

**InfrArtSonic INCO-CT-2005-015338 project:
Development of a Novel System for Non-Destructive Stratigraphy Determination of Artworks
using Acoustic Microscopy and UV/VIS/nIR/mIR spectroscopy**

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Abstract

The target of the current work is the development of an integrated non-destructive portable analysis-diagnosis system for the 3D “sampling”, archiving & reconstruction of painted artworks. The system will be of a unique and invaluable aid for art historians and restorers in the Mediterranean and pan European area. The *InfrArtSonic* system will unify different modalities such as acoustic microscopy and UV-VIS-nIR-mIR spectroscopy. Combining the information provided by these two modalities the system is expected to display the depth profile of the paint layers of an artwork as well as the local distribution of the pigments in each of these paint layers. The *InfrArtSonic* system will constitute an overall, bottom up, powerful tool for the non-destructive detailed documentation of artworks.

The system will provide information on:

1. The stratigraphy overview without any sampling.
2. The authenticity of the artwork.
3. Any original underpainting or previous restoration attempts.

The painting technique and materials (mainly the inorganic ones) used.

Description of the system

A main concern for the documentation of artworks is how to reveal the information that stems from the internal paint layers of artworks (stratigraphy). This information is up to now mainly acquired through analytical spectroscopic methods which require a micro-sampling operation. Most of the times, the objects under study are extremely valuable and therefore must not be subjected to any intervention. This fact constitutes the micro sampling a prohibitive procedure.

Taking into consideration the above mentioned issue the *InfrArtSonic* project deals with the development of a novel and integrated device for the determination of the paint layers of artworks without any sampling, using non destructive acoustic microscopy and UV/VIS/nIR and mIR spectroscopy (figure 1).

