

X-Ray Exposure Reduction Using Rare-Earth Oxysulfide Intensifying Screens¹

Robert A. Buchanan, Ph.D., Solon I. Finkelstein, M.D., and
Kenneth A. Wickersheim, Ph.D.

ABSTRACT—The characteristics of terbium-activated gadolinium oxysulfide and lanthanum oxysulfide are compared with those of calcium tungstate. The new rare-earth phosphors have higher x-ray absorption in the 38 to 70 keV region relative to calcium tungstate. They also exhibit a larger x-ray-to-visible light energy conversion efficiency. Rare-earth phosphors, however, emit primarily in the green spectral region and thus poorly match the spectral response of blue-sensitive x-ray film. Despite this disadvantage, gadolinium oxysulfide screens allow a significant x-ray exposure reduction relative to calcium tungstate screens providing equivalent resolution. An additional exposure reduction is anticipated when faster green-sensitive x-ray film becomes available.

INDEX TERMS: Screens • Diagnostic Radiology, apparatus and equipment

Radiology 105:185-190, October 1972

IN 1896, film intensifying screens made of calcium tungstate (CaWO_4) were used for the first time. Today, more than seven decades later, CaWO_4 remains the principal phosphor used in x-ray intensifying screens. To its credit, CaWO_4 has a relatively high x-ray absorption coefficient and is a physically hardy material. However, its x-ray-to-light conversion efficiency is poor, typically 3 to 5% (1, 2).

The physical properties of two green-emitting x-ray conversion phosphors, terbium-activated lanthanum oxysulfide ($\text{La}_2\text{O}_2\text{S:Tb}$) and terbium-activated gadolinium oxysulfide ($\text{Gd}_2\text{O}_2\text{S:Tb}$), have been described previously (3). The most noteworthy characteristics of these phosphors are their unusually high x-ray absorption coefficients, especially above the K -edge of the particular host metal ion, and their high x-ray-to-light conversion efficiencies of 13 and 18%, respectively.

It is the purpose of this communication to compare the properties of the new rare-earth phosphors with those of CaWO_4 and to evaluate these new phosphors as constituents in x-ray film intensifying screens. X-ray film is typically most responsive to blue light, being well matched to the emission from a CaWO_4 screen. It shows almost no response in the green. The two oxysulfide phosphors have very little emission in the blue, and thus the rare-earth phosphors have a significant disadvantage when used as intensifying screens with present x-ray film. Despite this fact, our experiments have shown that intensifying screens

made of $\text{Gd}_2\text{O}_2\text{S:Tb}$ allow x-ray exposure reduction of a factor of two even when used with conventional x-ray film at 70 kVp and above.

If a green-sensitive film, equivalent in all other respects to present blue-sensitive film, were to become widely available, the exposure reduction over CaWO_4 could possibly be increased by a factor of 20. The development of such an equivalent green-sensitive film is feasible,² but to the present time there has been no motivation for such a specialized development. In addition to the direct reduction in radiation exposure to the patient, such a screen-film combination would decrease the need for more powerful x-ray generators, would extend tube and equipment life and, if screen thicknesses were to be reduced in proportion to the gain in total sensitivity, would make possible much higher resolution than can be achieved with the CaWO_4 intensifying screens.

In the following sections, the relevant properties of the two rare-earth phosphors will be compared with those of CaWO_4 , and then detailed results of film exposure measurements will be reported. Finally, direct comparisons of radiographs obtained using the new $\text{Gd}_2\text{O}_2\text{S:Tb}$ screens with radiographs obtained using commercial intensifying screens will be presented.

COMPARISON OF PHYSICAL PROPERTIES OF PHOSPHORS

The rare-earth oxysulfide phosphors can be produced as crystalline powders of controlled

¹ From the Lockheed Palo Alto Research Laboratory, the Department of Radiology, Palo Alto Medical Clinic, Palo Alto, and the Spectrotherm Corporation, Sunnyvale, Calif. Presented in part at the Work-in-Progress session of the Fifty-seventh Scientific Assembly and Annual Meeting of the Radiological Society of North America, Chicago, Ill., Nov. 28-Dec. 3, 1971.

This work was supported by the USAEC, Division of Biology and Medicine.

