



Measurement of the X-ray luminescence and spectral compatibility of the $\text{CdPO}_3\text{Cl}:\text{Mn}$ phosphor

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Abstract

The absolute efficiency, the spectral matching factor, the effective efficiency and the detective quantum efficiency of $\text{CdPO}_3\text{Cl}:\text{Mn}$ phosphor were evaluated to test its suitability for use in medical or other type of radiation detectors. $\text{CdPO}_3\text{Cl}:\text{Mn}$ was tested for 21–275 mg/cm^2 coating weights and for 50–250 kVp X-ray tube voltages. $\text{CdPO}_3\text{Cl}:\text{Mn}$ was most efficient between 70 and 110 kVp and its emission spectrum extended from 450 to 650 nm, being compatible with modern optical detectors. Additionally, a theoretical model was used to fit experimental data and to estimate the intrinsic conversion efficiency (0.12) and the reciprocal diffusion length (27 cm^2/g). © 2001 Elsevier Science Ltd. All rights reserved.

1. Introduction

Phosphors used as radiation to light converters in X-ray or γ -ray detectors are characterized by their luminescence efficiency and their spectral compatibility to optical photon detectors (Hamaker, 1947; Ludwig, 1971; De Poorter and Bril, 1975; Stevels and Schrama-de-Pauw, 1976; Arnold, 1979; Giakoumakis, 1991; Gurwich, 1995; Cavouras et al., 1998, 2000; Kandarakis et al., 1998, 1999). Luminescence efficiency is often expressed by the absolute efficiency (AE), which is defined by the ratio of the light energy flux, emitted by an excited phosphor, over the incident X-ray or γ -ray exposure rate (Cavouras et al., 2000). AE is a parameter of primary importance since it is related to the sensitivity of a radiation detector to detect low levels of radiation. The use of high absolute efficiency phosphors in various medical imaging applications may significantly reduce the radiation dose to the patient. Spectral compatibility describes the degree of coincidence between the spectrum of the light emitted by the excited phosphor and the spectral sensitivity

distribution of the optical photon detectors (films, photocathodes, photodiodes, CCDs, etc.) coupled to the phosphor. Spectral compatibility is often quantitatively assessed by the spectral matching factor (SMF), defined in terms of the light spectrum and the spectral sensitivity (Giakoumakis, 1991; Cavouras et al., 1998, 2000). Since spectral compatibility expresses how well the optical detector can capture the light emitted by the phosphor it is also related to the sensitivity of the radiation detector and consequently to the patient dose. AE and SMF may be combined in one parameter, the effective absolute efficiency (EAE). EAE may be defined as the ratio of the emitted light energy flux that can be detected by an optical photon detector, over the incident exposure rate (Cavouras et al., 1998, 2000). Besides experimental determination, AE and EAE may be also theoretically evaluated using theoretical models describing the radiation and light transfer through the phosphor material (Hamaker, 1947; Ludwig, 1971; Swank, 1973). These models are based on the radiation absorption properties and on the optical properties (optical scattering, absorption, reflectivity, X-ray to light conversion) of the phosphor materials. Using the theoretical models the detective quantum efficiency (DQE), describing the output signal to noise ratio (SNR) of a detector with respect to the input SNR (Swank, 1973; Dick and Motz, 1981), may be also calculated.

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In the present study, AE, SMF, EAE and the optical properties of the CdPO₃Cl:Mn phosphor were determined by experimental and theoretical methods, employing X-ray radiation in the X-ray tube voltage range between 50 and 250 kVp. This range of X-ray energies is very useful for various applications in astrophysics, medical physics, crystallography, etc. The optical detectors considered for SMF and EAE evaluation were photographic emulsions, photocathodes and photodiodes used in modern digital or conventional X-ray detectors. Additionally using the theoretical models and the values of the optical properties DQE was calculated. We have found that CdPO₃Cl:Mn is an efficient phosphor material exhibiting a broadband spectral distribution. This provides adequate spectral compatibility with a large variety of optical detectors including amorphous silicon detectors used in modern digital radiography systems. However, due to its medium fluorescence decay time (Lumilux Data Book, 1989) it may be only considered for radiographic (radiographic cassettes, flat panel digital detectors) or other types of static imaging applications (crystallography, astrophysics). To our knowledge, CdPO₃Cl:Mn is a phosphor material that has never been studied or used in imaging applications.

