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EVALUATION OF UNEXPECTEDLY LARGE RADIATION EXPOSURES BY MEANS OF PHOTOGRAPHIC FILM

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FOREWORD

This Note covers the material presented by the author at the 1962 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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EVALUATION OF UNEXPECTEDLY LARGE RADIATION EXPOSURES BY MEANS OF PHOTOGRAPHIC FILM*

William L. McLaughlin

Conventional film types used in personnel monitoring film badges are suitable for measuring X- and γ -radiation exposures only up to 1000 R. By using special processing procedures, it is possible to extend the range of the less sensitive component of most commercial film packets up to at least 10,000 R. Limitations in precision of readings due to changes in rate dependence, energy dependence, and changes in the shape of the characteristic curve in this range are discussed.

Sometimes it is important to recover photographic dosimetry data that might be lost in routine experimental practice because the total radiation exposure happened to be higher than the upper limit of the useful range of the film. The useful range of photographic dosimetry can be extended beyond its conventional upper limit of about 1000 R by:

1. Use of Supplementary Emulsions of Low Sensitivity. Some of the very fine-grain, spectroscopic films show a useful response to 1-Mev X-rays up to 100,000 R [1]**, but these films are not always satisfactory for dosimetry, because their latent images are not very stable [2].

2. Developed Silver Analysis. One of the most accurate methods for determining the amount of silver in a developed film is X-ray fluorescence spectrometry [3]. One measures the silver K-fluorescence excited in a piece of developed film, which is being irradiated by X-rays. This method gives greater accuracy than routine densitometry and is now finding wide sensitometric use. It can be utilized to extend the upper limit of the useful range of some films by a factor of about two. Another way to analyze the silver content in a developed film is by activation of the silver by neutrons in a reactor thermal column, and measuring the induced activity with a scintillation counter [4].

3. Utilization of the Print-Out Effect. The print-out effect begins to be useful only when the exposure is well beyond 10,000 R [5], and in order to cover the range between 1000 and 40,000 R by this method, the films must be given special chemical treatments prior to exposure [6].

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**Figures in brackets indicate literature references at the end of this paper.

4. Special Development Procedures. By using conventional developing solutions containing certain chemical retarders, or by selective development of internal regions of the emulsion grains, it is possible to decrease the film sensitivity and retain sufficient contrast, so that some film types achieve useful ranges up to at least 10,000 R.

This report gives results of studies at the National Bureau of Standards on the fourth point; namely, the usefulness of several special processing procedures for extending film dosimetry ranges. Suppose a dosimeter film packet has been exposed beyond its normal useful range. One would like to know how reliable would be the exposure evaluation when one of the special development techniques is used. People interested in increasing photographic dosimetry ranges have tried using incomplete, localized development of the emulsion grains [7]; that is, they would use a very slow-acting developer for a relatively short time or they might decrease the developer concentration or temperature, such that only the surface of the grains would be affected. This often led to troubles in the form of very uneven density distributions, or a tendency for the characteristic curve shapes to vary widely with exposure rate, especially in the higher density regions. The results of this study show that these difficulties are not so severe if one uses certain chemical retarders in the X-ray developer, or if one develops the internal latent images rather than the surface latent images.

Some of the more successful special development techniques are listed in the table, along with the conventional technique. Adding 200 grams of sugar to a liter of commercial X-ray developer results in loss in film sensitivity at high density regions by a local exhaustion effect [8] (solution 2). One gram of 6-nitrobenzimidazole added to a liter of X-ray developer also acts to reduce the rate of development [9] (solution 3). Similar reductions take place with the addition of other development retarders, such as the mercaptotetrazoles [9,10].

Development localized in internal regions of emulsion grains was achieved by using solutions which etch away part of the grain, either during development (solutions 4 and 5) or prior to development (solution 6). Solution 4 is a fine-grain metol-hydroquinone developer buffered with borax and containing 2 grams per liter of potassium thiocyanate as a silver halide solvent. Solution 5 is an Amidol stabilization processing solution containing thiourea, described by Levinos and Ehrlich [11]. Solution 6 is an internal developer based on the method of Stevens [12,13]. It involves bleaching away the surface of the grains with an acid dichromate bath and then processing in a metol-hydroquinone developer containing a small amount of sodium thiosulfate as silver-halide solvent.