² We have already had discussions with several manufacturers of x-ray film who state that such a green-sensitive x-ray film can be manufactured.

keV	X-Ray Absorption in a 100 μm Screen (%)			Intrinsic Efficiency (%)
	40	60	80	
$\text{Gd}_2\text{O}_2\text{S:Tb}$	37	51	28	18
$\text{La}_2\text{O}_2\text{S:Tb}$	73	33	17	13
CaWO_4	33	13	27	4

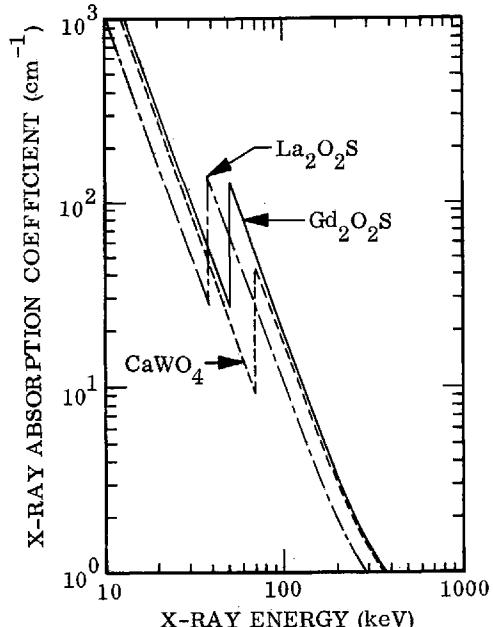


Fig. 1. Lower figure. X-ray attenuation coefficient of $\text{La}_2\text{O}_2\text{S}$, $\text{Gd}_2\text{O}_2\text{S}$, and CaWO_4 versus energy.

Upper figure. Absorption coefficient of screens 100 μm thick at three x-ray energies and the intrinsic x-ray-to-visible light conversion efficiencies.

particle size by a variety of techniques (4). Terbium-activated gadolinium oxysulfide and $\text{La}_2\text{O}_2\text{S:Tb}$ are closely related to $\text{Y}_2\text{O}_2\text{S:Eu}$, an important red-emitting color television phosphor. Such phosphors are now produced commercially on a large scale. The phosphors, once produced, are chemically stable and physically hardy. Thus screen life can be expected to be the same or better than that of presently used screens.

Figure 1 shows the x-ray absorption coefficients versus x-ray energy from 20 to 400 keV for $\text{La}_2\text{O}_2\text{S}$, $\text{Gd}_2\text{O}_2\text{S}$, and CaWO_4 . From 50 to 70 keV, $\text{Gd}_2\text{O}_2\text{S}$ has an advantage of 4 to 5 \times over CaWO_4 . Lanthanum oxysulfide has a somewhat lower x-ray absorption than CaWO_4 above 70 keV and below 38 keV but has a factor of two advantage over CaWO_4 between 38 and 70 keV. The distribution of x-ray energy which reaches the intensifying screen after passing through the patient depends on the kVp settings used, but in most practical situations a significant fraction of the energy available will fall in the 38 to 70 keV range. In

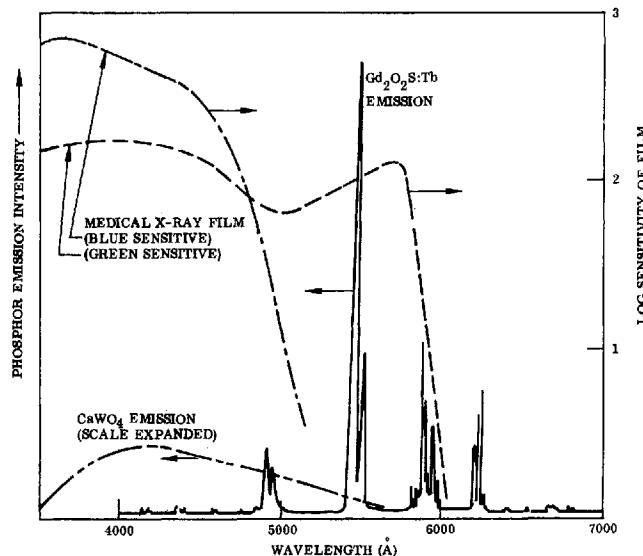


Fig. 2. Emission spectra of x-ray phosphors and x-ray film spectral response curves. The vertical scale has been expanded for CaWO_4 emission spectrum to make it visible on the same plot. It can be seen that the blue-sensitive film has no measured response at 5440 \AA whereas the green-sensitive film responds almost equally well to the blue CaWO_4 emission and the principal emission peak of $\text{Gd}_2\text{O}_2\text{S:Tb}$. Note how well the spectral sensitivity of the blue-sensitive x-ray film matches the emission spectrum of CaWO_4 .