2. Materials and methods

2.1. Theory

The light energy flux $\bar{\Psi}_\lambda$ emitted by a phosphor material when irradiated by an X-ray energy flux $\bar{\Psi}_X$, may be given as follows:

$$\bar{\Psi}_\lambda(E_0, w_0) = \int_0^{E_0} \bar{\Psi}_X(E) \bar{\eta}_Q(E, w_0) \eta_C(E) \times \int_0^{w_0} \bar{\psi}_Q(E, w) \bar{g}_\lambda(\sigma, w) dw dE, \quad (1)$$

where E_0 is the maximum energy of the spectrum of X-rays, E_0 is numerically equal to the high voltage of the X-ray tube, w_0 is the thickness (or coating weight) of the phosphor, which is considered to be in the form of a fluorescent layer (screen), E is the energy of an X-ray photon, η_Q is the quantum detection efficiency (QDE), which expresses the fraction of incident X-ray photons that are detected by the phosphor and η_C is the intrinsic X-ray to light conversion efficiency (ICE), which expresses the fraction of absorbed X-ray energy that is converted into light within the phosphor material. In general, ICE is energy dependent (Engelkemeir, 1956). ψ_Q is a function giving the probability of an absorbed X-ray photon of energy E to be absorbed at a depth $w < w_0$. g_λ is the fraction of light photons, created at depth w , that escape the phosphor following the absorption of one X-ray photon, and σ is an optical parameter accounting for the attenuation of light within the phosphor, which is called the reciprocal of the light diffusion length (Ludwig, 1971; Swank, 1973). The second integral in (1) expresses

the fraction of light photons, created at various depths within the phosphor, that escape of the phosphor layer. This fraction is defined as the light transmission efficiency (LTE) of the phosphor (Hamaker, 1947; Ludwig, 1971; Kandarakis et al., 1998, 1999). The first integral is used to integrate over the energies of X-ray spectrum. Mean values in (1) express averaging of the corresponding parameters over the area of the detector.

The absolute efficiency (η_A) of a phosphor is given by the relation (Cavouras et al., 2000):

$$\eta_A(E_0, w_0) = \frac{\bar{\Psi}_\lambda(E_0, w_0)}{\dot{X}(E_0)}, \quad (2)$$

where \dot{X} is the exposure rate incident on the phosphor, which is emitted by an X-ray tube with high voltage equal to E_0 .

The spectral matching factor is given by the relation (Giakoumakis, 1991; Cavouras et al., 2000):

$$f_S = \frac{\int_{\lambda_1}^{\lambda_2} \varepsilon_P(\lambda) S_{OD}(\lambda) d\lambda}{\int_{\lambda_1}^{\lambda_2} \varepsilon_P(\lambda) d\lambda}, \quad (3)$$

where $\varepsilon_P(\lambda)$ is the spectrum of the light emitted by the phosphor and $S_{OD}(\lambda)$ is the spectral sensitivity of the optical detector coupled to the phosphor.

The effective absolute efficiency (η_{eff}) of a phosphor-optical detector combination is then defined by the relation

$$\eta_{A,\text{eff}}(E_0, w_0) = \eta_A(E_0, w_0) f_S. \quad (4)$$

The detective quantum efficiency (η_D), which is defined by the ratio $(\text{SNR}_{\text{output}}/\text{SNR}_{\text{input}})^2$, has been also expressed by the relation (Dick and Motz, 1981)

$$\eta_D(E_0, w_0) = \bar{\eta}_Q(E_0, w_0) I(\sigma, w_0), \quad (5)$$

where I is a parameter expressing the statistical fluctuations in the number of emitted light photons per absorbed X-ray photon, often called the statistical factor or the information factor. I is defined by the relation (Swank, 1973)

$$I(\sigma, w) = \frac{M_1^2(\sigma, w_0)}{M_2(\sigma, w_0)M_0(\sigma, w_0)}, \quad (6)$$

where

$$M_i(\sigma, w_0) = \int_0^{w_0} \bar{\psi}_Q(E, w) [\bar{g}_\lambda(\sigma, w)]^i dw, \quad i = 0, 1, 2. \quad (7)$$

M_i are the zeroth, first and second moments of the statistical distribution of the number of light photons emitted per X-ray absorbed (Swank, 1973; Dick and Motz, 1981). M_2 increases with the width of the statistical distribution while for a perfect detector $I = 1$.

