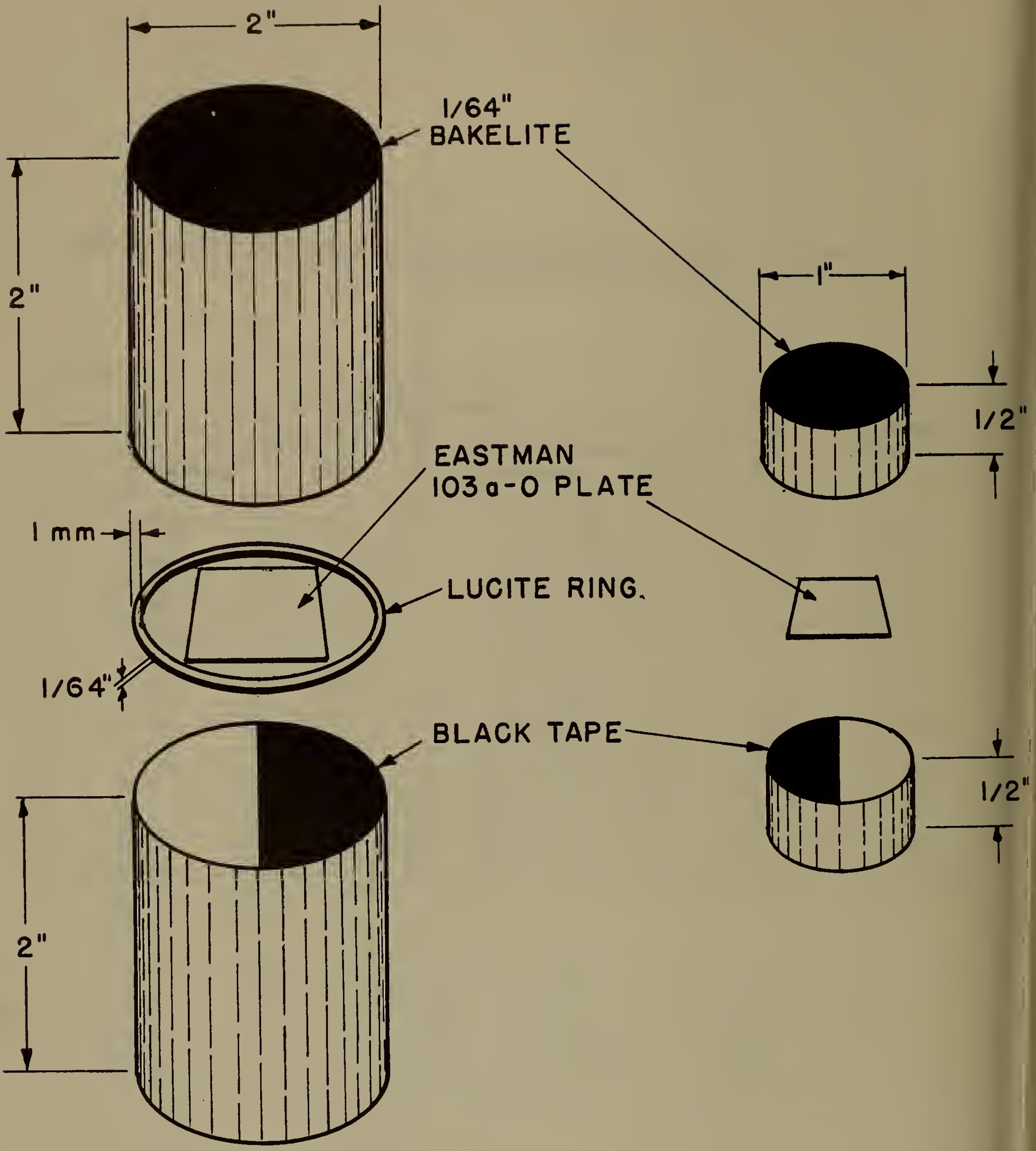

Figure 1. Assemblies of photographic plate and Pilot B plastic scintillato

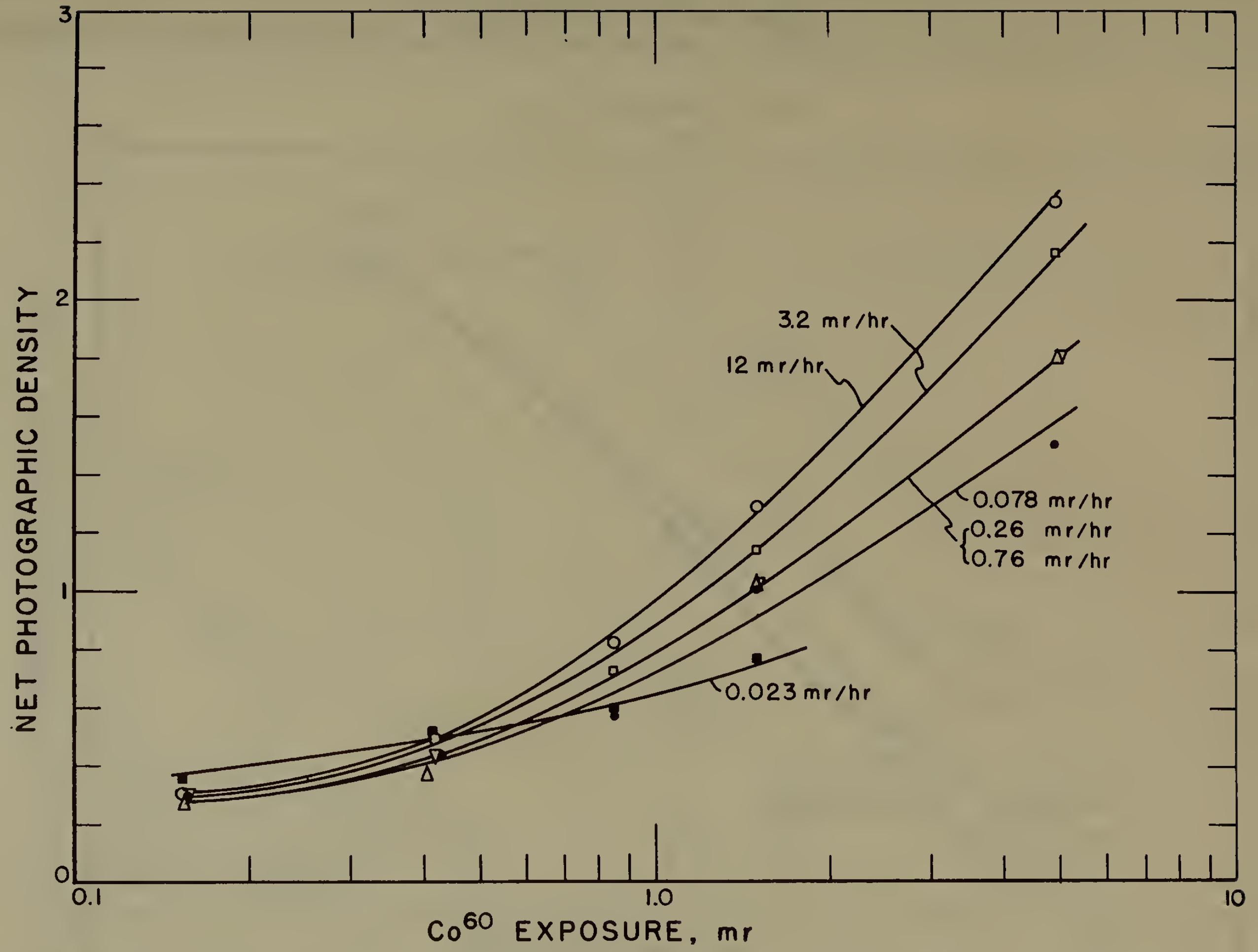

Figure 2. Response of Eastman Spectroscopic Plate 103a-0, sandwiched between two 2" x 2" scintillators, at various exposure rates.

