

## Article

# Dynamic Laser Speckle Technique Sensing Long-Term Changes Caused by Painting Treatments in Restoration of Paintings

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**Abstract:** Dynamic laser speckle is applied as a reliable sensor of activity in all sort of material. Traditional applications are based on a time rate that is usually higher than 10 frames-per-second (FPS). Even in drying processes, where there is a high activity in the first moments after the painting and a slow activity after some minutes or hours, the process is based on the acquisition of images in a time rate that is the same in both moments of high and low activity. In this work, we present an alternative approach to follow the drying of paint and the other processes related to restauration of paintings that takes long-term to reduce the activity. We illuminated, using three different wavelength lasers, an accelerator (Cyclododecane) and a varnish used in restauration of paintings and monitor them at long-term drying using an alternative fps, comparing the results to the traditional method. The work also presents a way to do the monitoring using a portable equipment. The results present the feasibility to use the portable device and show the improvement in the sensitivity of the dynamic laser speckle to sense long-term process regarding the drying of Cyclododecane and Varnish used in restauration.

**Keywords:** dynamic speckle; activity; temporal history speckle pattern; Varnish; Cyclododecane

## 1. Introduction

Digital holography [1,2], a well-known speckle interferometry, is generally used for the measurement of drying paints, however demanding complex experimental configurations that limit its use. [3,4]. One can see the use of other digital speckle pattern interferometry, such as the Shearography, being applied in art objects, particularly in canvas and panel paintings [5,6], but demanding a similar experimental configuration of holography.

In art restauration, usually are used treatments that change the visual aspect of the art objects, such as in paintings; the changes can be in the brightness and in the light saturation, as it is the case of Varnish application[7–9].

In other hand, the processes that provide fixation and consolidation of the restauration in paintings also use a substance, the most common is the Cyclododecane, that form bright artefacts in the surface, thus changing the final prime aspect of the object [10–12].

One way to control the interference of the restauration in art objects is the managing of the drying times, helping in the definition of the best manipulation time and the reduction of artefacts in the surface.

The timing of solvent evaporation present in inks can be measured using thermogravimetric [13], as well as, using the weighing process during drying [14], with some limitations in *in situ* restauration.

Dynamic laser speckle (DLS), or biospeckle laser (BSL), is a non-destructive technique that can be used to monitor biological and non-biological activity in many areas with application in different material, and thus with different behavior. A complex application is in fluids such as in the motility of bovine frozen semen [15], in blood flow [16,17], in bacterial chemotaxis [18] as well as in the reaction of cancer cells of line MEL-RC08 to the application of the drug Colcemid [19]. In fluids, the approaches must be biased to the high movement of the scatterers of light from those in colloid or in biological tissues.

The dynamic of the processes is critical in the DLS, demanding adjustments in the setup as well as in the choice of the best image processing method. In colloids, which is the case of paintings, one can see some reports linking DLS to paint drying [20–23], where the level of activity of the scatterers is highly different from the moment of the application of the ink, with high volatilization, to the end of drying. Thus, a way to monitor it without compromise the observation of the ongoing phenomenon must be evaluated and tested.

It is also possible to observe in the literature the use of a commercial equipment to evaluate ink [24], or drying paint, that also presented an alternative to use the technique out of optical laboratories, which is a challenge regarding many external influences [25], mainly when we are dealing with long-term drying paint processes.