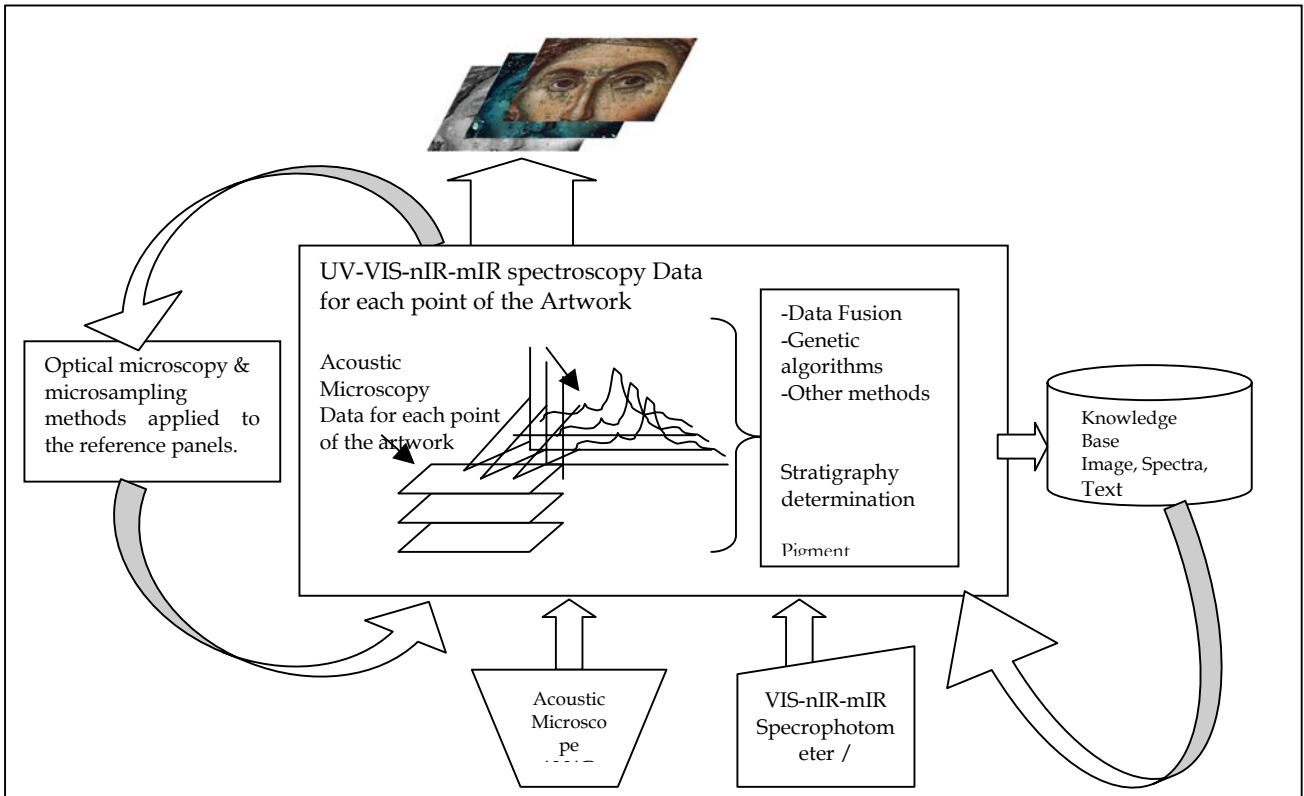


Figure 1: General conception of the system.

Using acoustic microscopy the depth profile of the paint layers' may be defined. Using UV-VIS-nIR-mIR spectroscopy the local distribution of the materials (pigments) will be identified (fig. 1 and fig. 2). Through the combination of this complementary information, the materials of each separate layer will be identified and thus the exact description of the local stratigraphy of the artwork will be obtained.

The applied methodology will be optimized, taking into consideration several special aspects for the acquisition of relevant information. Special methods will be developed in order to fuse all the available information and thus maximize the benefit obtained by the information that is provided from each of these methods. As soon as a definite and exact result is derived from the fusion of the data, a 3D model will be used in order to display the results in a user-friendly visual way.

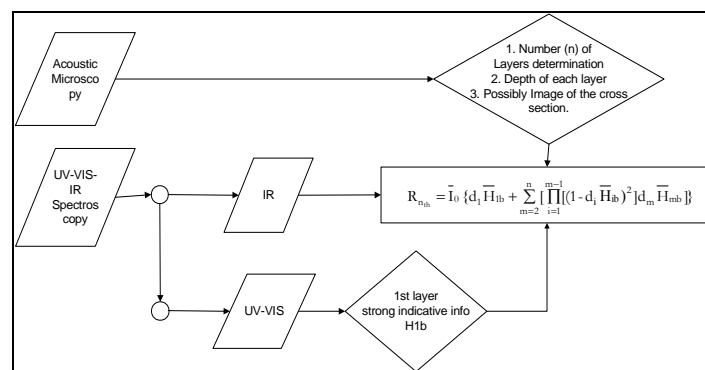


Figure 2: General description of the combination of the complementary information provided by the two devices in the InfrArtSonic system.

Description of the methods

Within the concept described in paragraph 2 , a macroscopic approach is applied in order to determine the material of each paint layer using signal processing and systems theory; The stratigraphies of the artwork is modelled as a sequence of successive layers. Each layer is treated as an independent sub-system. Using acoustic

microscopy we can obtain the number of the layers of the stratigraphy. Using the UV/VIS/nIR/ mIR spectroscopy the identification of the materials existing in each paint layers is achieved.

The proposed algorithm has been initially tested using painted stratigraphies existing in Byzantine artworks. These stratigraphies are additionally thoroughly studied using invasive techniques and optical microscopy. The measured spectra and the reconstructed ones, as calculated by the proposed algorithm are in good similarity.

The acoustic microscope module description and simulation

The acoustic microscope module will be used for revealing the depth profile of the stratigraphies in each point of the artwork, in a region of interest. In order to determine the system characteristics (operating frequency, pulse width etc) the propagation of the acoustic waves in similar materials was simulated. The method for the simulation of the high frequency (100MHz – 250MHz) acoustic waves propagation in the stratigraphies of artworks is the Finite Difference Method (FDM) [6]. The displacement of the material is provided by the equation 1.

