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OPTIMISATION OF THE X-RAY FLUORESCENCE IMAGING SYSTEM FOR MAPPING OF PIGMENTS IN HISTORICAL PAINTINGS

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The basic principle of operation of the X-ray fluorescence imaging system of cultural heritage painting has already been proven and presented elsewhere. In this paper, we report on optimisation of the pilot imaging system based on Gas Electron Multiplier (`GEM`) detector, a pinhole camera and a dedicated data acquisition system (DAQ). We focus on evaluation of detection capability of the system for various combinations of the pigments layouts. Carefully designed and prepared painting phantom with stripes, which overlay each other, has been investigated. The selected pigments and their composition were chosen in such a way that the energy resolution of the system was critical capability of reviling hidden layers. Even though the investigated pigments are based on elements which differ in Z by one, the system is capable to distinguish them well.

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1. Introduction

The basic principle of an X-ray imaging system based on a large area $10\,\mathrm{cm}\times 10\,\mathrm{cm}$ position sensitive `GEM` (Gas Electron Multiplier) detector equipped with 2-dimensional (2D) readout and its application for mapping of pigment distributions in historical paintings using X-ray fluorescence technique has already been demonstrated in our previous work [1].

In this paper, we report on further studies of the system performance with a particular focus on capability of pigment recognition. A dedicated phantom painting, prepared according to the XV century techniques with carefully selected historical pigments has been thoroughly examined in order to determine the detection capabilities of the overall system.

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2. Experimental setup

Experimental setup comprises an X-ray tube, pinhole camera, standard GEM detector, and a dedicated readout system with self-triggering front-end electronics and a software suite. The system together with the investigated object are placed inside a dedicated box where environmental conditions are constantly monitored and controlled. The goal is to limit fluctuations of ambient temperature and air humidity, which affect strongly the gain of the GEM detector. Maintaining the stable gain during measurements is important for achieving the best possible energy resolution,

The main hardware components of the readout electronics are: custom designed front-end boards hosting multichannel Application Specific Integrated Circuits (ASICs), custom designed Analog-to-Digital Converter (ADC) board, and a commercial FPGA mini-module mezzanine. More details on the readout system can be found in [2–4].

All measurements presented in this work have been performed using a standard triple GEM detector developed for the Compass experiment [5]. The GEM detector operates at 3.9 kV, which corresponds to the gas gain about 10^3 and is flushed with Ar/CO_2 (70%/30%) gas mixture with one detector volume exchange within about four hours.

The imaging system is based on an X-ray tube (with a copper anode), which delivers the excitation beam, and a pinhole camera, which projects the excited fluorescence radiation on the GEM detector. The system has already been used as described in [1], however, some geometrical arrangements have been changed as shown schematically in figure 1. A new pinhole camera with adjustable focal length and smaller external dimensions has been introduced. It allows to shorten the distance between the X-ray source and the object being imagined. The distance between the painting and pinhole camera was decreased resulting in small magnification of the obtained pictures ($\approx \times 1.25$).

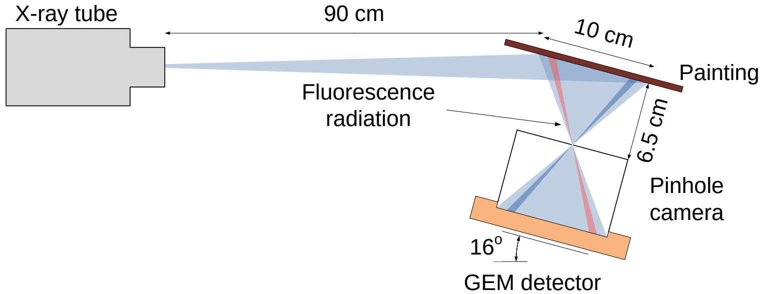


Fig. 1. Schematic view of the complete XRF measurement system.

3. Painting phantom with stripe pattern

Capability of the system to perform selective imaging of hidden painting layers has been evaluated using a dedicated phantom. The phantom shown in figure 2 (a) has an area of 8 cm×8 cm and was prepared following the XV century painting technique. The colour pigments were painted on a wooden panel, overlaying each other as illustrated in figure 2 (b). Such a composition of pigments with different combinations of overlapping areas allows us to determine the influence of the top layer pigments on detection of the shallow and background ones.

