Linköping University Medical Dissertation No. 1773

Radioluminescence:

A simple model for fluorescent layers
- analysis and applications

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Linköping 2021

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To all of those who never stopped believing!

According to a widespread legend,
a wise old monk on a Tibetan mountain,
從來沒有一次, uttered the following words:

“A grain of truth:
the simple model is,
theoretically and practically,
about something, next to nothing”
廢話很多 (1832-1914)

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ABSTRACT

A phosphor or scintillator is a material that will emit visible light when struck by ionising radiation. In the early days of diagnostic radiology, it was discovered that the radiation dose needed to get an image on a film, could be greatly reduced by inserting a fluorescent layer of a phosphor in direct contact with the film. Thus, introducing the step of converting the ionising radiation to light in a first step. Going forward in time, film has been replaced with photodetectors and there is now a variety of imaging x-ray systems, still based on phosphors and scintillators.

There is continuous research going on to optimise between the radiation dose needed and a sufficient image quality. These factors tend to be in opposition to each other. It is a complicated task to optimise these imaging system and new phosphor materials emerges regularly. One of the key factors is the efficiency of the conversion from x-rays to light. In this work this is denoted “extrinsic efficiency”. It is important since it largely determines the final dose to the patient needed for the imaging task. Most imaging x-ray detectors are based on phosphor or scintillator types where their imaging performance has been improved through tweaking of various parameters (light guide structure, higher density, light emission spectrum matching to photodetectors, delayed fluorescence quenching etc)

One key factor that largely determines the extrinsic efficiency of a specific phosphor is the particle size. Larger particles result in a higher luminance of the phosphor for the same radiation dose as does as a thicker phosphor layer (to a limit). There exists already a battery of models describing various phosphor qualities. However, particle size and thickness have not been treated as a fully independent variables in previous model works. Indirectly, the influence of these parameters is accounted for, but the existing models were either considered too general, containing several complex parameters and factors to cover all kind of cases or too highly specialised to be easily applicable to fluorescent detectors in diagnostic radiology.

The aim of this thesis is therefore to describe and assess a simple model denoted the “LAC-model” (after the original authors Lindström and Alm Carlsson), developed for a fluorescent layer using individual sub-layers defined by the particle size diameter. The model is thought to be a tool for quickly evaluating various particle size and fluorescent layer thickness combinations for a chosen phosphor and design. It may also serve as a more intuitive description of the underlying parameters influencing the final extrinsic efficiency.

Further tests affirmed the validity of the model through measurements. The LAC-model produced results deviating a maximum of +5 % from luminescence measurements.

During the development of the model various assumptions and simplifications were made. One assumption was the absence of a so called “dead layer”. This is a layer supposedly surrounding each particle decreasing the efficiency of converting x-rays to light. It is not completely “dead” as in inactive but is thought to have a reduced efficiency. This phenomenon was struggled with, when historically designing electron beam stimulated phosphors for various applications (i.e. displays, TV tubes etc). There are also articles reporting dead layer influence for x-ray detectors (usually spectrometers i.e. not for imaging). By introducing a dead layer in the LAC-model the effect of the layer was investigated and was found to result in a change of less than 8% for the extrinsic efficiency.

It was also noted that sometimes a dead layer effect may emerge at surfaces of a scintillator slab but not necessarily connected to the phosphor particles themselves. Due to differences between phosphor material and the surroundings, an interface effect arose to compete with the process of inherent dead layers of the individual particles. It was found to be mostly negligible for x-rays in the studied energy and material range. However, an effect was shown for electrons as incident ionising radiation which could shed some light on the strangely neglected apparent dead layer created this way. Finally, applications, one involving developing a prototype for checking the light field radiation field coincidence, were evaluated for overall performance and the optimisation level of the applied fluorescent layer. Interesting findings were made during the development process: for the first time to the knowledge of the author, *focus shift wandering* was quantified in the corresponding movement of the x-ray field edge and a non-trivial discussion on the concept of an apparent light field edge resulted in a modified definition of the same.

SVENSK SAMMANFATTNING

En fosfor eller scintillator är ett material som avger synligt ljus när det träffas av joniserande strålning. Inom diagnostisk radiologi upptäckte man i ett tidigt skede att stråldosen som behövdes för att få en bild på en röntgenfilm, reducerades kraftigt om man placerade ett fluorescerande skikt, en fosfor, i direkt kontakt med filmen. I nutid har film ersatts med fotodetektorer och det finns nu en mängd olika röntgenbildsystem men som fortfarande är baserade på fosforer och scintillatorer.

Det pågår en kontinuerlig forskning för att optimera mellan erforderlig stråldos och en tillräcklig god diagnostisk bildkvalitet. Dessa faktorer tenderar att motverka varandra. Det är en komplicerad uppgift att optimera röntgenbildsystemen och nya fosformaterial dyker ständigt upp. En av de viktiga egenskaperna är fosforns omvandlingseffektivitet från röntgen till ljus. I detta arbete används benämningen "extrinsisk (yttre) effektivitet". Denna egenskap är viktig eftersom den i stor utsträckning bestämmer den slutliga dosen till patienten som krävs för bilddiagnostiken. De flesta röntgendetektorer är baserade på fosfor- eller scintillatortyper där bildprestanda har förbättrats genom att utveckla olika parametrar (ljusledarstruktur, högre densitet, ljusemissionsspektrum som matchar fotodetektorer, minskad efterlysning etc.). En viktig faktor som i stor utsträckning bestämmer omvandlingseffektiviteten hos en specifik fosfor är partikelstorleken. Större partiklar resulterar i en högre luminescens (mer ljus) från fosforen för samma stråldos. Vilket också gäller för ett tjockare fosforlager (till en viss gräns!). Det finns redan fysikaliska modeller som beskriver olika fosforparametrar men partikelstorlek och fosfortjocklek har dock inte hanterats som fristående variabler i dessa modellarbeten. Istället har deras inverkan modellerats indirekt men det har gjort att de befintliga modellerna kan anses komplexa. De är antingen för generella som medför flera komplexa parametrar och faktorer för att täcka alla tänkbara varianter eller för specialiserade för att kunna tillämpas enkelt på fluorescerande detektorer i diagnostisk radiologi.

Syftet med denna avhandling är därför att beskriva och analysera en praktisk modell betecknad "LAC-modellen" (efter de ursprungliga författarna Lindström och Alm Carlsson). Den är utvecklad för ett fluorescerande block som består av flera underliggande skikt vars tjocklek bestäms av partiklarnas diameter. Avsikten med modellen är att den ska vara ett verktyg för att snabbt utvärdera olika varianter av partikelstorlek och tjockleks-kombinationer för en vald fosfor med i grunden samma design. Experiment har bekräftat modellens giltighet och mätresultat visar att modellresultaten avvek maximalt +5% från luminescensmätningar.

Utvecklingen av modellen krävde olika antaganden och förenklingar. Ett antagande var frånvaron av ett så kallat "dött lager". Det är ett skikt som antas omge varje partikel och som därför minskar omvandlingseffektiviteten från röntgen till ljus. Det är dock inte helt "dött" i meningen helt inaktivt men har en mindre förmåga att omvandla röntgen till ljus jämfört med fosforns huvudmaterial. Historiskt sett har man försökt åtgärda detta fenomen under lång tid och speciellt för applikationer där man använt sig av elektronstrålar (dvs olika typer av displayer, TV-rör etc.). Just för

elektroner har man sett att döda skiktet tenderar att växa med tiden. Det finns också artiklar som rapporterar en påverkan av röntgendetektorers funktion (vanligtvis dock för spektrometrar, dvs inte för avbildning).

Genom att införa ett dött skikt i LAC-modellen undersöktes skiktets effekt och visade sig resultera i en förändring på mindre än 8% för effektiviteten. Det noterades också att ibland kan en dödsliktsliknande effekt uppstå vid ytor av ett scintillatorblock men inte nödvändigtvis pga. av själva fosforpartiklarnas ljusomvandlingsegenskaper. Då det uppstår skillnader mellan fosformaterialet och omgivningen får man en s.k. gränsskiktseffekt som s.a.s. konkurrerar med kemiskt döda skiktet på de enskilda partiklarna.

De döda skiktens inverkan visade sig i princip försumbara för röntgenbild-detektorer - åtminstone inom det studerade energi- och materialområdet. En tydlig effekt kunde dock noteras för joniserande strålning i form av elektroner. Simuleringarna kunde ge en bättre bild av egenskaperna hos det döda skiktet som skapats på detta sätt.

Slutligen utvärderades två applikationer med hjälp av LAC-modellen: en prototyp för kontroll av ljusfältets och strålfältets överensstämmelse i läge och position. Samt en etablerad produkt med samma användningsområde. I båda fallen undersöktes det fluorescerande skiktets optimeringsgrad. Intressanta resultat noterades under utvecklingsprocessen av prototypen: för första gången, så vitt författaren vet, kunde man kvantifiera röntgenrörs s.k. fokusvandring.

LIST OF PAPERS

- I. Lindström, J., & Carlsson, G. A. (1999). A simple model for estimating the particle size dependence of absolute efficiency of fluorescent screens. *Physics in Medicine & Biology*, 44(5), 1353.
- II. Lindström, J., Carlsson, G. A., Wåhlin, E., Tedgren, Å. C., & Poludniowski, G. (2020). Experimental assessment of a phosphor model for estimating the relative extrinsic efficiency in radioluminescent detectors. *Physica Medica*, 76, 117-124.
<https://doi.org/10.1016/j.cjmp.2020.07.009>
- III. Lindström, J., Lund, E., Wåhlin, E., Tedgren, Å.C. (2021). Revisiting the dead layer in phosphors from a dosimetric perspective- assessment through Monte-Carlo simulations and modelling, - - to be submitted to *Journal of Luminescence*.
- IV. Lindström, J., Hulthén, M., Sandborg, M., & Tedgren, Å.C. (2020). Development and assessment of a quality assurance device for radiation field–light field congruence testing in diagnostic radiology. *Journal of Medical Imaging*, 7(6), 063501. Epub 2020 Nov 20.
<https://doi.org/10.1117/1.JMI.7.6.063501>

Related papers, not included in the thesis

Lindström, J., Hulthén, M., Carlsson, G. A., & Sandborg, M. (2014, March). Optimizing two radioluminescence based quality assurance devices for diagnostic radiology utilizing a simple model. In *Medical Imaging 2014: Physics of Medical Imaging* (Vol. 9033, p. 90333R). International Society for Optics and Photonics.

CONTRIBUTIONS

Paper I: The author developed the idea and theoretical framework for the model. Definitions and theoretical framework were added by the supervisor at the time; professor Gudrun Alm Carlsson. The paper was written by the author with help from the supervisor.

Paper II: The author set-up and conducted the experiments, including manufacturing the screens. The paper was written by the author with the help of the co-authors. Professor Emerita Gudrun Alm Carlsson helped with the writing and also made sure that the connection to the first paper was not broken. Medical Physicist Erik Wåhlin did the Monte-Carlo simulations based on the geometry given by the author. The current main supervisor, Associate professor Åsa Carlsson Tedgren coordinated the various efforts besides helping with the writing. PhD Gavin Poludniowski did some extensive quality assurance of the theoretical content in the paper, including the uncertainty budget and theoretical connections of the LAC-model to the Hamaker-Ludwig model. Gavin also contributed with help in the writing process.

Paper III: The author developed the idea and set-up the framework for the paper. The paper was written by the author with the help of the co-authors. Professor Emerita Eva Lund did extensive quality assurance of the comprehensiveness of the content. Medical Physicist Erik Wåhlin conducted all the Monte-Carlo simulations based on geometries and set-ups by the author. Main supervisor Åsa Carlsson Tedgren coordinated the efforts and scrutinised the text.

Paper IV: The author developed the idea and set-up the framework for the paper. The paper was written by the author with help from the co-authors. The paper is based on the content of the Master thesis of Medical Physicist Markus Hulthén. (Jan Lindström was the practical supervisor for that work.) Markus contributed with his knowledge on the theoretical background, programming and assessment of the prototype involved. Associate Professor Michael Sandborg was supervisor and co-author of the preceding conference paper on the subject. He quality assured the content of this paper after having a main impact on the preceding conference paper. The main supervisor Åsa Carlsson Tedgren, helped with the writing and also secured that the paper fell in line with the main subject of the PhD thesis.

ABBREVIATIONS

AE	Absolute Efficiency
AEC	Automatic Exposure Control
AU	Arbitrary Unit
CCD	Charge Coupled Device
CIE	Commission Internationale de l'Éclairage
CL	Cathodoluminescence
CMOS	Complementary Metal Oxide Semiconductor
CRT	Cathode Ray Tube
CT	Computed Tomography
DAP	Dose Area Product
EE	Extrinsic Efficiency
EL	Electroluminescence
ESSCR	Electron Beam Stimulated Surface Chemical Reaction
FPA	Field Position Analyser
FWHM	Full Width at Half Maximum
HL	Hamaker-Ludwig (model)
ICRP	International Commission on Radiological Protection
ICRU	International Commission on Radiation Units & Measures
IE	Intrinsic Efficiency
KAP	Kerma Area Product
K-M	Kubelka-Monk (model)
kVp	kilovolt (peak)
LAC	Lindström Alm Carlsson (model)
LED	Light Emitting Diode
LIS	Linear Imaging Sensor
LLG	Lambertian Light Guide (model)
LP	Line Pairs
LSF	Line Spread Function
LTE	Light Transmission Efficiency
MC	Monte Carlo
MTF	Modulation Transfer Function

OTF	Optical Transfer Function
PENELOPE	PENetration and Energy LOss of Positrons and Electrons in matter
PET	Positron Emission Tomography
PL	Photoluminescence
PMMA	Polymethyl methacrylate (Perspex, Plexiglas etc)
PVC	PolyVinylChloride
RL	Radioluminescence
SEM	Scanning electron microscope
SPECT	Single Photon Emission Computed Tomography
SPIE	The International Society for Optonics and Photonics
TLD	Thermoluminescence Dosimeter

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1.INTRODUCTION

1.1 Background

Applications of luminescence in the radiological contexts was introduced early on in the previous century. This was a major leap in medicine as well as in physics. This thesis started as a question during the 1990's when film cassettes were still around. As a part of the quality control program, the cassettes were a continuous source for attention. The main focus was on the film developers and the properties of the intensifying screens were something of a "black box". They worked obviously, but there was relatively little information on how they were optimised by the manufacturer. Extensive information was given on the film itself but very often your questions were answered with just a "trade secret" shrug. In hindsight, it was obvious that the factual information was in few hands. When mammography screening was about to start in the late 80's, special cassettes with only one intensifying screen were introduced. It was not only due to the lower energies of the x-rays in mammography (lower transmission), it was also to increase the resolution. During the procurement process this author asked many questions, one was about the phosphor material used. At the time, Gadolinium Oxysulphide was *the* material and the classical blue light emitting Calcium Tungstate was definitely on its way out. Hamaker-Ludwig was yet not ever heard of, or anything similar for that matter. But this author understood intuitively that once the phosphor material was chosen, one could optimise with the thickness and the particle size. Where did that idea come from? This author obtained some samples of the raw phosphor powder from a manufacturer and realised soon that the luminance produced from the imparted x-ray energy was connected to these parameters. But how? Unaware of any other approaches some very simple (perhaps over-simplistic) fitting functions were tried and one could see that for a fixed thickness, it seemed that the luminance varied proportionally to the square root of the particle size diameter times a fitting factor. The results were clear. During that time storage phosphors image plates and Cesium Iodine (needle-shaped) phosphors made their entrance. It was the beginning of the era of digital imaging.