$$\rho \frac{\partial^2 \vec{u}}{\partial t^2} = \left[\lambda + \mu + \phi \frac{\partial}{\partial t} + \frac{\eta}{3} \frac{\partial}{\partial t} \right] \nabla (\nabla \vec{u}) + \left[\mu + \eta \frac{\partial}{\partial t} \right] \nabla^2 \vec{u} \quad (1)$$

where

$$\vec{u} = \begin{bmatrix} u_x \\ u_y \\ u_z \end{bmatrix} = \text{the displacement of the material in three dimensions [m]}$$

ρ = the density of the material [kg/m³]

λ, μ = first and second Lamé constant [N/m²]

η = shear viscosity [N*s/m²]

ϕ = bulk viscosity [N*s/m²]

Special reference panels simulating the artworks paint layers of Byzantine iconography style were synthesized in order to test the proposed methods. On these reference panels optical microscopy was applied in order to have an exact image of their cross section for two main reasons:

1. To know exactly the stratigraphy (the layers, their thickness and the grain distribution in the layers (an example is displayed in figure 2).
2. Create digital models of grey level images (figure 3) of them in order to simulate the acoustic wave propagation in these stratigraphies.

The digital models of them based on the microscope images of cross sections of artworks paint layers were created in order to apply the FD method on them.

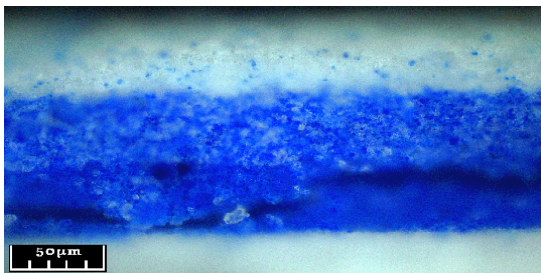


Figure 2: Real cross section of the paint layers (stratigraphy) of artwork

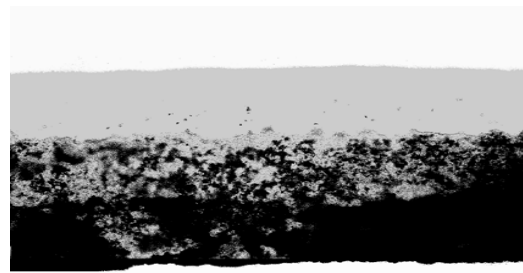


Figure 3: Digital model – grey level image of the cross section of the stratigraphy used for the simulation of the acoustic wave propagation in it.

The main problem is the development of grey-level digital images which will simulate the stratigraphies of the paint layers providing a different grey level for the different materials (figure 4). In order to determine the materials acoustic properties, the acoustic velocity of them was measured using the setup shown in figure 5. The results of the measurements are provided in table 1. Using acoustic waves of frequencies above 100MHz we can have a resolution in the specific pigments of approximately 16μm. The mean value of the acoustic velocities of the materials used in Byzantine hagiography is 1600m/s (table 1).

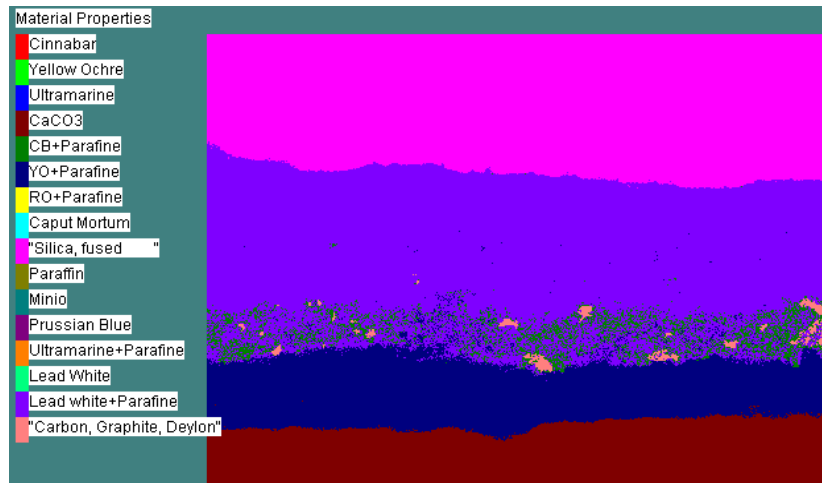


Figure 4: Image of the materials distribution in the stratigraphy used for the FDM application

The grain distribution in these materials is spread between pm and several decades of μm . Taking into consideration that the mean velocity in these materials is of 1660m/sec and the frequency is of 100MHz – 250MHz then the mean wavelength is of 16.6 μm . The grains of 1.6 μm and less are contribute mainly as wave scatterers.