2.2. Experiment

The phosphor material was supplied in powder form by Derby Luminescents Ltd. (code: CA2M). The mean size of the powder grains was approximately 7 μm . The size of the

grain is very crucial for both phosphor efficiency (Lindström and Carlsson, 1999) and image resolution. The latter is degraded with increasing phosphor grain while efficiency increases at the same time. However, it is generally accepted that sizes in the range from 5 to 10 μm give a satisfactory compromise between resolution and efficiency (Arnold, 1979; Gurwich, 1995). The phosphor powder was used to prepare fluorescent layers (screens), deposited on a fused silica substrate, by a sedimentation technique. A mixture containing 2.0 l of deionized water and 25 ml of Na_2SiO_3 (index of refraction 1.353) was used for the sedimentation. Na_2SiO_3 acting as a binder between the grains. The coating weight of the layers varied from 21 to 275 mg/cm^2 . X-rays, corresponding to tube voltages from 50 to 250 kVp, were used to irradiate the layers. To simulate X-ray attenuation by human body, the X-ray beam was filtered by 2 mm inherent and a 20 mm added aluminum filter. The emitted light energy flux Ψ_λ was measured by an EMI 9558 QB photomultiplier connected to a Cary 401 vibrating reed electrometer, while the incident exposure rate \dot{X} was measured by a PTW (ionization chamber No. 23333) dosimeter.

Light flux measurements were performed from both the front and the back side of each layer. These two measurements are referred to as the reflection and transmission observation mode, respectively. Reflection mode simulates the excitation and light emission from the rear side of a double-coated medical radiographic cassette. Transmission mode simulates all other types of radiation detectors. Light flux measurements were also corrected for: (1) the spectral mismatches between the emitted light and the spectral sensitivity of the photomultiplier, and (2) the optical losses due to incomplete light collection by the photomultiplier (geometric collection efficiency). These losses depend on the slight distance between the phosphor layer and the photocathode of the photomultiplier and on the angular distribution of the emitted light (Giakoumakis and Miliotis, 1985).

The spectrum $\varepsilon_p(\lambda)$ of the emitted light was measured by an Oriel grating monochromator while the spectral sensitivities of the optical detectors were obtained from manufacturers' data.

2.3. Calculations

Using relations (1) and (2) absolute efficiency may be also calculated as a function of the intrinsic physical parameters of the phosphor material. The physical quantities employed in relation (1) may be expressed by the following functions:

- (1) The incident X-ray energy flux Ψ_X was expressed and calculated using a theoretical model (Tucker et al., 1991) that describes the energy spectral distribution of the X-rays produced by a tungsten target X-ray tube.
- (2) The quantum detection efficiency η_Q was calculated by considering exponential X-ray absorption within the phosphor material, determined by the X-ray absorption coefficient and the thickness of the phosphor layer. The

absorption coefficient of $\text{CdPO}_3\text{Cl}:\text{Mn}$ was calculated using the corresponding coefficients of Cd, P, O, Cl obtained from data as tabulated by Storm and Israel (1967).

- (3) The absorption probability function ψ_Q , was calculated by the formula

$$\bar{\psi}_Q(E, w) = \frac{\bar{\Psi}_X(E)\mu(E)\exp[-\mu(E)w]dw}{\int_0^{w_0}\bar{\Psi}_X(E)\mu(E)\exp[-\mu(E)w]dw}, \quad (8)$$

where the numerator gives the probability for an X-ray photon to be absorbed at depth w within the phosphor and the denominator gives the total probability of X-ray absorption within the whole phosphor layer. $\mu(E)$ is the X-ray absorption coefficient of the phosphor material.

- (4) The function g_λ , giving the fraction of light escaping to the output per X-ray absorbed, was expressed by the formula (Ludwig, 1971; Swank, 1973)

$$\bar{g}_\lambda(\sigma, w) = \frac{\rho_1[(\beta + \rho_0)e^{\sigma w} + (\beta - \rho_0)e^{-\sigma w}]}{(\beta + \rho_0)(\beta + \rho_1)e^{\sigma w_0} - (\beta - \rho_0)(\beta - \rho_1)e^{-\sigma w_0}}, \quad (9)$$

where σ is the light attenuation coefficient of the phosphor, which is equal to the reciprocal of the light photon diffusion length (Hamaker, 1947; Ludwig, 1971; Swank, 1973) and it is given as a function of the optical scattering coefficient (s) and the optical absorption coefficient (a):

$$\sigma = [a(a + 2s)]^{1/2}. \quad (10)$$

ρ_0, ρ_1 are optical parameters expressing the reflection of light at the front and back phosphor surfaces defined as

$$\rho_n = (1 - r_n)/(1 + r_n), \quad n = 0, 1, \quad (11)$$

where r_n denotes the optical reflection coefficients at the front (0) and back (1) screen surfaces.