The main focus of this thesis is a radioluminescence model for x-ray imaging detectors and quality assurance applications. The model can describe (polycrystalline or semi-monocrystal) phosphor layer of the classical intensifying screens, Flat Panel Detectors (FPD) found in Digital Radiology (DR) or semi-single crystals in Computed Tomography (CT), Positron Emission Tomography (PET) or Single Photon Computed Tomography (SPECT).

Optimisation between patient dose and image quality involves many factors and has exhaustively been described in the scientific literature (see eg. Tsai and Matsuyama, 2015; Gingold, 2017; Morin and Frush, 2017; Sensakovic, Warden and Bancroft, 2017; Tootell, 2018; Tsapaki, 2020)

Focusing on the x-ray to light conversion process in a chosen phosphor means, among other things, a process of optimising the combination of particle size and thickness of the phosphor layer.

There are various established models for phosphors and scintillators. Depending on what the modelling task is, there is still a demand for different input parameters. These can be difficult to obtain and sometimes must be produced using highly specialised measuring equipment. Even when these obstacles are overcome, the model output results are then normally valid for the one modelled case, i.e. the results cannot effortlessly be applied and extended to other particle size and thickness combinations.

Liaparinos and Kandarakis (2009) investigated factors having an impact on resolution (and noise) for a $\text{Gd}_2\text{O}_2\text{S:Tb}$ phosphor layer in imaging detectors used in conventional diagnostics and mammography at the time. Utilising Monte-Carlo techniques for both the radiation transfer and the optical transport, they obtained Modulation Transfer Functions (MTFs) for various particle size and phosphor thickness combinations. The particle size was reduced from $13\text{ }\mu\text{m}$ to $4\text{ }\mu\text{m}$ for a fixed incident x-ray spectrum (mean energy 49 keV) and coating thickness (60 mgcm^{-2}). A corresponding variation in the (maximum) resolution of 11.9 to $13.4\text{ (mm}^{-1}\text{)}$ in the so-called reflection mode was calculated (observer or detector at the impinging side of the fluorescent layer). They also varied the packing density, showing that an increase will improve the resolution.

Even if Liaparinos and Kandarakis (2009) were able to simulate and obtain satisfactory results from their Monte Carlo modelling, it was done so after considerable effort. If a more practical approach is desired, an ideal model would contain the particle size and thickness of the phosphor layer as independent variables rather than indirect through for instance optical parameters difficult to obtain outside a specialised laboratory.

Instead of handling the phosphor as a bulk material it seemed reasonable to try to find a model where the particles were treated as individual objects of a mean particle size. Literature studies did however point in a different direction. Kuboniwa (1973) argued that 'it is not advantageous to make a microscopic consideration on such particle layers'. That was interpreted as a path leading nowhere. There was really no firm justification for this statement and another paper by Giakoumakis et al. (1991) did treat the particles as individual objects. This turned out to be a step to obtain the coating weight which may be seen as an indirect approach to account for the particle size (among other parameters). It was at this time a decision was made to try to develop a model with the same starting step as Giakoumakis et al. but for the different purpose of trying to preserve the discrete presence of the particles in the model of the collective slab.

As a medical physicist and being used to the stringent terminology and definitions of ICRU (International Commission on Radiation Units & Measures) and ICRP (International Commission on Radiological Protection), there was a moment of despair when encountering so many different terms for the same thing. It seemed that the terminology depended more on the application at hand rather than trying to create

a comprehensive terminology across the various fields of luminescence. This author counted to at least ten separate ways of expressing the luminescence from an excited phosphor or scintillator. It made going from one paper to another, sometimes difficult unless the current author was a part of the same research team. It underlined the importance of very clearly describing the terms to be used in the model.

Dead layers in luminescence and their elusive characteristics was something encountered early in the process of the model development. The work of Kuboniwa et al. (1973) founded the traditional explanation of particle size dependence for the luminescence of phosphor layers. Kuboniwa et al. stated that a dead layer of a fixed thickness surrounded each individual particle. (Denoted “intrinsic” dead layer in this work). When the particle size is decreased, the ratio of the fixed dead layer to the volume of the particle will increase and hence the luminance will be reduced accordingly. Avoiding the suggested influence of the dead layer at the time, the first theoretical framework was laid out in paper I, (Lindström and Alm Carlsson, 1999). However, the intrinsic dead layer was not forgotten and was further investigated in paper III (Lindström et al., 2021).

Finally, radioluminescence based applications developed by the author, are included in the thesis. These are devices for the light field – radiation field congruence testing of x-ray equipment (in versions of the first device, also for therapy equipment, i.e. denoted “X-lite” IBA Group, Louvain-La-Neuve, Belgium).

The suggested model plays a part in the assessment of the fluorescent layer optimisation of the applications. The devices are of two different technical solutions: One device is based on activation of phosphorescence from x-rays where a full field surface will show a visible afterglow for measuring any deviation from built-in scales for the light field position. (Field Position Analyser, FPA)

The other device is based on a one-dimensional, linear imaging sensor (LIS) that was used for detection and determination of field edges (of both light and ionising radiation fields). As is, the linear imaging sensor is not sufficiently sensitive for x-rays and therefore sensitised utilising a $\text{Gd}_2\text{O}_2\text{S:Tb}$ -strip. The particle size and thickness of the chosen strip was evaluated using the simple model and showed that potential improvements may be considered.

The target reading group of this thesis is assumed to have an appropriate background in the radiation physics involved. More emphasis is therefore placed on optical processes necessary for the understanding. Radiation physics and luminescence research has been tightly connected throughout time and the history section give some examples of important milestones on the way. The theory and materials/method sections are extensive but crucial to grasp the various approaches and benefits of the proposed model

The first paper of the thesis was published in 1999 and has some 30+ citations at the time of writing. Despite the time that has passed, the model keeps finding new applications and use in research. This has also been outside the field of optimising radioluminescent layers for x-ray imaging; i.e. LED (light emitting diodes), Chen et al.

(2010); Photodynamic drugs, Abliz et al. (2010); double layer scintillators, Song, Shim and Han (2018); and nanoparticle phosphors for lasers, Yordanova et al. (2018).

The simplicity of the proposed model facilitates thought experiments for potential future extension of its use. One such experiment is the possibility of obtaining the MTF of a modelled phosphor layer. Another utilisation is modelling (double-) multilayer detectors where the layers comprises different particle sizes (Song, Shim and Han, 2018). Even structural scintillators like CsI:Tl could be the target of modelling by introducing a well-defined unit cell. Some hints on these prospects (not previously published) are given in chapter 5; “future prospects”.

1.2 History of radioluminescence

Not widely known among medical physicists, studies of fluorescence and phosphorescence materials accidently led the way to the discovery of radiation in 1895. A barium platino-cyanide screen ($\text{Ba}[\text{Pt}(\text{CN})_4]$) alerted Wilhelm Conrad Röntgen by glowing when he switched on a discharge tube (Collier, 1974; Feldman, 1989; Thomas and Banerjee, 2013; Biduchak et al., 2019). This new, invisible, and hitherto unknown radiation was denoted the “x-ray”. Early on, it was realised that film had a rather low absorption of these new x-rays. Already in 1896, Mihajlo Pupin suggested the use of the phosphor Calcium Tungstate (CaWO_4) in the shape of a thin layer applied on the film. This increased the overall sensitivity to dose greatly in the terms of film density and was named “intensifying screen”. Thomas Edison (1896) utilised CaWO_4 further and developed fluoroscopy equipment claiming a six times higher luminescence than barium platino-cyanide (Edison, 1896). Calcium Tungstate survived as an intensifying screen material long into the 1980’s but already in the 1960’s, Buchanan, Tecotzky and Wickersheim (patent in 1973) discovered phosphors based on rare earth materials (Buchanan, Tecotzky and Wickersheim, 1973). The $\text{La}_2\text{O}_2\text{S:Tb}$ and $\text{Gd}_2\text{O}_2\text{S:Tb}$ materials, proved to be superior to previous phosphors and succeeded with the achievement in both providing better image quality and lowering the radiation dose in diagnostic radiology (Ludwig and Prener, 1972).

In another radioluminescence field Karl Ferdinand Braun; finished his development of the new cathode ray tube (CRT) in 1897. Accelerated electron beams strike a Zinc Sulphide (ZnS) phosphor and this energy is converted into visible light. The basic technical design gave the name to this special process: cathodoluminescence. Braun received the Nobel Prize in Physics for his achievements (1909). The ZnS -screen and cathodoluminescence were still a commonly used technique (as an output screen) in image-intensifiers until very recently (Barbin and Poulos, 2002).

In yet another branch, very close to radioluminescence, Alexandre Becquerel (Henri Becquerels father), published a paper on crystals “glowing in the dark” (phosphorescence). Henri Becquerel picked up this research and continued (Becquerel, 1866). He extended the research to among other phosphors, to ZnS . The scientist Sidot studied originally these phosphorescent ZnS (zinc-blende) crystals, which thereafter were called “Sidot’s blende”. A useful application of ZnS was developed by Sir William Crookes in 1903. The so called Spinthariscopes. Essentially

this was a microscope connected to a ZnS-screen where scintillations were observed (and counted) ocularly. This was followed by Erich Regener who used the Spinharscope to record alpha particles of Polonium (1908). Ernest Rutherford and his co-worker & Thomas Royd published a paper in 1909 describing their experiments using the method to gain evidence for alpha decay. Not until 1944, was an improvement made when Curran and Baker connected the recently developed photo multiplier tube (PMT) to the scintillator. The first modern scintillation detector was introduced (Kolar and Den Hollander, 2004).

ZnS:Ag also turned up in an application of phosphorescence when Radium-228 and Radium-226 were used to excite the phosphor for dials in clocks and other instruments. Lacking awareness of the hazards surrounding this painting process, the application went on from 1913 until 1950 (Sharpe, 1978).

There was an increasing demand to be able to detect high energy gamma-rays during the 1930's in the wake of research in nuclear physics. In response to this demand single crystal scintillators were designed as large blocks. In the late 1940's, Tl-doped NaI and CsI, were the primary materials made for this purpose and become widely used – and still are - because of their high extrinsic efficiency (Budinger, 2014). They have now been replaced in certain areas (PET and CT) by ceramic scintillators which were initially developed in the 1990's and based on polycrystalline (powder) phosphor materials (Wojtowicz, 1999). CsI is a common material in imaging detectors in diagnostic radiology. This is due to the fact of the successful change in the internal structure of the scintillator (Nagarkar et al., 1998). This was developed to counter the negative effect on resolution when increasing the radiation sensitivity by increasing the thickness.

1.3 Phosphors and Scintillators

This section introduces definitions of luminescence, later narrowing into the luminescence in phosphors and scintillators. The section includes a so-called Jablonski diagram which theoretically explains luminescence from excitons in general (Figure 2).

1.3.1 Definitions and theory

Luminescence

is defined as any emission of light not related to an emitting body's temperature (i.e. "cold light"). Luminescence can be further divided into the processes of *Fluorescence* and *Phosphorescence*, which is described more in detail further down in this section. The general term for energy of electromagnetic radiation (visible light in this context) is Radiant energy (J). The emitted Radiant energy per unit time is denoted Radiant flux or sometimes Radiant power ($J/s = W$). In this thesis, the theoretical parts implicitly refer to this terminology but once measurements or other photometric conditions are at hand, light is denoted as luminance (light apparent to standard human eye

sensitivity spectrum, i.e. C.I.E). Luminance is measured in the unit $\text{cd}\cdot\text{m}^{-2}$ (see figure 1).

Figure 1. Luminance is used to characterise emission or reflection from flat, diffuse surfaces. Luminance levels indicate how much luminous power is detected per unit surface and solid angle, by the human eye (Figure adapted from Wikimedia)

https://commons.wikimedia.org/wiki/File:Luminance_sch%C3%A9ma_Louvain.png

Physics of luminescence

Electronic band structures can be defined in molecules or crystals where scintillations may occur. When ionising radiation excites an electron from the valence band to either the conduction or exciton band (an energy level situated just beneath the conduction band); (see figure 2), a corresponding hole is created in the valence band. If there are impurities present, (also denoted *activator* or *dopant*) these may create further energy levels in what is known as the *forbidden* gap.

There may exist several energy levels for a phosphor or scintillator defined by various combinations of orbit and spin states. These are known as singlet states, (here denoted S_0 , S_1 , S_2 , in the figure 2.) and triplet or intermediate states, denoted T_1 and T_2). These can in turn be divided into further energy levels.

Figure 2. A Jablonski diagram showing an overview of the processes involved in radioluminescence. See text for detailed explanation (from Lindström, 2011).

Fluorescence

When a scintillator is struck by ionising radiation it is excited from the lowest energy level, denoted S_0 , to the excited level – known as the *singlet state*, S_1 . At this level there is a battery of choices for various processes to take place. If there is a return to the initial state by a photon emission it is defined as *fluorescence*. The lifetime and consequently the decay time of the excited singlet state, is c:a 10^{-9} to 10^{-7} s. The portion of crystals or molecules that are fluorescing, is defined as the *quantum efficiency*. The emitted quantum energy of the fluorescence is *lower* than the quantum energy absorbed by the crystal. This is due to so called *vibrational relaxation* (see figure 2). This change is referred to as the Stokes Shift where the photon energy always shows a shift to *longer* wavelengths (lower energy), relative to the absorption spectrum.

Phosphorescence Photoemission from the transition of a triplet (T_1) excited state and to a singlet ground state, S_0 , (or between any two energy levels that differ in their respective so-called spin states), is denoted *phosphorescence*. The average lifetime and consequently decay time, for phosphorescence can be from 10^{-4} up to $\sim 10^4$ s. The phosphorescence process also has a lower energy for the emitted photons than for the corresponding fluorescence in the same scintillator.

