Exploring the possibility of radiography in emission mode at higher energies: Improving the visualization of the internal structure of paintings

Olivier Schalm [*1,2], Diana Leyva Pernia [3,4], Peter Willems[5], Ana Cabal[6,4], Piet Van Espen[6]
[1] University of Antwerp, Conservation Studies, Blindestraat 9, B-2000 Antwerp, Belgium,
[2] Antwerp Maritime Academy, Noordkasteel Oost 6, 2030 Antwerp, Belgium,
[3] University of Antwerp, Department of Mathematics and Computer Science, Middelheimlaan 1, 2020 Antwerp, Belgium
[4] Centre of Technological Applications and Nuclear Development (CEADEN), Havana, Cuba
[5] GE Sensing & Inspection Technologies, Roderveldlaan 5, B-2600 Berchem
[6] University of Antwerp, Department of Chemistry, AXES, Groenenborgerlaan 171, 2020 Antwerp, Belgium
olivier.schalm@uantwerpen.be; olivier.schalm@hzs.be
*: presenting author

Abstract
We demonstrated in previous investigations that the internal structure of paintings can be visualized with conventional radiography in transmission mode when paintings have the proper stratigraphy. Unfortunately, there are many paintings that do not result in useful images. This problem can be solved by using radiography in emission mode. With this technique, the painting is irradiated with high energetic X-rays originating from an X-ray tube operating at 100 keV – 320 keV while inside the painting low energetic signals such as photoelectrons or characteristic photons are being generated. These signals escape from the top 10 μm of the painting and are able to illuminate the imaging plate. However, this technique has also some disadvantages. One of them is that it is not able to visualize underlying paintings. In this study, we explored the possibility to enhance the information depth by increasing the energy of the photon source from 100 keV up to 1.3325 MeV (i.e., ^{60}Co source). At the same time, we also studied how the contrast between pigments is generated in emission mode. For this, we used mathematical simulation of particle transport in matter to understand the relation between input particle (particle type such as photon, electron or positron and the energy of the particle), the material being irradiated (element from which it is composed, thickness and density) and the output signal (generated particle types and energy). Finally, we will show that it is possible to image paintings using a ^{192}Ir and even a ^{60}Co source.

Key words: *cultural objects; X-ray radiography; X-ray sources; Monte Carlo method; stratigraphy ; iridium 192; cobalt 60.*

Explorando las posibilidades de la radiografía en modo de emisión a altas energías: Cómo optimizar la visualización de la estructura interna en pinturas

Resumen
En investigaciones previas se ha demostrado que la estructura interna de las pinturas se puede visualizar satisfactoriamente con la radiografía convencional en modo de transmisión, siempre y cuando dichas pinturas tengan la estratigrafía adecuada. Desafortunadamente, hay muchos casos en los que la aplicación de este método no resultan en imágenes útiles. Este problema puede ser resuelto usando la radiografía en modo de emisión. Con esta técnica, la pintura se irradia con rayos X de alta energía originados en un tubo de rayos X trabajando entre 100 keV y 320 keV. Esto genera señales de baja energía (fotoelectrones o fotones característicos) en el interior de la pintura que, al escapar de las 10 μm superiores, pueden iluminar una placa de imágenes. No obstante, su aplicación también implica ciertas desventajas. Una de ellas es la incapacidad de visualizar las pinturas subyacentes. En este estudio, exploramos la posibilidad de incrementar la información obtenida a mayores profundidades aumentando la energía de la fuente de fotones desde 100 keV hasta 1.3325 MeV (fuente de ^{60}Co). También estudiamos el impacto de esta energía en el contraste obtenido entre los pigmentos. Para esto, utilizamos la simulación matemática del transporte de partículas en la materia para comprender la relación entre partículas de entrada (fotones, electrones o positrones y la energía de las partículas), el material que se irradia (elemento del que está compuesto, espesor) y la señal de salida (tipos de

partículas generados y energía). Finalmente, mostraremos que es posible crear imágenes de pinturas usando una fuente ^{60}Co.

