Old Man in Warnemünde (1907) colouring palette: A case study on the use of hyperspectral imaging for pigment identification

Hilda Deborah*[1]*, Sony George*[4]*, Jon Y. Hardeberg*[5]*; The Norwegian Colour and Visual Computing Laboratory, Department of Computer Science, NTNU – Norwegian University of Science & Technology; Gjøvik, Norway
Jin Strand Ferrer*[2]*, Irina C.A. Sandu*[3]*; Munch Museum; Oslo, Norway

Abstract

The paper presents results from hyperspectral identification of the colouring palette used by Edvard Munch in a canvas painting entitled "Old Man in Warnemünde". The painting is part of a collection at the Munch Museum in Oslo. A collaboration between the Conservation Department of the Munch Museum and the Norwegian Colour and Visual Computing Laboratory from NTNU has allowed to analyze several points of the painting by means of hyperspectral imaging and identify thus the pigments present. The hyperspectral pigment identification involves a use of two pigment databases (Kremer and ENST) which were created using different binding media. Some results from the hyperspectral analysis were also validated through elemental analysis by means of XRF. The pigment identification method employs the shape component of spectral Kullback-Leibler pseudo-divergence function, instead of the widely-used but inaccurate spectral angle mapper. In addition to the interest of this pigment analysis for conservation practice of this particular painting, an important contribution of this paper is the validation of hyperspectral imaging and processing methods for pigment identification.

Introduction

Edvard Munch is known as one of the fathers of expressionism. Many of his paintings are often a challenge for conservators in regards to the unstable condition they are in today. This is a result of the artist's exploration and use of paint, and of outdoor exposure since their creation and during their storage whilst in the artist's own care. One of the main research directions that the Munch Museum in Oslo is developing has to do with the study of materials and techniques of the artist, and of the degradation mechanisms of the paint materials. The impressive collection of the museum offers many interesting case studies. One of these is the canvas painting Old Man in Warnemünde (1907).

Literature in conservation science reports many analytical methods to characterize pigments and dyes from paint layers, i.e., optical microscopy, scanning electron microscopy, FTIR and Raman spectroscopic techniques, chromatography, X-ray diffraction and fluorescence, etc. [12, 6, 13]. However, in the past years, there is an increasing interest in the application of non-invasive and non- or micro-destructive tools [6], and hyperspectral imaging is one of them. The use of spectral imaging in the cultural heritage domain started in the early '90s as a mean to accurately color document paintings [9]. Since then, it has been increasingly popular in the domain, mostly for its capability of capturing both spatial and spectral information of objects. Applications can be found in pigment identification [1, 14], revealing hidden information such as underpaintings [4], and analyzing crack patterns [3].

The collaboration between the Munch Museum and NTNU aims at establishing a non-invasive methodology for the identification of colouring palette in the paint layers based on hyperspectral imaging. To this respect, several points on the surface of Old Man in Warnemünde were chosen for hyperspectral imaging based analysis. Additionally, X-ray fluorescence (XRF) spectrometry was also performed for elemental identification of components in the paint for two different coloured areas (dark blue, blackened edges in pastose paint stroke and red). The XRF analysis was conducted in order to validate the results provided by the hyperspectral pigment identification method.

Experimental approach

Hyperspectral image acquisition

The imaging system used for the spectral acquisition of the painting in the present study is a pushbroom based hyperspectral scanner HySpex VNIR-1800*. Spectral sensitivity of this imaging system is in the visible and near infrared (VNIR) ranges of the electromagnetic spectrum, from 0.4 μm-1.0 μm, with a spectral sampling interval of 3.6 nm, and captures 182 bands. In this system, images are formed by sequential capture of spectral details line by line from the painting [7]. After a proper calibration, the resulting image cube of dimensions (x, y, λ) provides spectral reflectance functions corresponding to pigment sample at any point in the image.