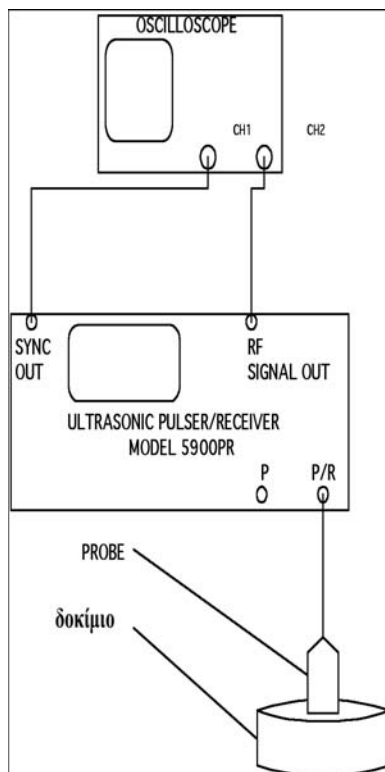


Figure 5: The acoustic velocity of the materials measurement setup

Table 1: The acoustic velocity of the materials used in byzantine iconography.

| Pigment (name) | Acoustic Velocity (m/sec) | Z (Kgr/m ² sec) |
|----------------------|---------------------------|----------------------------|
| 1. Minio | 801.9 | 4100546 |
| 2. Prussian Blue | 1257.1 | 1571870 |
| 3. Cinnabar | 1318.2 | 4083397 |
| 4. CaCO ₃ | 1407.6 | 2412732 |
| 5. Caput Mortum | 1470.6 | 4002477 |
| 6. Red Ochre | 1515.2 | 3059998 |
| 7. Cochineal | 1560.9 | |
| 8. Green Earth | 1608.7 | 2941475 |
| 9. Azurite | 1609.7 | 4259427 |
| 10. Ultramarine | 1666.7 | 2603551 |
| 11. Lead White | 1667 | |
| 12. Cobalt Blue | 1743.6 | |
| 13. Hematite | 1763.6 | |
| 14. Yellow Ochre | 1764.7 | 2615130 |
| 15. Titan White | 1782.6 | |
| 16. Lapis | 2000 | 4037000 |
| 17. Indigo | 2125 | |
| 18. Carbon Black | --- | --- |
| 19. Siena Burnt | --- | --- |
| 20. Malachite | --- | --- |

Having all these parameters calculated and using eq. 1 applied to the digital models of the stratigraphies the simulation of the propagation of the acoustic waves was performed. Some indicative examples of the simulation results are provided in figures 6 - 8 (stratigraphies, digital models and simulated a-scans).

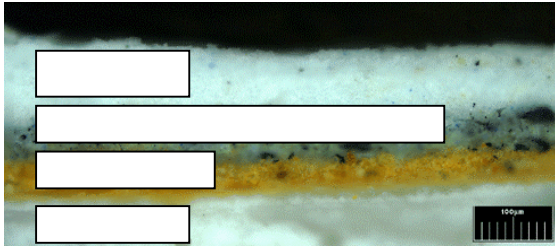


Figure 6: Microscope image of stratigraphy reference sample No 92

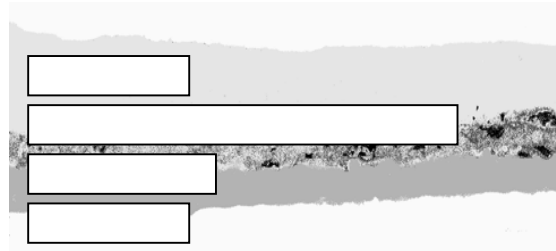


Figure 7: Corresponding digital model of the stratigraphy reference sample No 92 for the FDM computation of the acoustic wave propagation through it (stratigraphy No 92).