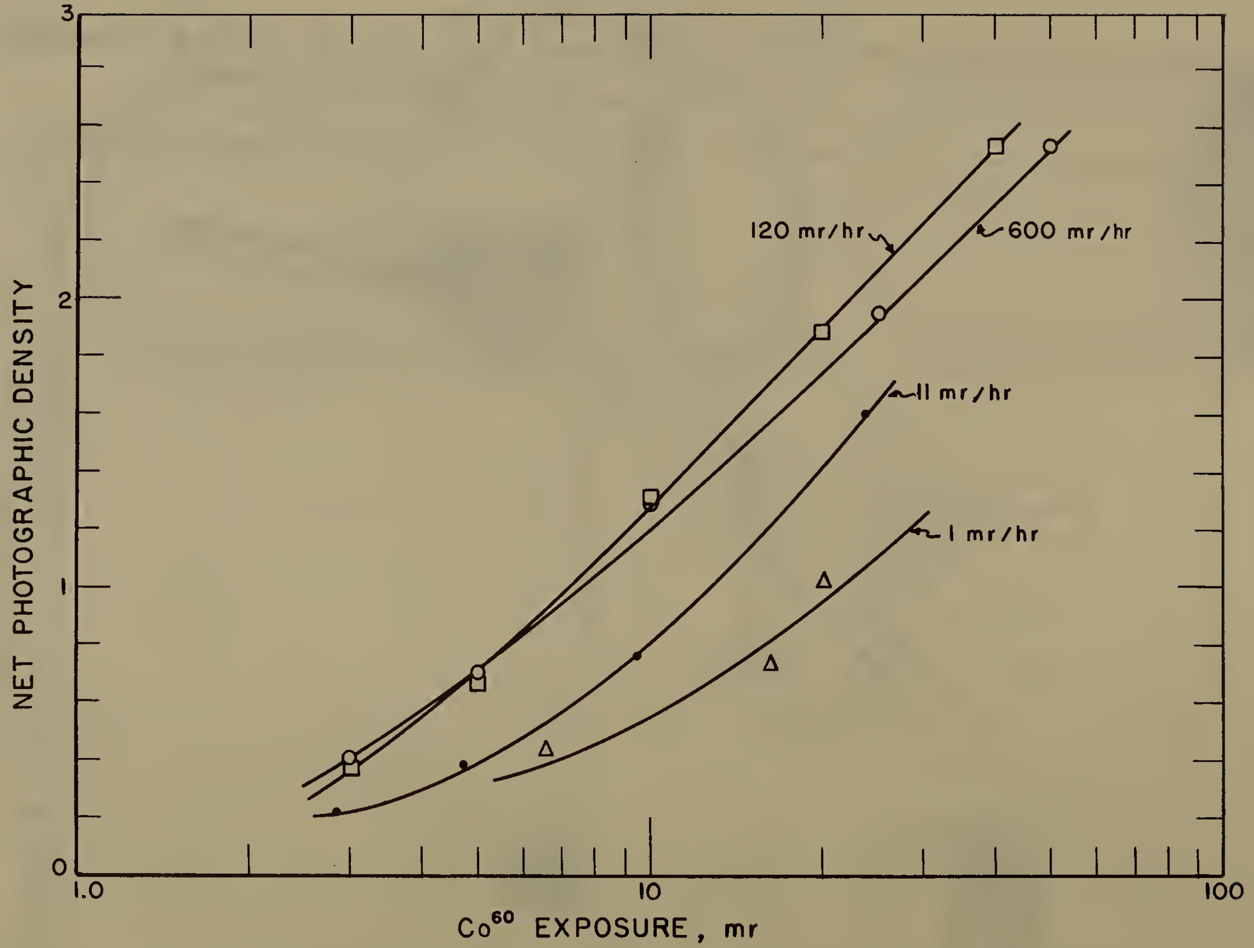

Figure 3. Response of Eastman Spectroscopic Plate 103a-O, sandwiched between two 1" x 1/2" scintillators, at various exposure rates.