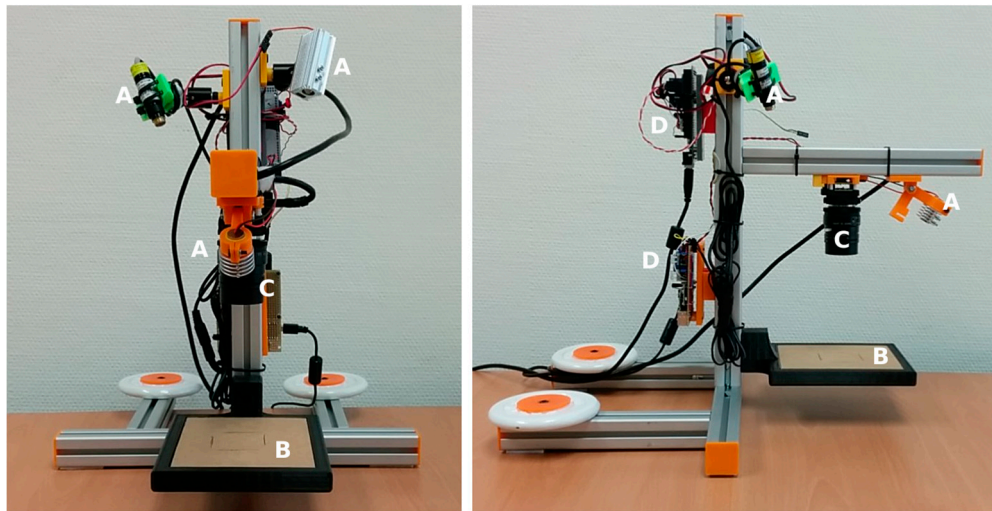
This work aimed to test a portable equipment to monitor the activity of painting treatments during the restauration of paint *in situ* by means of a modified image processing biased to a long-term measurement. Two types of chemicals were tested during drying, evaluated by three laser wavelengths and compared to a laboratorial setup as well as compared to the traditional image processing, all of them validated by a scale monitoring the weight.

## 2. Design and control of portable system

### 2.1. Structural design

Figure 1 shows the elements composing the portable system for dynamic speckle patterns. It can control up to four diode-lasers, allowing rear and frontal illumination by means of prisms and the removable platform. The images can be captured with a CCD camera (See3Cam 2304x1296 pixels, 2.2µm) with no IR filter.

All the elements are assembled together using aluminum T-slots. This structure is sturdy enough to capture stable images. In addition, cushion pads are glue in the base to absorb vibrations. Camera height can be adjusted, and so the laser orientation. The platform B (Figure 1) is removable to place translucent material. This allows a rear illumination setup by means of prisms and laser reorientation. As a portable equipment, it can be moved on to the place of the restoration, for example, and the orientation of camera and lasers adjusted to monitor horizontal and vertical disposition of the paintings.



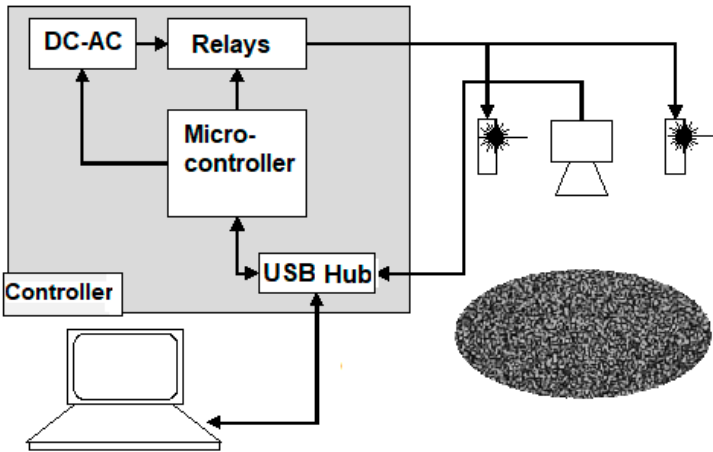
**Figure 1.** Portable experimental setup to get the dynamic laser speckle. A: lasers; B: removable platform; C: camera; D: Laser controller.

## 2.2. Electronic design

Figure 1 shows the elements composing the portable system for dynamic speckle patterns. It can control up to four diode-lasers, allowing rear and frontal illumination by means of prisms and the removable platform. The images can be captured with a CCD camera (See3Cam 2304x1296 pixels, 2.2 $\mu$ m) with no IR filter.

The portable system control was designed and developed at the Universidad Polit cnica de Valencia, Spain, as shown in Figure 2.