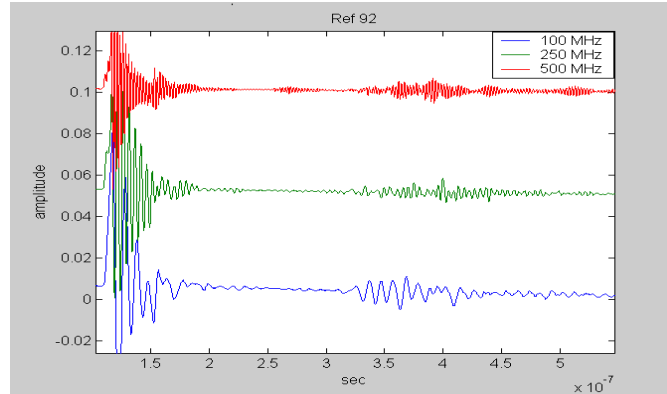


Figure 8: Results (A-Scans of the acoustic wave propagation for different frequencies 100MHz, 250MHz, 500MHz through the stratigraphy No 92 revealing the number of the layers existing in the stratigraphy.

The UV/VIS/nIR spectroscopy module description - simulation

From the UV/VIS/nIR module qualitative information complementary (according figure 1) to the acoustic module information is provided. Monochromatic light for each wavelength in the region between 200nm and 2400nm penetrates sequentially into the artwork from the above device [5]. The light passes through all the individual sub-systems, backscatters from each consecutive transition, and is received by the external probe of the device providing the corresponding diffuse reflectance spectra.

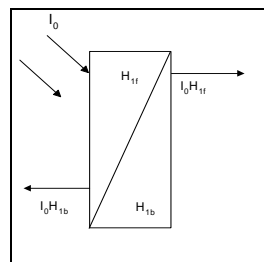


Figure 9: Single sub-system transfer function.

Each sub-system (paint-layer) is assumed to have a forward transfer function H_{if} and a backward transfer function H_{ib} (figure 12). Therefore, as light travels trough each layer, two light components are produced: The forward, or incident component i_{if} , $i_{if}=i_0 * h_{if}$ or in the frequency domain

$$I_{if}=I_0 H_{if} \quad (2)$$

and the backward, or reflected component i_{ib} ;

$i_{ib}=i_0 * h_{ib}$ or in the frequency domain

$$I_{ib}=I_0 H_{ib} \quad (3)$$

where * represents the convolution of the signals, i_0 the incident light and I_0 its corresponding spectrum.

In general, we assume that we have a n-th layered stratigraphy in any point of an artwork, that we may represent as a sequence of successive sub-systems (figure 13). If we have an incident radiation $i_o(\lambda)$ then the forward and backward output after the first sub-system will be $i_{1f}=i_o * h_{1f}$ and $i_{1b}=i_o * h_{1b}$ respectively. The radiation input for the second sub-system is i_{1f} . Similarly, the forward and backward radiation outputs of the second sub-system are $i_{2f}=i_{1f} * h_{2f}$ and $i_{2b}=i_{1f} * h_{2b}$, respectively.

The backward radiation of the second sub-system traverses also the first sub-system. Therefore, the total radiation output from the first and second sub-layers will be:

$$r=i_o * h_{1b} + i_{1f} * h_{2b} = i_o * h_{1b} + i_o * i_{1f} * h_{2b} * h_{1f} \text{ or } R = I_o H_{1b} + I_{1f} H_{2b} = I_o H_{1b} + I_o I_{1f} H_{2b} H_{1f}$$

The total response from a n-th layered system can therefore expressed as:

$$R_{n\text{-th}} = I_o H_{1b} + I_o H_{1f}^2 H_{2b} + I_o H_{1f}^2 H_{2f}^2 H_{3b} + \dots + I_o H_{1f}^2 H_{2f}^2 H_{3f}^2 \dots H_{(n-1)f}^2 H_{nb} \quad (4)$$

Given that the thickness of the paint layers is between 2-100 μ m and that the horizontally scattered power of the enclosed grains is negligible (Figure 11), we may assume that photon energy is practically conserved.