TABLE

Development to Increase Useful Range of Dosimeter Films
(all at 20°C)

<u>Solution No. ^a</u>	<u>Development</u>	<u>Dev. Time (min.)</u>
<u>CONVENTIONAL</u>		
1	Commercial X-ray developer	5
<u>RETARDED</u>		
2	Commercial X-ray developer, with sugar (200 g/l)	5
3	Commercial X-ray developer, with 6-nitrobenzimidazole (1 g/l)	5
<u>INTERNAL</u>		
4	Metol-hydroquinone developer buffered with borax, with thiocyanate (2 g/l)	10
5	Amidol stabilization developer, with thiourea	3
6	Metol-hydroquinone developer buffered with sodium carbonate, with thiosulfate, (0.5 - 2.0 g/l) ^b , preceded by acid dichromate bleach	6

a. These numbers serve as keys on the curves of the figures.

b. The optimum amount of thiosulfate was determined experimentally for each film type.

Figure 1 shows characteristic curves of DuPont Industrial X-ray film, type 510, exposed to Co^{60} γ -radiation and developed in the various solutions listed in the table. The curves marked "1" are for conventional commercial X-ray developer used in dosimetry, with both emulsion sides (solid curves) and with one emulsion side removed (dashed curves). The other curves are for the special development techniques. The slopes of these curves are still relatively high in the 1000 R to 10,000 R range, although the two curves (2 and 3) for chemically retarded development show a tendency to saturate at relatively low density levels. The curves obtained with internal development are shown by 4, 5, 6, and 7. Curves 6 and 7 are for two different concentrations of hypo in the developer; curve 6 was for 0.5 g/l and curve 7 was for 0.7 g/l. The fact that these curves are generally steeper than curves 2 and 3, particularly for exposures beyond 10,000 R, makes them more desirable from the standpoint of giving greater precision in the density measurements. If one assumes that the limits of precision in reading the density are ± 0.02 density units, then a 5,000-R exposure reading on curve 6 would have precision limits of about 5 percent. This certainly is not as good as the 1-percent precision limits of readings at a similar density level on curve 1, but is still useful.

Figure 2 shows similar curves for DuPont Ciné-Positive film, type 834.¹ Again, special processing provides readings from 1000 R to 10,000 R, with a precision nearly as great as that offered by routine processing. With some of the developers, however, this film shows solarization.

The less sensitive component film of Kodak Personnel Monitoring Packet, type 3, is represented in figure 3.² This film type has a wider useful range than the others; however, the slopes of the curves obtained with internal development are not quite as satisfactory, the precision limits of readings at 5000 R being as much as 10 percent for ± 0.02 density units. This film type also solarized in some instances above 10,000 R.

Variations in the energy dependence of these film types occurred when the various developers were used. Figure 4 shows relative sensitivity of a particular batch of DuPont Industrial X-ray film, type 510, over an effective photon energy range from 25 kev to 1000 kev, for the

¹ The same solution numbers apply as in figure 1. The concentrations of hypo in the developer for this film were 1.0 g/l for curve 6 and 2.0 g/l for curve 7.

² The hypo concentrations for the development of this film type (curves 6 and 7) were the same as those in figure 2.

different developers. It is interesting that with this very fine-grain emulsion, the internal developers provide smaller variations in spectral response than do the developers whose action is localized more on the surfaces of the grain. Also, with internal development, the spectral sensitivity is nearly constant in the lower part of the diagnostic X-ray range.

Figure 5 shows the energy dependence curves for DuPont Ciné-Positive film, type 834, which has larger emulsion grains.¹ In this case, the energy dependence is greater with internal development, but, as before, the low-energy response is flatter. A similar family of curves is seen in figure 6 for the less sensitive component of the Kodak Dosimeter film packet.¹

Figure 7 shows that, although there is no rate dependence with conventional X-ray development up to 10,000 R/hr, relatively wide variations in the film sensitivity occur with retarded and internal development. These curves are for DuPont Industrial X-ray film, type 510, and show variations in relative sensitivity, determined at about 1000 R of exposure (except for curve 1, which was made at a lower exposure level). The curves were plotted for different exposure rates, relative to the sensitivity at an exposure rate of 20 R/hr. Very similar rate dependence curves were found for the other film types. There are variations in curve shapes and maximum density levels with exposure rate, but these are not as severe as when slow surface development is used. Among these special development procedures, the one that produces the smallest rate dependence at exposure rates up to 1000 R/hr is the internal development with prior bleaching (curve 6). For the very high exposure rates, X-ray developer with sugar (curve 2) or with 6-nitrobenzimidazole (curve 3) is more suitable.

In summary, it should be pointed out that these special development techniques could be useful in emergencies when applied with some caution. A set of workable procedures should be established before an emergency arises.