In the spectral imaging setup, the camera and light sources were mounted on a translation stage while the painting remains stationary on a mounting frame. Horizontal and vertical movements of the camera and light sources were accurately controlled and synchronized by an image acquisition software. The translation stage moves the camera and light sources parallel to the painting, to capture the light reflected from the painting. The acquisition distance is set accordingly to coincide with the focal length of the camera's optics. Field of view of the camera depends on the focal distance and is 30 cm in this case. As dimensions of the painting is larger than 30 cm, the scanner had to scan 3 times to capture the whole painting. A broadband light source with spectral coverage over 400-2500 nm was used to illuminate the painting in a stable and repeatable way during the whole imaging process. Concurrent movement of the camera and light source with the hyperspectral camera attached to the same translator system ensures that the illumination stays constant for every acquisition. To make sure that the light intensity is not too high for the painting, temperature at the light exposed painting surface was constantly monitored using a thermal camera.

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*NEO – Norsk Elektro Optikk A/S, `www.hyspex.no`

X-ray fluorescence spectroscopy

A portable XRF Thermo-Niton XL3t 950 He spectrometer with a GOLDD+ detector, and Ag anode target, spot diameter 3 mm, voltage 50 kV and current up to 40 µA was rented from Holger Hartmann AS. The XRF instrument was used for the acquisition of spectra on a darkened blue area of the painting, and also on a red area.

Methods and materials

An overview of the hyperspectral pigment identification method used in this study is illustrated in Fig. 1. First, for every pigment of interest, its spectrum must be obtained from the hyperspectral image of the painting in question. Then, its matching scores to every pigment available in the spectral library will be computed using a spectral difference measure. Finally, the method will return three matching candidates having the highest matching scores to the pigment target. Note that each pixel in the image as well as pigments in the spectral library are represented in terms of spectral reflectance, at approximately 414-793 nm range.

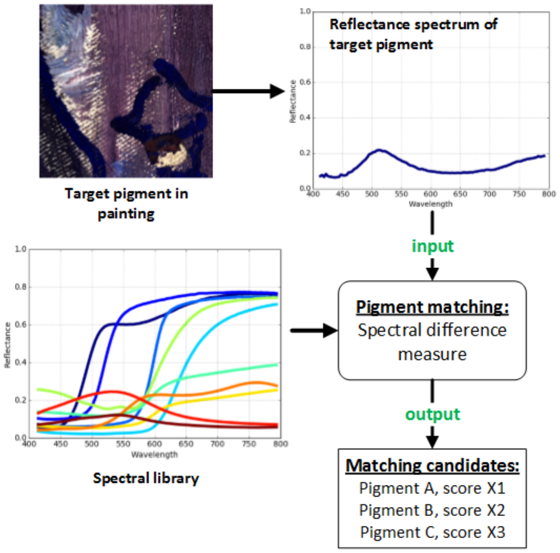

Figure 1. Overview of the hyperspectral pigment identification method used in this study. Pigments are represented in terms of spectral reflectance from approximately 414 to 793 nm.

The case study painting

Old Man in Warnemünde was painted in 1907, when Edvard Munch lived in a seaside village on the Baltic coast in Germany. The canvas painting measures 110.5 x 81 cm and it has the inventory number MM491 (Woll M 755). Based on data obtained from a previous campaign, the XRF analysis is focused on blue and red painted areas marked by samples #6 and #7 in Fig. 2. For the hyperspectral analysis, all 7 points shown in the figure are represented as spectral reflectance functions and will be studied.

Spectral library

A spectral library is a collection of spectral reflectance functions where each entry corresponds to and is characteristic of a certain pigment. Two pigment databases were employed in this study to build the spectral library, as follows:

Figure 2. Image of the painting with the points used for both hyperspectral and XRF analyses: Points 1-7 for hyperspectral pigment identification and points 6-7 for XRF analysis.