β is an optical parameter which is equal to ρ , corresponding to the case of a very thick phosphor with no light transmission through it. β has been also expressed as a function of a and s (Ludwig, 1971):

$$\beta = [a/(a + 2s)]^{1/2}. \quad (12)$$

β and ρ_n were determined as described previously (Ludwig, 1971; Cavouras et al., 2000; Kandarakis et al., 1999). These data were used in order to fit relation (1) to the experimental absolute efficiency measurements. Using the Levenberg-Marquardt technique (Press et al., 1990), best fit was obtained, for specific values of the parameters η_C and σ , in relations (1) and (2). These values, together with β and ρ_n , were then adopted as the intrinsic optical properties of the phosphor.

3. Results and discussion

Fig. 1 shows the variation of absolute efficiency of $\text{CdPO}_3\text{Cl}:\text{Mn}$ with X-ray tube voltage for various coating weights, measured in reflection mode. Points represent

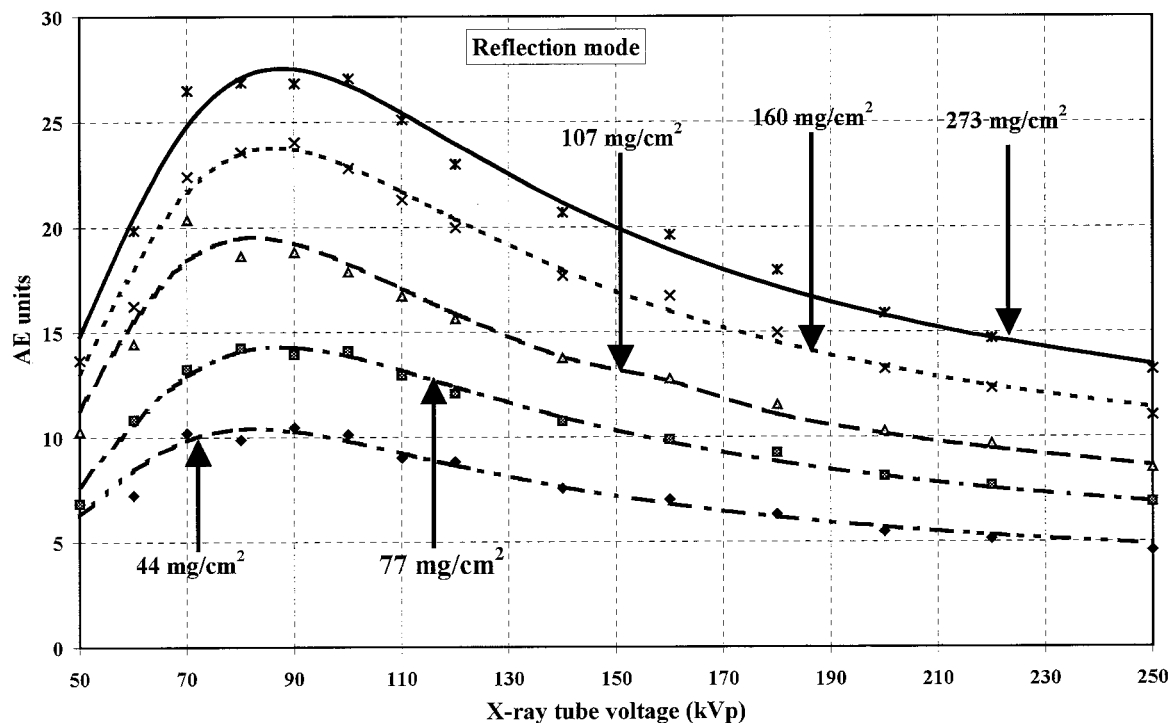


Fig. 1. Variation of the absolute efficiency (AE) of the $\text{CdPO}_3\text{Cl}:\text{Mn}$ phosphor with X-ray tube voltage, measured in reflection mode. Points correspond to experimental values and solid lines represent theoretical curves. AE units: $\mu\text{W s/mR m}^2$.