Palabras clave: *objetos culturales; radiografía por rayos X; fuentes de rayos X; método de Monte Carlo; estratigrafía ; iridio 192; cobalto 60.*

Introduction

Radiographic images obtained with X-ray transmission radiography of paintings can play an important role in the examination of the creation process of the artist. It can be used to detect changes in the composition made during the process of painting or to detect the presence of underlying paintings. Radiography can also be used to localize zones of older restorations or to solve authentication problems. Unfortunately, the visualization of the pictorial layers is sometimes hampered by two main factors. The first is the use of lead white ($2PbCO_3.Pb(OH)_2$) in the ground layer resulting in a dominating contribution of that layer to the radiographic image. The second factor is the introduction of low Z-pigments, such as zinc white (ZnO), titanium white (TiO_2), organic pigments and the gradual decrease of use of lead white in the 20th century [1].

Various X-ray-based techniques have been suggested and applied to overcome the shortcomings of conventional radiography, such as monochromatic radiography in transmission, energy-resolved X-ray radiography in transmission, radiography in emission mode, or scanning micro X-ray Fluorescence (i.e., macro XRF) [2-6]. Both macro XRF and radiography in emission mode are able to substantially enhance the contribution of the pictorial layer to the overall image. However, these alternative imaging techniques also entail significant drawbacks. Macro XRF requires about a day to analyse a painting and considerable post processing of the collected spectra is needed in order to obtain the element specific images, while radiography in emission mode, typically obtained with an X-ray tube at 320 kV, has a limited lateral resolution when compared to conventional radiography in transmission and a limited information depth so that underlying paintings cannot be visualized.

This contribution explores the possibility to enhance the image quality of radiography in emission mode by increasing the energy of the X-ray source from 100 keV up to 1 MeV using radioactive sources such as ^{192}Ir and ^{60}Co.

Materials, methods and results

Test paintings

We have asked the artist Peter Eyskens to make 4 oil paintings on oak panels with an identical pictorial composition. The main difference between the paintings is the build-up of the layers and the materials used. These differences have been well documented by the artist and are shown in figure 1. Heavy Z white paint refers to lead white; low Z white paints to zinc white and titanium white. Examples of heavy Z pigments are cadmium yellow (CdS), cadmium red (CdSe), yellow lead-tin oxide, or vermillion (HgS). Low Z pigments refer to pigments such as yellow ochre ($Fe_2O_3.nH_2O$), brown ochre (Fe_2O_3), Prussian blue ($Fe_4[Fe(CN)_6]_3.nH_2O$), ultramarine (a silicate) or cobalt blue ($CoAl_2O_4$). More information about the test paintings can be found elsewhere [4]. As can be seen in figure 1, the conventional radiography of painting 4 resulted in clear pictorial information while the other 3 are very challenging for conventional radiography. Paintings 1, 2 and 3 are challenging because the ground layer contains substantial amounts of lead white so that the contribution of the less interesting ground layer to the overall image dominates the contribution of the pictorial layer. An additional challenge for painting 2 is the use of low Z white pigments so that the absorption contrast between coloured zones and zones in which white paint has been used is reduced. However, all paintings resulted in reasonably good image when radiography in emission mode is used, except that lateral resolution is lower than for conventional radiography and the information depth is limited to a few µm so that underlying paintings cannot be visualized. The paintings can be considered as a good compromise between real artistic creations and well characterized test paintings with challenging characteristics for conventional analysis.

X-ray techniques

We used computed radiography to collect the images in emission mode [1]. For the experiments performed, we used a blue coloured IPU imaging plate with a BaFBr thickness of 55 µm ± 2 µm (density: 4.96 g/cm^3) without top coating. The irradiation was done using a radioactive ^{192}Ir source with an activity of 42.6 Ci. That source emits several lines between 9 keV and 885 keV. All lines below 295 keV have been removed using a combination of Fe and Pb filters of different thickness. The ratio of the lines between 300 keV and 600 keV are changed by the filters as well. The source to detector distance was set to 500 mm while the exposure time variated between 60 s (i.e., filter of 20 mm Fe plate) and 4200 s (i.e., 20 mm Fe plate + 5 mm Pb plate) in order to compensate the reduction of the intensity of the sources due to the increasing filter thickness.

A second irradiation source that has been used without any filtering is a radioactive ^{60}Co source with an activity of 50 Ci, emitting photons of 1.17 MeV and 1.33 MeV. An exposure time of 900 s and a working distance of 500 mm was used.