¹ The numbers shown in the graph refer to the same developing solutions as those of figure 4.

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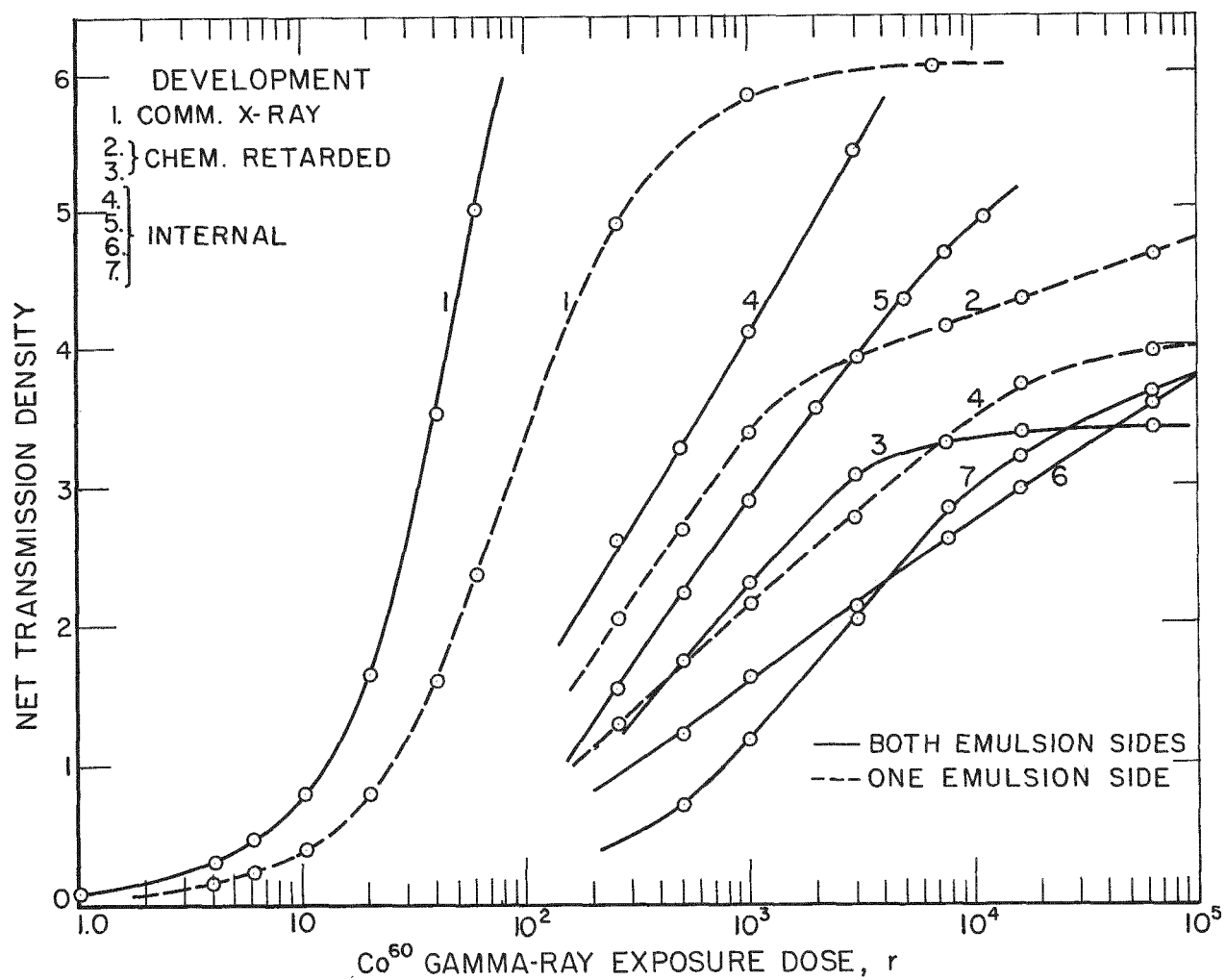


Figure 1. Characteristic curves of DuPont Industrial X-ray film, type 510.