many situations much of the energy will be in the narrower 50 to 70 keV range where $\text{Gd}_2\text{O}_2\text{S}$ has the advantage. Thus intensifying screens made of these two rare-earth phosphors will typically absorb and usefully convert to visible light more incident x rays than will CaWO_4 screens of the same thickness.

The x-ray-to-light conversion efficiencies of the rare-earth phosphors are also significantly greater than that of CaWO_4 . The numbers previously reported (3) for $\text{La}_2\text{O}_2\text{S:Tb}$ and $\text{Gd}_2\text{O}_2\text{S:Tb}$, using electron excitation techniques, are 13% and 18%, respectively, whereas the CaWO_4 efficiency falls in the range from 3 to 5% (1-2). The efficiency as defined here is watts of optical power output per watt of x-ray (or electron) power absorbed, expressed as a per cent. The maximum conversion efficiencies are obtained for materials containing approximately 0.3 atom-per cent of terbium replacing lanthanum or gadolinium.

The spectral emission of the rare-earth phosphors is produced by the terbium ion. The terbium emission has a very strong peak at 5440 \AA in the green with less intense emission peaks in the blue, blue-green, yellow, and red. The blue peaks in $\text{Gd}_2\text{O}_2\text{S:Tb}$ are stronger than in $\text{La}_2\text{O}_2\text{S:Tb}$, and can be enhanced somewhat further by lowering the terbium concentration below what would otherwise be the optimum value. The degree of exposure of blue-sensitive x-ray film by the oxysulfide screens depends strongly on the proportion of this relatively weak blue emission in the total output spectrum.

In Figure 2, the emission spectra of $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ and CaWO_4 are plotted *versus* the spectral response of both blue-sensitive x-ray film and a presently available, though less sensitive, form of green-sensitive medical x-ray film which is generally used for photofluorographic applications.

If one makes a comparison of the rare-earth phosphors and CaWO_4 using x-ray film having the spectral characteristics shown in Figure 2, one obtains two very important pieces of information. First, a comparison of the two phosphors in screen form made with blue-sensitive film provides quantitative data on any advantage to be gained by replacing CaWO_4 screens with the rare-earth oxy-sulfide screens and continuing to use the present blue-sensitive x-ray film. Second, a comparison of the same screens made using the green-sensitive x-ray film provides a quantitative measure of the advantage of the rare-earth phosphors relative to CaWO_4 which would be possible if a film could be manufactured with the spectral sensitivity of the green-sensitive film, but having a speed equivalent to that of the blue-sensitive film. These comparisons will be described in the following section.

COMPARISON OF X-RAY SCREEN PERFORMANCE

Of the two rare-earth phosphors under discussion, $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ has the higher x-ray absorption and the higher x-ray-to-light conversion efficiency. In addition, its emission makes a somewhat better match to the response characteristics of presently used x-ray film than does the emission of $\text{La}_2\text{O}_2\text{S}:\text{Tb}$. For these reasons we have chosen to make the majority of our direct comparisons between screens composed of $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ and CaWO_4 . Test screens were made by dispersing powders of $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ and x-ray phosphor grade CaWO_4 of essentially identical particle sizes in a binder of methyl methacrylate and drawing down the mixture with a doctor blade on a substrate of translucent Mylar. The phosphor coatings of the screens were five mils thick, quite comparable to commercial CaWO_4 Par-speed screens. These screens were used to conduct a direct comparison of film exposure.

To compare CaWO_4 and $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ screens, half of the x-ray film was covered with a CaWO_4 screen and the other half covered with a $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ screen. Each exposure was made in such a way that a portion of a narrow x-ray beam fell on the CaWO_4 screen while the remaining portion of the beam fell on the $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ screen. It was thus assured that the exposure for the two screens was identical. A representative example of an exposed green-sensitive film resulting from such an exposure series is shown in Figure 3.