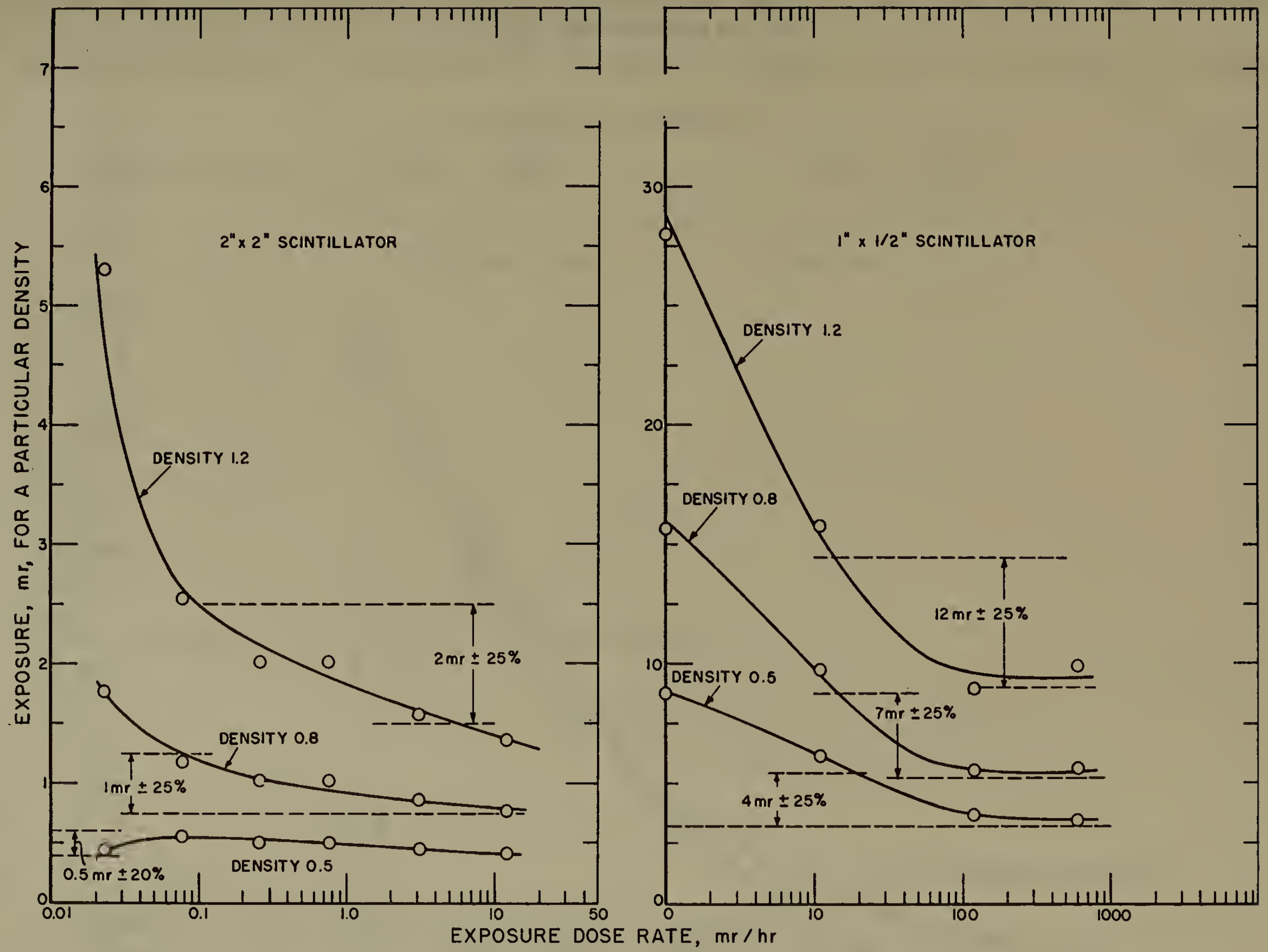

Figure 4. Rate dependence of scintillator assemblies exposed to Co^{60} gamma radiation.