Phosphor A substance that possesses the phenomenon of *luminescence*. This includes both phosphorescent and fluorescent materials. Phosphors are (inorganic) transition metal compounds or rare earth compounds of diverse types. Organic materials are usually not denoted phosphors.

Scintillator is any material that scintillates which is a property of luminescence when exposed to ionising radiation. *Luminescent* materials are characterised by re-

emitting (a fraction of) the imparted energy as light. Scintillators can be *both* of organic and inorganic origin.

Phosphors and scintillators have historically been divided into distinct categories where phosphors usually are polycrystalline materials with varying particle size and scintillators are referring to bulky, mono-crystal luminescent materials. Some authors still make this distinction. With the advent of ceramic techniques where polycrystalline phosphors are essentially manufactured to create near mono-crystalline properties, this distinction has no real practical meaning. Particularly since monocrystal scintillators are available in polycrystalline versions. Therefore, phosphors will sometimes be termed “scintillators” throughout this work. However, monocrystal materials will always be referred to as scintillators (not phosphors). (Lempicki et al., 2002; Gorokhova et al., 2005; Ayvacilı et al., 2014)

1.3.2 Properties

There is a vast scientific literature on scintillators and the importance of various qualities may vary depending on application.

Extrinsic efficiency

The extrinsic (absolute) efficiency, N , of a phosphor slab is defined as the ratio of the light energy per unit area of the phosphor layer surface, Λ (Wm^{-2}), to the impinging energy fluence rate $\dot{\Psi}_0$ (Wm^{-2}) of normally incident photons (Paper I and II: Lindstrm and Alm Carlsson, 1999; Lindstrm et al., 2020)

$$N \equiv \frac{\Lambda}{\dot{\Psi}_0} \quad (1)$$

This parameter is known under many names in the literature: *conversion efficiency*, *x-ray efficiency*, *light yield*, *light output conversion efficiency*, *the luminescence efficiency* (LE), *sensitivity* (also used as a synonym in this work) etc. It is therefore important to actually check the context since the parameter is sometimes confused with the next term:

Intrinsic efficiency

The intrinsic efficiency, η , is defined as the efficiency of the process of conversion to light energy from the imparted energy of ionising radiation of the luminescent material.

Many tabulated values of η are based on the so-called cathodoluminescent power efficiency (Alig and Bloom, 1977). Hence the “c” index. This can be expressed as

$$\eta_c = S \cdot \frac{E_{ph}}{\bar{W}} \quad (2)$$

where $\bar{W}$ is the average energy imparted to the phosphor per electron-hole pair created, E_{ph} is the energy of the emitted light photons and S the probability of a

photon emitted when an electron-hole pair recombines. The term “cathodoluminescent” means that the ionising radiation are electrons. The short range and total energy impartation of the electron energy in the phosphor, are thought to produce an extrinsic efficiency that is approximately equal to the intrinsic efficiency for an optically thin sample (where optical losses can be neglected). This approximation has been utilised to produce tabulated values of η for most radioluminescent materials.

Material characteristics

Some standard properties to categorise (inorganic) phosphors/scintillators for various applications are (Lindström, 2011):

- *Afterglow*, sometimes denoted persistence
- *attenuation coefficient* (and *stopping power* when impinging radiation is electrons)
- *decay time*
- *effective efficiency*: phosphor emission spectrum and photo-detector sensitivity spectrum matching,
- *hygroscopicity*
- *linearity of light response* with imparted energy and rate
- *material stability*
- *spatial resolution* of an imaging phosphor layer

Afterglow is *not* equivalent to decay time. Unfortunately, these terms are often mixed up in the literature but in this work, we will make the distinction between decay time as in fluorescence and afterglow (time) as in phosphorescence. The afterglow is usually not strictly quantified due to its inherent dependence on environmental factors such as temperature. Afterglow can sometimes be denoted “persistence”. (It should be noted though that some materials exhibiting long persistence do so through something called “delayed fluorescence”. This process is often initiated through some additional exciting process). The afterglow of phosphorescence can be partially quenched by introducing so called “killer” impurities of metal in the phosphor (Uppal, Cahturvedi and Nath, 1987). Such as Ce, Cu, Fe, Ni etc.

The x-ray **attenuation coefficient** of a given thickness of a material depends on its density ρ and atomic number Z .

Typical **decay times** are given in databases available from manufacturers and from measurements in the literature. These are defined as the time passed for the luminance to decrease to $1/e$ or $1/10$ of the initial luminance subsequent to an excitation. The decay time of a fluorescent material is an essential parameter for its characterisation and potential application in imaging and detection devices. The lower the time value, the “faster” the phosphor and hence the material can be used in applications, such as

computed tomography (CT) were fast changes in energy fluence of transmitted x-ray beams needs to be handled (Nakamura, 1996)

Effective efficiency It is not only important to choose a scintillator exhibiting a high extrinsic efficiency but also to match the emission spectrum to the spectral sensitivity of the photodetector. 100-450 nm (UV–blue) is the optimum for a photomultiplier tube (PMT) and the 530-580 nm (green–red), for a photodiode (Si). The match is given in percent (%) (Cavouras et al., 1998).

Hygroscopicity of some scintillators, limits the applications to sealed containers. (Examples are NaI:Tl, CsI:Na, LaBr₃:Ce). “Hygroscopicity” is a general term describing materials that readily take up moisture in a non-structured way. The scintillation process can be severely perturbed in the presence of humidity. (Yang et al., 2014)

Linearity of emitted light to incident x-ray energy fluence and/or energy imparted. Some scintillators show a non-linearity response in luminescence and this is attributed to non-homogeneous spatial distribution of the imparted energy. (Ferreira et al., 2004).

Stability of materials describes the potential changes in performance due to the imparted energy and the creation of so-called dead layers inhibiting a lower conversion efficiency. It has been observed in mono crystal scintillators, and notably in scintillators used for cathodoluminescence (Abrams and Holloway, 2004). In many applications, a low temperature dependence is also advantageous contributing to a good stability of the performance of the phosphor (Ajiro et al., 1986).

Spatial resolution in imaging applications - is determined mainly by the geometry and morphology of the phosphor layer itself, i.e. thickness, particle size, contaminants, properties of the binder material, reflective backing etc. Measuring the particle size retrospectively is next to impossible for a scientist without specialist knowledge. Therefore, the particle sizes stated by the manufacturer have been treated as correct in this work, within any uncertainties given. There is a substantial description on the shape, size and how to control these parameters given in the Phosphor Handbook (Ed: Shionoya, Yen and Yamamoto, 2006).

1.3.3 Common phosphors and scintillators

Polycrystalline (powder) phosphors

Summary of characteristics of common phosphor materials is given in Table 1. Data from Ludwig and Prener, 1972; Blasse and Grabmeier, 1994; Moharil, 1994; Nikl, 2006; Yanagida, 2018.

Table 1. Summary of some important properties of selected phosphor materials

Phosphor	Density (g/cm ³)	Decay time (ns)	Intrinsic efficiency (%)	Emis. max.(nm)	Afterglow
ZnS:Ag	3.9	~1000	17-20	450	Very high
CaWO ₄	6.1	6×10 ³	5	420	Very low
Gd ₂ O ₂ S:Tb	7.3	6×10 ⁵	13-16	540	Very low
Gd ₂ O ₂ S:Pr,Ce,F	7.3	4000	8-10	490	Very low
LaOBr:Tb	6.3	~10 ⁶	19-20	425	Low
YTbO ₄ :Nb	7.5	~2000	11	410	Low
Lu ₂ O ₃ :Eu	9.4	~10 ⁶	~8	611	Medium
SrHfO ₃ :Ce	7.7	40	2-4	390	?

1.3.4 Dead layer perturbation

So called dead layers are defects appearing on the surface of phosphor particles. Dead layers decrease the intrinsic efficiency (and consequently the extrinsic efficiency as well) and there is no strict definition what specifies a dead layer apart from a lower than expected conversion efficiency compared to the unperturbed material. They may originate from the manufacturing process and are then usually stable with time. In this work they will be denoted **intrinsic** dead layers differing from the dead layers appearing at the surfaces of a phosphor or scintillator slab. These dead layers, on the other hand, may also originate from intrinsic properties but will deteriorate with time due to chemical reactions (i.e. hygroscopicity, oxygen etc. See also sec 1.3.2 Properties - Stability). The latter type of dead layer will therefore be denoted **extrinsic**. Various measures have been suggested to try to eliminate these dead layers and particularly in cathodoluminescence applications, this has been a known and sometimes severe problem. Intrinsic dead layers are also thought to play a role in the particle size dependence observed from phosphors (Kuboniwa et al., 1973).

Figure 3. Adapted from Paper III (Lindström et al., 2021) Illustration of the two dead layer types.

1.4 Modelling phosphors

Phosphor layer optimisation for medical x-ray imaging purposes, involves the choice of type, thickness, and particle size. High extrinsic efficiency is only one feature of an optimised fluorescent layer and must be considered together with properties such as noise and resolution. Contradicting requirements, i.e. high spatial resolution (thin screen) and high x-ray attenuation (thick screen) are illustrated below in figure 4 and 5. (Adapted from Lindström, 2011).

Figure 4. Illustration of trade-off between resolution and extrinsic efficiency (sensitivity) of phosphor-based detector system when varying thickness. Increasing thickness from A to B. Everything else kept constant. (Lindström, 2011)

Figure 5. Illustration of trade-off between resolution and extrinsic efficiency (sensitivity) of phosphor-based detector system with varying mean particle size. Going from C to D, increasing particle size. Everything else kept constant (Lindström, 2011)

Modelling phosphors and scintillators from many approaches has led to an extensive body of scientific literature on the subject. Two major approaches are described superficially in the next section.

1.4.1 Two-flux theories

Light transport in matter has been studied extensively and Schuster (1905) is often regarded the starting point. Later on, Kubelka-Munk (K-M) (1931) derived a set of optical transport equations for a layer of paint (Kubelka, 1931). This model became widely used and is today still regarded as the standard model in printing industry. Hamaker (1947) developed these transport equations further. By utilising the effects of multiple scattering, he postulated an isotropical scattering of the light in the medium. From this, two directions can be used to describe the optical transport, hence the denotation “two-flux”-theory. Ludwig, using Hamaker’s transport equations, then focused on the special case of radioluminescence (Ludwig, 1971). (This was later known as the “Hamaker-Ludwig”-model in the scientific literature). Ludwig derived an expression for the so-called light transmission efficiency (LTE) of a phosphor layer. This describes the fraction of light produced from the imparted energy from the ionising radiation reaching the phosphor surface. The LTE depends on the optical properties of the phosphor layer, i.e., light absorption, light scattering, and reflectivity (boundary condition). These optical parameters are denoted σ , β and ρ and are formulated as follows:

$$\sigma = [a(a + 2s)]^{1/2} \quad (3)$$

$$\beta = [a/(a + 2s)]^{1/2} \quad (4)$$

$$\rho_j = \frac{1-r_j}{1+r_j} \quad (5)$$

The parameters a and s are light absorption and light scattering coefficients for the phosphor and depend on the properties of the phosphor layer. The parameter r_j is a boundary condition describing the light reflectivity at the screen entrance ($j=1$) and exit ($j=2$) and depends on the refractive index of the materials at the interfaces and any reflective coating at the opposing end of the phosphor layer. The σ parameter is obtained by fitting an expression for the extrinsic efficiency to experimental data (eg. Kandarakis et al., 1997). β and r_j are determined by a demanding series of measurements. This means that four input parameter values are needed and then inserted in relatively complicated theoretical expressions. (Ludwig, 1971)

1.4.2 Mie theory and Monte Carlo simulations

Another approach is through Mie theory and Monte-Carlo methods. Mie theory is an analytical solution to the Maxwell equations describing the propagation of electromagnetic waves and the light scatter from spheres (Mie, 1908). The step to apply this theory on phosphor and scintillator materials therefore makes it possible to estimate the optical parameters. The Mie theory includes the complex refractive indexes connected to the light interaction in the phosphor layer. These indexes are indirectly functions of the particle size and the wavelength of the emitted light. By using Monte Carlo methods, the phosphor specifications can then be simulated. This

is a powerful tool meaning that the various optical input parameters do not have to be measured. Despite this, there are drawbacks of the approach, i.e. the values of the optical parameters are based on spherical particles and the refractive index has a large uncertainty in the imaginary part. Furthermore, the optical absorption is limited to the phosphor host material. Finally, the overall equations are relatively complex and the Monte Carlo calculations simulating the optical parameters of a scintillator, takes time to process.

1.5 Special case: radioluminescence applications in quality assurance

The light field of an x-ray equipment illuminates an area of the patient which will be exposed by the x-ray field. Any misalignment between the two fields, may result in an unnecessary patient dose due to image retakes when there is missing diagnostic information in the x-ray image. The coincidence of the light field to the radiation field is therefore regularly checked in Quality Control programs.

Most national and international standards allow a maximum sum of misalignments of 2% of the SID (Source to Image detector Distance) between the light and radiation field at two opposing field sides. It is a tolerance level valid for both conventional radiology and mammography. However, in mammography, there is a special demand for the breast support edge: IEC (International Electrotechnical Commission) demands a maximum 2 mm deviation between the physical edge and the radiation field. (IEC 2009; IEC 2011)

In the task of checking the light field - radiation field coincidence, there are two radioluminescence applications, (i.e. quality control devices) described in this work. The proposed model has been used for checking the level of optimisation of the contained radioluminescent layers in the devices.

The two devices are a fluorescence-phosphorescence based field position analyser (FPA) (unpublished), and, a device based on a one-dimensional, Linear Imaging Sensor (LIS)-camera. The FPA is established in routine QA-work whereas the LIS-device is still in the prototype stage.

1.6 Aims and framework

Most models use optical absorption and scattering parameters to describe the propagation of light in phosphors. Together, these are commonly referred to as “optical attenuation”. These parameters are unique to the test material and show an indirect effect of particle size. A specific drawback is that measuring or calculating these parameters is not trivial (see section 1.4 on the Hamaker-Ludwig model and Mie-scattering). The presented model was developed without using conventional optical parameters and particle size was introduced into expression as an independent parameter. The input parameters can be obtained by measurements with standard instruments, which are likely to be found in a department of Medical Physics. The

model can be used to calculate changes in the extrinsic efficiency by simulating different thickness-particle size combinations and to find the optimal trade-offs. The model shows that a discrete approach can be fruitful, depending on the context.