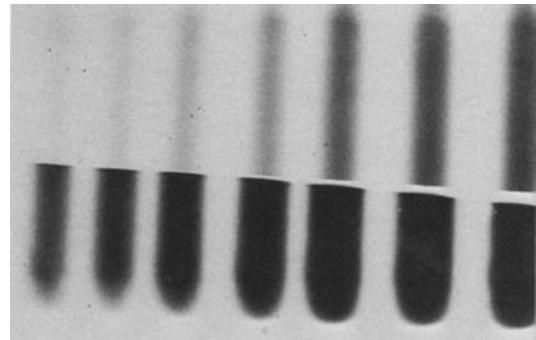


Fig. 3. Typical film exposure. The upper section of this film was exposed from the CaWO_4 screen, and the lower section was exposed from the $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ screen (Kodak medical x-ray, green-sensitive, photofluorographic film).

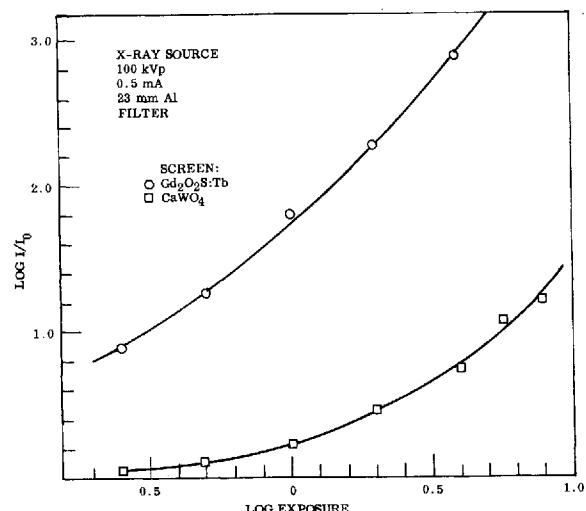


Fig. 4. Measured film density *versus* exposures; Kodak medical x-ray, green-sensitive, photofluorographic film (single coated).

The exposed film was next measured with a densitometer. The densitometer readings were plotted *versus* the log of the exposure. Such a plot is shown in Figure 4. By drawing a horizontal line across the figure at an optical density of 1, the relative exposure necessary to produce equivalent film darkening can be obtained. Relative intensification factor (RIF) will be defined as the ratio of the time to expose the film to unit density using the CaWO_4 screen to the time necessary to expose film to unit density using the $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ screen. From Figure 4 we deduce that the RIF is 20 at 100 kVp. That is to say, it takes approximately 20 times the amount of x-ray exposure using the CaWO_4 screen to obtain equivalent film darkening to that obtained using the $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$ screen. The RIF value at 80 kVp is 12.5. These values are indicated in TABLE I. The RIF values indicated in TABLE I are for an arbitrary film density of 1. These values would be smaller (less advantage for the rare-earth phosphor) at densities

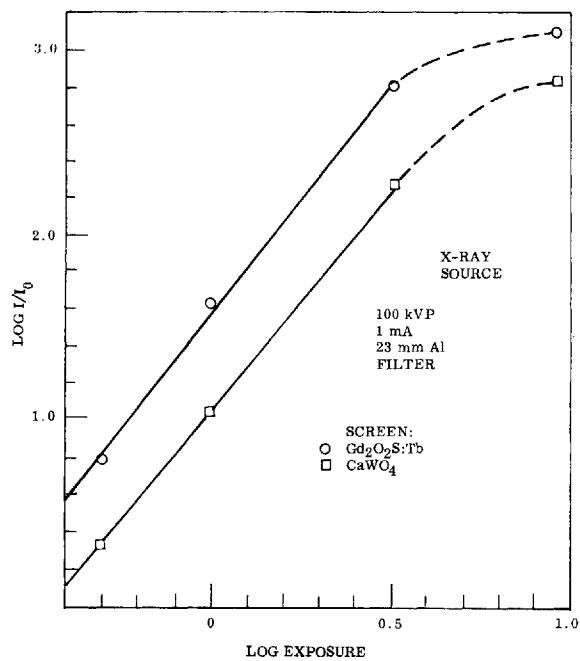


Fig. 5. Measured film density *versus* exposure; Kodak medical x-ray, blue-sensitive film (single coated).