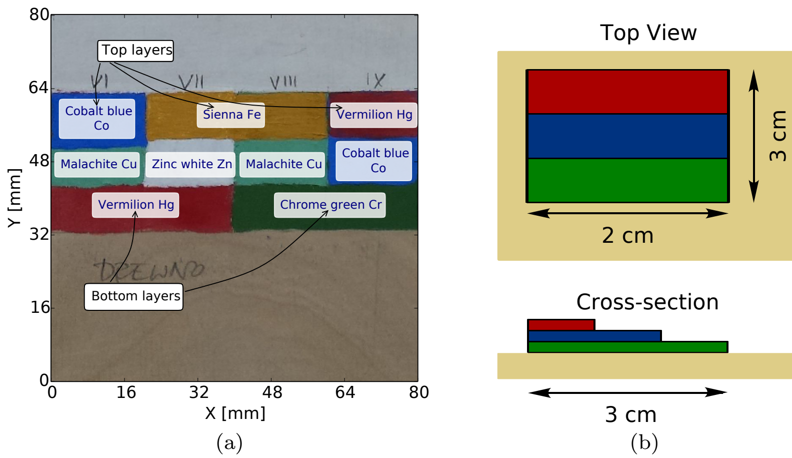


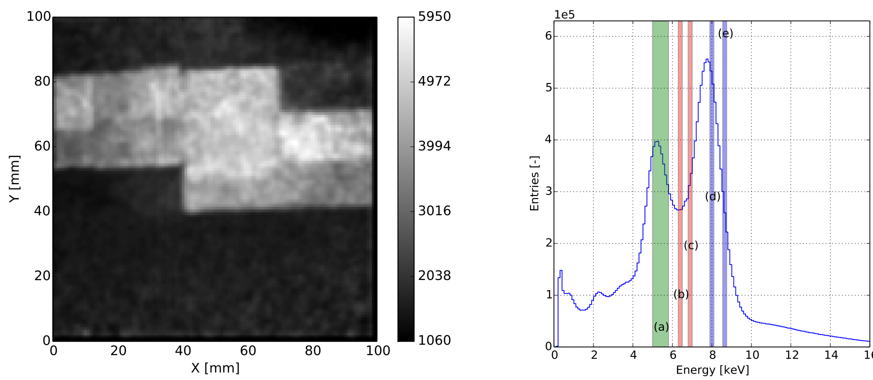
Fig. 2. Composition of the painting phantom. (a) Photograph of the painting phantom with labels showing pigments names. (b) Schematic view of the rightmost fragment of the phantom with one composition of overlapping pigments.

4. Imaging results of the phantom

In order to obtain proper results, the detection system has to be tuned before measurements as it was already presented in [4] and [1]. Besides the standard calibration of the electronic readout system and gain of the GEM detector, one has to correct the intensity of measured X-rays for the vignetting introduced by the pinhole camera. For a discussion of the test results, it is important to note that our system allows us to measure simultaneously the position and the energy for each photon absorbed in the detector volume.

4.1. Cumulative photon energy distribution

A gross result of the measurement is the intensity map of X-ray radiation excited in the painting and projected on the detector. Of course, some scattered photons from the primary beam may as well reach the detector and they will form a background. An example of the image of the investigated phantom is shown in figure 3 (a). The map helps to select interesting regions, especially those which have hidden layers not seen in visible light. From the collected data, we can build a cumulative energy distribution (see figure 3 (b)) for all photons detected across the whole detector area. The five colour shaded regions indicate the energy windows within which we expect the characteristic fluorescence radiation from the pigments used in the painting. The following energy windows have been defined: (a) 5.0–5.8 keV for chrome green, (b) 6.3–6.5 keV for sienna (iron rich), (c) 6.8–7.0 keV is used for cobalt blue, (d) 7.9–8.1 keV for malachite (copper based) and (e) 8.55–8.75 keV for zinc white.



(a) (b)