- **Kremer pigments**: A total of 304 pure pigments that came in color charts were hyperspectrally acquired by NEO. Each pure pigment was applied on cards with varying gray scales and, thus, each pigment comes with several different shades. Considering the shades as individual colors, the Kremer spectral library has a total of 1052 entries.
- **ENST pigments**: Another pigment spectral library used in this study is one that is obtained from a pigment target that consists of 64 pure and mixed pigments. Spectral imaging of this target was performed using a hyperspectral scanner at C2RMF, Louvre Museum, Paris.

It should be noted that the creation of the two pigments databases was different. Kremer pigments were prepared mostly with water-based binder made of Gum Arabic and were applied on paper by screen-printing, while for ENST the application is unknown. Nevertheless, they are employed based on the knowledge that spectral reflectance properties of paints and coatings in the VNIR range are dictated by pigment properties. Spectral characteristics of other materials present in the paint formulations (e.g., filler and binder) will affect other spectral ranges [5].

Spectral difference measure

In many studies of pigment mapping based on hyperspectral imaging, spectral angle mapper [8] is the most employed similarity measure. Due to its drawback as pointed out in [1, 2], in this study, the shape component of the spectral Kullback-Leibler pseudo-divergence (KLPD) function [11] is employed instead. The mathematical expression of KLPD measure between two spectral functions $KL'(S_1,S_2)$ is given in Eq. 1, where S_i is any arbitrary spectral function as a function of wavelength, λ. Then, as written in Eq. 2, KL is the Kullback-Leibler divergence function, $\bar{S}$ is a normalized spectral function, and k is the corresponding normalizing factor. With this measure of differences, a lower value means a higher similarity or higher matching score.

$$KL'_{shape}(S_1,S_2) = k_1.KL(\bar{S}_1,\bar{S}_2) + k_2.KL(\bar{S}_2,\bar{S}_1) \tag{1}$$

$$KL(\bar{S}_1,\bar{S}_2) = \int_{\lambda_{\min}}^{\lambda_{\max}} \bar{S}_1(\lambda).\ln\frac{\bar{S}_1(\lambda)}{\bar{S}_2(\lambda)}\,d\lambda,\ \text{where} \tag{2}$$

$$k = \int_{\lambda_{\min}}^{\lambda_{\max}} s(\lambda)\,d\lambda, \bar{S} = \left\{\bar{s}(\lambda) = \frac{s(\lambda)}{k}, \forall\lambda \in [\lambda_{\min},\lambda_{\max}]\right\}$$

Results and discussion

Hyperspectral pigment identification

The results of pigment identification based on hyperspectral imaging (see workflow in Fig. 1) are provided in Table 1, where best matching candidates for each pigment sample are highlighted in blue. For sample #1, which will simply be referred to as a green pigment, ENST database suggests that it is a scheele's green ($AsCuHO_3$) with a matching score of 0.829. The Kremer database suggests the presence of two cadmium greens (cadmium green is known with the chemical formula CdS), each with scores of 0.228 and 0.531. Despite their identical pigment names, they are not identical in their chemical nature. Observing the general matching candidate scores of both databases for this particular pigment, it can be said that better results are provided by Kremer database. See also Fig. 3, where Kremer suggested cadmium greens have similar reflectance peak to the target green pigment around 510 nm. On the other hand, the reflectance peak ENST's scheele's green is shifted to the right of the target pigment.

Table 1: Matching pigment candidates as suggested by both ENST and Kremer pigment databases. For each sample, the best matching candidate is highlighted in blue. 'LW' means that the corresponding pigment is mixed with lead white.