The lasers can be turned on/off by means of relays controlled by a microcontroller (Arduino) that is connected to a PC through an USB port. The camera is a USB 3.0 camera (See3Cam 2304x1296 pixels, 2.2 $\mu$ m) with no IR filter, also connected to a computer. A software has been developed to control image capture timing and the laser synchronization. Diode lasers are not heavy-duty components and keep standing still without the interference of external mechanical noise and vibration. Thus, the software can be configured to turn lasers on at a preselected time before the capture. This reduce laser deterioration, and allow its stabilization before a new capture. The software allows to capture burst of images with the different lasers during an experiment.



**Figure 2.** Diagram of portable system controller with the gray box representing the control system of the device connected to the computer and to the lasers and camera.

**3. Validation experimental**

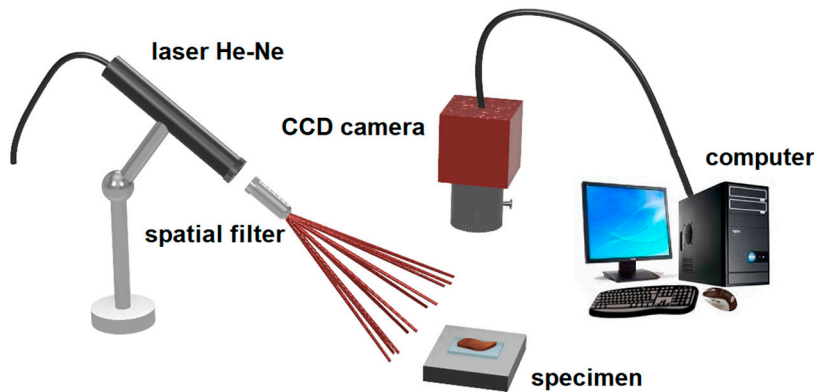
*3.1. Specimen*

A surface of canvas was painted with Varnish and Cyclododecane ink (accelerator), forming a homogeneous layer. The room had 22-23 °C and 51-53% of humidity.

*3.2. Experimental setup employed for non-portable dynamic laser speckle*

Figure 3 presents the experimental setup to acquire the dynamic laser speckle patterns generated in time by the illumination of the samples using a linearly polarized He-Ne laser beam (632.8 nm, 30 mW). The beam size was expanded using a microscope's objective with a 10X magnification in order to have a round illuminated area with 100 mm of diameter, covering a squared area with 40 mm of side.

The images were acquired by a TV zoom lens with a focal length of 50 mm, numerical aperture of f/11 (speckle size was 13.57µm), connected to an Allied Vision Technologies (AVT) CCD Camera (AVT Marlin F-145B, pixel size of 4.65 µm).



**Figure 3.** Experimental setup to get the dynamic laser speckle of a paint drying process.

### 3.3. Protocol to speckle patterns acquisition

First experiment: validation of the portable system; a collection of 64 images (8 bits, 640x480 pixels and auto-exposure 1/125s) was acquired with a traditional experimental system, at the beginning of each minute, along only 10 minutes with a time rate of 10 frames per second. While, with portable system at the beginning of each minute, along 8 hours with a time rate of 10 frames per second. In both cases, canvases of 120 x 120 mm were painted using the Cyclododecane and the Varnish.

Second experiment: comparison of back and forward scattering using the portable equipment. The portable device has the flexibility to get dynamic laser speckle using back and forward scattering approaches, related to reflection and transmission of the light on and through the sample respectively. Where the transmission only can be used when the sample allows the light pass through the sample to the camera. A same layer of Cyclododecane (accelerator) was applied in a glass surface that was illuminated using back and forward scattering approaches at each time, and using the same image time rate and processing presented in the first experiment.

Third experiment: changes in time rate and different lasers compared to weight. Uniform layer was painted on canvases. One of the canvas was placed in the dynamic laser speckle capture system and another placed in a scale to be weighted.

The canvas was weighted using a scale Smart Weight ( $\pm 1\text{mg}$ ) every 5 minutes first. Then every 10 minutes, and finally every 30 minutes.