- Hence, the major aim was to develop a model supporting the process of optimisation by treating the particle size as an independent variable thus facilitating model results covering an ensemble of varying thicknesses and size. The suggested model is using input data obtained from measurements utilising standard equipment normally found in a medical physics department. (Paper I and II; Lindström and Alm Carlsson, 1999; Lindström et al., 2020)
- Another aim is to investigate the perturbation of dead layers for the intrinsic and extrinsic efficiency of phosphors. (Paper III; Lindström et al., 2021)
- Finally, the last aim of this work is to describe two radioluminescence based applications. They are both devices for quality controls of the light field radiation field coincidence in a diagnostic radiology department. The level of optimisation of the phosphor layers of the devices, has been assessed using the LAC-model (Paper IV; Lindström et al., 2020b)

2. MATERIALS AND METHODS

2.1 The LAC-model

The proposed model is described in paper I, II and partly in III. Highlights of the approach will be described here. The approach of the LAC-model is concluded in figure 6.

Figure 6. The discrete approach of the LAC-model is illustrated where foremost, the particle size diameter, represented by ΔL , is preserved throughout the model calculations. L is the total thickness of the phosphor layer and n , the number of sub-layers consequently derived from the relationship. Figure from Paper I (Lindström and Alm Carlsson, 1999)

The particle size and thickness were to be preserved in the wanted model and indirect representation through other (complex) expressions or factors, were to be avoided. This led eventually to the assumptions described in the next section.

2.1.1 Basic approach and assumptions

Firstly, it was decided that the proposed model would describe phosphors and scintillators comprising a homogenous slab. As in the Hamaker-Ludwig model (1971), the LAC-model assumes that we only need to concern ourselves with two directions. Hence, the area of the slab is treated as infinite. Also, even though presumed homogenous, the phosphor layer is assumed to still be describable in discrete modules, i.e. “unit cells”. For a polycrystalline (powder based) phosphor this meant the individual particles.

Secondly, it is assumed that all of the individual phosphor particles are perfect spheres with the same diameter d . This is not true in real life, where there is a distribution of the particle size. To simplify for the development of the transport equations, the particles are considered ordered in a matrix configuration at a distance x from each other, i.e. $\Delta L = d + x$. In a square packed structure, $\Delta L = d$. (In reality, the structure

is closer to a so called “random close packed structure”) (Dullien, 1992). The sublayers defined in this way are numbered $i \in [1, n]$ in discrete steps to the last layer n . The first layer ($i = 1$), is defined as the entrance layer of the incident ionising radiation.

Figure 7. Illustration of square packed structure of phosphor particles

2.1.2 Energy imparted from ionising radiation

The phosphor layer is regarded as a “thin target” when it comes to the energy fluence of the ionising radiation propagating through the sublayers. This means that the average energy will not change during the process. We have now the tools to describe the energy imparted $\Delta\epsilon$ in a sublayer i :

$$\Delta\epsilon_i = \Psi_0 \left[\exp\left(-\left(\frac{\bar{\mu}}{\rho_c}\right)(i-1)\Delta L\rho\right) - \exp\left(-\left(\frac{\bar{\mu}}{\rho_c}\right)i\Delta L\rho\right) \right] \quad i \in [1, n] \quad (6)$$

where ρ (g cm^{-3}) is the packing density of the phosphor and $\bar{\mu}/\rho_c$ ($\text{cm}^2 \text{g}^{-1}$) is the average mass attenuation coefficient of the phosphor material weighted over the x-ray energy spectrum of the energy fluence rate, Ψ_0 , of impinging photons. Also, any K-fluorescence produced is obviously disregarded in this expression (Paper I; Lindström and Alm Carlsson, 1999).

2.1.3 Light production and optical transport

Quite the contrary to the energy imparted calculations, the overall phosphor layer is considered optically “thick”. This means that the conditions for an isotropic light distribution are fulfilled. In reality, an anisotropic distribution is often encountered for individual light scattering events and that can be described by a so called Henyey–Greenstein function (Binzoni et al., 2006). In the LAC-model, two basic assumptions are made for the light emission and optical transport:

Firstly, the light energy, Λ_i , produced in the sub-layer i is assumed to be proportional to the energy imparted, $\Delta\epsilon_i$, in this location, i.e.,

$$\Lambda_i = \eta \Delta \varepsilon_i \quad (7)$$

where η denotes the intrinsic efficiency (IE) of the phosphor. (IE is an unitless parameter). IE expresses the fraction of the imparted energy from ionising radiation converted to light energy. In this context, however, it is taken a step further. Since we are dealing with *discrete* target objects (spheres) the intrinsic efficiency is defined as the fraction of the imparted energy per unit volume, converted to light energy per unit volume (of the particle). This implies that the intrinsic efficiency is assumed to be *independent* of the thickness of the layer i.e. independent of the particle size. This also implies (indirectly) the absence of (intrinsic) dead layers on the surfaces of the phosphor particles. (We will return to this in later sections).

Light is produced and leaves the phosphor particles. Based on the assumptions of a laterally infinite phosphor layer and an isotropic light emission, we assume, that the produced light has two possible (vector-) directions:

towards the entrance and exit surfaces of the phosphor layer slab seen from the impinging ionising radiation.

Observing these surfaces are described as “modes”. When the entrance surface is the subject then it is denoted the *reflection* mode and consequently studying the exit surface, at the opposite side of the slab, is called *transmission* mode.

When the fluorescence light is passing through its neighbouring particles on the way to the surface, it is assumed in the LAC-model that the particles do not introduce any optical losses, i.e. they are perfect light guides. Obviously, there is an optical loss during the optical transport somewhere. In the model, for convenience, this can be associated with the light crossing in *between* the particles.

Figure 8. Illustration of the light propagation from one sub-layer to the next in the model phosphor. (Paper I: Lindström and Alm Carlsson, 1999)

The optical loss when the light is going from one sublayer to the next (from i to $i-1$ in figure 8) can be described as a unitless, fixed fraction in terms of an extinction factor, ξ . It is defined as:

$$\xi \equiv 1 - \frac{\Lambda_{i,i-1}}{\Lambda_i} \quad (8)$$

The model does not describe the optical loss in terms of absorption, scatter, or refraction. Instead **all** optical losses are handled in this single parameter.

The value of ξ , the extinction factor, depends on the build of the phosphor layer; i.e. choice of phosphor, binder material, packing density etc. The extinction factor has to be determined empirically from layers of different thicknesses but for the **same** build and design of the phosphor layer. (See next section 2.1.4 for details on this procedure)

Once determined, the value is then thought to be valid for most practical particle size and phosphor layer thickness combinations which is a unique feature of the LAC-model compared to other models where unique optical parameters have to be determined for each new case.

We will now turn to the *reflection* mode and describe the luminance contribution at the phosphor layer (entrance) surface from one excited particle in layer i :

$$\Lambda_{i,s} = \Lambda_i (1 - \xi)^i \quad (9)$$

The light passes through i layers until it reaches the surface, losing a fraction ξ , every time it passes a sub-layer. The luminance contribution at the surface is therefore $\Lambda_{i,s}$ where index s denotes the surface.

We may now derive an expression for the total light energy (per unit area), Λ , at the phosphor layer surface from all sub-layers and by inserting equation 7, we obtain:

$$\Lambda = \sum_i^n \Lambda_{i,s} = \sum_i^n \Lambda_i (1 - \xi)^i = \eta \sum_i^n \Delta \epsilon_i (1 - \xi)^i \quad (10)$$

2.1.4 LAC-model equation: extrinsic efficiency

In equation 1 the extrinsic efficiency was defined and by combining this equation with equation 6 yields,

$$N \equiv \frac{\Lambda}{\psi_0} = \eta \left(\exp \left(\left(\frac{\bar{\mu}}{\rho_c} \right) \frac{L}{n} \rho \right) - 1 \right) \sum_{i=1}^n \exp \left(\left(- \left(\frac{\bar{\mu}}{\rho_c} \right) \frac{L}{n} \rho \cdot i \right) (1 - \xi)^i \right) \text{ where } i \in [1, n] \quad (11)$$

Eq. 11, (Lindström and Alm Carlsson, 1999) is the expression for the extrinsic efficiency in the reflective mode. Correspondingly the expression for the *transmission* mode is given in equation 12 (Lindström et al., 2020).

$$N = \eta \left(\exp \left(\left(\frac{\bar{\mu}}{\rho_c} \right) \frac{L}{n} \rho \right) - 1 \right) \exp \left(-(n+1) \left(\frac{\bar{\mu}}{\rho_c} \right) \frac{L}{n} \rho \right) \sum_{i=1}^n \exp \left(+ \left(\frac{\bar{\mu}}{\rho_c} \right) \frac{L}{n} \rho \cdot i \right) (1 - \xi)^i$$

where $i \in [1, n]$ (12)

Note that Λ is the total light energy observed for either side depending on the chosen mode. Normally, opposite side is coated with a reflective layer in most applications. Implied in the expression 11 and 12, is that any real-life distribution of particle size should produce the *same* extrinsic efficiency as for the average particle size in the phosphor layer. This is denoted the *efficiency equivalence principle* (Lindström and Alm Carlsson, 1999). See figure 9.

Figure 9. Illustration of efficiency equivalence principle. Sample to the left have the same extrinsic efficiency as the ordered matrix sample to the right where all particles have been replaced by the average particle size (and distance)

To obtain the extrinsic efficiency for a series of phosphor samples of the same design, the extinction factor, ξ , has to be determined experimentally. A procedure may look like this:

- (I) Λ is measured in a fixed geometry for screen samples of different but known particle sizes and thickness combinations;
- (II) One screen sample is chosen as a reference. Λ , is calculated using the Eq.s 11 (or 12), together with an initial guess of the value of the extinction factor, ξ ,
- (III) This calculated value of Λ is then normalised to the corresponding measured value for the reference screen.
- (IV) The normalising factor is then multiplied to other samples in the series
- (V) The parameter, ξ , is then used as a fitting factor and is varied until a best fit is obtained between calculated and measures values for the studied series.

2.2 Assessment of LAC-model

2.2.1 Measurements: set-up and geometry

(A detailed description of the assessment is found in Paper II; Lindström et al., 2020). To assess the model, a very common phosphor was chosen: $\text{Gd}_2\text{O}_2\text{S:Tb}$. The properties of this phosphor are thoroughly investigated in the scientific literature and it is regarded as an excellent x-ray to light converter due to its high intrinsic efficiency (0.15-0.2), high Z (Atomic number, Gd (64)), consequently a high density, 7.34 g/cm^3 . Furthermore, the decay time is sufficiently short for many applications, $600 \mu\text{s}$. The emission spectrum has a prominent peak in the visible range: 545 nm. Another feature of the phosphor is the shape of the particles themselves, i.e. they are usually spherical. There are also versions of the phosphor, using the host material, such as $\text{Gd}_2\text{O}_2\text{S:Pr}$ (ceramic scintillator) seen in some Computed Tomography equipment (Nakamura 1996, Lempicki et al., 2002; Gorokhova et al., 2005; Ayvacikli et al., 2014)

Screens were then in-house manufactured from $\text{Gd}_2\text{O}_2\text{S:Tb}$ polycrystalline powder of 7.5 and $20 \mu\text{m}$ average particle size. (The details of the manufacturing process can be found in the licentiate thesis, Lindström 2011). Due to the high-quality demands, only a few screens passed the quality controls following the manufacturing process. In total four, these were:

Table 2. Manufactured test screens

Screen	Particle size, ΔL (μm)	Phosphor layer thickness L (μm)	Number of sub-layers, $L/\Delta L$
A	7.5	400	53
B	20	220	9
C	20	420	17
D	20	830	33

All screens had a 0.1 mm transparent protective coating at the surface and a TiO_2 induced PVC-backing (for enhanced reflectivity).

The next step was then to set-up the geometry for the assessment of the screens and choose equipment suitable for the job.

Some prior testing showed that the mode of fluoroscopy was preferred since there were to be steps of manual surveillance in the assessment process. Ordinary exposures produced higher output than for the fluoroscopy mode, but their short time span made it next to impossible to measure the corresponding produced light from the screens to any high degree of accuracy. This was due to the light measuring equipment available: a Hagner Photometer (Universal Photometer Hagner, Solna, Sweden). It measures luminance adapted to the human eye response curve (C.I.E). That meant that it did not measure the total light energy produced from the screen surface. Knowing the light emission spectrum of $\text{Gd}_2\text{O}_2\text{S:Tb}$ and the sensitivity curve of the

Hagner photometer, it was initially thought useful to convert the luminance values to true light energy. However, the main point was to show how a reference luminance value changes according to the LAC-model for a fixed exposure and varying particle size and thickness combinations. Therefore, it was decided to instead aim for the best possible reproducibility of the measuring conditions and focus on the relative changes in luminance from the screens samples.

Eventually, a Varian Ximatron, Palo Alto, USA, x-ray equipment (therapy simulator) was chosen for its relatively high output in fluoroscopy mode and mechanical stability. The x-ray generator was checked using a non-invasive x-ray multimeter and found to be within standard requirements. A KAP meter (Kerma Area Meter, PTW Freiburg, Germany) (see figure 10) was put on the collimator of the x-ray tube assembly (1) to check the stability of the output during fluoroscopy. The x-ray focus to phosphor sample distance was set to 1 m. The luminance, L_{eff} (cd/m^2) was measured using a photometer (Universal Photometer Hagner, Solna, Sweden) angled at 45 degrees from the central axis beam of the ionising radiation at a fixed distance of 30 cm from the phosphor sample. The photometer was mounted on a camera stand. The screen samples (A-D in table 2) were cut to fit squares of about $60 \times 60 \text{ mm}^2$. The screen sample to be measured was then centred in the central beam and the field size was collimated to $40 \times 40 \text{ mm}^2$. To reduce scatter, an extra collimation was added at the screen plane, comprising lead sheets bordering to the indicated field.

As seen from the figure 10, all screens were sequentially measured in the reflective mode. There were two reasons for this: 1) the luminance from the screen samples in the transmission mode was deemed too low when checking and that was for the maximum settings of the generator. 2) the Hagner is equipped with a photodiode for the luminance measurements. These are known to be susceptible to scattered radiation in some circumstances. To minimise the risk for x-ray perturbations, the transmission mode was not considered further. The possible interactions of x-rays with the photometer (the photodiode) due to its proximity to the beam, was checked in *reflectivity* mode by alternately placing a black, opaque sheet, replacing the screen samples. Scattered x-rays could not be detected. The geometry was now set and the luminance of the four phosphor samples (screen A-D) was measured using the fluoroscopy mode with the same generator settings throughout the whole session. (100 kV_p and 6 mA). Knowing that some phosphors have an efficiency dependence on temperature, the environmental temperature was recorded throughout the session (21 °C). No variations were noted during the measurements. In the geometry set-up it is implicitly assumed that the screens have a representative and identical light distribution on the emissive surface. This is known to be an approximation (Kandarakis et al., 2006).

