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## Four-flux model combined with optical coherence tomography technique for non-destructive testing of the colour ground layers of the paintings

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#### ABSTRACT

The colour of the ground layers of a painting has an influence on its visual appearance. In addition to the commonly used white ground layers, other colour ground layers have been used, for example, the grey ground layer used in Peter Paul Rubens's painting Portrait of Clara Serena Rubens helps the colour transition of the skin tones. Understanding the effects caused by the colours of the ground layers is of significance for both technical art history and conservation. Optical non-destructive testing (NDT) techniques are useful tools for the investigation of paintings, for example, optical coherence tomography (OCT) can be used to study the surface and subsurface layers non-destructively. In this work, the interaction of light with paint and ground layers is modelled to supplement OCT measurements of paintings with ground layers. A previously described near-infrared light range OCT system provides high spatial and depth resolution measurements. A four-flux model has been developed for analysing the light interaction in the paint and ground layers. This model considers forwards-propagating collimated light, backwards-propagating collimated light, forwards-propagating diffuse light and backwards-propagating diffuse light. The model uses the optical material properties, including refractive index (RI), absorption and layer thickness, as input. This paper describes the construction of the model and an evaluation of its performance by comparison with OCT data.

Keywords: Four-flux model, non-destructive testing, colour, painting, ground layer

#### 1. INTRODUCTION

The colour of the ground layers of a painting has an influence on its visual appearance. Figure 1 (a) shows a painting named Portrait of a Young Girl by Peter Paul Rubens.<sup>1</sup> The ground layers (see Figure 1 (b)) are a cool grey layer and a reddish layer and thet help the colour transition of the skin tones.



Figure 1. The structure of the painting and the structure of the model. (a) The painting Portrait of Clara Serena Rubens (Liechtenstein Museum, Vienna);<sup>1</sup> (b) The structure of the painting; (c) The structure of the proposed optical model.

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In the Down to the Ground Project,<sup>2</sup> the researchers are interested in the colour of the ground layers including the geographical spread and the role of the colour ground layers in the paintings and non-destructive techniques for the measurement of the ground layers. Optical coherence tomography (OCT) has been used for the detection of paintings to explore spectral and depth information for different layers non-destructively.<sup>3,4</sup> In this work, an optical model to analyse the transition of the colour of the ground layers with the assistance of an OCT system will be proposed. The OCT system is used for measuring the thickness of the layers of the paintings. Although in this work, the thicknesses of the samples' layers are well-controlled by a draw-down bar to achieve fixed values of single layers, the thickness and thickness variation for real painting without thickness information would be a challenge for implementing optical models based on a four-flux model. The reason for using the four-flux model to simulate the light propagation in different layers of the paintings is that the model only considers four fluxes in forward and backward directions which has the advantage of high-speed calculation. The simulated reflected light spectra from the painting will be compared with the measured spectral data by a hyperspectral imaging system. The proposed optical model will be shown in Section 2. In Section 3, the optical and geometrical parameters will be measured. Results and Discussion and Conclusions are in Sections 4 and 5.

#### 2. FOUR-FLUX MODEL

The structure of the model is shown in Figure 1 (c). There are two layers to be measured in the ground layers. They are a cool grey layer (lead white in linseed oil) and a reddish layer (red ochre in linseed oil). In this simplified model, the upper and lower ground layer is in contact with the air. The incident collimated light propagates into the grey ground layers at normal incidence from the air to the cool grey layer and then transmits to the reddish layers. At the interfaces of air and the cool grey layer and the interfaces of the reddish layer and the air layer, the Fresnel Reflection is taken into consideration. In the ground layers, four flux<sup>5</sup> (collimated light in forward  $(F_c^+)$  and backward  $(F_c^-)$  direction, diffused light in forward  $(F_d^+)$  and backward  $(F_d^-)$  direction) are considered. The scatters (pigments) in the ground layers scatter light and transfer collimated light to diffused light. The relationship between the four fluxes may be expressed in Equations 1-4.

$$\frac{\mathrm{d}F_{c}^{+}(z)}{\mathrm{d}z} = -(k + S_{f} + S_{b})F_{c}^{+}(z) \tag{1}$$

$$\frac{\mathrm{d}F_c^{-}(z)}{\mathrm{d}z} = (k + S_f + S_b)F_c^{-}(z)$$
(2)

$$\frac{\mathrm{d}F_d^+(z)}{\mathrm{d}z} = -(k + S_f + S_b)F_d^+(z) + (S_f + S_b)F_d^-(z) + S_fF_c^+(z) + S_bF_c^-(z)$$
(3)

$$\frac{\mathrm{d}F_d^-(z)}{\mathrm{d}z} = (k + S_f + S_b)F_d^-(z) - (S_f + S_b)F_d^+(z) - S_fF_c^-(z) - S_bF_c^+(z) \tag{4}$$

where z is the depth. k is the absorption coefficient,  $S_f$  and  $S_b$  are the forward and backward scattering coefficients, which are calculated by Mie theory when the pigments are assumed as spherical particles.

