LA--10290-MS

DE85 007376

Inorganic Fluorescent Screen Properties Important for MeV Radiation Imagery

G. J. Berzins
A. H. Lumpkin*
H. L. Smith**

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

*Present address: DERS/SES CEN Cadarache, 13115 St. Paul-lez-Durance, FRANCE.
**EG&G/EM, Inc., Las Vegas, Nevada.

MASTER

Los Alamos

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

CONTENTS

	Page
ABSTRACT	1
I. BACKGROUND	1
II. IMPORTANT CHARACTERISTICS FOR OUR APPLICATIONS	2
A. Sensitivity	2
1. Different Screen Types	2
2. Screen Thickness Effects	4
3. Screen Binder Effects	4
4. Optical Self Absorption	5
B. Optical Spectra	6
C. Fluorescence Decay	6
D. Resolution	8
III. EXAMPLE OF AN APPLICATION	11
IV. MONTE CARLO CALCULATIONS	11
V. SUMMARY	15
ACKNOWLEDGMENT	17
REFERENCES	17

INORGANIC FLUORESCENT SCREEN PROPERTIES IMPORTANT FOR MeV RADIATION IMAGERY^{*}

by

G. J. Perzins
A. H. Lumpkin
H. L. Smith

ABSTRACT

This report summarizes observed properties of inorganic fluorescent screens that are important in imaging experiments that rely on MeV photons and neutrons. The summary is based on our earlier, more comprehensive report. Recent, preliminary results of Monte Carlo calculations that complement that work are also discussed.

I. BACKGROUND

We became interested in the properties of inorganic fluorescent screens several years ago while we were involved in imaging experiments using MeV neutrons and gamma rays. An example of these experiments is the observation of a prototype fuel element under stress in a test reactor. We used pinhole techniques to image the test element with its own fission neutrons and gamma rays onto a scintillator, whose output we then photographed with intensified TV cameras and with film.

In the reactor experiments and in other field exercises, we typically encountered three concerns: (1) The plastic scintillators that had been our workhorse were intolerant of the physical-handling abuses arising frequently, though inadvertently, in a field environment. (2) In many instances we wanted

^{*}Summary of presentation at the Los Alamos Conference on Image Processing and Tomographic Reconstruction, November 26-28, 1984.

to improve sensitivity, both in terms of counting statistics and in light-conversion efficiency. (3) In some cases where high-energy photons carried the primary information, the spatial resolution with plastic converters was inadequate. Of course, in some experiments we simply had to use plastics because of their superior time resolution.

Although inorganic screens have served the medical-imaging and the nondestructive-testing communities well for many years, there appeared to be very little information available regarding MeV radiation. As we became more involved with these screens, we learned some interesting features and also found that extrapolation of low-energy response to MeV energies was not always reliable.

We therefore decided to systematically characterize some of the properties of inorganics that were of most importance to our applications. Eventually we summarized the principal results in the September/October 1983 issue of Optical Engineering.¹ Much of this report is a review of those results and is a summary of our presentation at the Conference held in Los Alamos November 26-28, 1984, with the scientists of the French Commissariat à l'Energie Atomique.

Since the writing of that report, all three of us have become involved in other projects that have redirected our attention, if not our interest. In the course of other work, we were able to perform a limited set of Monte Carlo calculations. Some of these results, presented in the last section, are rather gratifying.

II. IMPORTANT CHARACTERISTICS FOR OUR APPLICATIONS

A. Sensitivity

1. Different Screen Types. We selected a set of readily available screens that spanned a fairly large range of chemical compositions. The response of these to 1.2-MeV gamma rays and to 14-MeV neutrons, as measured with an S-20 photosensitive element, is given in Table I. The plastic, 10-mm-thick NE102, is used for normalization.

In imaging experiments it is common to use a TV camera as a signal-sensing medium. Results for these screens viewed by conventional and intensified vidicons are given in Table II. Unfortunately a silicon-based camera was not available when these tests were done, so we have not directly measured the

TABLE I

SENSITIVITY OF SEVERAL COMMERCIALY AVAILABLE FLUORESCENT
SCREENS RELATIVE TO THAT OF 10-mm-THICK PLASTIC

Trade Name	Composition	relative Sensitivity	
		14-MeV Neutrons	1.2-MeV Gamma Rays
NE102	Plastic	1.0 ^a	1.0
Rarex 100/18	Gadolinium oxysulfide	0.96	1.42
Rarex 250/18	Gadolinium oxysulfide	1.44	2.60
E-2	Zinc cadmium sulfide	1.17	0.67
Quanta II	Barium fluorochloride	0.48	0.71
Quanta III	Lanthanum oxybromide	0.55	0.68
Lightning Plus	Calcium tungstate	0.34	0.64
Hi-Plus	Calcium tungstate	0.27	0.48

^aSubsequent measurements performed by others using a different facility appear to disagree with our data for neutrons. Hence there may be an unresolved question regarding backgrounds, normalizations, or other experimental details.