Each screen was exposed by fluoroscopy for approximately 10 seconds and the luminance was simultaneously measured. This was repeated five times for each screen sample. The KAP values were monitored and if the KAP-value was determined to vary less than 2% of the mean value for the sample series, the calculated mean value was approved. The mean luminance value of the five measurements was then normalised by the mean KAP-rate value.

To maintain a good repeatability standard, all measurements, were made in one session. No TP-corrections were applied to the KAP-meter as the corrected, absolute value, was not the focus in this relative measurement.

Figure 10 (Adapted from Paper II: Lindström et al., 2020) Experimental set-up (reflection mode) for measurement of the luminance change (relative extrinsic efficiency) (see text for explanation) of a phosphor screen sample. Field-size at screen surface; $4 \times 4 \text{ cm}^2$

2.2.2 Monte-Carlo simulation of energy imparted

The LAC-model has a number of simplifying assumptions. One is the assumption of a constant mean energy of the x-ray spectrum transmitted through the phosphor layer. Another is ignoring any possible dosimetric interface effects between the entrance and exit surfaces of the phosphor layer and the surroundings. It was also well-known that Gadolinium introduced a K-edge at 50.2 keV which means that for most conventional and CT x-ray spectrum there will be a K-fluorescence production in the phosphor layer to an unknown degree (Michail et al., 2018).

Rather than trying to introduce correcting factors in the LAC-model we decided to do Monte-Carlo simulations of the energy imparted in order to do estimations of the impact of the simplifications done in the model. Would the resulting luminance change and by how much?

The calculation of energy imparted was improved by using the MC code PENELOPE (Baro et al., 1995) with the x-ray spectrum from SpekCalc. Details about the use of the code are given in papers II and III (Lindström et al., 2020; Lindström et al., 2021).

2.2.3 Introducing a dead layer in the LAC-model: analysis

Taking a panoramic view of the phosphor research field for a moment, there is an ongoing effort to control the morphology of nanosized phosphor which have a potential of new interesting applications (Kang et al., 2020) where high resolution imaging is also one of them (Jin et al., 2018). This will most probably have an impact also on imaging systems of diagnostic radiology in the near future.

The quest for a controlled morphology of a phosphor, also includes the so called dead layer. From a paper on nano-phosphors by Dinsmore et al. (1999), it is stated: “Some experiments have suggested (and it is commonly assumed) that as particles become smaller, the efficiency decreases because of the increasing surface area, which is thought to contain nonradiative recombination sites or some other kind of “dead layer.””

The intrinsic dead layer of polycrystalline phosphors is thought to decrease the intrinsic efficiency of the individual particle. The origin of the intrinsic dead layer may vary but, in this section, it is assumed to be fixed and not varying with time. Additionally, the dead layer thickness is assumed constant regardless of particle size. This means that the ratio of the dead layer to the particle size will vary accordingly and is the foundation of a suggested explanation of the particle size dependence of the extrinsic efficiency (Kuboniwa et al., 1973). Turning to the LAC-model, an introduction of the dead layer to the individual particles will not change the number of sub-layers of the observed phosphor layers. The number of sub-layer and the consequential inter-crossings are central to the LAC-model optical loss concept as manifested in the extinction factor, ξ . However, by modifying the individual particle by introducing a dead layer, it will inevitably make the LAC-model deviate from the assumption of an identical conversion efficiency per unit volume – i.e. intrinsic efficiency.

Based on the paper by Kuboniwa et al. (1973), the change in luminescent loss can be calculated by introducing a factor $(1-S_A\rho l)$, where S_A denotes the surface-area to volume ratio of the average particle diameter d , ρ is the specific gravity, i.e. the ratio of the density to a 100% bulk material, and l is the average of the dead layer thickness. The dead layer thickness is assumed to be $\sim 0.1 \mu\text{m}$. The work of Kuboniwa et al. is widespread and is referred to in the following examples of papers on extrinsic efficiency (or the equivalent); Nomicos et al., 1978; Giakoumakis et al., 1980; Giakoumakis, 1988; Giakoumakis and Lagaris, 1988; Giakoumakis, 1989. Finally, we assume that the intrinsic efficiency, η , can be modified using the same factor to determine relative changes in the extrinsic efficiency, in the LAC-model.

2.3 Dead layer assessment

The second type of the dead layer, i.e. surface dead layer of scintillator slabs, here denoted extrinsic, is mapped through literature studies on the subject and Monte-Carlo simulations in this section. The object being to show that interface dosimetric irregularities may at occasion appear as a dead layer without excluding the possibility of an existing chemically based dead layer.

2.3.1 Monte-Carlo simulation of apparent (entrance surface, extrinsic) dead layer

The absorbed dose in a medium depends on the fluence and type of ionising radiation energy and the attenuation and scattering properties of the medium. In a region between two dissimilar materials secondary electrons are scattered in both a forward and backward direction. This result in absorbed dose irregularities described by the term “interface dosimetry” (Carlsson, 1973; Regulla and Leischner, 1983; Verhaegen and Seuntjens, 1995; Das et al., 2001). To verify these irregularities through measurements is difficult and therefore Monte Carlo simulation techniques are used instead. These interface regions may give rise to the same lower light production in a scintillator as a dead layer emanating from structural defects. It would be difficult to distinguish the two types from a luminescence measurement alone unless the region types differ in dimensions. Thus, the region behaving like an extrinsic dead layer will be denoted *apparent* dead layer.

The light production is assumed to be directly linked to the energy imparted throughout the scintillator. To map any reduction in energy impartation (and the consequential light production) the energy imparted in the interface region was once again simulated by using PENELOPE (Baro et al., 1995). Input simulation parameters were the source to scintillator surface distance, field size, type, and energy spectrum of the impinging ionising radiation. The x-ray spectrum was obtained from the code SpekCalc (Poludniowski, 2007; Poludniowski and Evans, 2007; Poludniowski et al., 2009) and Siemens homepage (Siemens, 2019). In one case, impinging monoenergetic electrons were to be simulated and could be specified directly in PENELOPE.

It was decided to simulate a set of three common applications and any consequential apparent dead layer. From the literature it was known that the chemically based dead layers could be expected to be of relatively small dimensions compared to total thickness of the studied phosphor layers (Abrams and Holloway, 2004). Therefore, a step size of 0.05 μm (in depth) was considered adequate to be able to catch dosimetric irregularities posing as apparent dead layers. Up to 10^7 histories were simulated per phosphor/scintillator.

Case 1: **Case 2:** **Case 3:**

Figure 11. (Adapted from Paper III, Lindström et al., 2021) Geometries for Monte-Carlo simulations. Case 1 and 2 are x-rays. Case 3 is monoenergetic electrons. Phosphors are homogenous slabs with density of main compound. Parallel beams and field sizes are given at the entrance plane of the phosphor surface. Note: Case 2 geometry also with simulation of photodetector in beam.

2.4 Radioluminescence applications

Routine Quality Control (QC) programs for diagnostic radiology equipment includes the action of checking the light field – radiation field alignment. Misalignment may lead to additional exposures and unnecessary patient doses. Historically, this test was carried out by exposing film but in a modern, digital department, this is performed by using various dedicated devices.

Therefore, two radioluminescence based quality control devices for this purpose, are in the scope of this thesis. One is a fully working, commercial tool (the FPA, a.k.a. “Visi-X”, RTI Group, Mölndal, Sweden). The other, is still in its prototype stage (the LIS-method). Both of the devices are described in Paper IV (Lindström et al., 2020b) and in Lindström et al. 2014.

2.4.1 Field Position Analyser

The general version of the Field Position Analyser (FPA) consists essentially of two PMMA (Polymethylmethacrylate) plates sandwiching a 0.7 mm layer of a radioluminescent phosphorescent polycrystalline phosphor based on a concept originally introduced by Crooks and Ardran (1976). Normally, phosphorescence is minimised in various imaging techniques. In this device, the process is instead

essential. The phosphor is excitable from a wide range of the electromagnetic spectrum (i.e., from infra-red light to high-energy gamma rays) and also of particle radiation (i.e., alpha, beta, electrons, and protons). The sensitivity to visible light introduces a problem and is solved by using a semi-transparent, light filter blocking a substantial proportion of the exciting wavelengths of the ambient light (i.e. $< \sim 580$ nm). The semi-transparency allows use of the field-markings without removing the lid and it is normally removed only when doing the read-out to reduce the risk of excitation from ambient light (see figure 12.)

The FPA is used as follows: the device is placed at a maximum 1 m from the x-ray tube focus. The light field is adjusted to the chosen markings and two to three exposures at 100 kVp and 100 mAs are made. Any deviation between the radiation field and the light field is measured directly using the built-in scales by watching the field defined by the afterglow.

Figure 12. View of the device. The FPA is prepared for an exposure when the light filter is in place for later removal prior to the read-out. (Photo courtesy of RTI Group, Mölndal, Sweden)

The phosphor thickness of the FPA is 200 mg/cm^2 , which also is a manufacturing limit. The thickness influences the spatial resolution of the phosphor, such that the field edges appear blurred when observed on a too thick screen. Using a Hüttner resolution pattern, two line pairs per mm (LP/mm) can be resolved using a 200 mg/cm^2 layer.

The LAC-model can be used to evaluate the optimisation level of the phosphor layer in the FPA. Knowing that a maximum available particle size is already in use, we may focus on the luminance dependence on the phosphor layer thickness.

2.4.2 Field edge measurement device

Another quality control equipment application is a prototype denoted the LIS-device or LIS-method. (Linear Imaging Sensor) (Lindström et al., 2020b). While the device in section 2.4.1 is a full field detector, the LIS-device measures the location of the edges of the radiation and light fields. The device is based on an existing one-dimensional sensor camera board. Originally, it was designed for light spectrum measurements. The light sensor covers a length of about 30 mm and has been of several types during the development process. The sensor comprises small pixel elements ($8\ \mu\text{m}$) in an array (of between 2048 to 3200 pixels depending on version). When the LIS-device is placed in the light field - covering one of the four edges, immediately a profile of the light field edge will be registered. This is stored in a computer and without moving the device, an exposure is made and another profile, the radiation field edge, is detected and stored. Markings or rulers are deemed unnecessary on the LIS-device, since it keeps track on the light field and radiation field edges without further assistance of the user. A software processes the profiles

Figure 13. Photos showing the prototype in use. Light edge profile (upper picture) radiation edge profile (lower). Note the noise present in the x-ray profile.(Adapted from Paper IV: Lindström et al., 2020b)

and localises the edge positions along the sensor length. Any deviation between the edges position is detected, calculated, and presented in mms in the software. This procedure is then repeated until all four sides are checked.(see figure 13 and 14).

The LIS-sensor originally has a low sensitivity to x-rays and is therefore modified using a strip of a $\text{Gd}_2\text{O}_2\text{S:Tb}$ phosphor. The strip is without any reflective layers since the same sensor is also used for the light field edge detection. Accordingly, the strip needs to be sufficiently transparent to the visible light. This is an optimisation challenge and by knowing the properties of the applied phosphor layer we may use the LAC-model to see whether there is room for improvement for the LIS-device in terms of sensitivity to light and x-rays. The particle sizes were $7.5\text{ }\mu\text{m}$ and $20\text{ }\mu\text{m}$ and thicknesses ranged from 100 to $300\text{ }\mu\text{m}$.

The LIS device shows a susceptibility to noise when recording the x-ray profile (see figure 13). To improve the location process of finding the edge in the x-ray profile, a noise suppressing algorithm was used to uncover the underlying profile and eliminate the induced noise peaks.

Figure 14. Basic LIS-method. Close-up, side view of profiles and sensor. Note the phosphor strip covering the linear sensor. Phosphor layer thickness thin enough to transmit light field light but hick enough to produce sufficient light from x-ray energy impartation. 50% (of maximum intensity) edges indicated. Later changed to 25% to better represent the human eye edge detection (see Paper IV: Lindström et al., 2020b).

Being a previously untested method, the LIS-device underwent extensive testing for checking its functionality and uncertainty. (Lindström et al., 2020b). Location of the edges being the central process, the dependence from kVp , mAs (air Kerma), exposure time and repeatability was tested. The device was finally compared to both the FPA (in previous section 2.4.1) and two other established devices and methods for the checking of light field radiation field coincidence. The feature of a Bluetooth connection has been introduced to the prototype version currently in focus.

3. RESULTS AND DISCUSSION

3.1 Extrinsic efficiency comparison

In paper I and II (Lindström and Alm Carlsson 1999; Lindström et al., 2020), extensive calculations and measurements were carried out to test the validity of the LAC-model. Chosen series of model results are presented in table 3. The measured values are luminance values. The first series (phosphor: $\text{Gd}_2\text{O}_2\text{S:Tb}$), is based on screens designed, manufactured, and measured by the author. The other series are taken from the scientific literature and are compiled in paper I (Lindström and Alm Carlsson, 1999).

Table 3. Results from measurements and modelling work (Adapted from Paper 1 and II; Lindström and Alm Carlsson, 1999; Lindström et al., 2020). “Reference” indicating chosen reference screen or layer, for the LAC-model. See section 2.1 for explanation of reference screen. This screen is normally the one showing the highest luminance in a series. The luminance of the *reference screen* is normalised by a factor to be equal to the corresponding *measured* luminance in the same series. (Hence no deviation). This factor is then applied to the remaining calculated screen luminance values. The extrinsic parameter, ξ , is then varied to achieve best fit for the remaining modelled screens to the measured screens of the same series.

Phosphor	Particle size (μm)	Thickness (μm)	Sub-layers	Measured Value	LAC-model	Deviation (%)	Hamaker-Ludwig	Deviation (%)
(Paper II; Lindström et al., 2020)								
Gd ₂ O ₂ S:Tb	7.5	400	53	44.1	44.3	0	N/A	N/A
	25	220	9	79.4	77.3	-3		
		420	17	91.2	94.2	3		
	reference	830	33	100	100	----		
(Kandarakis et al., 1996)								
La ₂ O ₂ S:Tb	7	77	11	7	7	0	7	0
		147	21	12	12	0	12	0
		197	28	16	16	0	16	0
		250	36	19	19	0	19	0
		343	49	23	23	0	24	4
		367	52	24	25	4	25	4
		403	58	27	26	-4	26	-4
		490	70	28	30	7	29	4
reference		540	77	31	31	----	31	0
(Wickersheim et al., 1970)								
La ₂ O ₂ S:Tb	1.5	1000	667	0.8	0.8	0	N/A	N/A
	5		200	1.8	1.9	2		
	15		67	2.8	3.3	18		
	reference	50	20	4.4	4.4	----		
(Nomikos et al., 1978)								
ZnCdS:Ag	2.5	530	212	2.3	2.7	17	N/A	N/A
reference	10		53	10	10	----		

Overall, it can be seen from table 3, that applying the LAC-model retrospectively will introduce larger deviations when not in full command of measurements, phosphor properties and uncertainty estimates. The largest deviation between modelled results and measurements of extrinsic (absolute) efficiency are for the series found in the scientific literature (i.e.: Wickersheim et al., 1970; Nomicos et al., 1978; Kandarakis et al., 1996). It is also notable that the Hamaker-Ludwig model-based series (Ludwig, 1971) ($\text{La}_2\text{O}_2\text{S:Tb}$), produces results close to that of the LAC-model. That would be expected, considering the two models sharing many of the same basic assumptions. Taking the known overall uncertainty into account, $\sim 5\%$ for the extrinsic efficiency of the LAC-model, (Lindström et al., 2020) there is really no practical difference in the result between the models.