Equations 1-4 can be solved with a similar process from Slovick et al.'s work<sup>5</sup> with our Matlab code and transfer matrix is used for multiple layer cases. By the four-flux model, the reflected light  $(F_c^-(0) + F_d^-(0))$  will be calculated by matrix  $M = M_i M_l M_i$ , where  $M_i$  is the transfer matrix for light at interfaces according to Fresnel equations and  $M_l$  is the transfer matrix for light in the layers. The relationship between the incident light and the output light can be express as:

$$\begin{bmatrix} F_c^+(d) \\ F_c^-(d) \\ F_d^+(d) \\ F_d^-(d) \end{bmatrix} = \begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{34} \\ M_{41} & M_{42} & M_{43} & M_{44} \end{bmatrix} \begin{bmatrix} F_c^+(0) \\ F_c^-(0) \\ F_d^+(0) \\ F_d^-(0) \end{bmatrix}$$
(5)

The reflectance (R) can be calculated by:

$$R = \frac{F_c^-(0) + F_d^-(0)}{F_c^+(0)} = \frac{M_{12}M_{44} + M_{32}M_{44} - M_{34}M_{42}}{M_{22}M_{44} - M_{24}M_{42}}.$$
(6)

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#### **3. PARAMETER MEASUREMENT**

The parameters for the model that need to be measured are the thickness of the samples by an OCT system and the reflected light from the samples by a hyperspectral imaging system.

The thickness of the red ochre layer was measured by an OCT system. The central wavelength of the OCT system was around 1550 nm (a superluminescence diode, FESL-1550-20-BTF, Frankfurt Laser Company). Figure 2 shows the experimental setup of the OCT system used for the depth measurement of a painting. The measured light signals were monitored by a computer in real-time. The refractive index (RI) of the red ochre used is 2.682 (Fe<sub>2</sub>O<sub>3</sub> at 1550 nm (Iron sesquioxide, Hematite)<sup>6</sup>). The locations of the front and back surfaces are determined by the reflected light peaks by the OCT system. In this OCT system, the direct output of locations are based on the refractive index of air (RI=1) and the calibrated locations based on the RI of red ochre are shown in Figure 3. The location of the front surface is at 0.063 mm and the location of the back surface is at 0.150 mm. The distance between the front and the back surface is about 0.087 mm. This result is close to the manufacturing data of this red ochre sample (0.100 mm thickness),<sup>7</sup> so the thickness of 0.100 mm was used for simulation in the model.

The reflected light was measured by a hyperspectral imaging system (SPECIM, Spectral Imaging Ltd.). As shown in Figure 4, the green and the blue regions were measured by the system to obtain the light reflection from the cases of the red ochre layer and lead white on top of the red ochre layer respectively. The mean values of the measured spectral data were obtained by averaging the randomly selected pixels in the blue regions and the green regions. Table 1 shows the parameters used for the light propagation in the ground layers with the four-flux model.



Figure 2. Measurement setup for optical coherence tomography.



Figure 3. The result of the red ochre sample by the measurement by the optical coherence tomography. The front and back surfaces of the paint layer are labelled.



Figure 4. Measurement by a hyperspectral imaging system. The reflected light is collected within the regions of green and blue rectangular in the photo. The lead white pigments are painted on a glass slide which is put on top of the red ochre sample.

Parameter	Value	Source
The thicknesses of cool grey layer	0, 1  or  10,  and	used in simulation
	$100\mu{ m m}$	7
The thickness of reddish layer	100 µm	7
Diameter of lead white	$0.1, 0.5\mu{ m m}$	used in simulation
Diameter of red ochre	$0.3, 0.6\mu{ m m}$	used in simulation
Volume ratio of lead white	49.5%	
	(mean density $= 6.78 \mathrm{g/mL}$ )	8,9
Volume ratio red ochre	31.3%	estimated by author by using
		similar red ochre paints' data
	(mean density $= 4.75 \mathrm{g/mL}$ )	10
The refractive index of the medium (linseed oil)	1.48	11
The mean density of linseed oil	$0.93\mathrm{g/mL}$	12

Table 1. The parameters used for the simulation.