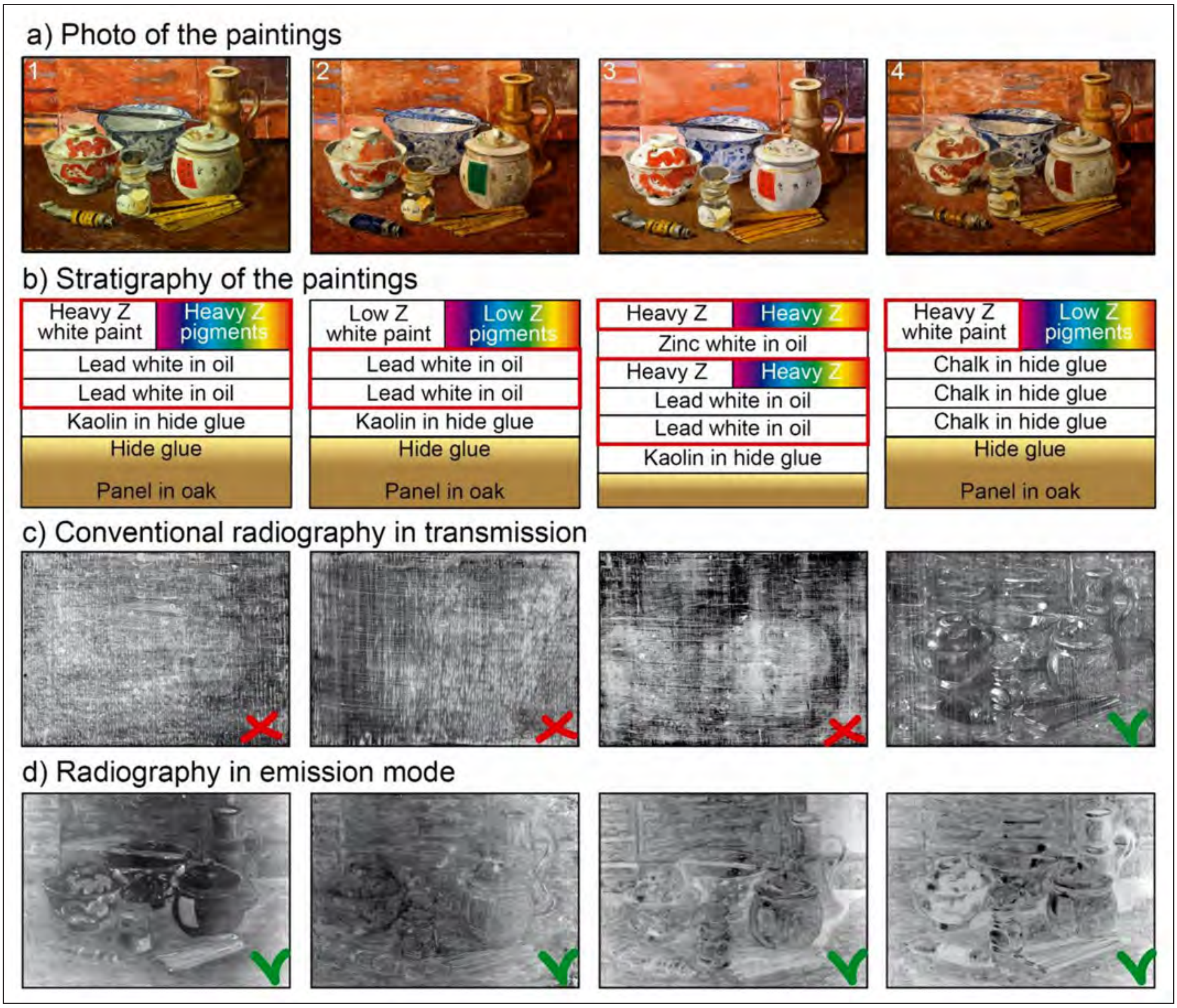

Figure 1. Photo of the 4 paintings with nearly identical composition but with a different stratigraphy and material use and the impact of these differences on conventional radiography in transmission and on radiography in emission mode as collected with an X-ray tube operated at 320 kV.