For the series of in-house manufactured screens, a Monte-Carlo simulation was done to improve on the energy impartation of the phosphor layer (the optical transport part of the LAC-model was left intact). From this simulation, all the possible interface dosimetry perturbations, K-fluorescence and change of mean energy with depth, were included. Interestingly enough, the original simple model using an exponential energy imparted showed the lowest discrepancy between measurements and model result in this particular case. The largest RMSE error was calculated for the Monte-Carlo energy imparted simulation. At 6.3%, it is still a very acceptable result. The LAC-model does produce results consistent with the given conditions and measurements.

One may argue that the $\text{Gd}_2\text{O}_2\text{S:Tb}$ series (see table 3) does not have that many screens available to check the validity of the LAC-model but by compensating the relatively low number with a smaller uncertainty it is believed that the assessment shows that the LAC-model possess good predictive powers. At the same time, it clearly shows that it is important to keep track on all factors influencing the uncertainty of the calculation of the (relative) extrinsic efficiency. It is also important to point out that the extrinsic efficiency is not measured *per se* in this series, but it is still expected that any shortcomings in the LAC-model would manifest themselves if they were severe.

With the advent of nano sized phosphor particles manufacturing techniques, it also followed that the control of the size distribution has increased (Phosphor handbook, Ed: Shionoya, Yen and Yamamoto, 2006) meaning that the given average particle size is based on a very narrow size distribution. Some of the average particle sizes given in table 3 are based on unknown distributions that may suspected to be larger than those expected today. Furthermore, these distributions may not follow the Poisson distribution implicitly required for the *efficiency equivalence principle* (Lindström and Alm Carlsson, 1999) to be valid. The particle size is crucial for the LAC-model as it determines the number of sub-layers and hence the value of the extinction factor, ξ .

The LAC-model requires investigated phosphor layers to be of the same design. In practice, this is not a problem since the design is usually determined before doing the modelling. Even if it has not been validated it should be pointed out that varying the (packing-)density of the screen should not pose a problem as long as the binder

substance is the same. Changing the “boundary conditions”, i.e. different reflective backing (or no backing), different protective coating etc, for a single screen in a series, make this sample unique and the LAC-model can no longer be applied on this particular screen. There is a possibility to change the boundary conditions in the Hamaker-Ludwig model (Ludwig, 1971) for the same session, but the changed values still need to be known with good accuracy. In practice, this is not thought to be an essential advantage over the LAC-model since the design (i.e. setting the boundary conditions) is usually already determined.

3.2 Dead layer analysis and simulation

3.2.1 Intrinsic dead layer

The relative extrinsic efficiency values are deviating less than 5% between the measured and LAC-model results. Using the factor suggested by Kuboniwa et al. (1973) (see also sec 2.2.3) the reduction in intrinsic efficiency is 8% and 2% for the particle size 7.5 μm and 25 μm respectively, assuming a dead layer of thickness 0.1 μm . A comparison is shown in Table 4 (corrections according to Kuboniwa et al. (1973) are given in column “Model, N_{corr} ”).

Table 4. Relative Extrinsic Efficiency. (Paper III: Lindström et al., 2021)

Screen	Thick- ness (μm)	Particle size (μm)	Measured, N_{meas} (arb.units)	Model, N_{simple} (arb.units)	Model, N_{corr} (arb.units)
1	400	7.5	44.1	44.3	40.9
2	420	25	91.2	94.2	92.0

Introducing a dead layer in the LAC-model have an impact on the calculated extrinsic efficiency. However, the suggested explanation of a luminance change with varying particle size cannot alone be attributable to a dead layer surrounding each particle (type intrinsic). The influence is too small.

3.2.2 Extrinsic dead layer

The Case 1 simulation has been chosen for display and is an interface between air and Gd_2O_3 for 100 kVp x-rays. The simulated distribution of absorbed dose across the interface region of case 1 is seen in figure 15. A “build-up region” of dosimetric origin within the scintillator material is clearly shown in the vicinity of the air-scintillator interface. Values of D_{50} and D_{90} , are the corresponding depths, (usually μm), given by the normalised dose value for 50 and 90 %, of the maximum given dose in the scintillator. D_{50} and D_{90} are used to describe the interface region on the scintillator side for comparison of size with any given chemical dead layer. D_{50} and D_{90} are at approximately 0.2 μm and 1.5 μm depth in Gd_2O_3 . Further, a substantial dose

increase is noted in the air in the proximity of the interface. It has similar origin, and stems from particles created in the $\text{Gd}_2\text{O}_3\text{S}$ being back-scattered into the air.

Figure 15. Dose as function of depth normalised to the maximum dose along central axis. D_{50} and D_{90} are indicated at 0.2 μm and 1.5 μm depths respectively (calculated from underlying numbers in MC simulation (not shown)). The magnified insert shows D_{50} and left column at “0”, indicating interface border. Y-axis: indicating fraction of normalised dose. X-axis: distance from interface border in μm . (Paper III: Lindström et al., 2021)

The results of the Cases 1-3 are summarised in Table 5, where the D_{50} and D_{90} estimated in the simulations of this work are also compared to any given dead layer estimates found in the literature (these are provided in the second last column with the corresponding literature reference in the last column).

Table 5. List of simulated cases 1-3, type and energy of ionising radiation, scintillator material, effective atomic number of the scintillator Z_{eff} , D_{50} and D_{90} as obtained in this work, dead layer estimation from literature with corresponding literature reference. (From Paper III; Lindström et al., 2021)

Case	Impinging Energy	Scintillat or/ phosphor	Z_{eff}	D_{50} μm	D_{90} μm	Dead layer estimate, μm	References
1	100 kVp, x-rays	$\text{Gd}_2\text{O}_3\text{S}$	60	0.2	2	0.15 0.2	Chappel et al., 1970 Izumi et al., 2012
2	28 kVp, x-rays	CsI	54	0.1	0.5	>2 ^{*)}	Goodman, 1976
3	10, 20, 50, 100 keV electrons	ZnS	32	N/A	N/A 0.1, 1, 2.5	0.03	Abrams and Holloway, 2004

*) dead layer estimates are for spectrometer applications only. No imaging detector estimates found in scientific literature.

The results of Monte Carlo simulations of absorbed dose as function of depth in the scintillators of cases 1-3 all show an absorbed dose in the vicinity of the interface air-scintillator that is lower than the maximum dose. A clear dependency on scintillator material and particle type (photon, electrons) and particle initial energy is also seen. This reduced dose will lead to a corresponding reduction in x-ray to light conversion near the interface. The reduction shown here is of a dosimetric origin and not due to a real conversion efficiency ionising radiation to light.

It should be noted that the simulations of the homogenous slabs of the various materials and are not capable of revealing any information on dead layers surrounding individual particles (“intrinsic” of figure 3). Such effects of dosimetric origin can exist when the particles are resolved in a binder or surrounded by thin layers of air, but they are likely considerably smaller than the encountered entrance dead layers (“extrinsic”).

From the results, we may conclude that in most cases of interest for medical *imaging*, interface dosimetry effects are negligible, the phosphor being much thicker than the dead layer itself.

3.3 Applications: assessment of devices

This section starts with the results from the assessment of the phosphor layer used in the devices using the LAC-model and continues with the functionality for the task at hand. Finally, a comparison between the devices is made as well as to other available devices. In the chapter 5, some ideas for the future are discussed for the devices.

3.3.1 Optimisation level of phosphor layer in the Field Position Analyser (FPA)

From equation 11 and the phosphor of the FPA, results are shown in figure 16 for $\xi = 16.5\%$, $\rho_c = 2.2 \text{ gcm}^{-3}$ (50% packing density), $\overline{\mu/\rho_c} = 2 \text{ cm}^2 \text{ g}^{-1}$ (100 kVp x-ray spectrum). The light output is normalised to the maximum luminance of the curve for the $40 \mu\text{m}$ particle size. Arrows indicating thickness ($700 \mu\text{m}$) and relative light output for the commercially applied FPA phosphor screen. (Paper IV: Lindström et al., 2020b). The thickness to luminance ratio of the phosphor layer has a monotonic increase to about $300 \mu\text{m}$ ($20 \mu\text{m}$ particle size) and $700 \mu\text{m}$ ($40 \mu\text{m}$ particle size). Using a Hüttner resolution pattern, two line pairs per mm (LP/mm) can be resolved which is thought sufficient to capture most deviations between radiation and light field. In practice there was no room for improvement to be found; the FPA phosphor is close to optimum within practical limits.

Figure 16. Light output with increasing phosphor layer thickness. (particle sizes; 20 and 40 μm). Arrows indicating thickness (700 μm) for the commercial FPA phosphor screen. (Paper IV: Lindström et al., 2020b)

3.3.2 Optimisation level of phosphor layer in the Linear Imaging Sensor (LIS)-device

The modelling results for the phosphor strip used in the LIS-device ($\text{Gd}_2\text{O}_2\text{S:Tb}$) are shown in figure 17. $\xi = 8.8\%$, $\rho_c = 3.7 \text{ g cm}^{-3}$ (50% packing density), $\frac{\bar{\mu}}{\rho_c} = 8 \text{ cm}^2\text{g}^{-1}$ (100 kVp x-ray spectrum). The light output is normalised to the maximum light output of the curve obtained for 25 μm average particle size. The 100 μm phosphor strip used in the current LIS-method prototype is indicated by an arrow.

Figure 17. Light output change in arbitrary units with increasing phosphor layer thickness shown for two particle sizes; 7 and 25 μm . The currently applied phosphor strip in the prototype is indicated in the diagram (25 μm particle size, 100 μm thickness) (Paper IV: Lindström et al., 2020b)

Thickness (and particle size) of available strips were 100 μm (25 μm); 200 μm (7 μm); and 400 μm (7 μm). The model results yield a maximum luminance at about 200 μm for 25 μm particle size and 100 kVp x-ray generator setting. In practice, due to the

high overall sensitivity of the tried out LIS-sensor and strip combination, the 100 μm thick phosphor was judged to be sufficient.

3.3.3 Functionality of devices

In this section a brief description of the functionality of the devices follows. It should be noted that the first device, the FPA (a.k.a. Visi-X) is an established device (RTI Group, Mölndal, Sweden) and the second, the LIS-device, is under development where the focus is still on encountered technical issues not mentioned here. Another two, commercially available devices, were tested: RaySafe DXR+ (edge detector: Raysafe, Billdal, Sweden) and Gafchromic XR-MR2 (self-developing film strip, edge detector: Gafchromic, Bridgewater, USA). “Edge detector” indicates method where four individual measurements have to be performed for one x-ray field. The functionality test of the devices was configured by checking the following parameters: 1) dose requirements 2) uncertainty in measurements 3) overall set-up and read-out procedure. The LIS-method, being recently developed, was additionally tested for kVp, dose and dose-rate dependence, for the localisation of the edges.

The results have been disclosed and discussed elsewhere (Lindström et al., 2014; paper IV: Lindström et al., 2020b). During the development process of the LIS-device, the phenomenon of focal spot wandering was encountered and to the knowledge of the author, could be quantified for the first time for x-ray diagnostic radiology equipment (Stein, 2012). This was not expected and initially, the phenomenon was not identified correctly but was rather thought to emanate from the sensor physically moving during exposures. The magnitude of the focal spot wandering can be seen in figure 18, where the pixel address of the edge in the radiation field can be seen to move quite dramatically in the beginning (cold tube) to finally come to rest (warm tube).

Figure 18. Edge location results on mammographic equipment. X-axis showing the sequential exposures in numerical order. Y-axis; pixel address of radiation field edge. Opposite Y-axis: pixel address of edge in mm's along sensor entrance plane. Figure from Lindström et al., 2014 and Paper IV: Lindström et al., 2020b.

Below are the results from the comparison and based on this test (Table 6), the LIS-method came out well even if not fully developed. The LIS-method has the lowest dose requirements and the lowest uncertainty.

Table 6. Comparison of devices for light field and radiation field coincidence testing. Parameters: Uncertainty (1σ) in read-out, Dose (Air-Kerma), measured at entrance of sensor/detector for typical set-up. Set-up and read-out functionality from real tests. Opinion of author. (From Lindström et al. 2014 and Paper IV; Lindström et al., 2020b)

Device	LIS-method	FPA (Visi-X)	Gafchromic	Raysafe DXR+
Uncertainty (+/- 1σ)	0.09 mm	0.5 mm	0.5 mm	1.25 mm
Dose required (0.7m*, 1m focus to device distance)	< 1 mGy	~15 mGy	~12 mGy	~12 mGy (2 mGy*)
Set-up and read-out	Set-up time and read-out: initial long start-up time for software etc. Quick read-out and stores results	Short set-up time: needs dimmed lights in x-ray lab, 3-4 exposures. Gives results for all four edges in one session. Do not store results	Long set-up time and requires 3-4 exposures for readability. Gives results for all four edges in one session. Strips can be stored as a permanent record.	Short set-up time (2 devices) requires 2 exposures and repositioning in between for all four edges. Uncertainty in mammography an issue for breast support edge. Do not store results.

*) for mammography.

4. CONCLUSIONS

The LAC-model was developed to simplify the modelling of radioluminescent phosphor layers with regards to variations of the particle size and thickness combinations. The suggested model is using input data obtained from measurements utilising standard equipment normally found in a medical physics department. Results from experiments support the proposed model. Theoretical comparisons with existing models suggest that the physics assumption involved in the present model has some validity beyond simple curve fitting. Optimising the phosphor particle size and thickness combination for an imaging task, is a key process where the outcome will largely determine the patient dose. Previously available models require input parameters either produced from rather complex measurement or time consuming computer simulations. Furthermore, the results are sometimes valid for a specific case only. On the other hand, the LAC-model has showed to be able to yield results for a variety of particle size and phosphor thickness combination, all with a reasonable uncertainty.