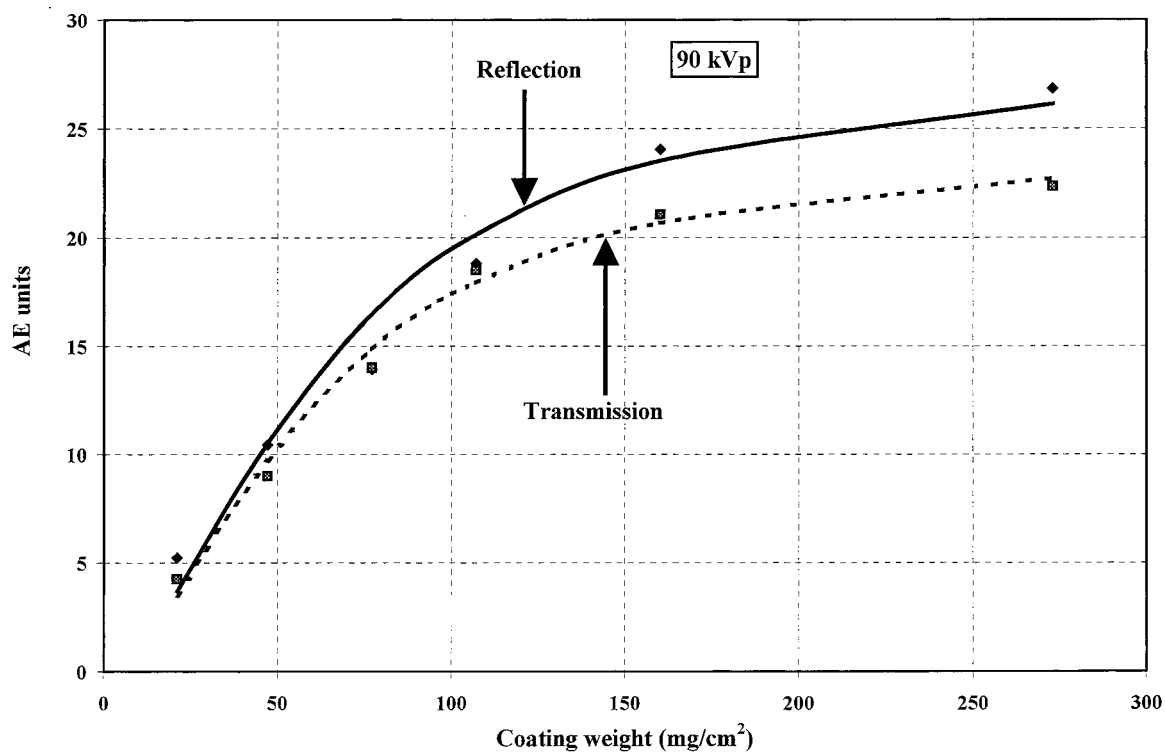


Fig. 2. Variation of the absolute efficiency (AE) of $\text{CdPO}_3\text{Cl}:\text{Mn}$ phosphor with coating weight measured in transmission and reflection mode at 90 kVp. AE units: $\mu\text{W s/mR m}^2$.