2.3. X-ray simulations

We simulated the X-rays transport using the software package Monte Carlo N-Particle eXtended (MCNPX) [7]. MCNPX is a general-purpose radiation transport code based in the Monte Carlo method that includes 3-D geometry, continuous-energy transport, transport of 34 different particle types, and a variety of source and tallies. In our research we used the detailed physics treatment which includes, among others, coherent (Thomson) scattering and accounts for fluorescent photons after photoelectric absorption. The energy considered goes from 1 keV to 100 GeV for photons, and from 1 keV to 1 GeV for electrons. Despite of this detailed treatment the code still simplifies the physical model in relation to the characteristic fluorescence lines. This is done by considering only 1 L-line as average of all transitions to L, and omitting all M-lines.

The simulations were performed using the Tally F1 for determining the number of particles crossing a surface. We applied specific cosine binning in the analysed surfaces to differentiate the contribution of particles originated from the source from the secondary particles emitted from the studied material. Energy binning was also used for obtaining the energy spectra of electrons and photons with a resolution of 0.5 keV. All the simulations were carried out with a number of tracks larger than 10^6 in order to achieve results with a relative error below 10%.

Results

It is known that the images obtained by radiography in emission mode are generated by weak signals that escape the paintings [2]. To improve the image quality, more knowledge of these weak signals is needed. Such knowledge can be obtained by simulating the interaction between photons and materials [8]. In this contribution, the electron and photon spectra emitted by a Pb film of 10 µm thickness when irradiated by a mono-energetic photon beam have been simulated (see figure 2). The Pb