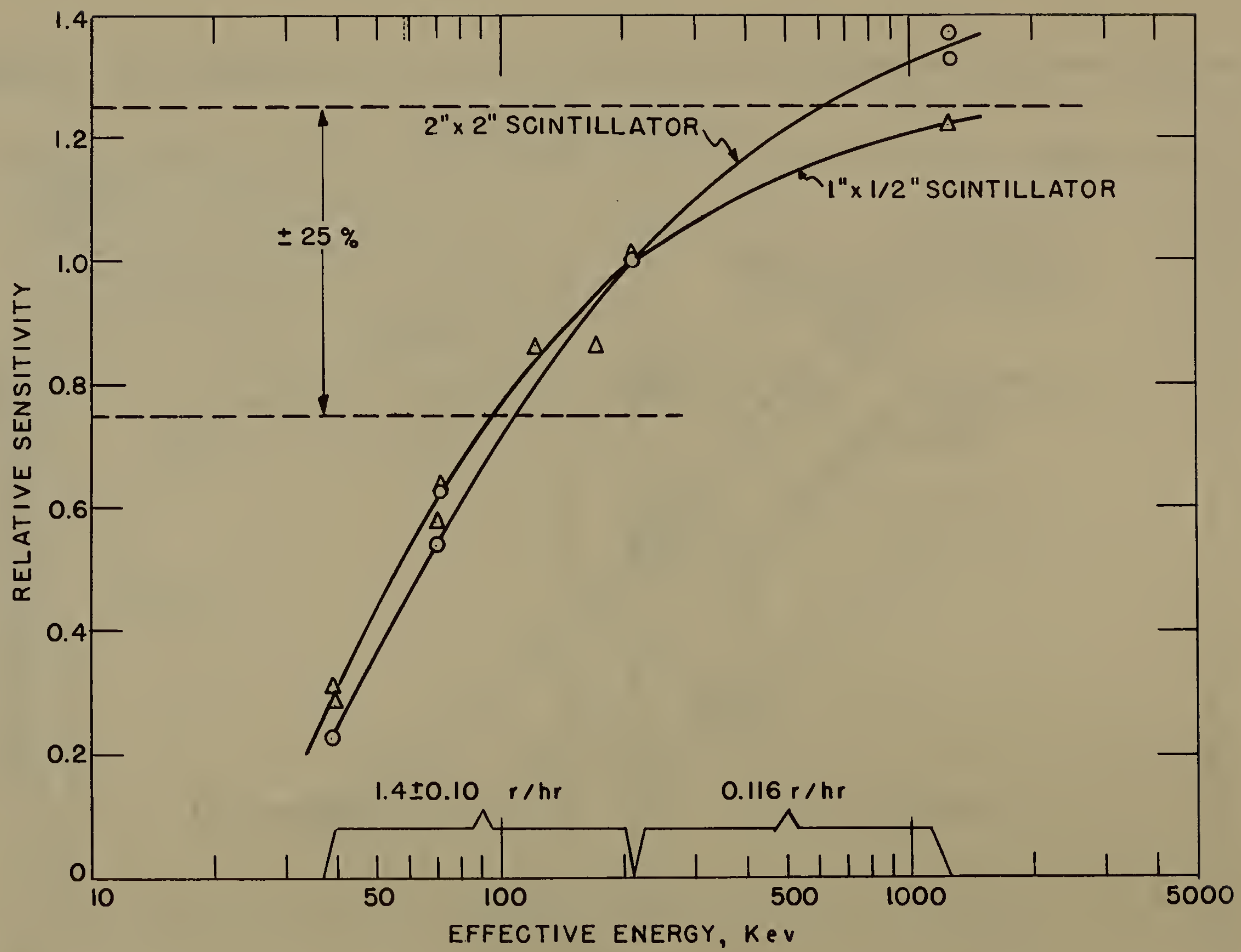

Figure 5. Energy dependence of scintillator assemblies, exposed to narrow bands of X-radiation and to Co^{60} gamma radiation.