The canvas was illuminated every minute by the three lasers (Infrared, 808nm, 50mW; Red, 650nm, 20mW and Green: 432nm, 20mW) alternatively, and 20 images were burst at 10 fps (frames per second). Before each image acquiring, the corresponding laser has been connected 5 seconds before starting the burst to assure light stability.

Five sets of images were created after the capture process for every laser:

- Set A: burst of 20 images at 10fps every minute (for fast dynamics).
- Set B, C, D, E: burst of {30, 15, 6, 3} images at {1 image at each 1 minute, 1 image at each 2 minutes, 1 image at each 5 minutes, 1 image at each 10 minutes} every 30 minutes (for slow dynamics).

This allow in the same experiment to measure the drying process by means of: In all cases, image quality was tested to avoid speckle grains with unworthy information about the phenomena. Therefore, the setup was biased to avoid speckle with blurred appearance with saturated areas and to avoid inhomogeneity in accordance with the proposed Quality Test Protocol [26,27].

### 3.4. Methodology to process dynamic speckle images

The dynamics of the speckle variation was monitored by second-order statistics [28], building the matrices THSP (Temporal History Speckle Pattern) and COM (Co-occurrence matrix) using the selection of random points in the prime image to create the THSP [29]. From the COM, we obtained the Absolute Values of the Differences (AVD) method [30], expressed by the Equation 1.

$$AVD = \sum_{ij} COM_{ij} |i - j| \quad (1)$$

where the COM is the Co-Occurrence Matrix related to the THSP, and the  $i$  and  $j$  variables represent the line  $i$  and the column  $j$  of each point of the COM matrix.

$$COM = [N_{ij}] \quad (2)$$



The entries are the number of occurrences (N) of a certain intensity value  $i$  that is immediately followed by an intensity value  $j$ .

In Fig 4, it is possible to see the THSP created instead of using random points in the prime image in the selected ROI.

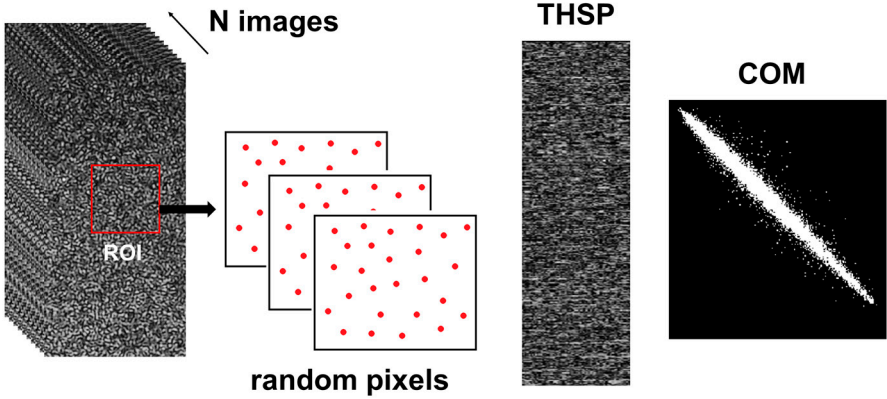
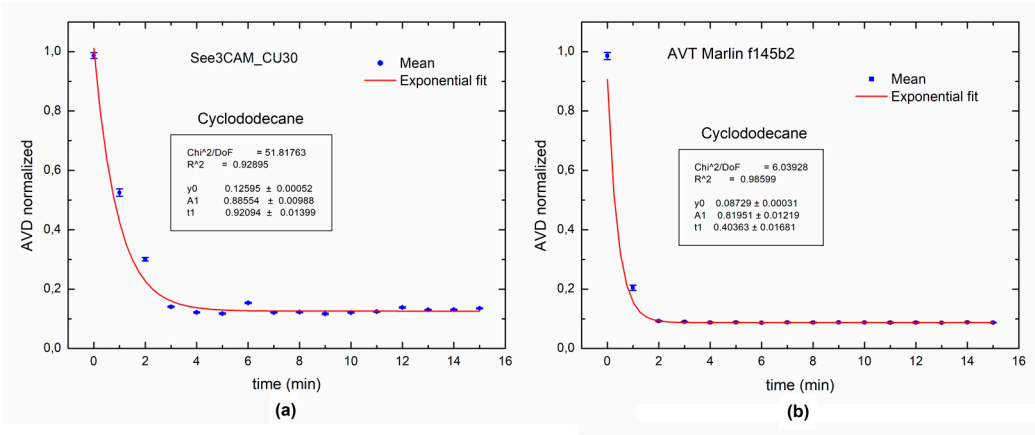