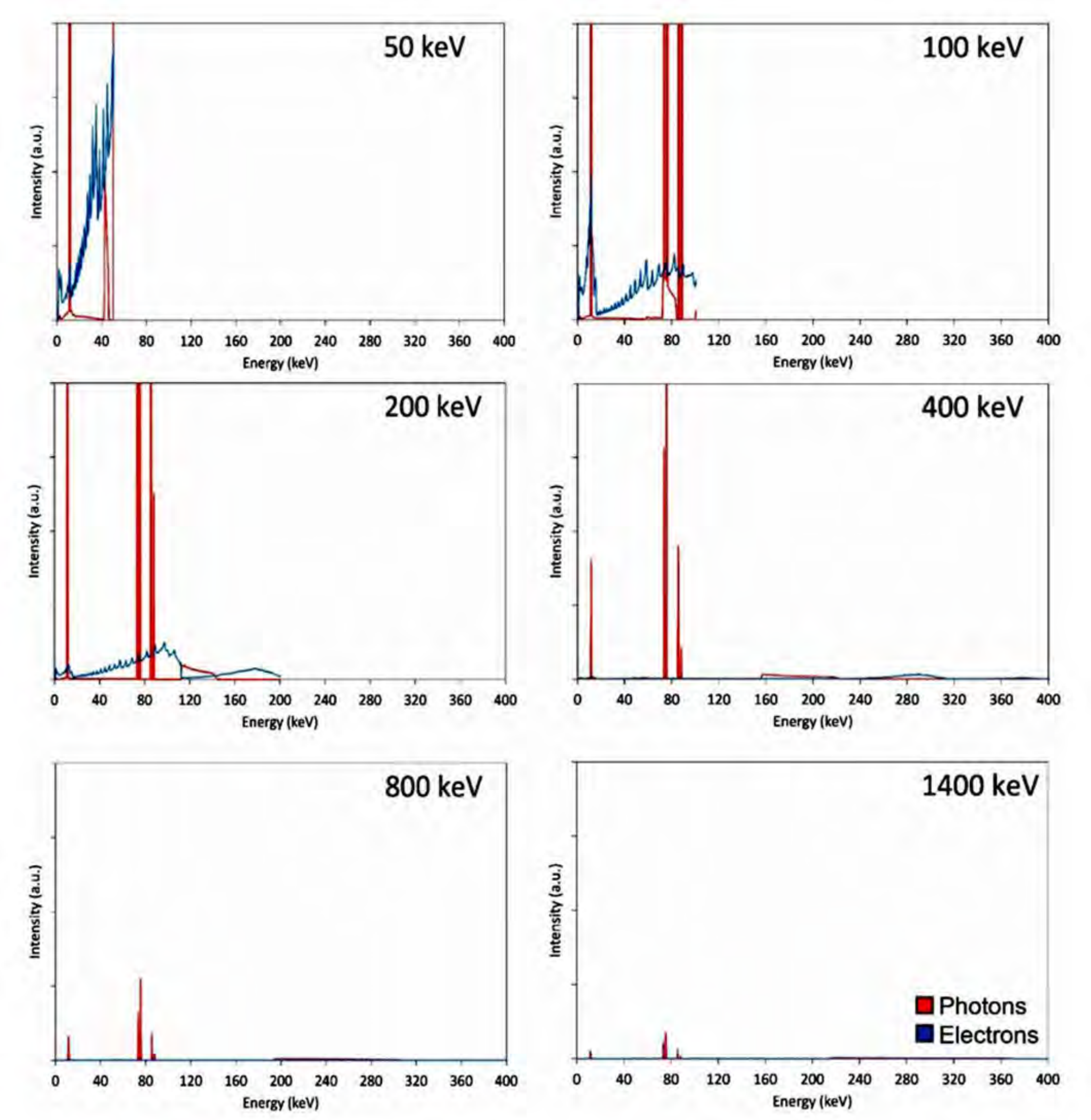

Figure 2. Simulated photon and electron spectra emitted by a Pb film of 10 µm thickness and a density of 11.34 g/cm^3 when irradiated by a monochromatic photon beam. The energy of the mono-energetic photons irradiating the film is stated in the top right corner of the spectra.

film can be considered as a simplification of a lead white paint layer. The low energy part of the photon spectra is clearly dominated by the characteristic X-ray lines. Bremsstrahlung generated by the electrons appear to play a minor role. This means that the low energy part of the emitted X-ray spectra cannot be tuned by increasing the energy of the incident photons. However, the overall intensity of the emitted photon spectra drops with increasing energy of the incident photons. The energy of the emitted electrons increases with the energy of the incident photons but their intensity drops at the same time. This means that only the emitted electron spectra can be tuned in order to obtain information from deeper zones in the pictorial layer.

In order to understand why radiography in emission mode is able to generate material contrasts, the emitted photon and electron spectra have been simulated for different materials when irradiated with 100 keV photons (see figure 3). For C, the emission of both electrons and low energy photons is low. The simulation shows that the emitted signals increases with atomic number. Pb clearly emits considerable amounts of photons and electrons. The spectra show that both emitted photons and electrons should be considered in the image formation.