Therefore, for each layer we may accept that $\bar{H}_{1f} = (1 - \bar{H}_{1b})$. Thus, the total response of a stratigraphy containing n layers, R_{nth} , is a polynomial with coefficients only the backward transfer functions of the consecutive sub-systems:

$$R_{n\text{-th}} = \bar{I}_o \{d_1 \bar{H}_{1b} + \sum_{m=2}^n [\prod_{i=1}^{m-1} [(1 - d_i \bar{H}_{ib})^2] d_m \bar{H}_{mb}]\} \quad (5)$$

where \bar{I}_o is the spectrum of the incident radiation, $\bar{H}_{1b}, \bar{H}_{mb}$ are vectors and d_i represents the thickness of each sub-layer/ sub-system.

The simulated spectra are calculated using equation 5. We used the transfer functions, obtained from the spectral responses of the reference panels, the number of the successive paint layers and their thickness values, which had been measured using invasive methods. The reference (transfer functions) are displayed in figure 15.

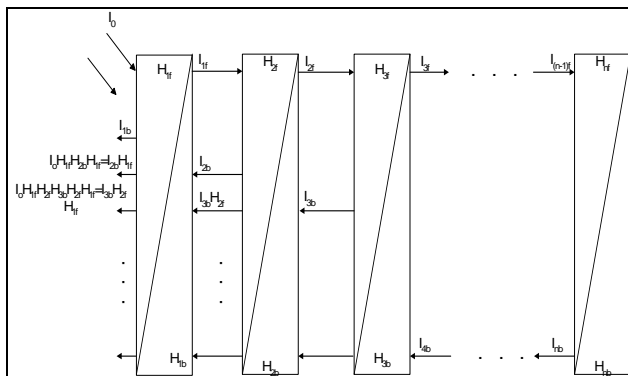


Figure 10: System theory simulation of the UV/VIS/nIR backscattered light from artworks paint layers.

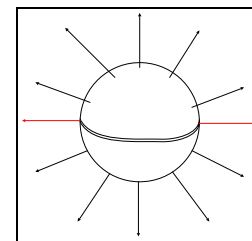


Figure 11: The negligible scattered power from a thin slice of the grain.

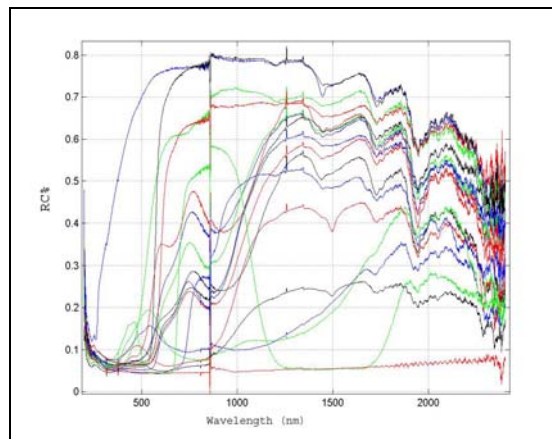


Figure 12: All the transfer functions (spectra of the pigments that constitute the paint layers) of the reference panels.

The simulation of the response of the UV/VIS/nIR module from the stratigraphies that were developed (see experimental setup and measurements) using eq. 5 is displayed in several figures below:

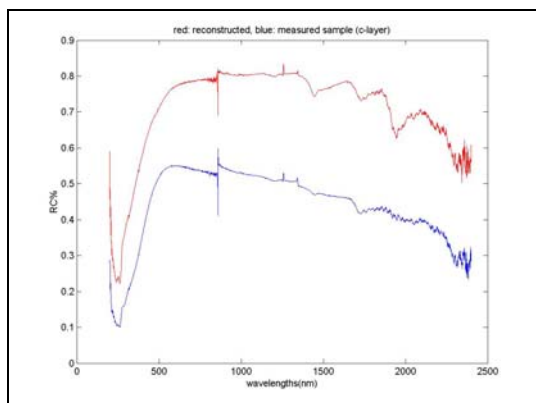


Figure 13: Simulated spectra (red) of the stratigraphy No 104 and the measured one (blue).

