Evaluation of Dosimetry Film by Neutron Activation and X-Ray Fluorescence

Dick DUFFEY,
*University of Maryland*

W. L. BECK, G. I. GLEASON,
*Oak Ridge Associated Universities**

P. F. WIGGINS
*U. S. Naval Academy***

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Photographic film was exposed to γ-rays from 226Ra and 60Co to levels of 5 mR to 5,000 R. Measurements of the Ag in the developed films by both (a) activation with neutrons from 252Cf and counting 110Ag (half life 24.4 sec) and (b) X-ray fluorescence correlated with the known exposures. In the lower ranges conventional optical density measurements were made, and the correlation with the activation and X-ray methods was good.

KEYWORDS: dosimetry film, silver 110, neutrons, silver, activation, radiation doses, X-ray fluorescence, accuracy, sensitivity

I. INTRODUCTION

Photographic film is extensively used for personnel dosimetry for ionizing radiation. The more significant reasons are that the film provides information on the direction of exposure, the type of radiation, and its energy, and the film is inexpensive and provides a record for reevaluation if necessary. The exposure information is left as elemental silver (Ag) in the developed film. The determination of the amount of Ag is usually done by measuring the optical transmission of the film. Unfortunately, film dosimetry accuracy suffers at levels below 30 mR of γ-radiation, which is a range of much interest in health physics and environmental studies. Also, at several hundred roentgens, a region of interest in serious accidents, the film is too dark to read with a densitometer.

Other means of measuring the exposure to the film have been suggested, and some years ago, activation by neutron bombardment in a reactor followed by β- and γ-counting of the induced Ag activity was tried(1)~(3). Recently, we have used two other methods, namely: (a) activation of the Ag with neutrons from 252Cf and counting the 110Ag (half life 24.4 sec) and (b) measurement of the Ag by X-ray fluorescence.

II. BACKGROUND

1. Current Methods

Traditionally, dosimetry film is evaluated by an optical reader which uses a photo-

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* College Park, Md. 20742, USA.
** Oak Ridge, Tenn. 37830, USA.
*** Annapolis, Md. 21402, USA.

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multiplier tube to measure the attenuation of a light beam passing through the film. The
response is in units of optical density, defined as the negative logarithm to the base 10
of the fraction of light transmitted. (Fast neutrons leave film tracks that can be counted;
also incorporation in the film of materials like B or Li can exchange thermal neutrons for α-
particles that can darken the film). For exposures below about 30 mR, the film is too
transparent for any significant attenuation of the light. Also at optical densities of 5 or
above, the film is so dark that little information is obtained; with ordinary Kodak Type
2 film, this corresponds to an exposure of perhaps 5 R. To avoid this insensitivity at
high exposures, the film has a thin and a thick layer of emulsion on the opposite sides of
the support base. To measure high exposures, the thick, or sensitive layer of emulsion
is removed by a chemical and mechanical procedure; by such stripping, measurement
through the thin layer of emulsion can extend the range to about 1,000 R(2)(4). However,
the stripping is time consuming, and attaining reproducibility and therefore accuracy, is
not easy.

For comparison, we mention here that thermoluminescent dosimeters (TLD) now com-
pete with film. With these, a small piece of CaF₂ or LiF, about 1/8” square, absorbs
energy from the exposing radiation. Later, light is released on heating, and the amount,
as measured by a photomultiplier tube, can be related to the roentgens of exposure(5).
The CaF₂ or LiF pieces can be reused. The procedure is relatively rapid, and the small
crystals permit more convenience in location, e.g. on fingers. The exposure range for
TLD is long, from a few milliroentgens to perhaps a 1,000 R, but the heating destroys
the information. Use of ⁶Li can provide thermal neutron measurements.

2. Activation Method

Some years ago at the Argonne National Laboratory, dosimetry film evaluation was
attempted by neutron activation of the Ag followed by β-counting of the ¹⁰⁸Ag (2.4 min)(1)
and by γ-counting of the isomer ¹¹⁰Ag (252 day)(2). Some radiation degradation of the film
by reactor environment, perhaps also heat, was noted. Although quantitative results were
obtained, the characteristics of the decay of the activation products and the radiation
damage were not favorable to this application. The reactions responsible for activation
are shown in Fig. 1(6).

Examination of the nuclear characteristics of Ag and its activation products reveal
that the ¹¹⁰Ag (24.4 sec) has advantages over the other isotopes. Use of this short lived
isotope requires less irradiation time which limits film radiation degradation and residual
radioactivity. The short lived isotopes in activated Ag are ¹¹⁰Ag (24.4 sec) and ¹⁰⁸Ag (2.42
min). The ¹¹⁰Ag yields about 100 times as many photons as does ¹⁰⁸Ag from a 1 min
irradiation for a 2 min count.

3. X-ray Fluorescence Method

X-ray fluorescence analysis has been applied to a variety of problems(7)(8). Excitation
of characteristic X-rays in the sample to be analyzed is accomplished by irradiating the sample with low energy photons from radioactive sources or from X-ray tubes. The energy of the excitation photons must be somewhat above the energy of the characteristic X-ray to be measured, because at least that amount of energy is necessary for excitation, and the yield is a maximum when the excitation energy is just above the characteristic X-ray energy. Photons from particle accelerators have also been used for excitation. But usually the accelerators have been designed for other purposes, and X-ray work is an ancillary application. The characteristic X-rays are conveniently measured with a Si(Li) detector.

Recently, this technique has been used to measure Ag in film as a gaging device in its manufacture. Our second method is to use this analytical technique for dosimetry.

III. PROCEDURE

Our techniques of activation and X-ray fluorescence were applied to developed Kodak Type 2 personal monitoring film by the procedure outlined in Fig. 2.