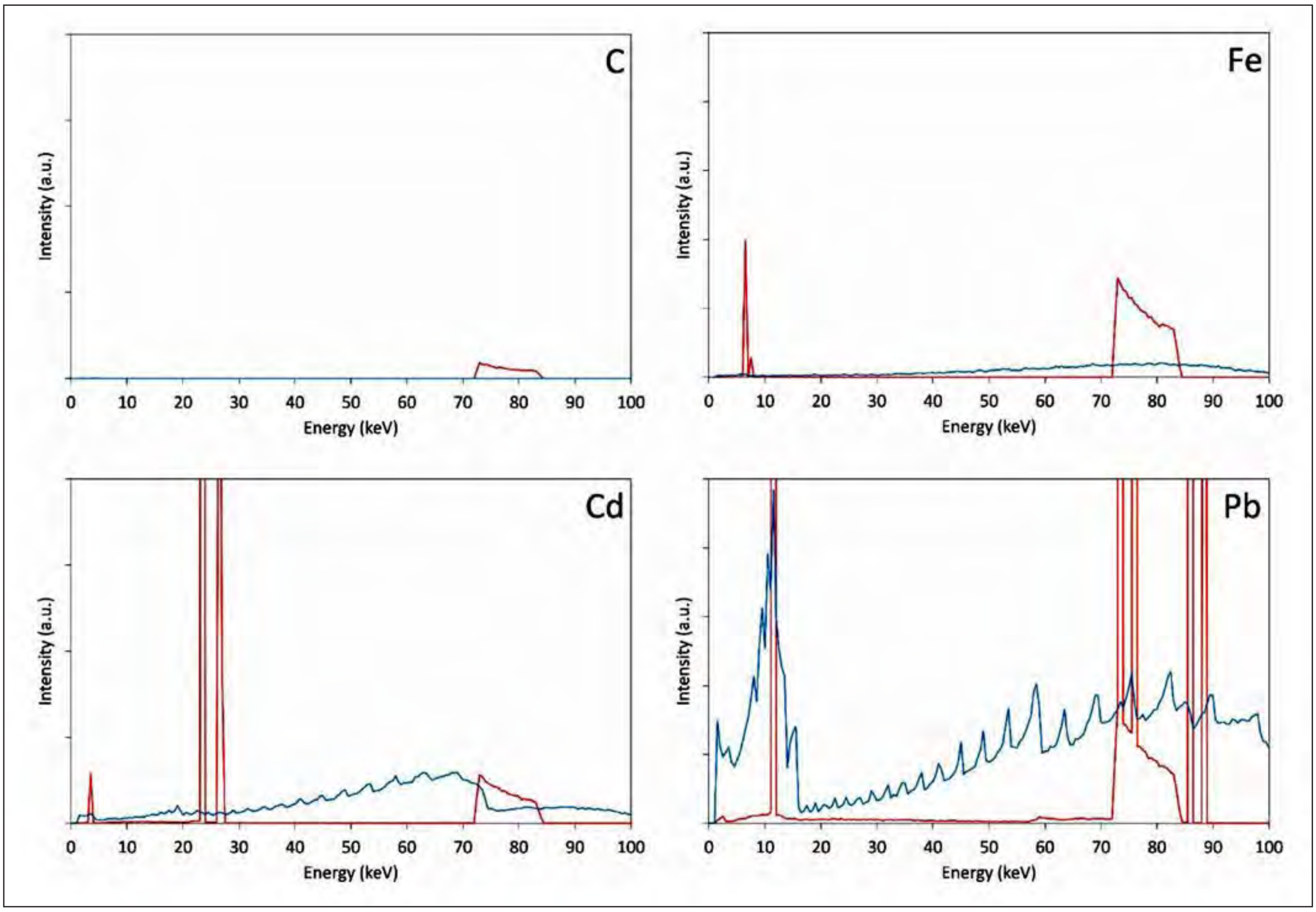

Figure 3. Simulated photon and electron spectra emitted by different materials when irradiated by photons of 100 keV.

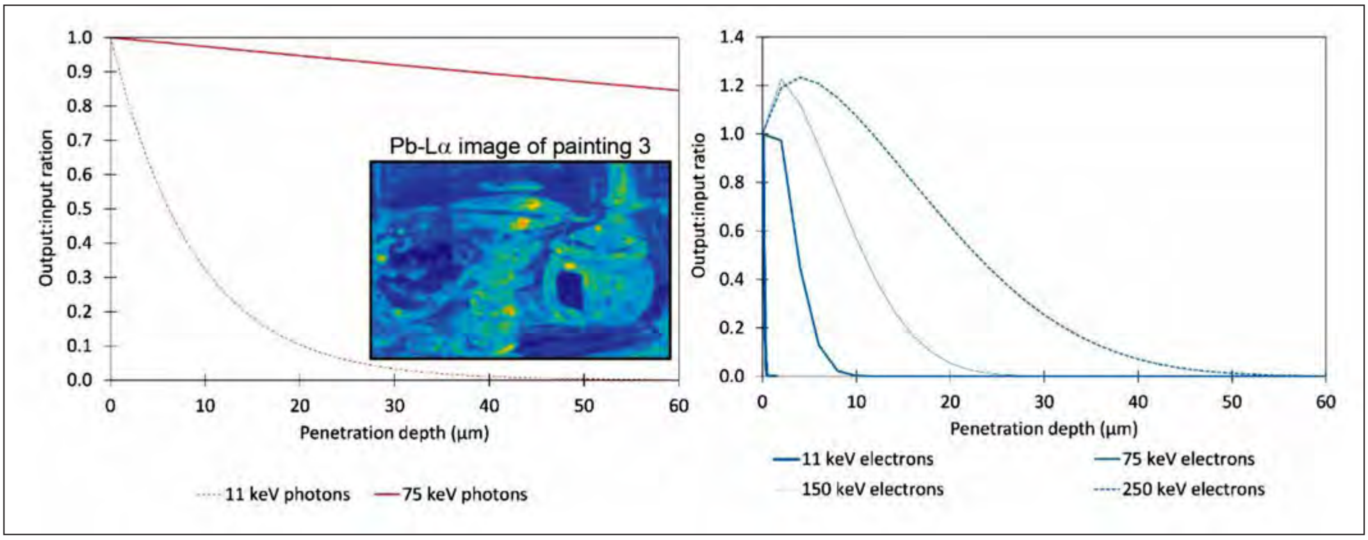

Figure 4. The input/output ratio of photons and electrons escaping at the other side of the film as a function of film thickness. This ratio is calculated for different photon and electron energies.