A comparison of the relative extrinsic efficiency between measurements, LAC-model results, and dead layer corrected LAC-model results, showed that introducing a dead layer may have an impact on the calculated extrinsic efficiency. However, as a suggested explanation of a luminance change with varying particle size, it was shown that this effect cannot alone be attributable to a dead layer surrounding individual particles. The effect was too small in magnitude.

Monte-Carlo simulations of interface regions of scintillator slabs and air showed that the energy impartation of impinging ionising radiation to the scintillator was lower in the region closest to the interface and displayed similar characteristics as of a chemical dead layer. i.e. a lower light production.

The LAC-model has been utilised in the development process of two radioluminescence based devices for quality control in a diagnostic radiology department. In the first case, phosphorescence has been indicated by the model to be adequate to be able to present the complete x-ray field position represented by an afterglowing surface. In the second case, a partially transparent phosphor strip has been used to sensitise a linear array sensor to x-rays and in the same time preserve the light transmission capabilities. The model run presented available alternatives. The use of the simple model for radioluminescence has greatly reduced the time for the optimisation and evaluation of the devices. The model results have indicated further areas of improvement but also when the optimisation level has been adequate.

4.1 Limitations, words of caution

The LAC-model must be approached carefully and acknowledged as a useful but simplistic tool. The extent to which the ξ -value can be considered a physical (i.e. a

true transport coefficient) rather than an empirical parameter is unclear. The degree to which the ξ -value changes with a particular phosphor layer design (e.g. protective and reflective coatings etc.) remains to be investigated. The model has not been tested for other unit cell features than particles. It is believed it may be feasible in ceramic versions of polycrystalline phosphors but also for structured scintillators like CsI (see next section). There is a limitation to the particle size where it is known that when the size is approaching the wavelength of the emitted light of the phosphor the assumption of scattering being dominating the absorption, no longer holds. It can therefore be expected that the LAC-model breaks down for very small (nano-sized) phosphor particles.

5. FUTURE PROSPECTS

In this section future prospects will be presented. Some are not considered to be more than subjects of discussion at the moment, others are in a more advanced stage.

5.1 Modelling structural scintillators

The LAC-model exhibit god results when modelling polycrystalline phosphors with easily definable individual spherical particles. But what about structural phosphors like CsI where the light is produced in needle-shaped crystals (Nagarkar et al.,1998)? Well, in the theory section (see sec. 2.1.1) “unit cells” are mentioned as the smallest building blocks. The LAC-model has not yet been applied on CsI scintillators but a hint that it may be fruitful, is derived from a very similar approach implemented by Evans et al. (2006). They defined a unit cell of a columnar CsI crystal by dividing the crystal into discrete volumes, much in the same way as for the discrete sub-layers in the LAC-model (see figure 19). Evans et al. then assigned optical parameters to each unit cell to describe the optical transport (Lambertian Light Guide model, LLG). At this time, it is not clear what modifications are needed for the LAC-model to be used for a columnar scintillator. A plausible approach would be to assign an effective extinction factor, ξ , to each unit cell.

Figure 19. Based on Evans et al. (2006), showing a scintillator column divided into discrete blocks exhibiting specific optical conditions.

5.2 Imaging approaches: MTF and dual-layers

The LAC-model does not handle imaging parameters. Originally assigned to be a tool for the optimisation between phosphor extrinsic efficiency and radiation dose, there are no obvious image quality parameters in the LAC-model. However, two examples of utilising the LAC-model for imaging will be given here. The first is a thought model, not fully brought to completion. The other is the work of Gavin Poludniowski (2021) based on the connection between the LAC-model, Hamaker-Ludwig (Ludwig, 1971), and Swank parameters (see Appendix of Paper II: Lindström et al., 2020). Both approaches are aiming for the MTF of a phosphor screen. The third approach is to improve resolution properties of a polycrystalline phosphor utilising two layers of different particle size.

5.2.1 MTF approach 1

Wang et al. (1997), monitored the light distribution on the surface of phosphor layers to check the agreement with Lambertian light distribution. In the same manner, the LAC-model can be used to calculate the light distribution at the surface (in reflective mode). Since we are dealing with resolution, it is decided that the smallest detectable object equals a (excited) column in the perfect matrix of the LAC-model. The particles are considered “binary”, i.e. the full particle volume is either on or off, with a light energy proportional to the energy imparted. The light produced in each grain is projected to the optical receiver plane. The light contribution at the surface from each particle in the activated column, is calculated by converting the original extinction factor, ξ , for each inter-crossing, to an extinction factor per unit length. The optical extinction of the light for each activated particle is then calculated for all angles to produce the light distribution for each particle at the surface. Taking the total light distribution for a complete row of columns can then be described by the light distribution $L(x)$, (see figure 20), where x is the lateral position on the surface of the slab. Assume that $L(x)$, is identical to the line spread function (LSF) which mathematically can be converted into an MTF-curve using the Fourier transform. i.e.

$$MTF(\nu) = \int L(x)e^{-i2\pi\nu x} dx \quad (14)$$

Where ν is the frequency.

Figure 20. Illustration of calculated light-distribution at phosphor surface based on the LAC-model for two different particle sizes but identical phosphor layer thickness. Full Width at Half Maximum (FWHM), is indicated by the arrows.

5.2.2 MTF-approach 2

The second approach on the MTF has been proposed by one of the co-authors, Gavin Poludniowski (2021). In a private communication, he outlined the following argument, noting the similarities between the LAC-model (Lindström and Alm Carlsson, 1999), Hamaker-Ludwig (Ludwig, 1971) and the Swank (1973) approach. Swank derived the Optical Transfer Function for a phosphor screen, $\text{OTF}(\nu)$. The solution was based on a 3D time-independent diffusion equation and a screen of infinite lateral extent. The normalisation of the OTF is such that the fraction of light emitted from the screen at the sensor side is given by $\text{OTF}(0)$. As Swank demonstrated, the light output predicted from his approach is equivalent to that of the theory of Hamaker (1947), which we will denote H . It has also been shown that in the continuous form of the LAC model, the light output, Λ , is equivalent to Hamaker's in the limit that (Lindström et al., 2020)

$$\frac{\xi L}{d} \equiv \sigma L \gg 1 \quad (15)$$

where ξ is the LAC extinction factor, L is the screen thickness and d is the particle size (diameter). The parameter σ appears in Hamaker and Swank's models and can be interpreted as the reciprocal of the mean diffusion length. It resembles an effective attenuation coefficient for light photons, arising out of the combined effects of absorption and scatter. In summary:

$$\text{OTF}(\nu) \xrightarrow[\nu \rightarrow 0]{} H \xrightarrow[\sigma L \rightarrow \infty]{} \text{LAC} \quad (16)$$

Strictly, the normalisations are defined differently for the three models, but we will gloss over this unimportant detail. So, the LAC-model can be obtained from Swank's OTF by taking the zeroth frequency component and taking the thick screen limit, although it also requires that we interpret the product σd as *independent* of particle size.

We could, instead of taking the successive limits, only take the thick screen limit of the OTF and obtain what one could interpret as the MTF corresponding to the LAC model. This could be done starting from Swank's (rather complicated) expression. However, it is simpler to start at the other end of the chain: with the LAC model. Typically, it is not possible to recover a more general model (all spatial frequencies) from a less general one (zeroth frequency result). However, in this case, we can use a simple insight. Due to the structure of the diffusion equation, the OTF only depends on spatial frequency and σ through the following variable:

$$q = \sqrt{\sigma^2 + \nu^2} = \sqrt{(\xi/d)^2 + \nu^2} = \frac{1}{d} \sqrt{\xi^2 + d^2 \nu^2} \quad (17)$$

The consequence of this is that we can recover the whole spectrum of spatial frequencies by substituting:

$$\xi \rightarrow \sqrt{\xi^2 + d^2 \nu^2} \quad (18)$$

That is:

$$\text{MTF}_{\text{LAC}}(\nu) = \frac{A\left(\sqrt{\xi^2 + d^2\nu^2}\right)}{A(\xi)} \quad (19)$$

where, in the continuous limit, for reflection mode,

$$A(\xi) = \eta\bar{\psi}_{\mu+\xi/d} \left[1 - \exp\left(-\left(\mu + \frac{\xi}{d}\right)L\right) \right] \approx \eta\bar{\psi}_{\mu+\frac{\xi}{d}} \quad (20)$$

We will assume the exponential term in the square brackets is negligible, since this is in the spirit of the thick screen approximation. Inserting (20) into (19), we obtain:

$$\text{MTF}_{\text{LAC}}(\nu) = \frac{\mu + \frac{\xi}{d}}{\mu + \frac{1}{d}\sqrt{\xi^2 + d^2\nu^2}} \quad (21)$$

Equation 21 has not yet been evaluated.

5.2.3 Dual-layer phosphors

Another interesting feature of the LAC-model is that it allows for a phosphor layer to contain different particle sizes for different depths by maintaining an identical extinction factor, ξ . It would be possible to design a screen to have reduced particle size layers opposite to the receiver output side. This could increase the spatial resolution without the same degree of impairment in extrinsic efficiency that would follow from a screen of identical reduced particle size throughout. This feature is not trivial to perform in other models. Song, Shim and Han (2018) have modelled a two-layered phosphor, by using parts of the LAC-model.

Figure 21. Illustration of a two-layered phosphor of different particle sizes.

5.3 Linear Imaging Sensor (LIS)-method

Finally, some words on one of the applications, the LIS-method. During the development project, the LIS prototype was also used in CTs to see if it was possible to produce the profile of the narrow field. The test results were encouraging (see figure 22) and there are plans to produce a prototype with a longer sensor to be able to cover most CT fields. The phosphor $\text{Gd}_2\text{O}_2\text{S:Tb}$ is known to have an energy dependence which has not been fully investigated for the CT profile, and it may well be interesting to try other phosphors better suited for the typical CT x-ray spectra.

Figure 22. A CT-profile measured with the LIS-prototype. To the right, the calculated centre of the CT-profile in terms of pixel-address from ten consecutive measurements. (Lindström et al. 2014)

The LIS-method obviously works, but it takes time to move around the device between the four edges to be measured, in conventional radiology. Therefore, it is planned to have at least *two* devices in opposite position of each other. Ideally, *four* devices would cover all edges at once but at the moment there are technical obstacles preventing that from materialise.

Figure 23. Vision of a four LIS-device measuring set-up. Light field location is measured and determined at the edges (pixel address). LIS-device(s) do not have to be exactly aligned by the centre of its sensor.

REFERENCES

- Abliz E, Collins, J. E., Bell H, & Tata, D. B. (2011). Novel applications of diagnostic X-rays in activating a clinical photodynamic drug: Photofrin II through X-ray induced visible luminescence from "rare-earth" formulated particles. *Journal of X-ray science and technology*, 19(4), 521-530.
- Abrams, B. L., & Holloway, P. H. (2004). Role of the surface in luminescent processes. *Chemical reviews*, 104(12), 5783-5802.
- Ajiro F, Hitosi N, Tamatani M, Tsuda, N. (1986). Projection CRT with a green emitting terbium activated lanthanum oxychloride phosphor exhibiting nearly constant light-output of elevated temperatures, United States Patent 5115306 (1986)
- Alig, R. C., & Bloom, S. (1977). Cathodoluminescent efficiency. *Journal of the Electrochemical Society*, 124(7), 1136.
- Ayvacıklı, M., Canimoglu, A., Karabulut, Y., Kotan, Z., Herval, L. K. S., De Godoy, M. P. F., & Can, N. (2014). Radioluminescence and photoluminescence characterization of Eu and Tb doped barium stannate phosphor ceramics. *Journal of alloys and compounds*, 590, 417-423.
- Barbin, R. L., & Poulos, A. S. (2002). Cathode Ray Tube Display Technology. *Encyclopedia of Imaging Science and Technology*.
- Baro, J., Sempau, J., Fernández-Varea, J. M., & Salvat, F. (1995). PENELOPE: an algorithm for Monte Carlo simulation of the penetration and energy loss of electrons and positrons in matter. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 100(1), 31-46.
- Becquerel H C, R. Acad. Sci. 62 (1866) 999 (1995)
- Biduchak, A., Grytsiuk, M., Chornenka, Z., & Domanchuk, T. (2019). The history of the development of radiology. *Current issues of social sciences and history of medicine*, 24(4), 24-26.
- Binzoni, T., Leung, T. S., Gandjbakhche, A. H., Ruefenacht, D., & Delpy, D. T. (2006). The use of the Henyey–Greenstein phase function in Monte Carlo simulations in biomedical optics. *Physics in Medicine & Biology*, 51(17), N313.
- Blasse, G., & Grabmaier, B. C. (1994). A general introduction to luminescent materials. In *Luminescent materials* (pp. 1-9). Springer, Berlin, Heidelberg.

- Buchanan, R., Tecotzky, M., & Wickersheim, K. (1973). *U.S. Patent No. 3,725,704*. Washington, DC: U.S. Patent and Trademark Office.
- Budinger, T. F. (2014). 1.01 History of Nuclear Medicine and Molecular Imaging.
- Carlsson GA. (1973) Dosimetry at interfaces. Theoretical analysis and measurements by means of thermoluminescent LiF at plane interfaces between a low Z-material and Al, Cu, Sn or Pb irradiated with 100 to 200 kV roentgen radiation. *Acta Radiol Suppl.* 1973;332:1-64. PMID: 4519462.
- Cavouras, D., Kandarakis, I., Bakas, A., Triantis, D., Nomicos, C. D., & Panayiotakis, G. S. (1998). An experimental method to determine the effective luminescence efficiency of scintillator-photodetector combinations used in X-ray medical imaging systems. *the british journal of radiology*, 71(847), 766-772.
- Chappell, J. H.; Murray, S. S. (1984) Relative efficiencies and physical characteristics for a selected group of x-ray phosphors. *Nuclear Instruments and Methods in Physics Research*, 221.1: 159-167.
- Chen, K., Zhang, R., & Lee, S. R. (2010, August). Integration of phosphor printing and encapsulant dispensing processes for wafer level LED array packaging. In *2010 11th International Conference on Electronic Packaging Technology & High Density Packaging* (pp. 1386-1392). IEEE.
- Collier Jr, B. D. (1974). A short history of diagnostic radiology.
- Crooks H. E., Ardran G. M., (1976). Checking X-ray beam field size *Radiography* Vol. XLII, No. 503
- Das, I. J., Kassae, A., Verhaegen, F., & Moskvina, V. P. (2001). Interface dosimetry: measurements and Monte Carlo simulations of low-energy photon beams. *Radiation Physics and Chemistry*, 61(3-6), 593-595.
- Dinsmore, A. D., Hsu, D. S., Gray, H. F., Qadri, S. B., Tian, Y., & Ratna, B. R. (1999). Mn-doped ZnS nanoparticles as efficient low-voltage cathodoluminescent phosphors. *Applied Physics Letters*, 75(6), 802-804.
- Dullien, F. A. L. (1992). *Porous Media: Fluid Transport and Pore Structure* (2nd ed.). Academic Press. ISBN 978-0-12-223651-8.
- Edison, T. (1896). Communication to Lord Kelvin. *Nature*, 53, 470.
- Evans, P. M., Mosleh-Shirazi, M. A., Harris, E. J., & Seco, J. (2006). Monte Carlo and Lambertian light guide models of the light output from scintillation crystals at megavoltage energies. *Medical physics*, 33(6Part1), 1797-1809.