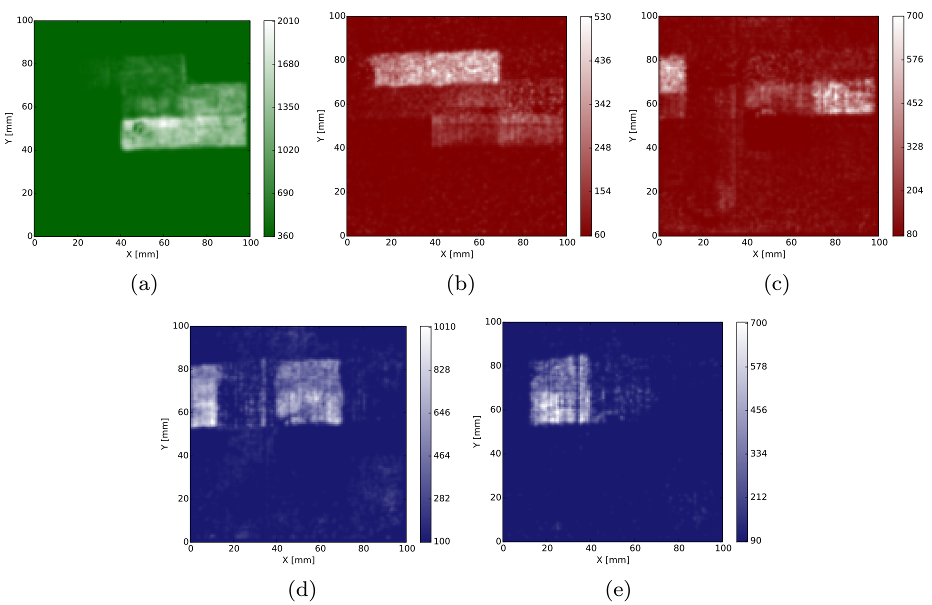
Fig. 3. (a) 2D intensity map of all detected photons. (b) Photon cumulative energy spectrum with characteristic energy windows.

4.2. Pigment recognition results

In order to get pigment (element) specific images, the data should be filtered according to the energy ranges shown in figure 3 (b). One can notice that the proposed energy windows do not match any noticeable peaks in the spectrum. The windows have been selected around the expected characteristic fluorescence X-ray lines. The window widths have been defined in such a way that they should not overlap with characteristic energies of elements with neighbouring atomic numbers. Such an approach applied to a painting, of which elemental composition is not known *a priori*, results in revealing of most of the pigments being used.

Chrome green has been painted as a bottom-most layer on top of which the other pigments are placed. This green pigment occupies half of the coated area, it is covered by malachite and cobalt blue (the second layer of pigments) and on top of them sienna and vermilion are painted (the top-most layer of pigments). The configuration of pigments composition is well reflected in the obtained results presented in figure 4 (a). The area where the chrome green pigment is the most intense matches the uncovered part of the pigment at the bottom of the investigated fragment. The middle part covered from left by copper and cobalt still can be seen, however due to photon absorption the intensity is lower and reflects the different attenuations between the two pigments. The upper part shows almost no signal because third layer of pigments makes the attenuation of soft X-ray fluorescence photons even stronger.

Similar approach has been applied in order to recognize the other pigments. However, narrower energy windows have been selected due to smaller energy differences between characteristic X-ray energies of the characteristic elements of given pigments. The pigment distribution maps for sienna and cobalt blue are depicted in figures 4 (b) and 4 (c) respectively. In those maps, expected signal from the sienna and cobalt blue pigments can be clearly seen. Much lower, but still detectable signal can be observed in the other parts of the maps, which can be attributed to a moderate energy resolution of the `GEM` detector. Better results have been obtained for malachite and zinc white pigments as it is visible in figures 4 (d) and 4 (e). In this case, the pigments are apparently visible almost without any traces of the others.



(a) (b) (c)

(d) (e)

Fig. 4. Intensity maps of all investigated pigments. (a) Chrome green pigment (energy window 5.0–5.8 keV). (b) Sienna pigment (energy window 6.3–6.5 keV). (c) Cobalt blue pigment (energy window 6.8–7.0 keV). (d) Malachite pigment (energy window 7.9–8.1 keV). (e) Zinc white pigment (energy window 8.55–8.75 keV).

5. Conclusions and future plans

In the paper, we have presented further studies on capabilities of the X-ray fluorescence imaging technique using position sensitive GEM detector equipped with a dedicated readout electronics, pinhole camera, and a wide field excitation X-ray beam. In order to demonstrate the performance of the system, a mock-up painting composed of overlapping pigment strips has been used for measurements. The presented 2D maps of pigment distributions confirm good recognition capabilities of the overall system. The system is capable to detect signals from the top-most and shallow layers painted with different pigments. Signals from shallow layers must not be attenuated too much by the high-Z top-most layers in order to be detected. The measurement system offers possibility of quick surveying painting compositions, while maintaining high safety standards of the investigated object. A collection of 2D maps of the pigment distributions allows for rapid painting analysis, giving the opportunity to focus only on the most interesting details, which can be further studied with more accurate but slower techniques, like micro-XRF.

Although, the system has demonstrated a good pigment recognition efficiency, there is still some room for improvements. More comprehensive data analysis can be employed to determine the optimal energy windows for different combinations of pigments distributions. Another aspect is to determine an appropriate (not necessary the best achievable) contrast and spatial resolution, which in a particular case are good enough to make decision if the object is worth of further investigation. Therefore, a procedure how to study the objects efficiently has to be established tougher with art conservators.

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