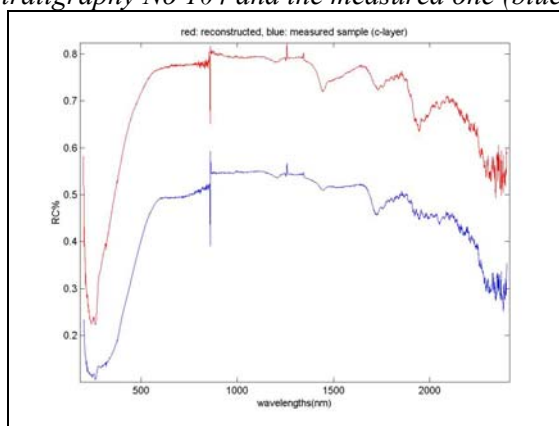


Figure 15: Simulated spectra (red) of the stratigraphy No 125 and the measured one (blue).

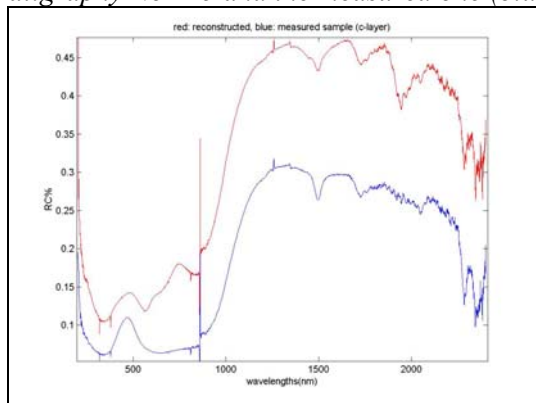


Figure 17: Simulated spectra (red) of the stratigraphy No 122 and the measured one (blue).

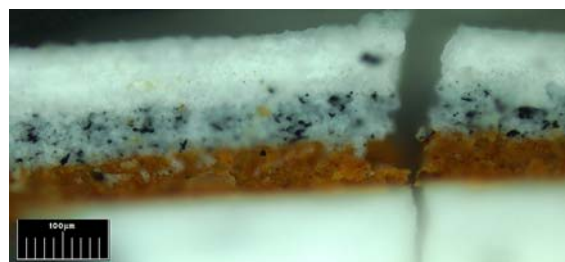


Figure 14: Corresponding stratigraphy of the measured and simulated spectra in figure 9a.
1st layer starting from bottom: Warm Ochre
2nd layer starting from bottom: Lead white + Carbon Black
3rd layer starting from bottom: Lead White

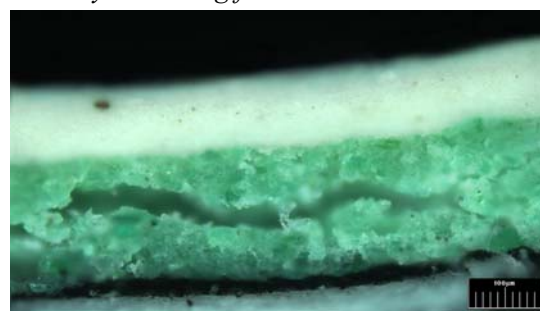


Figure 16: Corresponding stratigraphy of the measured and simulated spectra in figure 10a.
1st layer starting from bottom : Carbon Black
2nd layer starting from bottom : Malachite
3rd layer starting from bottom : Lead white

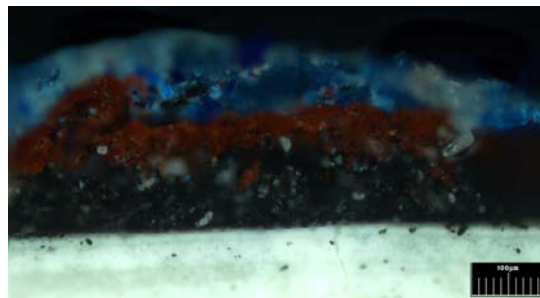


Figure 18: Corresponding stratigraphy of the measured and simulated spectra in figure 11a.
1st layer starting from bottom : Carbon Black
2nd layer starting from bottom : Caput Mortuum
3rd layer starting from bottom : Azurite

The measured UV/VIS/nIR spectra are labelled in blue and the corresponding reconstructed spectra using the proposed algorithm are labelled in red.