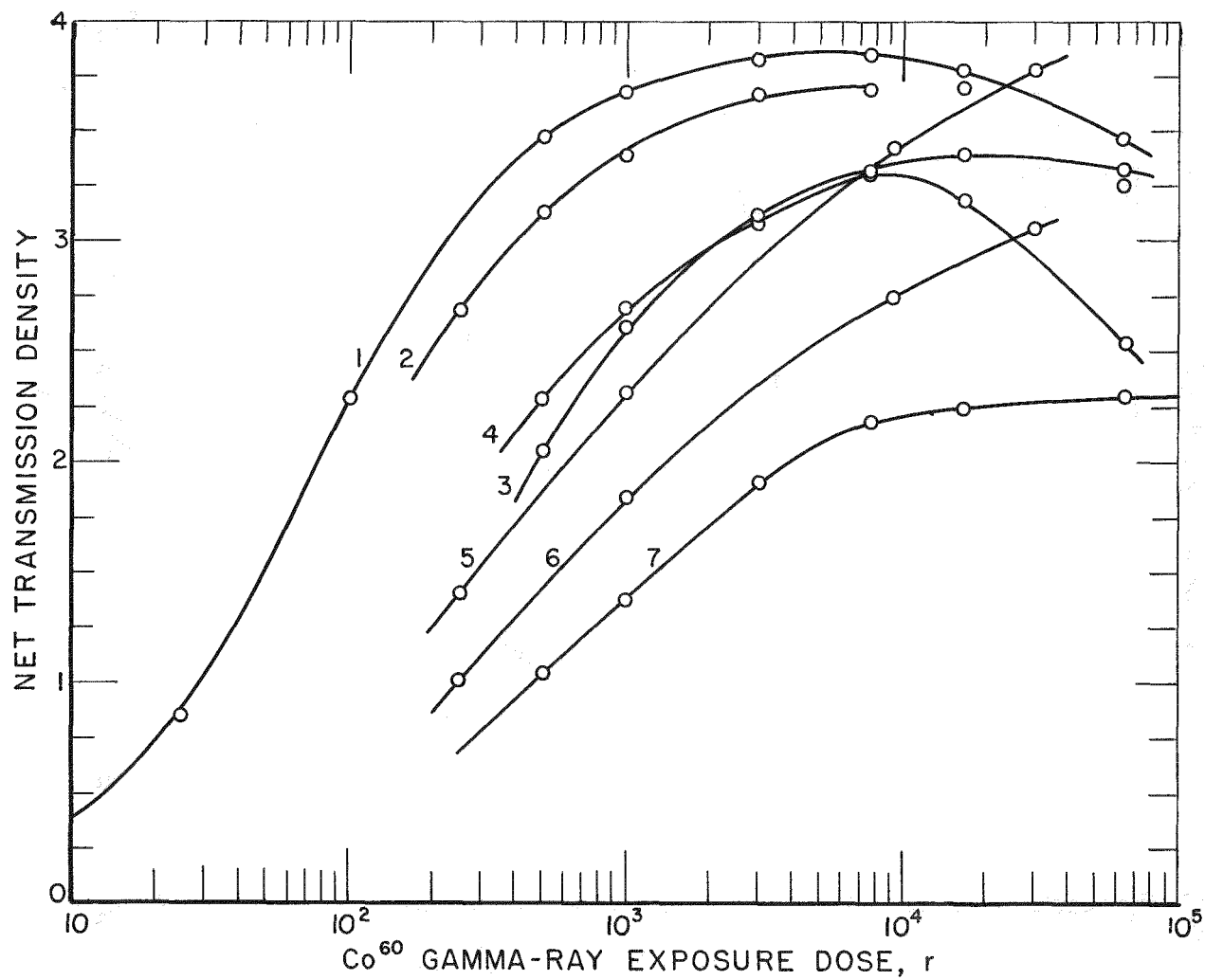


Figure 2. Characteristic curves of DuPont Ciné-Positive film, type 834.