TABLE II

RELATIVE RESPONSE OF FLUORESCENT SCREENS TO MeV
GAMMA RAYS^a WHEN VIEWED BY DIFFERENT PHOTSENSORS

Trade Name	Photomultiplier (S-20 photocathode)	Vidicon Sb Target	Intensified Vidicon (S-25 photocathode)
NE102	1.0 ^b	1.0 ^b	1.0 ^b
Rarex 100/18	1.5	5.1	4.2
Rarex 250/18	2.7	8.6	7.8
E-2	0.7	6.9	3.0
Quanta II	0.7	c	1.2
Quanta III	0.7	c	0.9
Lightning Plus	0.7	c	0.8
Hi-Plus	0.5	c	1.0

^aData in column 1 are obtained with a ⁶⁰Co source; data in columns 2 and 3 are obtained with a 2.5-MeV bremsstrahlung source.

^bNormalization.

^cSignal too small for measurement.

expected signal increase from the improved spectral match between some inorganics and such a sensor.

2. Screen Thickness Effects It appeared from early tests that Rarex* was one of the more promising screens. We were fortunate to obtain custom samples in a variety of thicknesses (tt) and binder concentrations (bb). It was therefore relatively easy to explore a variety of effects in Rarex. Analogous results should hold for the other material types, but the individual differences could be interesting.

The sensitivity as a function of Rarex thickness is compared in Fig. 1 for 1.2-MeV gamma rays and 14-MeV neutrons. Note that the slopes of both curves decrease with increasing thickness, an apparent consequence of optical self-absorption in the screen.

3. Screen Binder Effects All of the inorganic screens are held together with a hydrogen-rich "glue," and are deposited on a hydrocarbon backing. We reasoned that an increase of hydrogen should enhance the total neutron interaction probability and thus increase the sensitivity. We did indeed observe an ~10% sensitivity increase to 14-MeV neutrons as the binder concentration was increased from the usual 12% to about 20%. These data are shown in Fig. 2.

Several comments are in order about the data in Fig. 2. (1) In fission-neutron experiments, the net sensitivity gain was 20-30% over a comparable binder-concentration range. Because of the energy-continuum nature of the radiation and because of greater experimental difficulties, these data are less well documented and less complete. The improvement, attributable to the $1/\sqrt{E}$ nature of the neutron cross section, was rather noticeable in the overall signal (neutrons + gammas) to background (mainly gammas) ratio. (2) The gamma-ray sensitivity decreased by ~10% over the same binder range, suggesting opacity effects of the binder. This decrease suggests a potential for improving signal-to-background ratios in applications involving radiation from a fissioning source. (3) We do not know whether the abrupt change at 24% is real, results from a screen sample anomaly, or is attributable to experimental error.

*The use of trade names is preferred for reasons of accuracy and does not necessarily imply endorsement by the Laboratory.

Fig. 1. Relative sensitivity of Rarex as a function of thickness, normalized to the 200-mg/cm² sample. Neutron and gamma-ray energies were 14 MeV and 1.2 MeV, respectively.

Fig. 2. Effect of hydrocarbon binder on Rarex response to 14-MeV neutrons and to 1.2-MeV gamma rays.

4. Optical Self Absorption As a complement to the sensitivity measurements, we examined the signal from Rarex resulting from a low-energy bremsstrahlung source. Since the soft radiation (incident from the back side) is quickly absorbed, an increase in screen thickness contributes nil to energy absorption but adds to self-absorption of the emitted light. The results, Fig. 3, suggest an optical half thickness of ~150 mg/cm². We have not attempted to analytically unfold the curves in Fig. 3, nor to quantitatively reconcile the data in Fig. 3 with the "rolling off" in Fig. 1.

Fig. 3. Relative sensitivity of Rarex as a function of thickness to low-energy photons. The beam current of the x-ray generator was maintained constant for all voltages.

B. Optical Spectra

In view of the importance of the spectral response of the photosensitive surface, we recorded the output spectra from the various screens. Some of these data are shown in Fig. 4. Results from two Rarex samples are shown because there was a subtle but unmistakable shift in relative line strengths with thickness.