Figure 4 clearly shows that the Pb-La image of painting 3 is not able to reveal the underlying painting. This means that the information depth of the Pb-La photons at 10.55 keV is insufficient. For pure lead 11 keV photons penetrate about 40 μm. Electrons with the same energy only have a penetration depth of about 10 μm. This means that high energy electrons are needed to obtain information from deeper zones. The simulations suggest that this is possible with electrons of energies higher than 250 keV. For the situation of incident electrons, the output/input ratio can be higher than 1 due to multiple ionization of atoms by a single electron.

The possibility to use high energetic X-ray sources to improve the image quality of radiography in emission mode is not only explored by means of Monte Carlo simulations but also by 2 experiments. In a first experiment, radiography in emission mode of painting 3 (i.e., the one with the underlying painting) is performed with a

^{192}Ir source. Figure 5a show only a small detail from that image. The image collected with an X-ray tube at 320 kV filtered by a plate of 10 mm Cu and 2 mm Al demonstrated that this technique can be used in combination with computed radiography. Although the image is reasonable good, the lateral resolution is rather poor. Brush strokes are not well visible and details such as the signature are not very sharp. The images obtained with the ^{192}Ir source appear to have a better lateral resolution and that resolution appears to improve with increasing filter thickness. This means that by increasing the energy of incident photons, the lateral resolution can be improved.