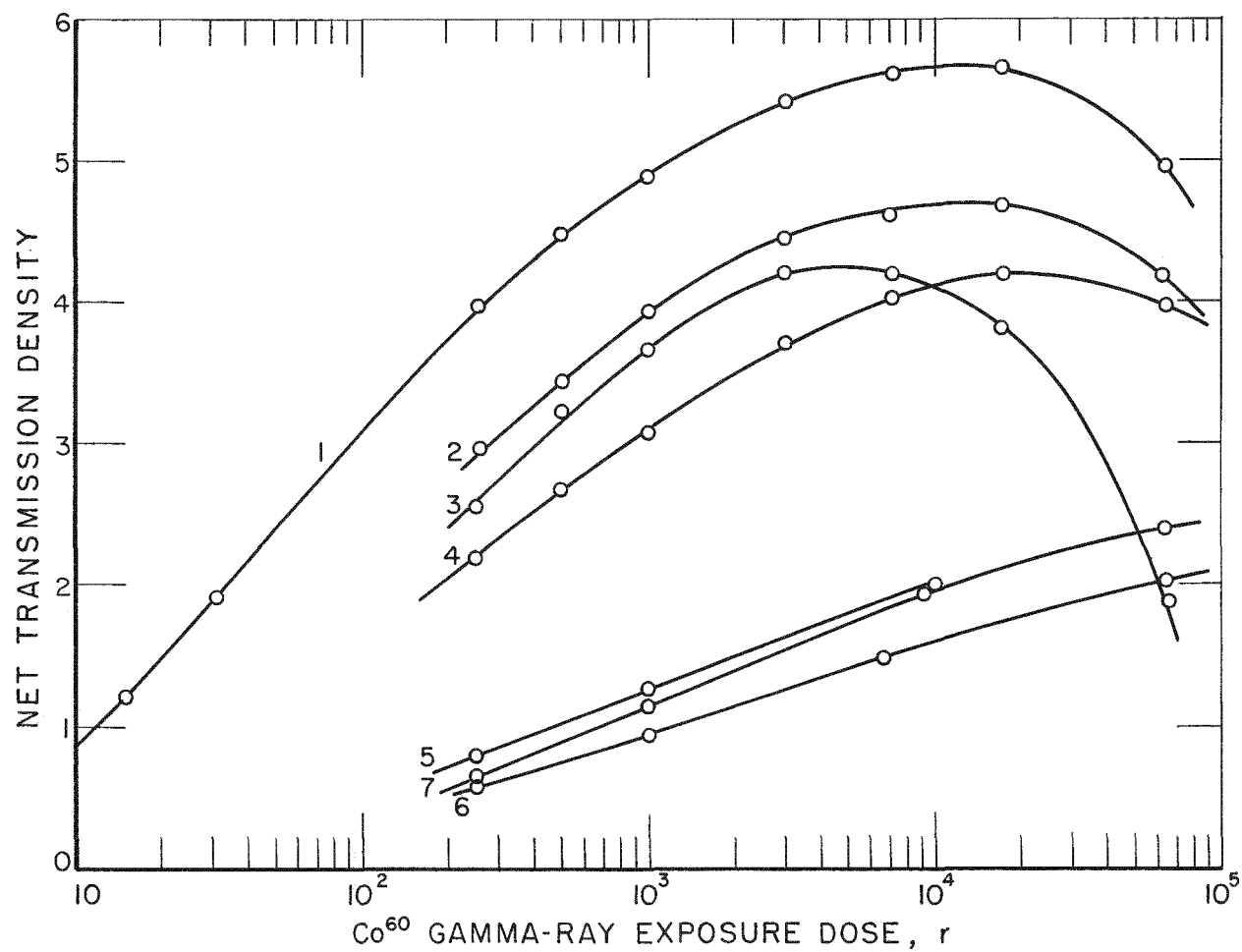


Figure 3. Characteristic curves of insensitive film of Kodak Personnel Monitoring Packet, type 3.