Pigment samples	ENST pigments		Kremer pigments	
	Name	Score	Name	Score
#1, green (gr.)	scheele's gr.	0.829	cadmium gr.	0.228
	prussian bl.	1.013	cobalt gr.	0.356
	lamp black	1.200	cadmium gr.	0.531
#2, yellow (yl.)	zinc yl.	0.764	isoindole yl.	0.302
	lemon yl.	0.766	cobalt yl.	0.346
	aureolin	0.985	permanent yl.	0.362
#3, vivid blue (bl.)	ultramarine	1.313	ultramarine bl.	0.164
	cerulean bl.	1.749		0.211
	monastral bl.	3.140		0.229
#4, orange	red ochre	0.258	terra ercolano	0.081
	burnt sienna	0.385	terra pozzuoli	0.127
	brown madder	0.528	French ochre soforouge	0.154
#5, turquoise	viridian, LW	0.389	indigo	0.516
	indigo, LW	0.643	cobalt gr.	0.638
	viridian	1.133	cobalt bottle gr.	0.711
#6, darkened blue	indigo, LW	0.393	indigo	0.328
	monastral bl.	0.760	ultramarine bl.	0.513
	ultramarine, LW	1.432	ultramarine bl.	0.522
#7, red	burnt umber	0.468	caput mortuum	0.089
	brown madder, LW	0.550	iron glimmer violet	0.097
	brown madder	0.641	gubbio red	0.177

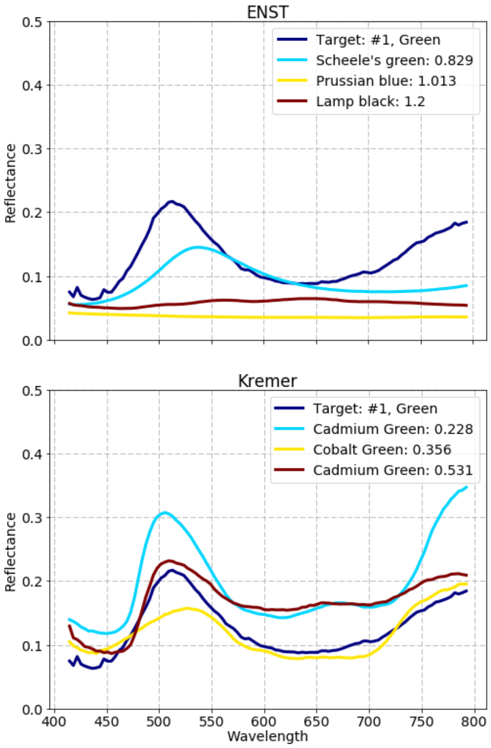

Figure 3. *Spectral reflectance functions of pigment sample #1 green and its matching ENST and Kremer candidates. The best matching candidate is cadmium green with a score of 0.228 and with a peak at approximately 510 nm. X-axes of both figures are identical and is also the case for Fig. 4-7.*

For sample #2 yellow, Table 1 shows that the best matching is isoindole yellow from Kremer database, providing a matching score of 0.302 and aureolin (potassium cobalt nitrite, also known as cobalt yellow) from ENST database with a score of 0.985. Considering that both databases have cobalt yellow in their list of candidates, this pigment can be considered as the right assignment for the yellow sample. As for sample #3 vivid blue pigment, the two databases conclude that it is highly probable to be an ultramarine blue. Kremer database, which has multiple entries of ultramarine blues, give matching candidates of only this pigment. For ENST database, its ultramarine pigment is found to be the best matching candidate with 1.313 score. Fig. 4 shows spectral reflectance functions of the vivid blue pigment and its matching candidates from both databases. For ENST, the most similar shape is indeed given by ultramarine, and not by the cerulean blue and monastral blue. Spectral reflectance functions of all Kremer's ultramarine blue pigments are almost identical in shape.

Fig. 5 allows to observe the shapes of spectral reflectance functions of sample #5 turquoise pigment and its matching ENST