C. Fluorescence Decay

Almost all screens produced a small, short "spike" when exposed to an x-ray pulse lasting a few nanoseconds. The spike contained only a small fraction of the total light output. Figure 5 shows the light output as a

Fig. 4. Optical output spectra obtained from various types of screens. We have not identified the source of the increased signal above 800 nm, but believe it to be instrument related.

Fig. 5. Persistence of optical output after very short (<10 ns) input. The data have been arbitrarily displaced vertically to facilitate comparison.

function of time for most of these screens. The measurement was started 20 nanoseconds after exposure to ensure that the spike had decayed away.

D. Resolution

Of all parameters, resolution was by far the most troublesome to measure and its data the most difficult to interpret. Because we have misgivings about some of our resolution measurements, we are avoiding conventional terminology, opting, for example, for "contrast" (as determined in our experiments) instead of "MTF" (as determined under carefully controlled conditions). We feel, however, that it is important to make available an index whereby screens can be compared.

Figure 6 displays microdensitometer scans of bar target images on film that was in contact with the screen on which the bar target was placed. Figures 7 and 8 contain contrast information deduced from data such as that in Fig. 6. These data were obtained with an ~ 50 -keV bremsstrahlung source.

It was essentially impossible to accurately interpret high-energy (2.5-MeV bremsstrahlung) resolution data obtained in our crude measurement. Two limits were a high noise level in the microdensitometer traces and an overall fogging of the film as a result of the general, scattered x-ray, room background. Though these limits preclude a good resolution measurement, they serve to

Fig. 6. Densitometer scans of representative test pattern images recorded on film in contact with the given screen. Data were taken with low-energy (~ 50 keV) bremsstrahlung x rays.

Fig. 7. Resolution of several screens compared to plastic and to film. (Low-energy x rays.)

Fig. 8. Resolution of Rarex as a function of thickness. (low-energy x rays).

illustrate, perhaps emphasize, the difficult experimental conditions often encountered in MeV imaging.

Some general comments and conclusions include:

1. With soft (≤ 100 keV) photons, the plastic (even 10 mm thick) was appreciably better than any of the inorganic screens.
2. With hard (≥ 1.0 MeV) photons, all of the screens were appreciably better than the plastic (even 2 mm thin).
3. Except the zinc sulfides, all screens of comparable thickness tended to compare in resolution, especially to the higher photon energies. The zinc sulfides were poorer.
4. In the case of Rarex, a gain in efficiency in the thicker screens was offset by a loss in resolution.

III. EXAMPLE OF AN APPLICATION

Figure 9 shows two images of a reactor fuel pin in the center of a test reactor. The data were taken through a pinhole aperture onto Rarex 250/18 in contact with ASA 1200 speed film. The fuel is the vertical, white band near the center of each image. The fuel column is straight before the test (left), and has expanded to about 1.6 times its original diameter (right) as a result of the experiment-induced stress. Large central voiding is evident in the "after" image.

These images provided information immediately (1-2 hours) after the test, in contrast to conventional after-the-fact examination, which is normally delayed days and weeks because of hazardous radiation levels. The two sets of data were consistent. A valuable feature of the in situ image is the potential for identifying structural changes induced by mechanical handling of the test pin assembly as it is removed from the reactor core and transported to the examination facility.

IV. MONTE CARLO CALCULATIONS

About two years after drafting our initial report, we had an opportunity to perform some energy-deposition calculations in some of these phosphor screens. We were particularly interested in tracking the resolution as a function of scintillator type and as a function of thickness. In addition, we were curious about the comparison we had observed between plastic and the screens at high and at low-photon energies.

Fig. 9. Example of the application of Rarex 250/18 to image a reactor test element with fission neutrons and gamma rays.

The Monte Carlo code CYLTRAN² performs coupled electron-photon transport in cylindrical geometry. Besides imposing a requirement of cylindrical symmetry, the code contains several practical limitations, including number of materials, number of zones, and low-energy (few keV) cutoff--none of which was a problem in our case. The code tracks particles incident in a prescribed beam until they either stop or escape through the problem's boundaries. It tallies (among other parameters) energy deposited in the various zones and subzones.

We set up each screen as a 10-mm-diameter disk, of given thickness and composition, and subzoned it radially into 0.25-mm annuli. The photon beam was a thin pencil incident along the disk axis. The profile of energy deposited within central "sub-disk" and (as a result of scatter and secondary processes) into the successive annuli would then represent a point-spread function. The complete point-spread function is, of course, a convolution of this energy-deposition profile with the spreading of the light transmitted out of the screen.