The 215-kev point was obtained both at the low rate and at the high rate. No correction for reciprocity failure was applied to the Co^{60} point, although it was obtained at the low rate only.

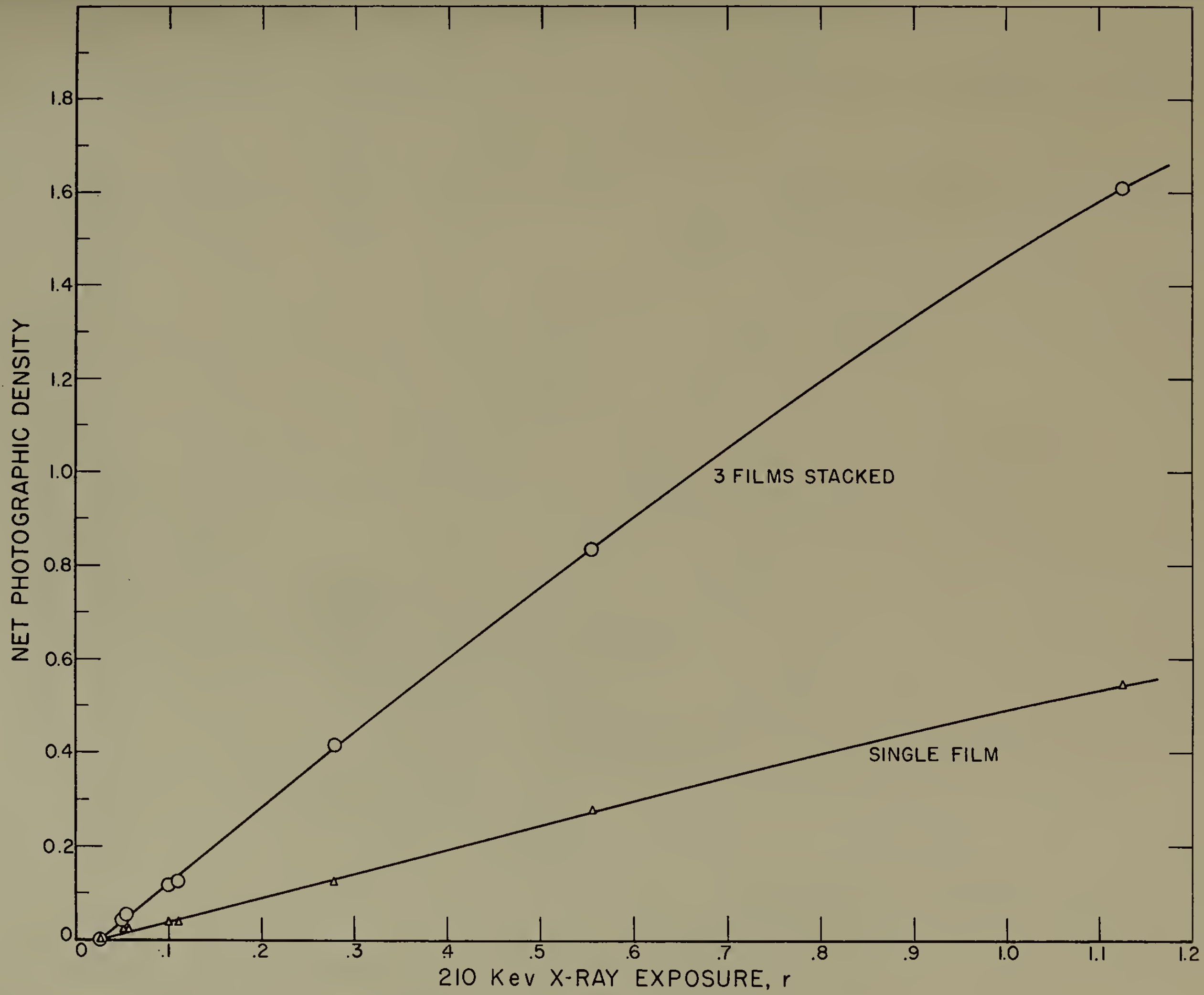

Figure 6. Improvement in precision through stack densitometry of DuPont Film Type 502.