- Feldman, A. (1989). A sketch of the technical history of radiology from 1896 to 1920. *Radiographics*, 9(6), 1113-1128.
- Ferreira, L. R., Ferreira, H. M. N. B. L., Veloso, J. F. C. A., & Dos Santos, J. M. F. (2004). Energy non-linearity effects in the response of ionic crystal scintillators to X-rays with energy in the region of the K-absorptions edges: experimental results. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 516(2-3), 486-491.
- Giakoumakis, G. E. (1988). X-ray efficiency of phosphor screens in the 20–60 kV range. *Journal of applied physics*, 64(2), 940-942
- Giakoumakis, G. E. (1989, November). On The X-Ray Luminescence Of Phosphor Screens Under Fluoroscopy Conditions. In *X-Ray Instrumentation in Medicine and Biology, Plasma Physics, Astrophysics, and Synchrotron Radiation* (Vol. 1140, pp. 253-255). International Society for Optics and Photonics
- Giakoumakis, G. E., Katsarioti, M. C., Lagaris, I. E., & Panayiotakis, G. S. (1991). A theoretical model for the x-ray luminescence of granular phosphor screens. *Journal of applied physics*, 69(9), 6607-6611.
- Giakoumakis, G. E., & Lagaris, I. E. (1988). A theoretical model for the cathodoluminescence of granular phosphor screens. *Journal of applied physics*, 64(10), 5106-5109
- Giakoumakis, G. E., Nomicos, C. D., & Euthymiou, P. C. (1980). Absolute efficiency of ZnCdS: Ag screens excited by an electron beam. *Journal of Applied Physics*, 51(9), 4976-4980.
- Gingold, E. L. (2017). The medical physicist's role in radiation optimization. *Journal of the American College of Radiology*, 14(10), 1335-1336.
- Goodman, N.B. (1976). The growth of deactivated layers on CsI(Na) scintillating crystals. *Space Science Instrumentation*, 2(4), 425-435.
- Gorokhova, E. I., Demidenko, V. A., Mikhlin, S. B., Rodnyi, P. A., & Van Eijk, C. W. E. (2005). Luminescence and scintillation properties of Gd/sub 2/O/sub 2/S: Tb, Ce ceramics. *IEEE transactions on nuclear science*, 52(6), 3129-3132.
- Hamaker, H. C. (1947). Radiation and heat conduction in light-scattering material. *Philips Research Reports*, 2(6), 420-425.
- International Electrotechnical Commission (IEC), (2009). IEC 60601-2-54, International Standard, Medical electrical equipment – Part 2-54: Particular requirements for the basic safety and essential performance of x-ray equipment for radiography and radioscopy.

- International Electrotechnical Commission (IEC), (2011). IEC 60601-2-45, International Standard, Medical electrical equipment – Part 2-45: Particular requirements for the basic safety and essential performance of mammographic x-ray equipment and mammographic stereotactic devices.
- Izumi, N., Emig, J., Moody, J., Middleton, C., Holder, J., Glenn, S., Chelli, S. J. (2012). Measurement of cathode luminescence efficiency of phosphors for micro-channel plate based x-ray framing cameras. In Target Diagnostics Physics and Engineering for Inertial Confinement Fusion (Vol. 8505, p. 85050I). International Society for Optics and Photonics.
- Jin, D., Xi, P., Wang, B., Zhang, L., Enderlein, J., & van Oijen, A. M. (2018). Nanoparticles for super-resolution microscopy and single-molecule tracking. *Nature methods*, 15(6), 415-423.
- Kandarakis I, Cavouras D, Panayiotakis G, Agelis T, Nomicos C and Giakoumakis G (1996) X-ray induced luminescence and spatial resolution of $\text{La}_2\text{O}_2\text{S:Tb}$ phosphor screens *Phys. Med. Biol.* **41** 297–307
- Kandarakis I, Cavouras D, Panayiotakis G, Nomicos, C. (1997). Evaluating X-ray detectors for radiographic applications: a comparison of ZnSCdS:Ag with $\text{Gd}_2\text{O}_2\text{S:Tb}$ and $\text{Y}_2\text{O}_2\text{S:Tb}$ screens. *Phys. Med. Biol.* **42**, 1351–1373.
- Kandarakis, I., Cavouras, D., Nikolopoulos, D., Episkopakis, A., Kalivas, N., Liaparinis, P., & Panayiotakis, G. (2006). A theoretical model evaluating the angular distribution of luminescence emission in X-ray scintillating screens. *Applied radiation and isotopes*, 64(4), 508-519.
- Kang, T., Lee, S., Kim, T., & Kim, J. (2020). Efficient Luminescence of $\text{Sr}_2\text{Si}_5\text{N}_8\text{:Eu}^{2+}$ nanophosphor and its film applications to LED and Solar cell as a downconverter. *Scientific reports*, 10(1), 1-7.
- Kolar, Z. I., & Den Hollander, W. (2004). 2003: A centennial of spinthariscopes and scintillation counting. *Applied radiation and isotopes*, 61(2-3), 261-266.
- Kubelka, P. (1931). Ein Beitrag zur Optik der Farbanstriche (Contribution to the optic of paint). *Zeitschrift für technische Physik*, 12, 593-601.
- Kuboniwa, S., Hoshina, T., Narahara, T., & Kanamaru, M. (1973). Effects of comminution on the luminescence of phosphors. *Journal of The Electrochemical Society*, 120(12), 1734.
- Lempicki, A., Brecher, C., Szupryczynski, P., Lingertat, H., Nagarkar, V. V., Tipnis, S. V., & Miller, S. R. (2002). A new lutetia-based ceramic scintillator for X-ray imaging. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 488(3), 579-590.

- Liaparinis, P. F., & Kandarakis, I. S. (2009). The Monte Carlo evaluation of noise and resolution properties of granular phosphor screens. *Physics in Medicine & Biology*, 54(4), 859.
- Lindström, J., & Carlsson, G. A. (1999). A simple model for estimating the particle size dependence of absolute efficiency of fluorescent screens. *Physics in Medicine & Biology*, 44(5), 1353.
- Lindström, J. (2011). *Radioluminescence: A simple model for fluorescent layers* (Doctoral dissertation, Linköping University Electronic Press).
- Lindström, J., Hulthén, M., Carlsson, G. A., & Sandborg, M. (2014, March). Optimizing two radioluminescence based quality assurance devices for diagnostic radiology utilizing a simple model. In *Medical Imaging 2014: Physics of Medical Imaging* (Vol. 9033, p. 90333R). International Society for Optics and Photonics.
- Lindström, J., Carlsson, G. A., Wählin, E., Tedgren, Å. C., & Poludniowski, G. (2020). Experimental assessment of a phosphor model for estimating the relative extrinsic efficiency in radioluminescent detectors. *Physica Medica*, 76, 117-124.
- Lindström, J., Hulthén, M., Sandborg, M., & Tedgren, Å.C. (2020b). Development and assessment of a quality assurance device for radiation field–light field congruence testing in diagnostic radiology. *Journal of Medical Imaging*, 7(6), 063501.
- Lindström, J., Lund, E., Wählin, E., Tedgren, Å.C. (2021). Revisiting the dead layer in phosphors from a dosimetric perspective- assessment through Monte-Carlo simulations and modelling, - to be submitted to *Journal of Luminescence*.
- Ludwig, G. W. (1971). X-ray efficiency of powder phosphors. *Journal of The Electrochemical Society*, 118(7), 1152.
- Ludwig, G. W., & Prener, J. S. (1972). Evaluation of Gd₂O₂S: Tb as a phosphor for the input screen of x-ray image intensifier. *IEEE Transactions on Nuclear Science*, 19(4), 3-8.
- Michail, C., Valais, I., Fountos, G., Bakas, A., Fountzoula, C., Kalyvas, N., & Kandarakis, I. (2018). Luminescence efficiency of calcium tungstate (CaWO₄) under X-ray radiation: Comparison with Gd₂O₂S: Tb. *Measurement*, 120, 213-220
- Mie, G. (1908). Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen. *Annalen der physik*, 330(3), 377-445.
- Moharil, S. V. (1994). A review of X-ray imaging phosphors. *Bulletin of Materials Science*, 17(1), 25-33.
- Morin, R. L., & Frush, D. P. (2017). An introduction to radiation sensibilities. *Journal of the American College of Radiology*, 14(1), 137.

- Nagarkar, V. V., Gupta, T. K., Miller, S. R., Klugerman, Y., Squillante, M. R., & Entine, G. (1998). Structured CsI (Tl) scintillators for X-ray imaging applications. *IEEE transactions on nuclear science*, 45(3), 492-496.
- Nakamura, R. (1996). Personal communication
- Nikl, M. (2006). Scintillation detectors for x-rays. *Measurement Science and Technology*, 17(4), R37.
- Nomicos, C. D., Giakoumakis, G. E., Euthymiou, P. C., & Sandilos, P. X. (1978). Absolute efficiency of (ZnCd) S: Ag phosphor under fluoroscopy conditions. *Journal of Applied Physics*, 49(6), 3636-3638.
- Poludniowski, G. G. (2007). Calculation of x-ray spectra emerging from an x-ray tube. Part II. X-ray production and filtration in x-ray targets. *Medical physics*, 34(6Part1), 2175-2186.
- Poludniowski, G. G., & Evans, P. M. (2007). Calculation of x-ray spectra emerging from an x-ray tube. Part I. Electron penetration characteristics in x-ray targets. *Medical physics*, 34(6Part1), 2164-2174.
- Poludniowski, G., Landry, G., Deblois, F., Evans, P. M., & Verhaegen, F. (2009). SpekCalc: a program to calculate photon spectra from tungsten anode x-ray tubes. *Physics in Medicine & Biology*, 54(19), N433.
- Poludniowski, G. (2021). *Personal communication*
- Regulla, D. F., & Leischner, U. (1983). Comparing interface dosimetry with conventional methods and TSEE. *Radiation Protection Dosimetry*, 4(3-4), 174-176.
- Sensakovic, W. F., Warden, D. R., & Bancroft, L. W. (2017). The link between radiation optimization and quality. *Journal of the American College of Radiology*, 14(6), 850-851.)
- Schuster, A. (1905). Radiation through a foggy atmosphere. *The astrophysical journal*, 21, 1.
- Sharpe, W. D. (1978). The New Jersey radium dial painters: a classic in occupational carcinogenesis. *Bulletin of the History of Medicine*, 52(4), 560-570.
- Shionoya, S., Yen, W. M., & Yamamoto, H. (Eds.). (2006). *Phosphor handbook*. CRC press. ISBN 0-8493-3564-7
<http://research.mrl.ucsb.edu/~ngeorge/downloads/Phosphor%20books/Phosphor%20Handbook.pdf>

- Siemens homepage (2019), <https://www.oem-products.siemens-healthineers.com/>
 Note: spectrum simulator no longer available (checked 2021-02-01), Previous address URL:
<https://www.oem-xray-components.siemens.com/x-ray-spectra-simulation>
- Song, J., Shim, H. B., & Hahn, J. W. (2018). Analytical model for designing a high-energy-efficiency granular double-layer X-ray scintillator with a diffuse reflection layer. *Optics express*, 26(17), 21642-21655.
- Stein A M (2012). Analysis of a focus shift during the operation of an X-ray unit. *The Russian journal of nondestructive testing*, 48(6), 378-383
- Swank, R. K. (1973). Calculation of modulation transfer functions of x-ray fluorescent screens. *Applied optics*, 12(8), 1865-1870.
- Thomas, A. M., & Banerjee, A. K. (2013). *The history of radiology*. OUP Oxford.
- Tootell, A. K. (2018). *Radiation dose assessment: measurement, estimation and interpretation* (Doctoral dissertation, University of Salford).
- Tsai, D. Y., & Matsuyama, E. (2015). Recent advances of quality assessment for medical imaging systems and medical images. In *Visual Signal Quality Assessment* (pp. 157-183). Springer, Cham.
- Tsapaki, V. (2020). Radiation dose optimization in diagnostic and interventional radiology: Current issues and future perspectives. *Physica Medica*, 79, 16-21.
- Uppal, A. K., Chaturvedi, S. N., & Nath, N. (1987). Effect of killer impurities on the phosphorescence of CaS: Cu and CaS: Ce. *Physica Status Solidi. B, Basic Research*, 143(1), K89-K93)
- Verhaegen F and Seuntjens J, (1995) Monte Carlo Study of Electron Spectra and Dose from Backscattered Radiation in the Vicinity of Media Interfaces for Monoenergetic Photons of 50-1250 keV, *Radiation Research*, Vol. 143, No. 3 (Sep. 1995), pp. 334-342
- Wang, P. C., & Cargill III, G. S. (1997). Optimization of phosphor screens for charge coupled device based detectors and 7–34 keV x-rays. *Journal of applied physics*, 81(3), 1031-1041.
- Wickersheim K A, Alves R V and Buchanan R A 1970 Rare earth oxysulfide x-ray phosphors *IEEE Trans Nucl. Sci.*
 17 57–60
- Wojtowicz, A. J. (1999). Some aspects of solid state radioluminescence. *Acta Physica Polonica-Series A General Physics*, 95(1), 165-178.

- Yanagida, T. (2018). Inorganic scintillating materials and scintillation detectors. *Proceedings of the Japan Academy, Series B*, 94(2), 75-97.
- Yang, P., Harmon, C. D., Doty, F. P., & Ohlhausen, J. A. (2014). Effect of humidity on scintillation performance in Na and Tl activated CsI crystals. *IEEE Transactions on Nuclear Science*, 61(2), 1024-1031.
- Yordanova, A., Iordanova, R., Koseva, I., Nikolov, V., & Kukeva, R. (2018). Spectroscopic investigations of nanosized Cr³⁺ doped Sc_{2-x}In_x (WO₄)₃: A new promising laser material. *Luminescence*, 33(7), 1185-1193.

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