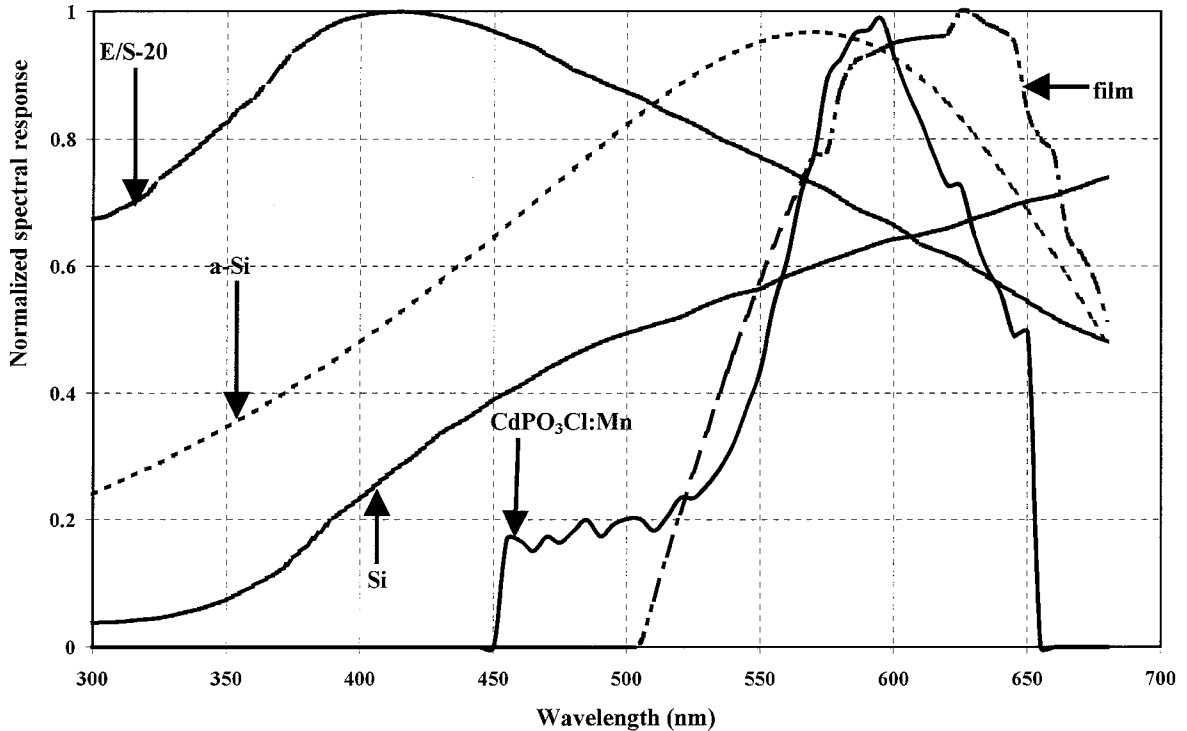


Fig. 3. Normalized light emission spectrum of $\text{CdPO}_3\text{Cl}:\text{Mn}$ phosphor and spectral sensitivities of optical photon detectors: red sensitivity film, crystalline silicon, amorphous silicon and Extended sensitivity S-20 photocathode.

experimental data while solid lines correspond to fitted theoretical curves. Best fitting was obtained for the values $\eta_C = 0.12$ and $\sigma = 27 \text{ cm}^2/\text{g}$. The value of the intrinsic conversion efficiency (η_C), as estimated by the fitting, is higher than the corresponding value of CaWO_4 ($\eta_C = 0.04$), which is used in conventional radiographic cassettes, but lower than that of rare earth phosphors ($\eta_C = 0.18$) (Cavouras et al., 2000). It is also slightly higher, or approximately equal to the η_C values of $\text{CsI}:\text{Na}$, $\text{CsI}:\text{Tl}$ and $\text{NaI}:\text{Tl}$ phosphors ($\eta_C = 0.10$), used in a large variety of radiation detectors including image intensifiers, flat panel detectors, γ -cameras, computed tomography detectors, etc. (Arnold, 1979; Haque and Stanley, 1981). The optical attenuation coefficient σ was found lower than in rare earth phosphors ($\sigma = 30 \text{ cm}^2/\text{g}$) (Cavouras et al., 2000). This was expected since the mean wavelength of the $\text{CdPO}_3\text{Cl}:\text{Mn}$ spectrum (see Fig. 3) is higher than that of the rare earth spectra (Cavouras et al., 2000). An important observation from Fig. 1 is that absolute efficiency maintains high values within a wide range of X-ray tube voltages from 70 to 100 kVp. This property is very interesting for medical imaging, since this range of tube voltages is used in a large number of radiographic examinations. As tube voltage increases further, from 110 to 250 kVp, absolute efficiency decreases in a rather slow rate, showing that $\text{CdPO}_3\text{Cl}:\text{Mn}$ may also be employed in other applications.

Table 1
Spectral matching factors

Optical detectors	$\text{CdPO}_3\text{Cl}:\text{Mn}$	$\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$
Agfa Scopix LT2B	0.737	0.519
c-Si	0.608	0.533
a-Si	0.885	0.875
ES-20	0.706	0.784

Fig. 2 shows a comparison between the reflection and transmission mode measurements of absolute efficiency. As it is expected from previous studies (Cavouras et al., 2000), reflection mode measurements give higher AE-values, due to the fact that a larger fraction of the incident X-rays are absorbed in the first half of the phosphor layer. However, the difference between the two modes is not significant for layers up to $110 \text{ mg}/\text{cm}^2$. This may be useful in digital radiography, where radiation detectors are employed only in transmission mode and layers thicker than $120 \text{ mg}/\text{cm}^2$ are rarely used, because of a significant degradation in the spatial resolution. As it is also observed from Fig. 2, absolute efficiency increases continuously with increasing phosphor-coating weight. However, the two curves show a tendency to saturate after $150 \text{ mg}/\text{cm}^2$. Hence, use of very thick phosphors may not be justified because the