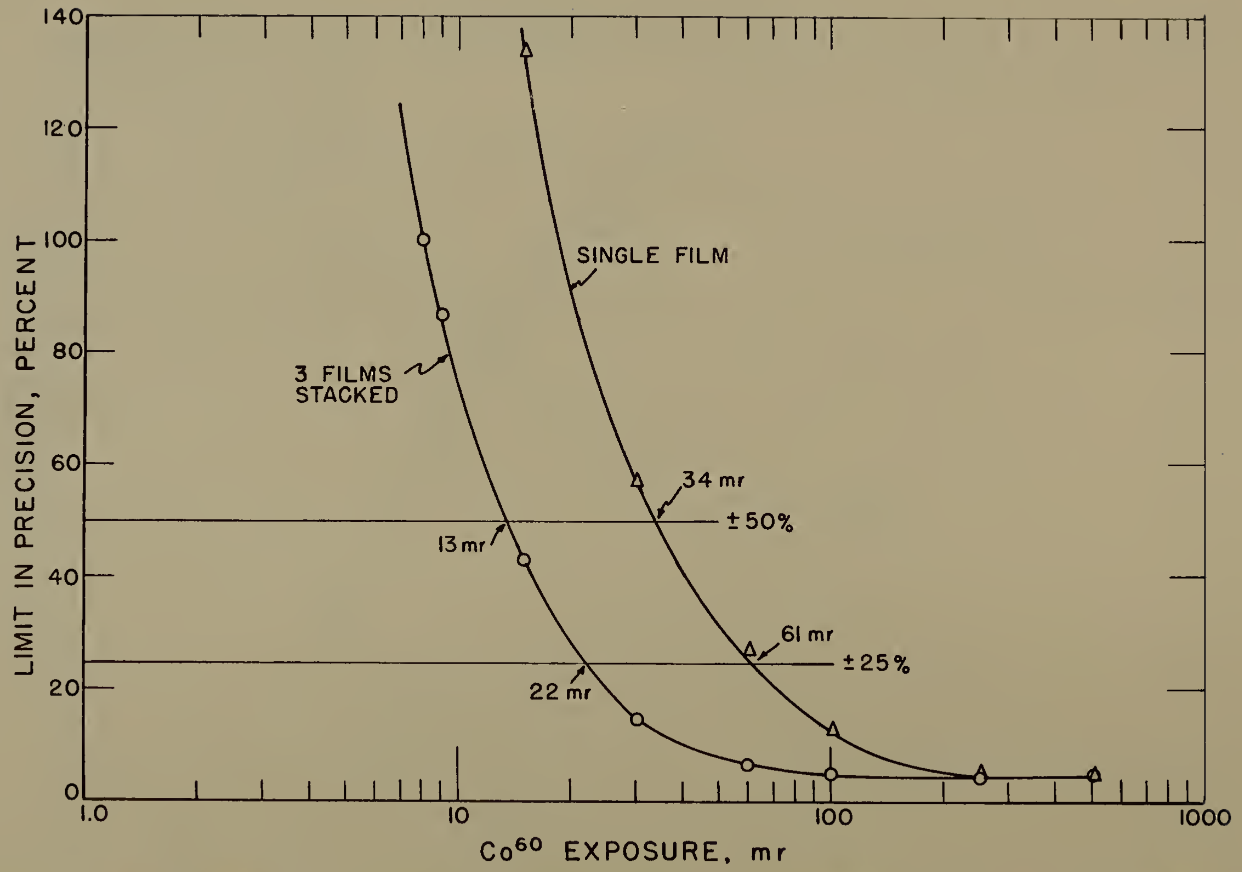

Figure 7. Improvement in precision through stack densitometry of DuPont Film Type 555.