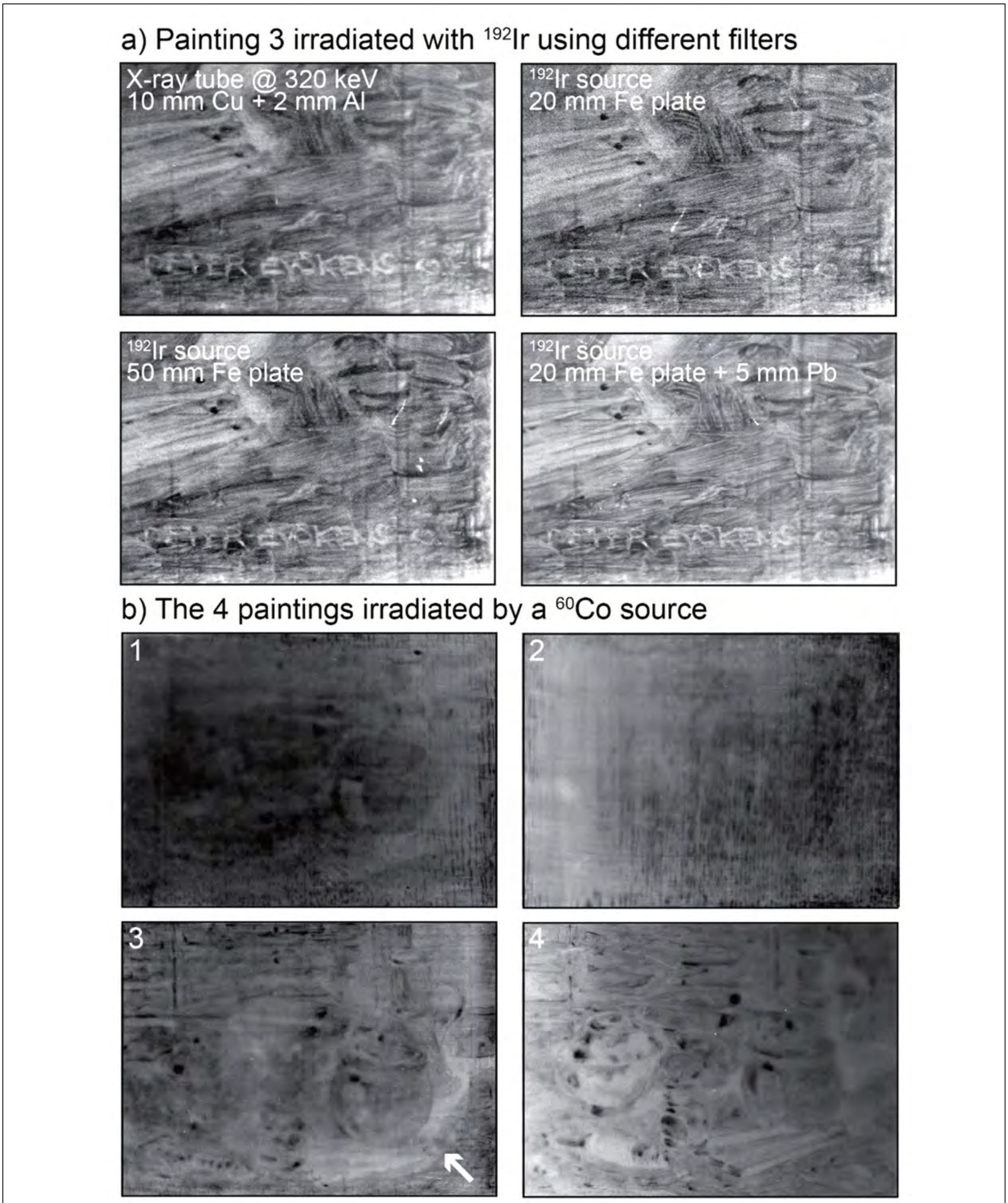

Figure 5. Radiography in emission mode as obtained by radioactive X-ray sources. a) A detail of painting 3 with the underlying paintings has been analysed using a 192Ir source in combination with different filters; b) Radiographic images in emission mode of the 4 paintings when irradiated with a ^{60}Co source.

Unfortunately, the ^{192}Ir source did not reveal the underlying painting. Therefore, it was decided to perform a second experiment at even higher energies by using a ^{60}Co source (figure 5b). Even at such high energies, a signal can be detected using radiography in emission mode. A more detailed description of the observation is given in the list below. The most important conclusion is that the underlying painting (i.e., painting 3) can be faintly seen.

- **Painting 1:** The image clearly shows the topography of the wooden panel. When the ground layer is applied on top of a wooden panel, a negative of the panel's topography might be formed in the lead containing ground layer. The wooden structure is the result of thickness variations in the ground layer. Also the pictorial layer containing heavy Z pigments can be faintly seen;
- **Painting 2:** The structure of the wooden panel can be seen while the pictorial layer composed of low Z pigments remained invisible. It is clear that the ground layer dominated the image and that the attenuation of that signal by the pictorial is negligible;
- **Painting 3:** In the image, the contour of the underlying portrait can be seen. This means that it is possible to generate high energy electrons that are able to escape from deeper zones, zones that cannot be visualized by MAXRF (see figure 4). However, the image quality of the pictorial layer is rather poor.
- **Painting 4:** In this painting, high-Z pigments are only found in the white containing parts of the pictorial layer. It explains why this painting gives the best image quality, although the contrast is rather poor.

Conclusions

Concerning radiography in emission mode, the Monte Carlo simulations has given a more profound understanding in the signals that are responsible for the image formation. The contribution of the low energy photons is clearly dominated by the characteristic photons. This means that the energy of the emitted X-ray signals cannot be tuned by increasing the energy of the incident X-ray source. However, there is also a contribution of electrons. Although the penetration depth of electrons is much lower than photons of the same energy, the energy of the electrons can be enhanced by using X-ray sources of higher energy. The simulations suggest that the information depth can be enhanced in by using electrons with an energy higher than 250 keV.

In this study radiographic images in emission mode have been collected at elevated energies by using ^{192}Ir and ^{60}Co sources. The ^{192}Ir source in combination with filtering to remove the low energy lines and to enhance that contribution of the high energetic lines resulted in improved lateral resolution but not in enhanced information depth. The ^{60}Co source resulted in a poor but readable signal with the required information depth.

Recommendations

Future research is needed to find the optimum energy of the X-ray source to obtain good quality images with high lateral resolution and sufficient information depth. For this, the production of electrons inside the painting with an energy higher than 250 keV appears to be needed.

Acknowledgements

The authors would like to thank the artist Peter Eyskens for the paintings he made. Without his paintings, our research would have been impossible.

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Recibido: 13 de febrero de 2018
Aceptado: 29 de mayo de 2018