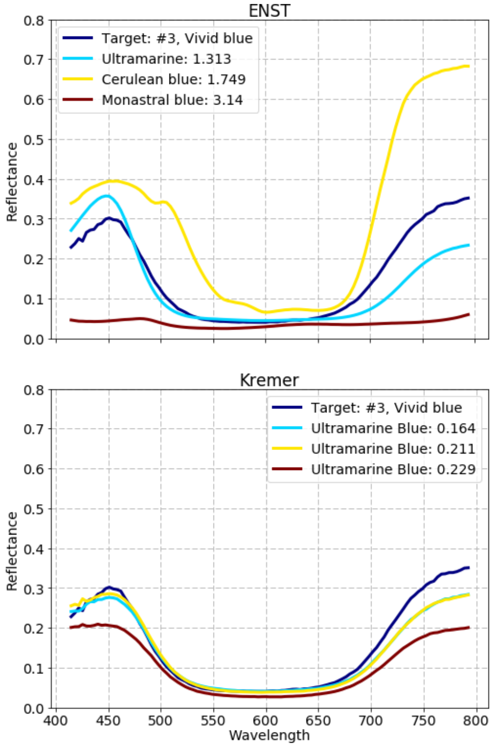

Figure 4. *Spectral reflectance functions of sample #3 vivid blue pigment and its matching ENST and Kremer candidates. Both databases agree on ultramarine blue as the most probable pigment and high similarity in spectral shape can also be observed.*

and Kremer candidates. For ENST's suggestions, the most similar shape by visual observation is given by the pure and mixed (with lead white) viridian pigments (viridian is a blue-green pigment, a hydrated chromium(III) oxide). As for the mixture of indigo and lead white, despite giving a better matching score than the pure viridian, it is evident that it lacks the reflectance peak at around 510 nm. Observing the results given by Kremer database, indigo (violet organic dye) is suggested as its best candidate, with a matching score not significantly different to ENST's indigo, LW. Similar to the case of Kremer-suggested cobalt green pigments, the reflectance peak is at different location than the turquoise pigment sample in question. Finally, for this pigment sample, it can be concluded that the sample is highly likely to be a viridian pigment, possibly mixed with lead white.

Further validation with X-ray fluorescence spectrometry

The hyperspectral identification of pigments faced a limitation that several candidate pigments are obtained but there is no certain way to validate the matching results. To address this issue, a preliminary trial elemental XRF analysis was performed for pigment samples #6 and #7, previously shown in Fig. 2. These two samples are selected based on data points available from a previous campaign carried out before the hyperspectral one.

The elemental analysis performed with XRF on pigment sample #6, which is approximately located at the outline of a human figure outline in the background on the left side of the painting, detected the presence of a lead based pigment (lead white probably), and of ultramarine for blue area (natural or synthetic pigment known for being an aluminium silicate zeolite with a sodalite structure; the synthetic ultramarine has the general chemical formula: $Na_{6\text{-}10}Al_6Si_6O_{24}S_{2\text{-}4}$) [10]. Table 1 suggests that ultramarine blue pigments are being the best matching results for both pigment databases. Fig. 6 also visually confirms the similarity between the sample and its candidates in terms of spectral shape. Furthermore, the ENST suggestion that the ultramarine candidate is a mixture with lead white pigment, was confirmed by the elemental analysis.

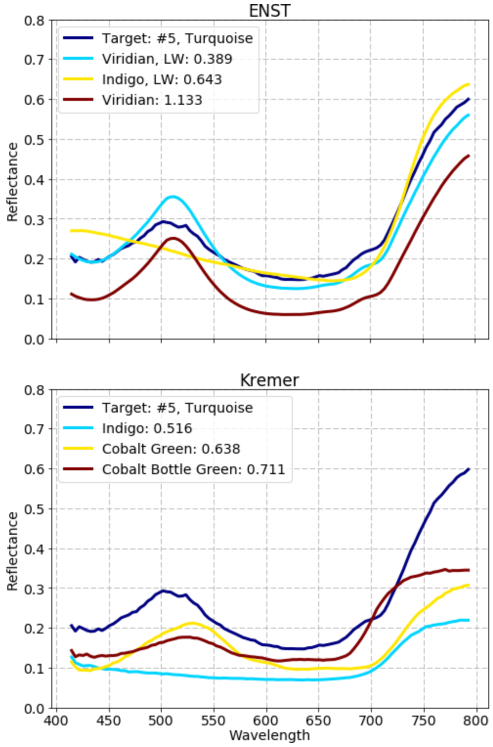

Figure 5. *Spectral reflectance functions of pigment sample #5 turquoise and its matching candidates. Better similarities can be observed for ENST's pure viridian and its mixture with lead white. Kremer's suggested cobalt greens have similar shapes but slightly different peak location at around 530 nm, unlike the pigment sample which is approximately at 500 nm.*