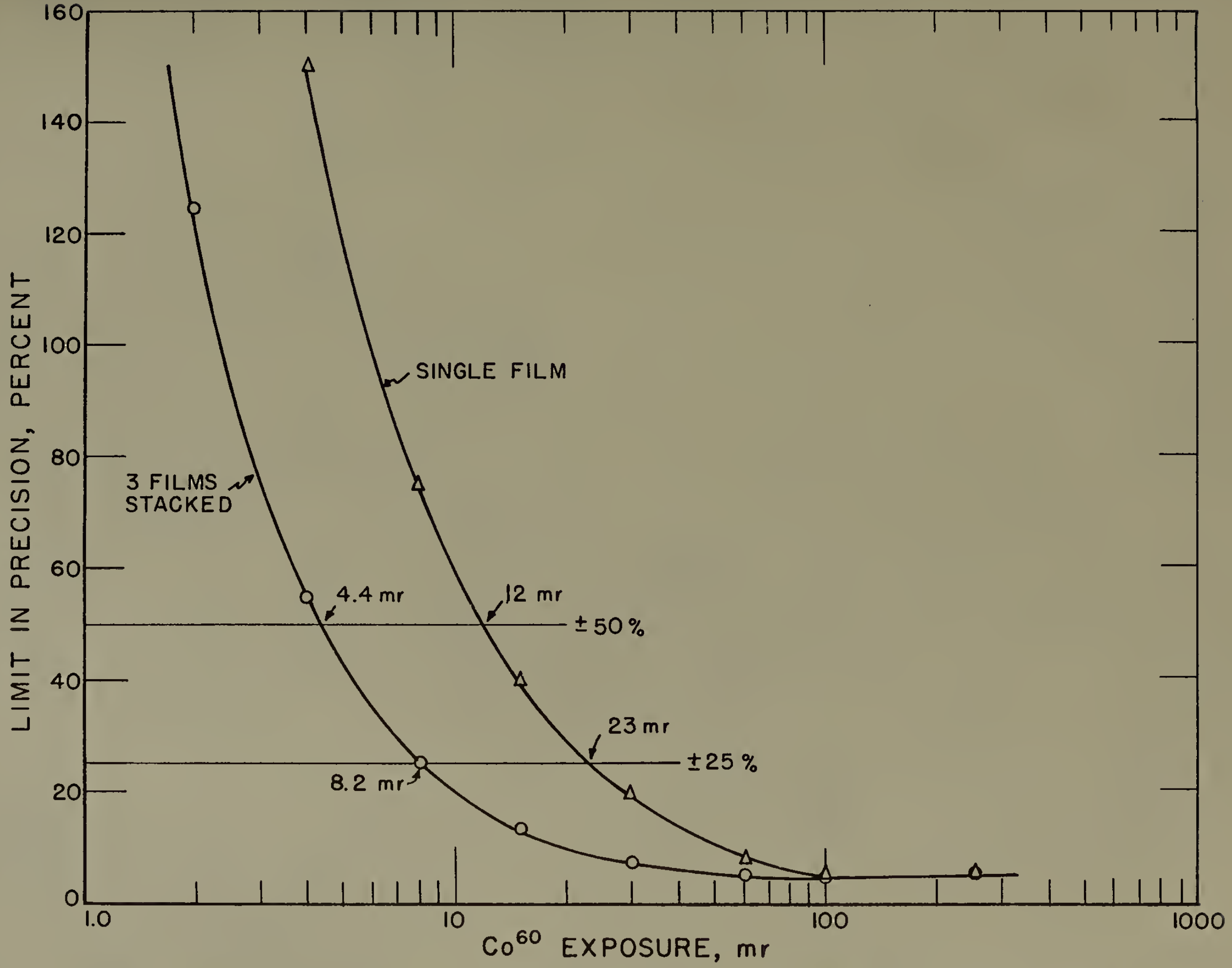

Figure 8. Improvement in precision through stack densitometry of Kodak Film Type KK.

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