TABLE I: EXPERIMENTAL DETERMINATION OF RELATIVE INTENSIFICATION FACTORS FOR BLUE- AND GREEN-SENSITIVE FILM AS A FUNCTION OF kVp SETTINGS

Medical X-Ray Film Type	kVp	RIF = T(CaWO ₄)/ T(Gd ₂ O ₃ :Tb)
Blue-sensitive	80	2
Green-sensitive	80	12.5
Green-sensitive	100	20

less than 1 and would be larger (more advantage for the rare-earth phosphor) at densities greater than 1. The increase of RIF values with increasing kVp is the result of the stopping power of Gd₂O₃:Tb relative to CaWO₄ as more and more of the x-ray emission intensity falls within the 50 to 70 keV range.

It can be observed from these data that because of the increased x-ray stopping power and the higher intrinsic luminescent efficiency of Gd₂O₃:Tb relative to CaWO₄, significant x-ray exposure reductions can be achieved using Gd₂O₃:Tb screens. The exact amount of exposure reduction that can be achieved in practice by using green-sensitive film will be determined by either the ultimate sensitivity limit (speed) of the green-sensitive film which film manufacturers will be able to achieve or the limit imposed by quantum mottle. At present we do not know which limit will be reached first. Use of the presently available green-sensitive photofluorographic film results in RIF of about 2.5. Exposure curves of Gd₂O₃:Tb and CaWO₄ screens using this higher speed blue-sensitive medical x-ray film are shown in Figure 5. From these data it can be seen that there is suf-

ficient overlap between the sensitivity of the blue film and the emission spectrum of the Gd₂O₃:Tb to yield some advantage over CaWO₄. The RIF value for blue film is 2. This means that one can reduce exposure by one-half simply by changing from CaWO₄ to Gd₂O₃:Tb screens and continuing to use conventional blue-sensitive x-ray film.

COMPARISON OF RADIOPHGRAPHS

Radiographs of a skull phantom have been obtained with a commercially prepared (Du Pont) Par-speed CaWO₄ screen and with a Gd₂O₃:Tb screen of comparable thickness and resolution. Figure 6 shows such a comparison taken with a 3-phase generator at 80 kVp using Cronex 4 film and 90-second processing. As can be seen in the figure, 40 mAs gives a satisfactorily exposed film using the Par-speed screen. This same exposure produces an overexposed film when the Gd₂O₃:Tb screens are used. Reducing the Gd₂O₃:Tb screen exposure to 20 mAs still gives a slightly darker radiograph than the Par-speed screens provide at 40 mAs.

Figure 7 shows a similar series taken at 60 kVp. At this lower voltage fewer x-ray photons have energies in the 50 to 70 keV range, and the advantage of the Gd₂O₃:Tb screen is diminished accordingly. Nonetheless, it can be seen that the Gd₂O₃:Tb screen still provides a reduction in exposure of at least 1/3 relative to the Par-speed screen.

These radiographs were made with experimental screens of Gd₂O₃:Tb of the same front-and-back thickness which did not have reflecting layers or protective coatings. The binder material has not yet been optimized to give the maximum light output from the screen, and no dyes were added to improve definition. Therefore, it may be anticipated that commercially prepared screens of Gd₂O₃:Tb could be improved considerably by incorporating many of the improvements that have been made to CaWO₄.

Because these rare-earth oxysulfides have different x-ray absorption spectra than CaWO₄, slight differences in contrast might be anticipated. Their relatively greater sensitivity to the higher energy x-ray photons passing through a dense media would tend to diminish contrast. They would also be expected to have a relatively lesser sensitivity to scattered low-energy x-ray photons and thus show improved contrast. Thus far, in trial radiographs, no significant differences in contrast have been observed.