Conclusions and further steps

The measured spectra and the reconstructed ones using the proposed algorithm are satisfactorily similar. The next issue is the identification of each paint layer of the stratigraphy. In order to optimize the paint layers identification procedure, we divided the detected spectrum into sub-spectral areas between 200nm-400nm, 400nm-1500nm and 1500nm-2400nm. In these sub-spectral areas, the transfer functions of the pigments appear

to have considerable uniqueness. This happens mainly in the sub-spectral areas between 200-800nm and 800-1550nm. We observed that in this spectral area all the transfer functions of the pigments have significant differentiations among them (figure 15).

We simulated all the possible stratigraphies-combinations using the transfer functions for each layer from the reference panels. Each of these simulated spectra was then compared with the measured spectra of the experimental painted stratigraphies. The maximum correlation coefficient between the measured spectra and the simulated ones was obtained in the case where the corresponding layers of the stratigraphies were of the same material. The final target is the identification of each sub-layer without simulating all possible stratigraphy combinations.

Towards this target the use of the acoustic microscopy in the non destructive stratigraphy determination of artworks is proposed for the first time. The results of the acoustic microscopy are very promising for the determination of the number of the paint layers in the algorithm. The comparison of the time of flight of the echoes of the paint layers are in accordance with the calculated velocity of the sound in these material and presented in table 1. Even higher frequency acoustic signals must be used and examined as well, after devising an optimum way of coupling the transducer with the artwork without damaging it.

The combination of the Acoustic microscopy method with the nIR spectroscopy method is also applied for the first time in this field.

More work must be done in the interpretation of the nIR information using signal processing techniques. The parameterization of the thickness of the layers and the depth in Eq. 1. will improve the results. For the time, the first layer is determined with a success of 90%, the second layer with a success of 66% and the subsequent layers with low success.

Last but not least the outcome of this work is the basis for the development of an innovative device in the field of NDT of artworks will be integrated combining acoustic microscopy with Vis-nIR-mIR spectroscopy. This scientific instrument will be capable to study the existing stratigraphy of the artworks. This will constitute a unique research device for art historians and restorers in the Mediterranean and in pan-European area.

The *InfrArtSonic* system will combine two modalities (figure 1):

1. An Acoustic Microscope for Art Diagnosis;
2. A Visible-near Infrared – mid Infrared (VIS-nIR-mIR) reflectance spectroscope (Infrared System for Art Diagnosis -ISAD);

Both sub-modules of the system (AMAD and ISAD) will be incorporated in real time using common hardware where it is needed as far as the acquisition and storage process is concerned. The information that these subsystems (AMAD-ISAD) will acquire will be complementary. The AMAD will provide with an “image” of the paint layers distribution in depth and the ISAD the materials, mainly the inorganic ones, in each layer. This information will be fused using data fusion techniques. The final result will be the 3D stratigraphy reconstruction.

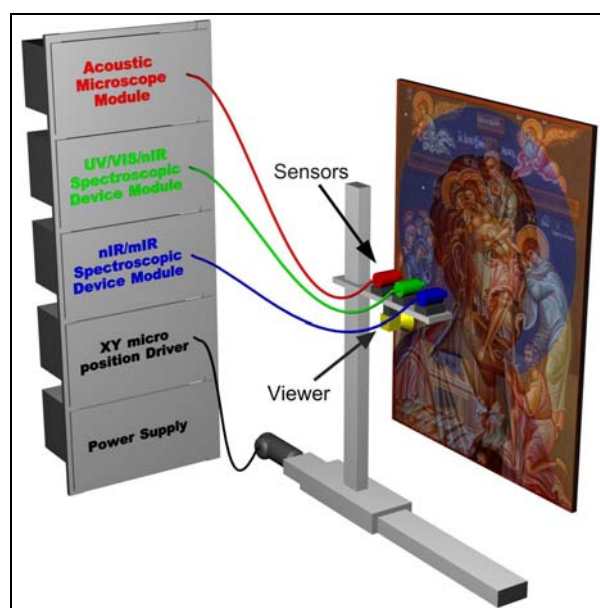


Figure 19: Initial setup design of the system.

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