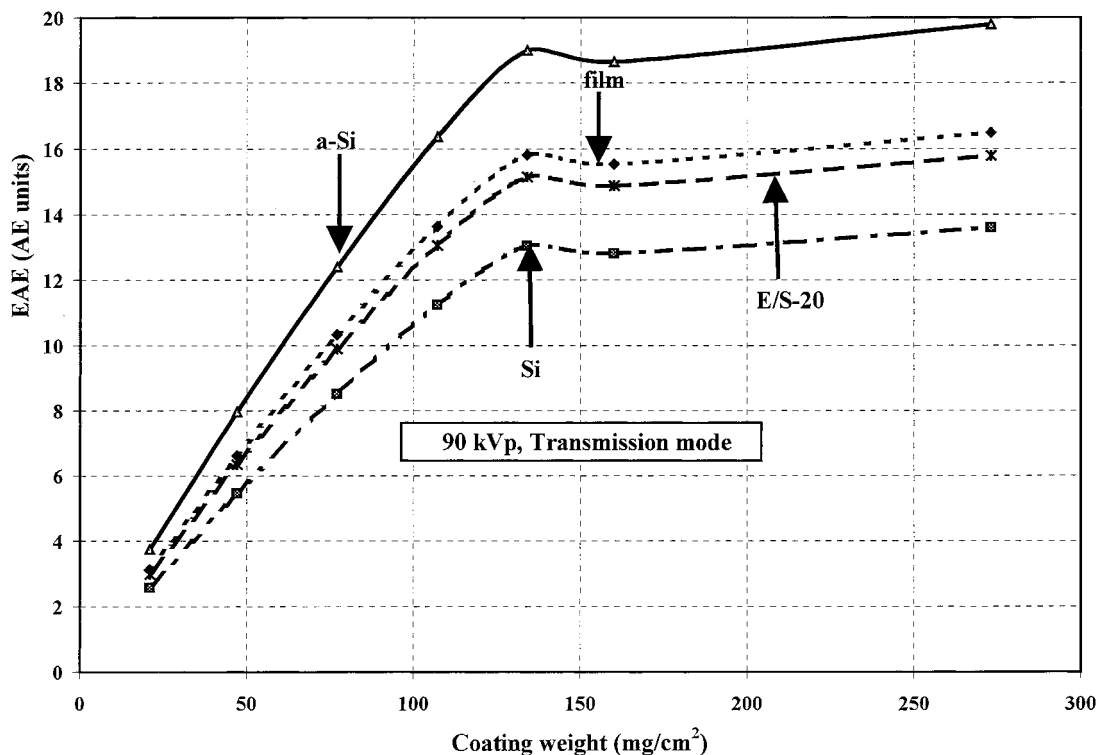


Fig. 4. Variation of the effective absolute efficiency (EAE) of $\text{CdPO}_3\text{Cl}:\text{Mn}$ with coating weight, for four optical photon detectors. AE units: $\mu\text{W s/mR m}^2$.

increase in absolute efficiency may not counterbalance the large loss in spatial resolution.

Fig. 3 shows a plot of the $\text{CdPO}_3\text{Cl}:\text{Mn}$ emission spectrum measurements including four spectral sensitivity curves corresponding to optical detectors currently used in a large variety of modern X-ray or γ -ray detectors. The optical detectors considered were the following: (1) A photographic emulsion (Agfa Scopix LT2B) sensitive to light wavelengths from 500 to 700 nm often used with laser cameras in medical imaging departments, (2) The crystalline silicon (c-Si) used in photodiodes and CCD arrays of various detectors for digital radiography, computed tomography, astronomy, crystallography etc., (3) The amorphous silicon (a-Si) used in recently introduced flat panel detectors for digital radiography and in computed tomography detectors (Haque and Stanley, 1981; Gurwich, 1995; Gambaccini et al., 1996; Spahn et al., 1997; Neitzel, 1997), (4) The extended sensitivity S-20 (ES-20) photocathode used in image intensifiers and in photomultipliers of nuclear radiation detectors. As it is observed from Fig. 3, the $\text{CdPO}_3\text{Cl}:\text{Mn}$ spectrum is lying well within the sensitivity limits of a-Si, c-Si and film.

Table 1 shows the values of the spectral matching factors, corresponding to the aforementioned detectors, calculated according to relation (3). In the same table and for comparison reasons, values related to the very often-employed

rare earth ($\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$) phosphor are also shown. $\text{CdPO}_3\text{Cl}:\text{Mn}$ shows very high spectral compatibility with a-Si, which is also higher than that of the rare earth phosphor. Additionally, the spectral compatibility with c-Si (and hence with CCDs) is adequately high and also higher than the corresponding compatibility of $\text{Gd}_2\text{O}_2\text{S}:\text{Tb}$. $\text{CdPO}_3\text{Cl}:\text{Mn}$ is also very well compatible with the photographic emulsion and, thus, it may be used in conventional radiographic applications.