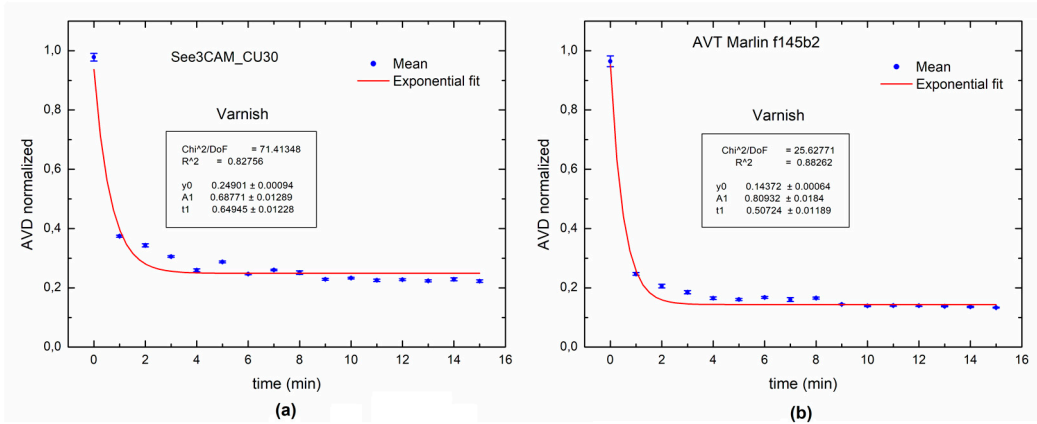
Figure 4. Scheme of THSP and COM construction using random pixels.

4. Results

4.1. Validation of portable device to monitor drying processes in paint restauration

In Figure 5, one can see the AVD index during the drying process of Varnish and Cyclododecane (accelerator) using the traditional experimental configuration and the proposed portable system. The data was fit in an exponential tendency curve, that is the expected behavior of a drying process.

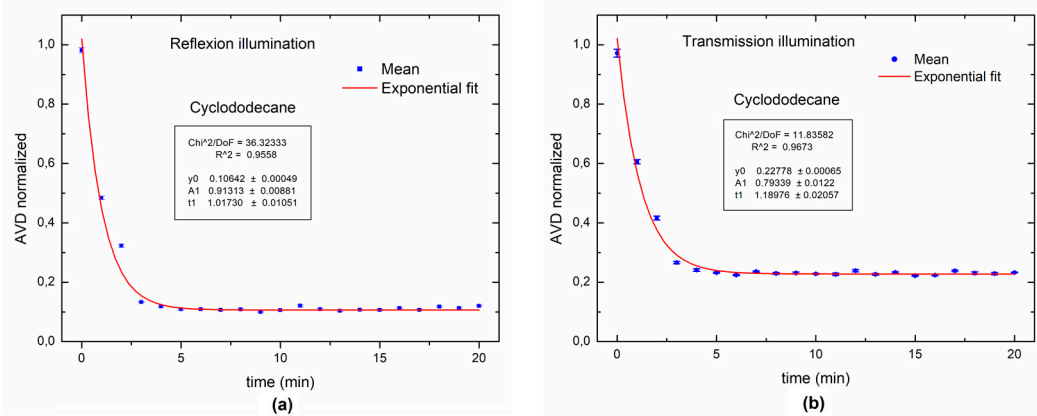




**Figure 5.** Dynamic laser speckle activity levels in paint drying process for each painting treatments (a) Portable system (b) Lab system.