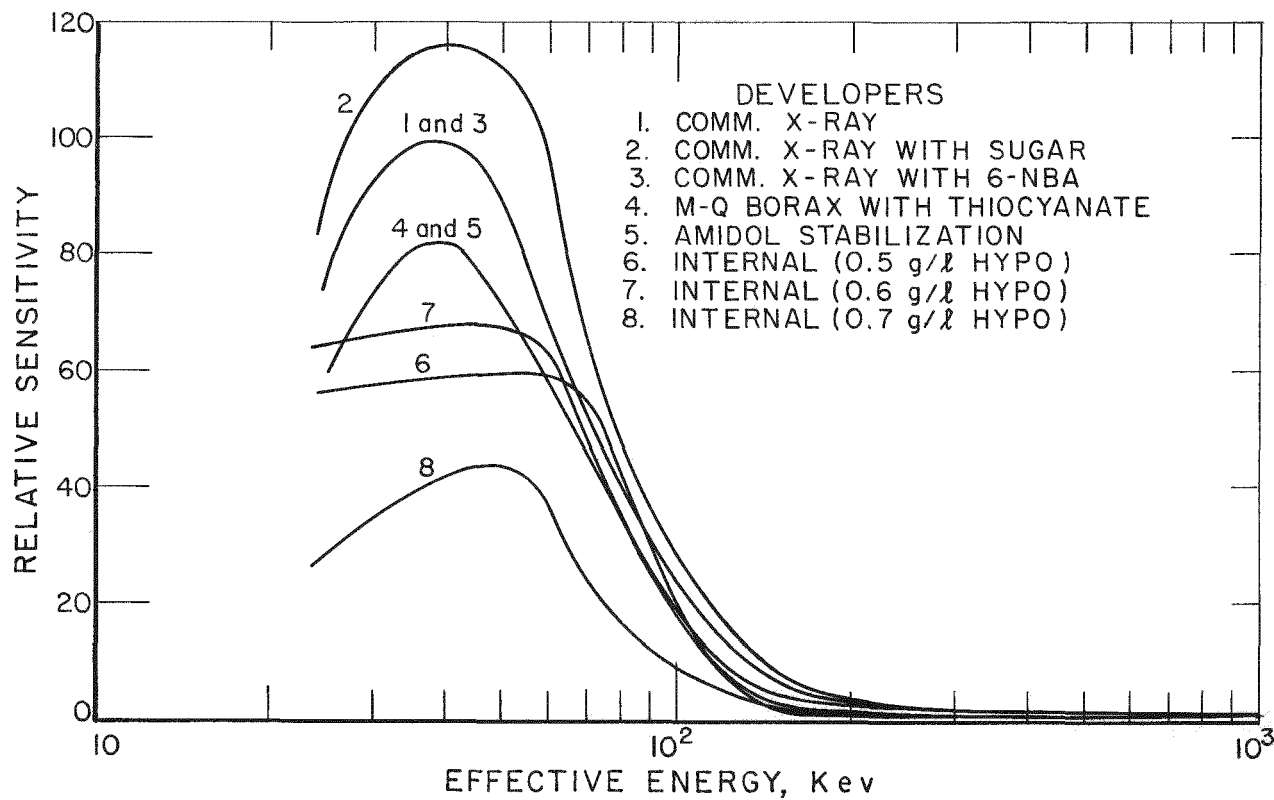


Figure 4. Spectral sensitivity curves of DuPont Industrial X-ray film, type 510.