The films were exposed at 5, 10, 50, 100, 1,000 and 5,000 mR with a 5 mg Ra source National Bureau of Standards (NBS), certified suspended in air. The films were positioned radially from the source on a cardboard box to reduce scattering; the total exposures were calculated using the specific γ-ray constant, the distance, and the exposure time. Dose rates were either 500 or 250 mR/hr.

A 60Co medical irradiator (450 Ci) was used for levels of 5, 10, 50, 100, 250, 500, 750 and 1,000 R. NBS calibrated ionization chambers were used to measure exposures to the film. Exposure rates with the 60Co were either 50 or 9.3 R/min. Twenty unexposed films were used as controls. Five films were exposed at each level for both the 226Ra and 60Co series. Developing of this film was by typical hospital procedures.

1. Neutrons Activation and Counting

Activation was accomplished with neutrons neutrons from 15 mg of 252Cf. The source was in a 2 ft.² by 1 ft. high block of graphite at the bottom of a 5 ft. diameter by 4 ft. high tank of water. The film was placed in ½" diameter by 1" long polyethylene capsule which was positioned near the source by a pneumatic transfer system for 1 min irradiations.
Two 3" diameter \times 3" long sodium iodide (NaI(T1)) crystals in the center of an Fe shield, 30" diameter \times 36" long, were used to count the films. Counter background was low, being only 100 counts for 80 sec. Previous measurements showed the system to have a counting efficiency of 10\% for 550 keV photons. A counting time of 80 sec for the 658 keV line of Ag was chosen. A 1024 channel analyzer with printer and X-Y plotter was used to acquire the counting data.

2. X-ray Fluorescence Method

The same films were also analyzed by an ORTEC model 7016-14/5 X-ray fluorescence unit. The Si(Li) detector had an active diameter of 6 mm with a ½ mil Be window. The resolution was 175 eV FWHM at 14.4 keV. The detector was inside a vacuum chamber about 6" radius \times 6" high; however, the vacuum system was not necessary with the 22 keV photons (the Ka line) of Ag measured. A thin Mylar film was above the window of the detector for it’s protection, and the films were laid, by tweezers, on the Mylar over the Si(Li) detector’s window.

Excitation of the film was by a 25 mCi annular shaped \(^{241}\text{Am}\) source which emits 60 keV photons; also, a \(^{153}\text{Gd}\) 25 mCi source (97 and 103 keV) was used. Four minute counts were sufficient for the low exposure film and 2 min for the higher exposure films.

3. Optical Measurement

An optical densitometer (Macbeth) TD-100A was used on the films in the conventional manner.

IV. RESULTS

All three measurement methods, namely neutron activation, X-ray fluorescence and optical transmission, were easily accomplished without special preparation, except for the activation; here films had to be cut slightly in size to fit the irradiation capsule. This same piece of film was also used for the X-ray measurement.

The activation counts on \(^{110}\text{Ag}\) for 80 sec gave about 5,500 (658 keV) for the controls and 60,000 for the 1,000 R films. The 5 mR films gave about 300 counts above the control value. Standard deviations of the counts (5 films) at each level were from 0.5 to 3\%. The activation counts vs. optical density was linear in the lower range (Fig. 3). A plot of the counts vs. log of exposure gave a sigmoid curve over the long range from 5 mR to 1,000 R (Fig. 4).

The X-ray fluorescence results were much like the activation. A 4 min count of the controls gave 1,400 counts (22 keV) and 17,000 counts for the 1,000 R films. The 5 mR films gave about 100 counts above the control value. A typical X-ray spectrum is Fig. 5. The counts vs. optical density were linear in the lower range, as were the activation results (Fig. 3). Similarly counts for both activation and fluorescence vs. exposure over the long range gave a sigmoid curve, as with the activation results (Fig. 6). The standard deviation of the fluorescent counts at each level was about 3\% which was somewhat more than for the activation results.

The optical density could not be read above 5 R because the film was too dark.
At somewhat above 500 R both the activation and the X-ray counts tended to saturate; perhaps all the Ag was being used. The stripping method(1) could, presumably, raise the useful limits of both the activation and X-ray methods as it does with the usual optical transmission method.

The somewhat poorer statistics with the X-ray fluorescence method may have stemmed from the lower penetration of the 22 keV X-rays compared to the 658 keV photon from $^{110}$Ag activation. Slight changes in geometry, e.g. the location of the film over the Si(Li) window and self-shielding of the film, may also have contributed to the variation.

V. CONCLUSIONS

The results of this study show some advantages for both the activation and X-ray techniques compared to current practice. The activation method with the $^{110}$Ag allows a
very short analysis time. Residual radioactivity and radiation degradation of the film are minimal. In addition, sealed $^{252}$Cf sources are low in cost, compact, portable, and have more simple licensing and operational requirements when compared with other sources of neutrons, e.g. reactors.

The X-ray fluorescence analytical units are widely available, and their licensing, registration and operation are also relatively simple. No residual activity remains.

Both the activation and the X-ray method provide results in a ready numerical form for computer processing and reporting as personnel exposures in roentgens. Both methods leave the film essentially unchanged as a permanent record. Both allow easy extension of the range of film dosimetry to lower and to higher levels of exposure than does the optical reading.

In summary, the $^{110}$Ag and X-ray fluorescence methods may be attractive to the nuclear industry for environmental monitoring and personnel monitoring. In addition the techniques may be useful in obtaining information from film used for other industrial and scientific purposes.

--- REFERENCES ---

(2) Berlman, I.B.: Determination of film exposure by activation of $^{109}$Ag, Nucleonics, 11[2], 70 (1953).