4.2. Comparison of back and forward scattering using the portable equipment

In Figure 6, it is possible to see the AVD index with an exponential tendency curve and using a red laser. Two experimental configurations were used to acquire the data, back and forward scattering. In Figure 6a, the drying process of Cyclododecane was sensed from 1 to approximately 0.1 in normalized values, while in Figure 6b the drying process was sensed from 1 to approximately 0.3 in normalized values.

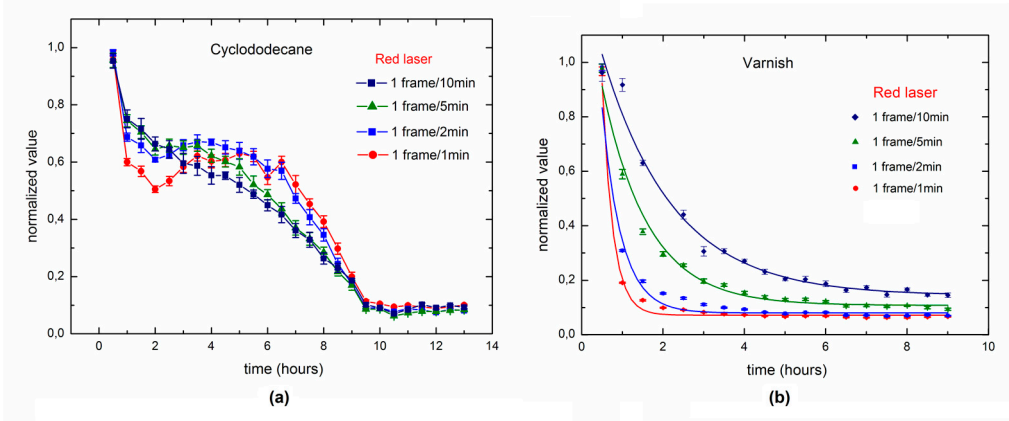


**Figure 6.** Dynamic laser speckle activity levels in paint drying process for Cyclododecane (a) reflection illumination (b) transmission illumination.

4.3. Third experiment: Changes in time rate using different lasers compared to weight

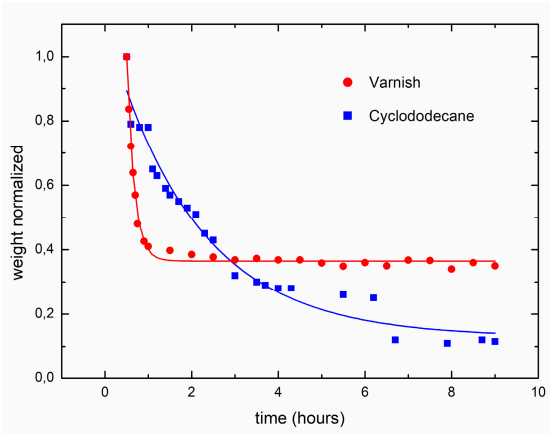
The results of the proposed method to monitor long term activities in the drying process can be seen in Figure 7, where the Cyclododecane (Figure 7a) and the Varnish (Figure 7b) presented their drying in time. In a time-rate of 1 frame per minute, 1 frame at each 2 minutes, 1 frame at each 5 minutes and 1 frame at each 10 minutes, the drying process was monitored during 16 hours. The AVD index were based on 30, 15, 6 and 3 images respectively. In Figure 7a, we can divide the curve in two parts, the first from zero to 3 hours, and the second part from 8 to 16 hours. The zone between 3 and 8 hours can be considered as transient. In the first part, the curve behaves as the drying process presented in traditional measurements of dynamic laser speckle using higher time rate, in the case of Figure 6, for example, 10 fps. It is comparable with the loss of mass monitored by the scale, Figure 8.

However, the similarity was higher in the higher time rates, such as 1 frame per minute (1frame/1min) and 0.5 frame per minute (1frame/2min). While in the second part of the curve, the behaviour is completely different from the traditional curve, presented in Figure 6, with an ability to sense the small changes in the drying surface of the Cyclododecane, where the traditional method presented a flat curve. In Varnish, otherwise, the behaviour presented is like the traditional methods of drying, that we can validate using a scale to weight the loss of mass.