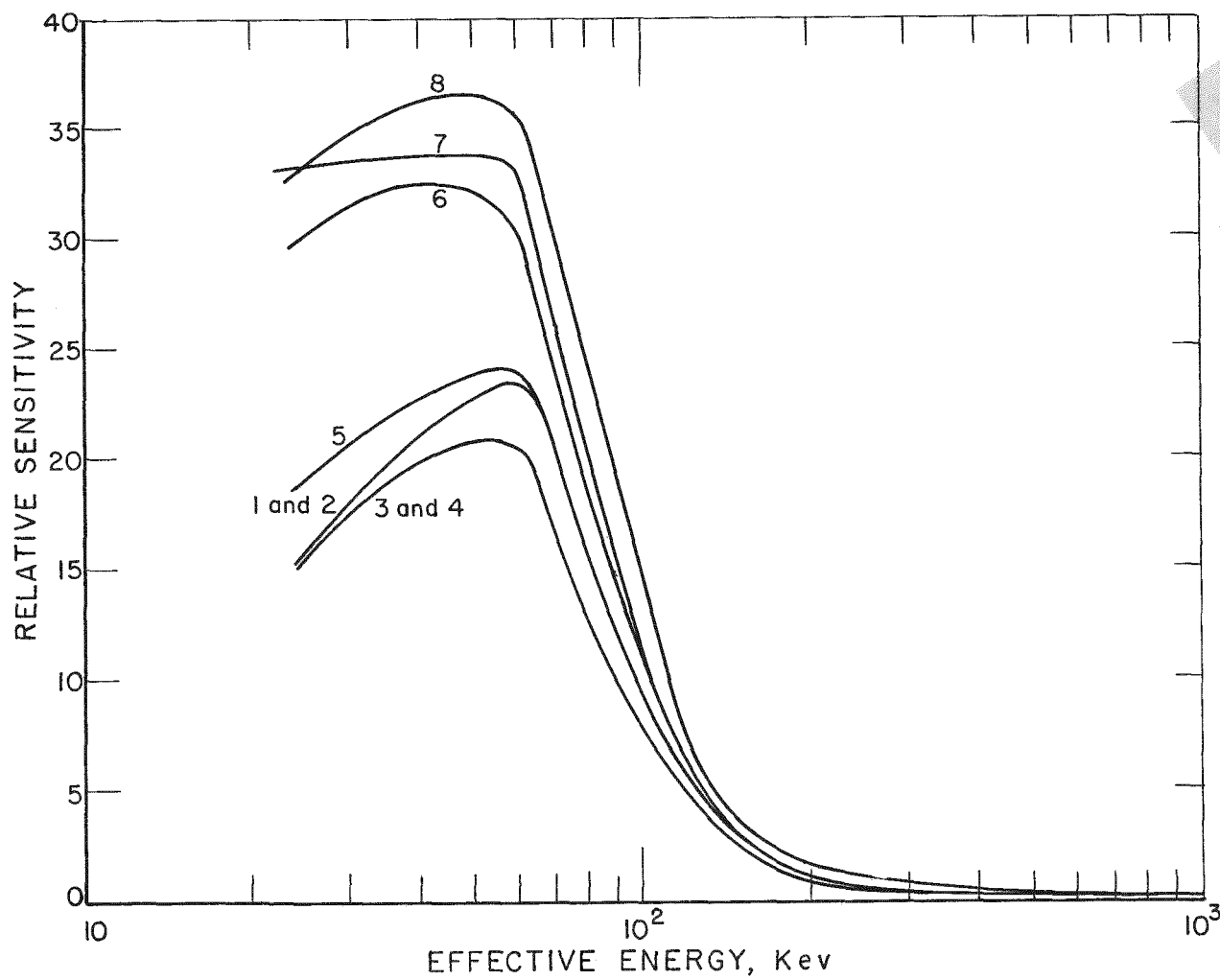


Figure 5. Spectral sensitivity curves of DuPont Ciné-Positive film, type 834.

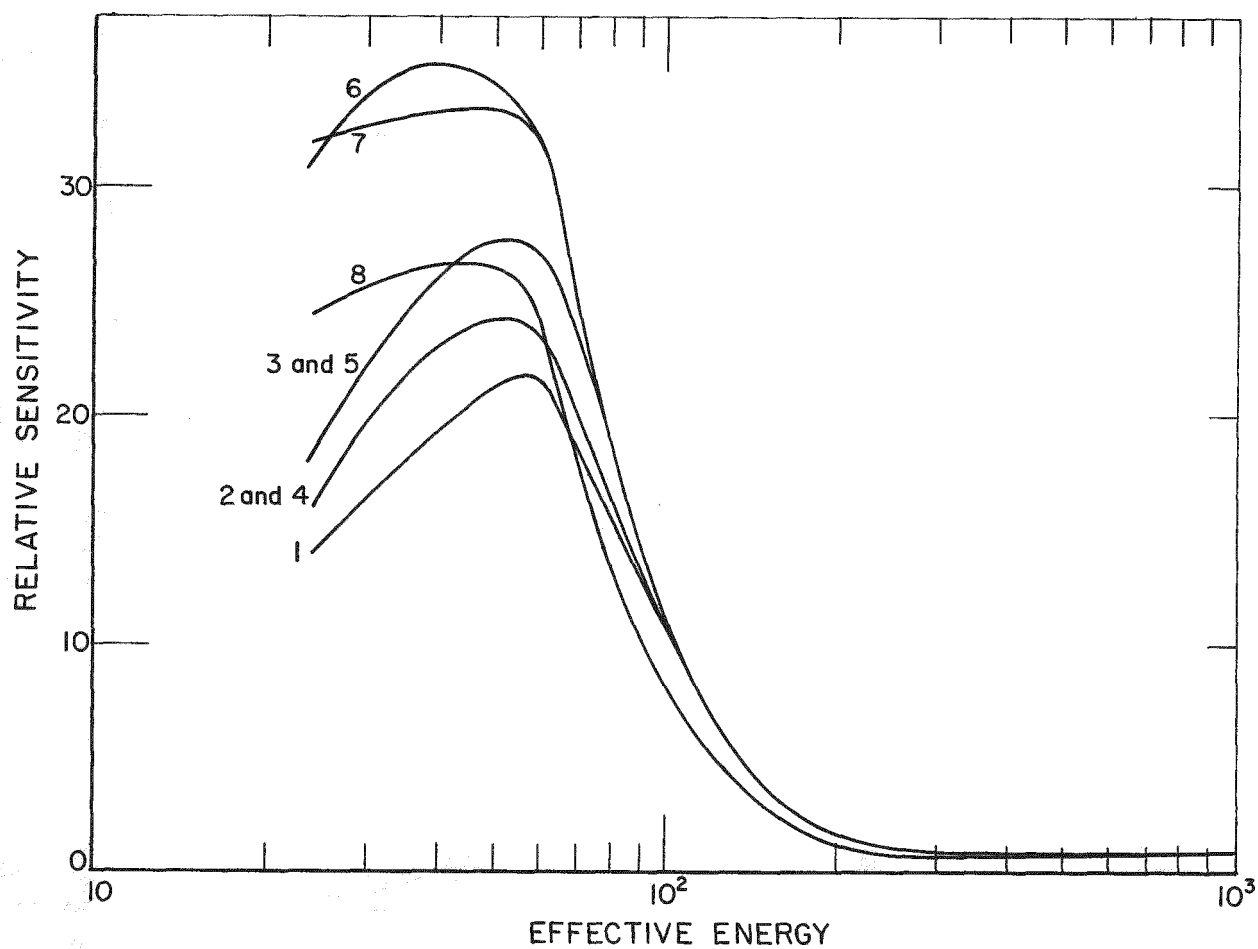


Figure 6. Spectral sensitivity curves of insensitizing film of Kodak Personnel Monitoring Packet, type 3.