DISCUSSION

It has been shown that the rare-earth oxysul-

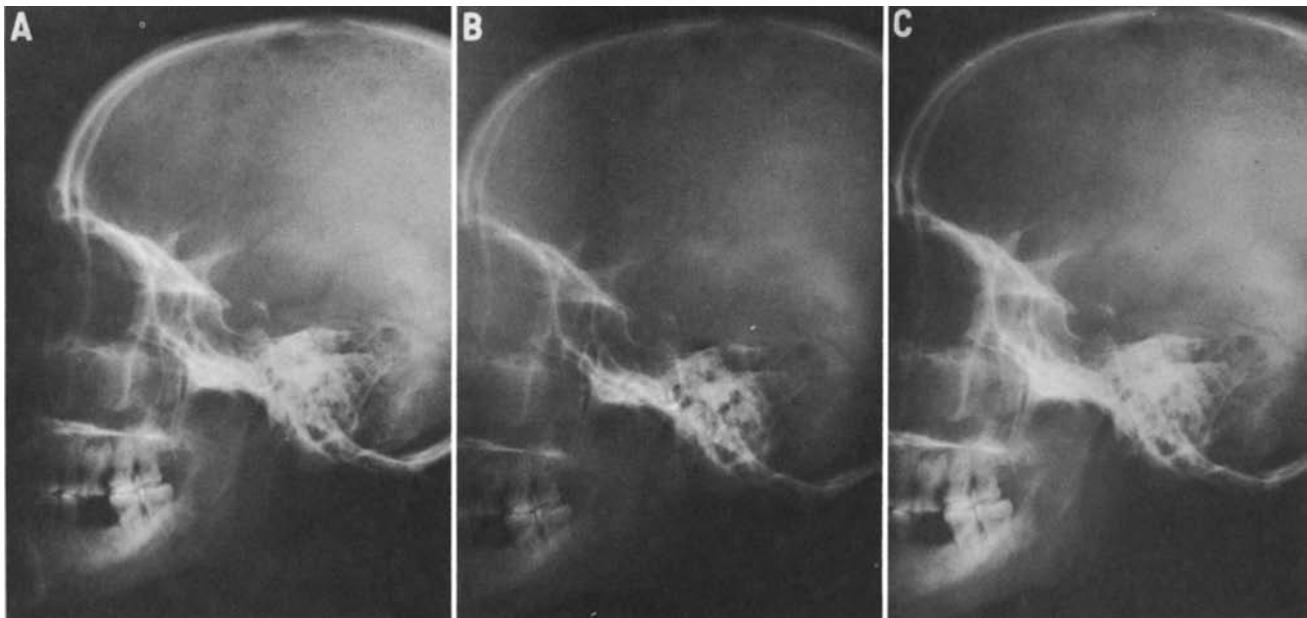


Fig. 6. Comparison of radiographs of a skull phantom taken with a 3-phase generator at 80 kVp using Cronex 4 film and 90-second processing.
 A. Par-speed screen, 40 mAs.
 B. $\text{Gd}_2\text{O}_2\text{S: Tb}$ screen, 40 mAs.
 C. $\text{Gd}_2\text{O}_2\text{S: Tb}$ screen, 20 mAs.