Fig. 4 is a plot of the variation of the effective absolute efficiency with coating weight, showing that the best phosphor-optical detector combination of $\text{CdPO}_3\text{Cl}:\text{Mn}$ phosphor is achieved with the amorphous silicon photodiode.

Fig. 5 shows the variation of DQE with coating weight for various tube voltages. As it is observed DQE decreases with increasing X-ray tube voltage and is higher for medium thickness phosphor layers in the range between 110 and 150 mg/cm^2 . At very thick layers DQE shows a tendency to decrease, which becomes more apparent for low voltages. This may be physically explained by considering that at low voltages the absorption of X-rays (η_Q) is higher. Also, at very thick layers statistical fluctuations in the number of emitted light photons are high, since the sites of light creation within the phosphor are largely dispersed. This causes an

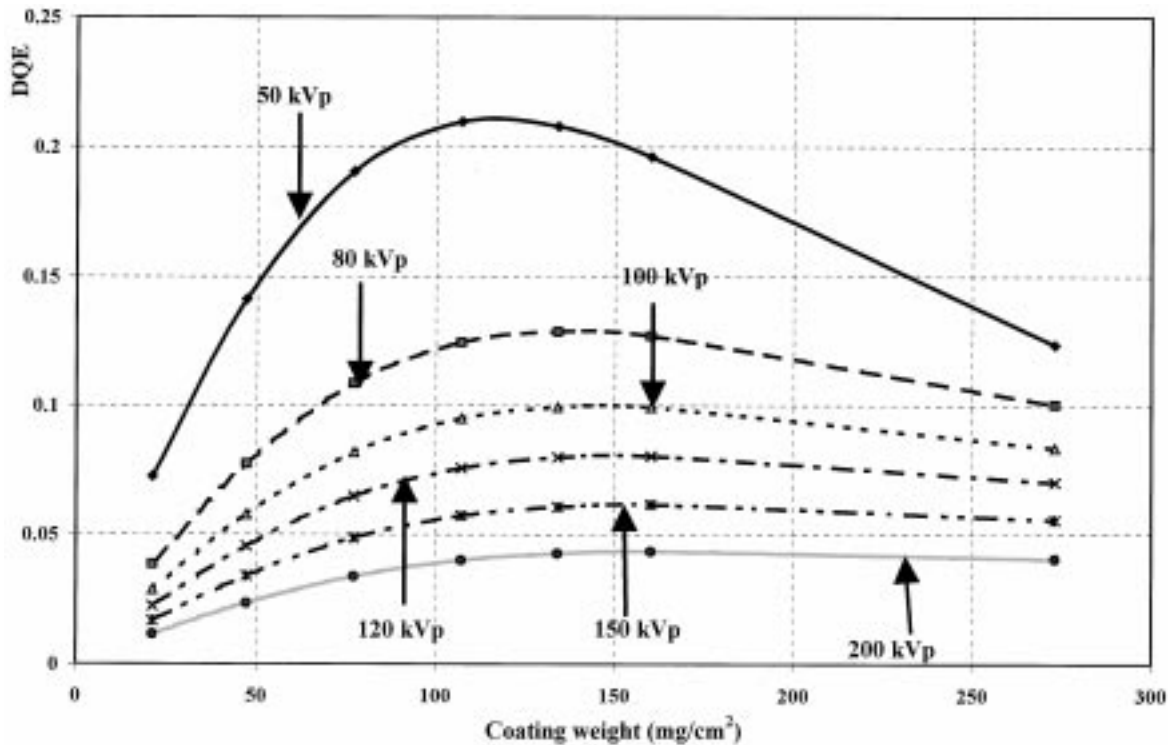


Fig. 5. Variation of the detective quantum efficiency (DQE) of CdPO₃Cl:Mn with coating weight and X-ray tube voltage.

increase in the second statistical moment M_2 (see relation (6)) and, hence, a decrease in the statistical factor I .

In conclusion, these data demonstrate the suitability of the CdPO₃Cl:Mn phosphor for use in modern digital detectors of medical radiography, in conventional radiographic applications, or in other types of applications, provided that very fast detector response is not necessary.

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