For the red area (sample #7), the following elements have been identified: Ca (calcium based compound), Pb (lead white), Fe (clay minerals or haematite pigment), Zn (zinc white), Hg (vermillion), Cl, Si, P (organic red, possibly a phosphorous containing mordant). The hyperspectral pigment identification in Table 1 suggests the presence of organic pigments (brown madder) but also of a burnt umber (brown or reddish color, obtained by calcination fo raw umber), an earth pigment (iron based oxide), which are both pure and mixed with lead white. Fig. 7 shows as the best matching in terms of shape the Kremer's caput mortuum pigment (variety of haematite/ iron oxide pigment), with a matching score close to 0. Therefore, the hyperspectral attribution of the main pigment for this area to a red-brown earth (burnt umber, or caput mortuum) mixed with some organic pigment is validated by XRF characterization.

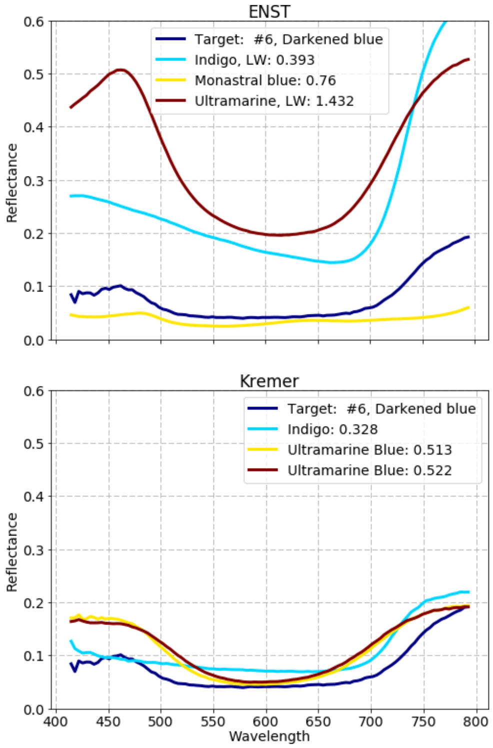

Figure 6. *Spectral reflectance functions of pigment sample #6 darkened blue and its corresponding matching ENST and Kremer candidates. Agreeing with the elemental analysis with XRF, both databases suggest ultramarine blue and in one case its mixture with lead white. Despite being the best matching candidates from both databases, indigo pigments lack the peak reflectance at approximately 460 nm.*

To summarize, the two cases of complementary identification of pigments using hyperspectral imaging and XRF analysis on blue and red areas of the painting showed that the proposed hyperspectral imaging based method is valid. The use of two databases, Kremer and ENST, is a first step and further improvements can be done, especially in creating more complex mixture of pigments with different binding media (oil, tempera, other techniques) and between different pigments.

Conclusion

The preliminary case study of hyperspectral imaging presented in this paper focused on the complementary use of this tool and of XRF spectrometry for pigment identification of a painting by Edvard Munch, *Old man in Warnemünde* (1907). The employed methodology showed the usefulness of two pigment databases, where one consists of hundreds of pure pigments (Kremer) and another of 64 pure and mixed pigments (ENST).

From the results of this study, it can be concluded that pigment databases made of different binding media and with different ways of application are useful for the identification of pigments based on the hyperspectral technology. It can also be concluded that spectral characteristics in the VNIR range are dictated by pigment characteristics which is represented in terms of spectral shape. XRF analysis on areas of the paintings was also useful to validate results obtained by the hyperspectral-based method.