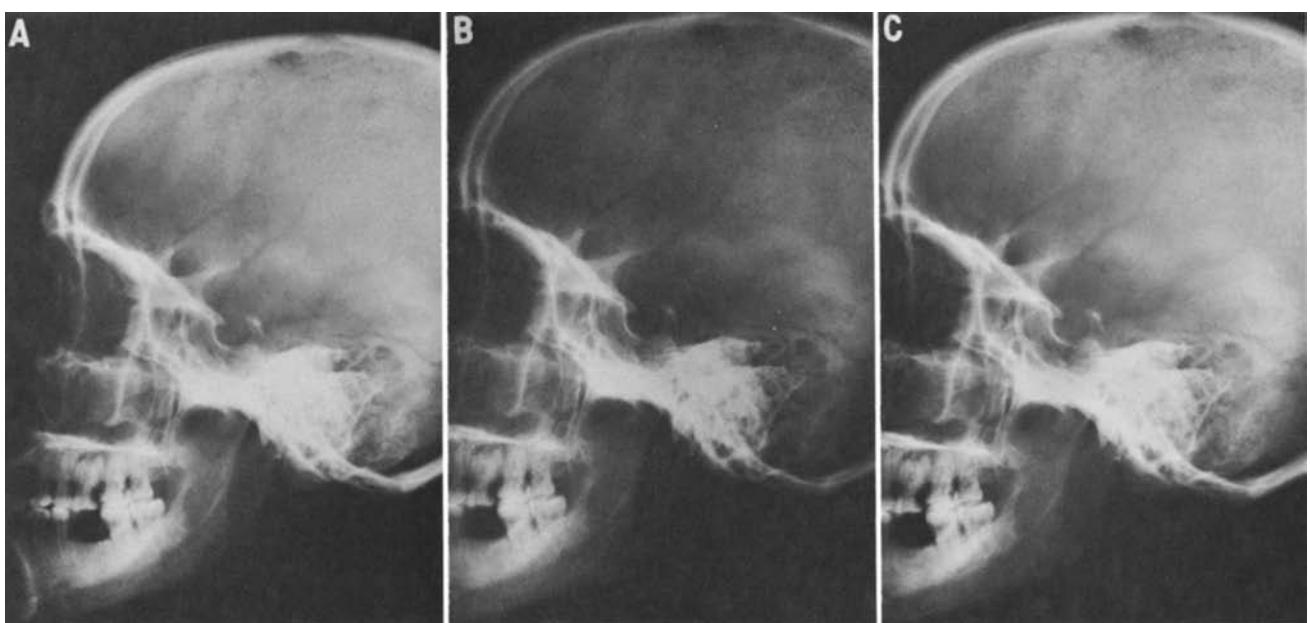


Fig. 7. Comparison of radiographs of a skull phantom taken with a 3-phase generator at 60 kVp using Cronex 4 film and 90-second processing.
 A. Par-speed screen, 200 mAs.
 B. $\text{Gd}_2\text{O}_2\text{S: Tb}$ screen, 200 mAs.
 C. $\text{Gd}_2\text{O}_2\text{S: Tb}$ screen, 140 mAs.

fides have great promise for making improved x-ray intensifying screens because of their increased x-ray stopping power and higher intrinsic luminescent efficiency. Exposure reductions of two or greater are achievable at 70 kVp and above using screens of $\text{Gd}_2\text{O}_2\text{S: Tb}$ and standard blue-sensitive x-ray film. Significantly larger exposure reductions are anticipated with improved manufacturing techniques and the development of a faster green-

sensitive film. The ultimate exposure reduction may depend upon the limit set by quantum mottle. This refers to the fact that as fewer and fewer x-ray photons are used to produce the radiographic image, the random statistical fluctuation of photons will eventually result in a loss of image detail.

The exposure reduction could also be traded for an increase in resolution by using thinner screens of smaller particle size. Very high resolution

screens of rare-earth oxysulfides may find application in areas where intensifying screens are not currently considered acceptable because of insufficient resolution.

These new rare-earth phosphors also have a considerable advantage in x-ray stopping power over cadmium zinc sulfide (Cd,ZnS) and they also have a comparable conversion efficiency. This means that the new phosphors should be useful for making higher efficiency fluoroscopic screens than can be made using Cd,ZnS. Recently, Gd₂O₃:Tb has replaced Cd,ZnS as the input phosphor in the Machlett x-ray image intensifier tube.

In the more than seven decades since CaWO₄ intensifying screens were introduced no new phosphor material has been developed which improves screen performance as dramatically as these rare-earth phosphors.

ACKNOWLEDGMENTS: The Gd₂O₃:Tb material used in this study was obtained from Dr. Melvin Tecotzky of the U. S. Radium Corporation. The Gd₂O₃:Tb screens were fabricated by Dr. T. Grant Maple of the Lockheed Research Laboratories.

REFERENCES

1. Bril A, Klasens HA: Intrinsic efficiencies of phosphors under cathode ray excitation. *Phillips Research Reports*. 7:401-420, Dec 1952
2. Coltman JW, Ebbinghausen EG, Altar W: Physical properties of calcium tungstate x-ray screens. *J Appl Phys* 18:530-544, 1947
3. Wickersheim KA, Alves RV, Buchanan RA: IEEE Nuclear Science Symposium, San Francisco, Calif., Oct. 29-31, 1969
4. Wickersheim KA, Buchanan RA, Sobon LE, et al: Study of rare earth activated materials for radiation-sensing applications. Third Annual Report. U. S. Atomic Energy Commission, Division of Biology and Medicine, Jun 1969, Lockheed Report No. 4-17-69-1

Lockheed Palo Alto Research Laboratory
Palo Alto, Calif. 94301