The calculated-energy deposition in gadolinium oxysulfide as a function of thickness is shown in Figs. 10 and 11 for 0.1-MeV and 1.0-MeV photons, respectively. The sample thickness is given by the row of numbers across the top. The axis for successive samples is displaced four divisions to the right to facilitate comparison. Thus the tall bar in each case represents energy deposited in the central 0.5-mm-diameter disk. Each successive step represents energy deposited in the next annulus of 0.25 mm width. The numbers across the bottom of each figure represent the amplitude ratio between the central zone and the first annulus.

We note the general increase in the amplitude of the deposition profile with thickness, associated with an increase in efficiency. Simultaneously the height of the shoulder increases relative to that of the central bar, reflecting a degradation of resolution.

The calculation for the 1.0-MeV case suffers from a large statistical error, imposed by a small interaction probability coupled with practical limits on computer running time. Typical error bars are $\leq 1\%$ and $\leq 12\%$ in the central zone for the 0.1-MeV and 1.0-MeV cases, respectively. Corresponding errors in the first few annuli are $\leq 15\%$ and $\leq 35\%$. The greatest uncertainties are in the thinnest samples.

It is interesting to compare the total deposited energy at 1 MeV from CYLTRAN with the measured efficiency vs thickness for 1-2 MeV gamma rays (Fig. 1). The two data sets are compared in Fig. 12. We have attempted to normalize the two sets so that the fit to the calculated data is an asymptote

Fig. 10. Results of Monte Carlo (CYLTRAN) calculations of energy deposited by a pencil of 0.1-MeV photons incident on various thicknesses of Rarex.

Fig. 11. Results of Monte Carlo (CYLTRAN) calculations of energy deposited by a pencil of 1.0 MeV photons incident on various thicknesses of Rarex.

Fig. 12. Comparison between the deposited energy as calculated by CYLTRAN, and the measured signal. We attribute the difference, increasing with thickness, as due mainly to optical self-absorption in the screen.

to the measured curve. Under these conditions the divergence between the two curves is at least roughly consistent with the assumption that (1) the Bouguer-Lambert law [$I/I_0 = \exp(-\mu x)$] applies, and (2) $\sim 150 \text{ mg/cm}^2$ (from our low-energy x-ray measurements) represents the half-thickness for optical self-absorption.

Results of two other calculations of interest are shown in Figs. 13 and 14, where we compare the deposition profile in plastic at 0.1 MeV and 1.0 MeV for thicknesses of 1 mm and 10 mm. These profiles, when compared to those for Rarex in Figs. 10 and 11, support our earlier observation regarding spatial resolution superiority of plastic at low-photon energies, and its inferiority at $\geq 1 \text{ MeV}$. For example, note the amplitude ratios between the central zone and the first annulus for plastics with 0.1-MeV photons (354, 205) and with 1.0-MeV photons (1.79, 1.51) in comparison to those for $\text{Gd}_2\text{O}_2\text{S}$.

V. SUMMARY

In summary, we believe that inorganic phosphor screens may play an expanding role in the fields of high-energy x-ray imaging, nuclear-fission-source imaging, and nuclear-fusion-source imaging. Neutron and/or photon response for specific applications can be improved by adjusting the binder concentration and the phosphor thickness. In addition, the higher binder concentrations may provide sufficient phosphor-screen-mechanical

Fig. 13. Results of Monte Carlo (CYLTRAN) calculations of energy deposited by a pencil of 0.1-MeV photons incident on plastic scintillator.

Fig. 14. Results of Monte Carlo (CYLTRAN) calculations of energy deposited by a pencil of 1.0-MeV photons incident on plastic scintillator.

strength so that either high-Z or hydrogen-enriched backing might further improve the response. Calculational efforts (such as that with CYLTRAN) to model the interplay of Compton electron ranges, proton ranges, detection probabilities, energy conversion, and self-absorption should lead to a better understanding of these inorganics and to their optimization in applications.

ACKNOWLEDGMENT

We are grateful to T. A. Garcia for help with the Monte Carlo calculations.

REFERENCES

1. G. J. Berzins, A. H. Lumpkin, and H. L. Smith, "Characterization of fluorescent screens for imaging applications with MeV neutrons and photons," *Optical Engineering* 22, p. 633 (1983), and references cited therein.
2. J. A. Hableib, Sr. and W. H. Vandevender, "CYLTRAN: A Cylindrical-Geometry Multimaterial Electron/Photon Monte Carlo Transport Code," Sandia National Laboratories report SAND 74-0030.