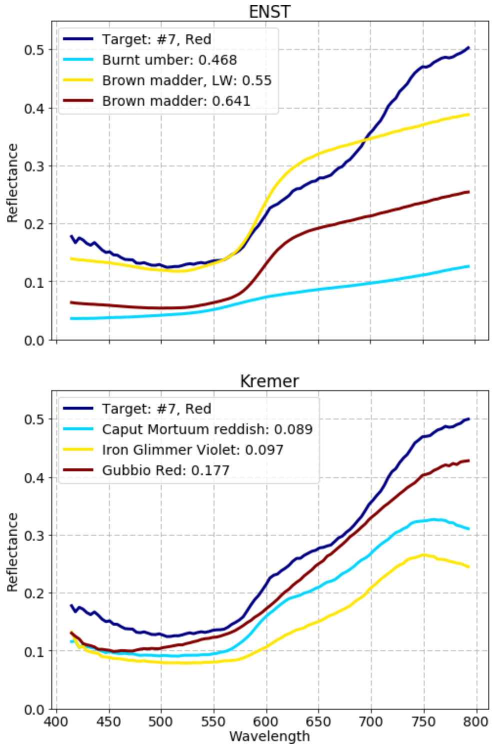

Figure 7. *Spectral reflectance functions of pigment sample #7 red and its corresponding matching ENST and Kremer candidates. Visually, the best match does agree with the matching score, i.e., caput mortuum.*

Another novelty of this study is the use of a new spectral difference function in the matching process instead of the widely used spectral angle mapper, which has previously been found to be inaccurate for pigment identification. In this study, the shape component of spectral Kullback-Leibler pseudo-divergence was employed and the results were shown to be promising. A limitation was found in the function, which measures overall differences in spectra. This causes the measure to be less sensitive to pigment characteristics located in narrow range of bands.

As a further work, in addition to improving the spectral difference measure, a more extensive study on artists' pigments using both hyperspectral and XRF or other compositional analyses are required in order to have the full range of parameters useful for pigment identification. There is also a need to extend the spectral range of the hyperspectral-based method in order to account for the effects of other materials such as binders and fillers on the color changes of the pigments.

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Author Biography

Hilda Deborah received her BS in computer science from the University of Indonesia (2010), MS from Erasmus Mundus Color in Informatics and Media Technology (2013), and PhD in computer science from the University of Poitiers in France and NTNU in Norway (2016). She is currently a researcher at the Norwegian Colour and Visual Computing Laboratory, NTNU. Her work focuses on the development of spectral mathematical morphology framework and applications in the cultural heritage domain.

Jin Strand Ferrer is working as a paintings conservator at the Munch Museum in Oslo. She is currently writing on a project-based Master thesis (University of Oslo). Her research focuses on colour changes in art works in the Munch Museum collection.

Irina C.A. Sandu received her PhD in Chemistry in 2003 from the Gh. Asachi Technical University in Iasi, Romania and since October 2016 is the new Conservation Scientist of the Munch Museum in Oslo. Her research interest focuses on the study of art materials and techniques, degradation mechanism of modern art materials, and also on finding and testing new materials and methods for the characterization and conservation of cultural heritage. She is author of several books and chapters and more than 150 publications in peer-reviewed journals and conference proceedings in heritage and conservation science fields.

Sony George received his PhD in Photonics in 2012 from Cochin University of Science and Technology, India. After working as a researcher from 2012 to 2016 at the Norwegian Colour and Visual Computing Laboratory, he has joined (since 2017) as an associate professor in the same group. His research interests include colour science, multi-spectral and hyper-spectral imaging, spectral image quality, and digital documentation of cultural heritage artifacts.

Jon Y. Hardeberg received his sivilingeniør (MSc) degree in signal processing from the Norwegian Institute of Technology in Norway (1995) and his PhD from Ecole Nationale Supérieure des Télécommunications in France (1999). He is currently Professor of Colour Imaging at the Dept. of Computer Science at NTNU and a member of the Norwegian Colour and Visual Computing Laboratory. His current research interests include multispectral colour imaging, print and image quality, colorimetric device characterisation, colour management, and cultural heritage imaging, and he has co-authored more than 200 publications within the field.