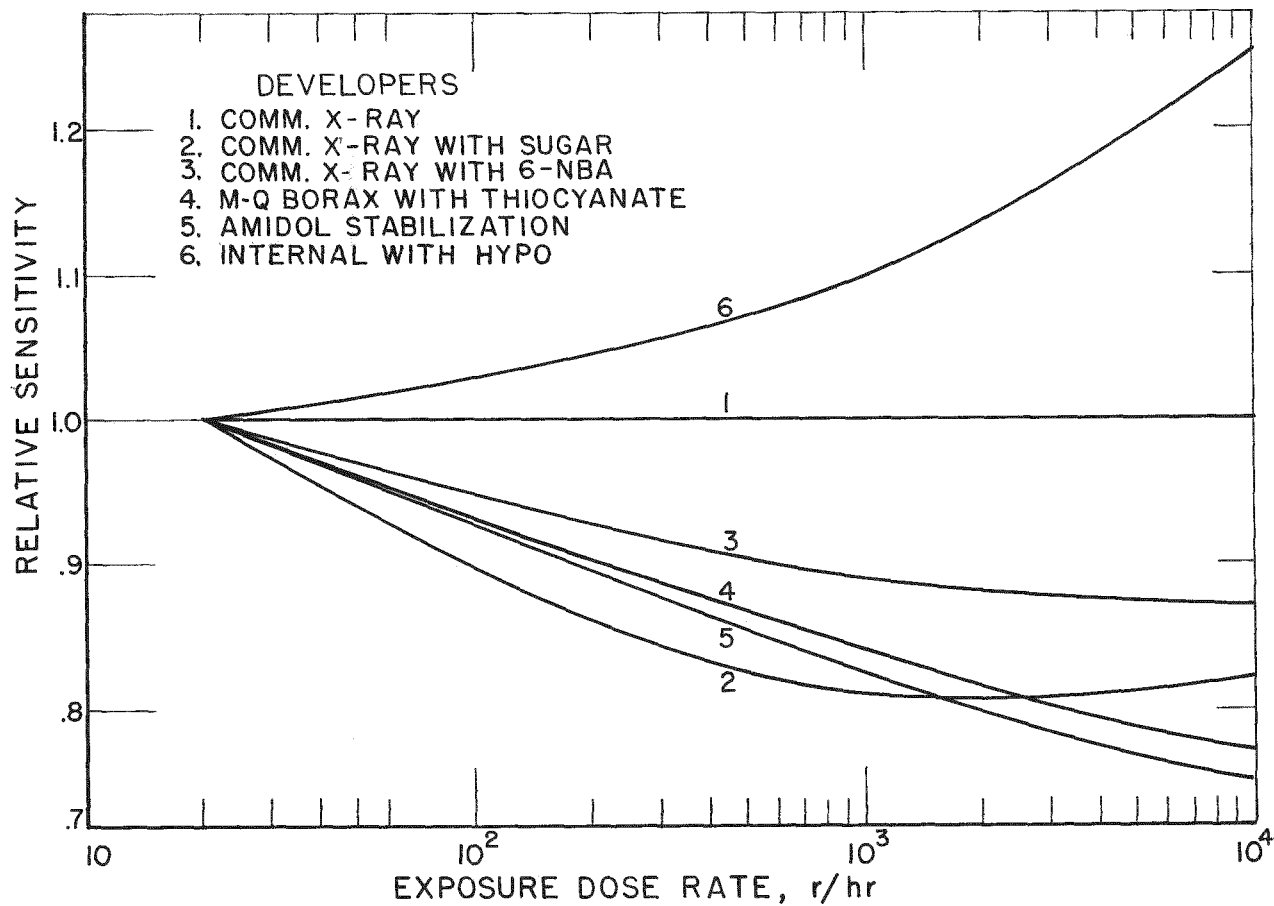


Figure 7. Rate dependence curves of DuPont Industrial X-ray film, type 510.