#### 4. RESULTS AND DISCUSSION

#### 4.1 Experiment

The red ochre sample, the lead white sample on a glass slide, and the lead white sample on a glass slide on top of the red ochre sample were measured by the hyperspectral imaging system. The results of the reflected spectra are shown in Figure 5. The red line shows the spectra reflected from the red ochre sample. There is a higher reflection in the red light region and near-infrared light range as would be expected from a red pigment. The lead white sample has a relatively flat spectra and the reflection values decrease from about 35% at a wavelength around 450 nm to about 20% at a wavelength around 1000 nm. When the lead white sample was put on top of the red ochre sample, the final visual appearance is influenced by both of the two colour ground layers, see the blue line in Figure 5.



Figure 5. The experimental results by the reflected light measurement with a hyperspectral imaging system. The red line shows the reflection from the red ochre sample. The green line shows the reflection from the lead white sample on the glass slide. The blue line shows the reflection from the lead white sample on the glass slide on top of the red ochre sample. The layers' thicknesses are 100 µm.

#### 4.2 Model

With the parameters shown in Table 1, the simulated reflected light signals were modelled using the four-flux model introduced in Section 2 and are shown in Figure 6. The simulated spectral range for the red ochre sample is from 400 nm to 1000 nm which is identical to the measured spectral range by the hyperspectral system. The simulated spectral range for lead white is restricted to up to 800 nm because part of the optical properties of lead white was obtained by a UV(Ultraviolet)-vis(visible) spectrometer and the spectral range of this UV-vis spectrometer restrict the simulated spectral range when lead white is taken into account. The red ochre layer (100 µm thickness) is beneath a lead white layer. The thicknesses of the lead white layer were set at 0 µm, 1 µm or 10 µm, and 100 µm. The diameters of red ochre and lead white are set at 0.3 µm and at 0.1 µm in Figure 6 (a) and are set at 0.6 µm and at 0.5 µm in Figure 6 (b) to show the influence by the setting of the sizes of pigments. The simulated spectra of the red ochre layer (in red dots) (Figure 6 (a)) have a similar spectra shape when the spectra are compared with the experimental result of the red ochre layer shown in Figure 5 but the intensity of the simulated lead white layer (1 or 10 µm in Figure 6 (a) and (b)) is on top of the red ochre layer, the spectra were influenced by the spectra reflected by the lead white layer. When the lead white layer increases to 100 µm, the lead white layer has the majority influence on the visual appearance of the ground layers.



Figure 6. The simulated results of the lead white layer on top of the  $100 \,\mu\text{m}$  thickness red ochre layer (red lines and blue dots) and the simulated results of the single lead white layers (green dots) and red ochre layers (red dots). (a) The diameters of the red ochre pigments and the lead white pigments are set at  $0.3 \,\mu\text{m}$  and at  $0.1 \,\mu\text{m}$ . (b) The diameters of the red ochre pigments and the lead white pigments are set at  $0.5 \,\mu\text{m}$ .

Compared with the experimental results of the lead white layer in Figure 5, the intensity of the simulated results is higher than the experimental results, especially for the case in Figure 5 (b). The blue dots and green dots show the different thicknesses of the lead white layers used for simulations and the intensity varies for different thicknesses, which infers that the simulated results will be influenced by the parameters used. For example, the thickness variation of the paint samples has an influence on the reflected spectra. Materials' optical

parameters, for example, the absorption of the pigment, also need accurate thickness measurement. These are the reasons that the OCT system is used for thickness measurement. Besides, the sizes of the pigments are also important parameter for the simulation. As shown in Figures 6 (a) and (b), layers with 0.6  $\mu$ m diameter red pigment reflects less light than the cases of layers with 0.3  $\mu$ m diameter red pigment for the same thickness. However, more light reflects from the lead white layer with lead white pigment sizes of 0.5  $\mu$ m diameter than the cases of 0.1  $\mu$ m diameter pigment. Therefore, it is important to choose suitable parameters for the simulations. In this model, pigments are assumed as spherical particles, which may be a reason for the simulated results deviating from experimental results. Therefore, optimizing the model will be a next step of this research.

#### 4.3 Conclusions

A four-flux model was proposed to analyze the light propagation in the painting's ground layers with the assistance of the OCT technique. The simulated results showed a similar spectra tendency compared with the spectral results by a hyperspectral imaging system and the results showed the colour of the ground layers of a painting has an influence on its visual appearance. Optimizing the model to appeal to the pigments used for the paintings will be a next step of this research.

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