**Figure 7.** Dynamic laser speckle activity levels in paint drying process to each painting treatments (a) Cyclododecane (b) Varnish.

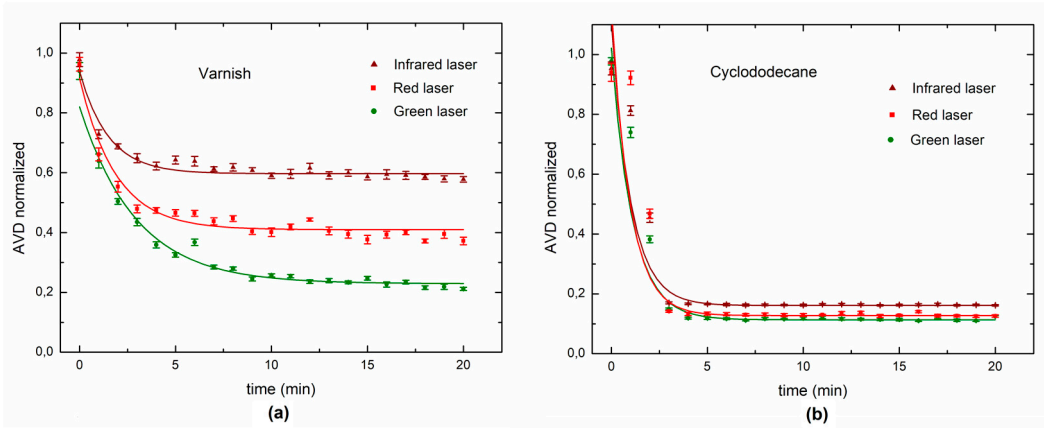
In Figure 8, we can see the weighting process of the evolution of the drying process using a scale. The expected speed of the drying process for the two types of material used in restauration (Varnish and Cyclododecane) is expressed, where the Cyclododecane, despite it is known as accelerator, it takes more time to dry in deep layers.



**Figure 8.** Weighing of drying paint treatments in time.

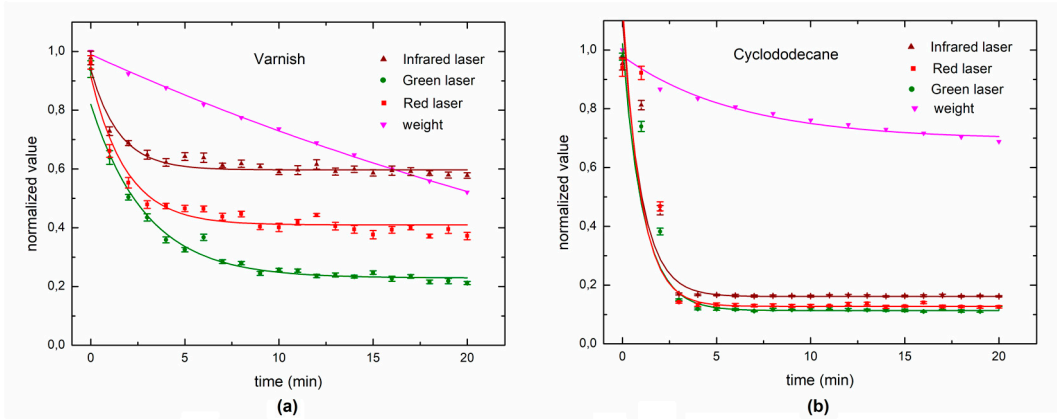
The application of lasers with different wavelength in the Varnish and Cyclododecane is presented in Figure 9. Normalized AVD indexes were obtained and the Varnish presented higher sensitivity to the wavelengths than the Cyclododecane. All the cases using what we defined as fast dynamics using 10 fps to acquire the images. And in this case, the behavior of Cyclododecane presented to the fast dynamics as the colloid that dried first, in the opposite observation of the weighing monitoring (Figure 8).





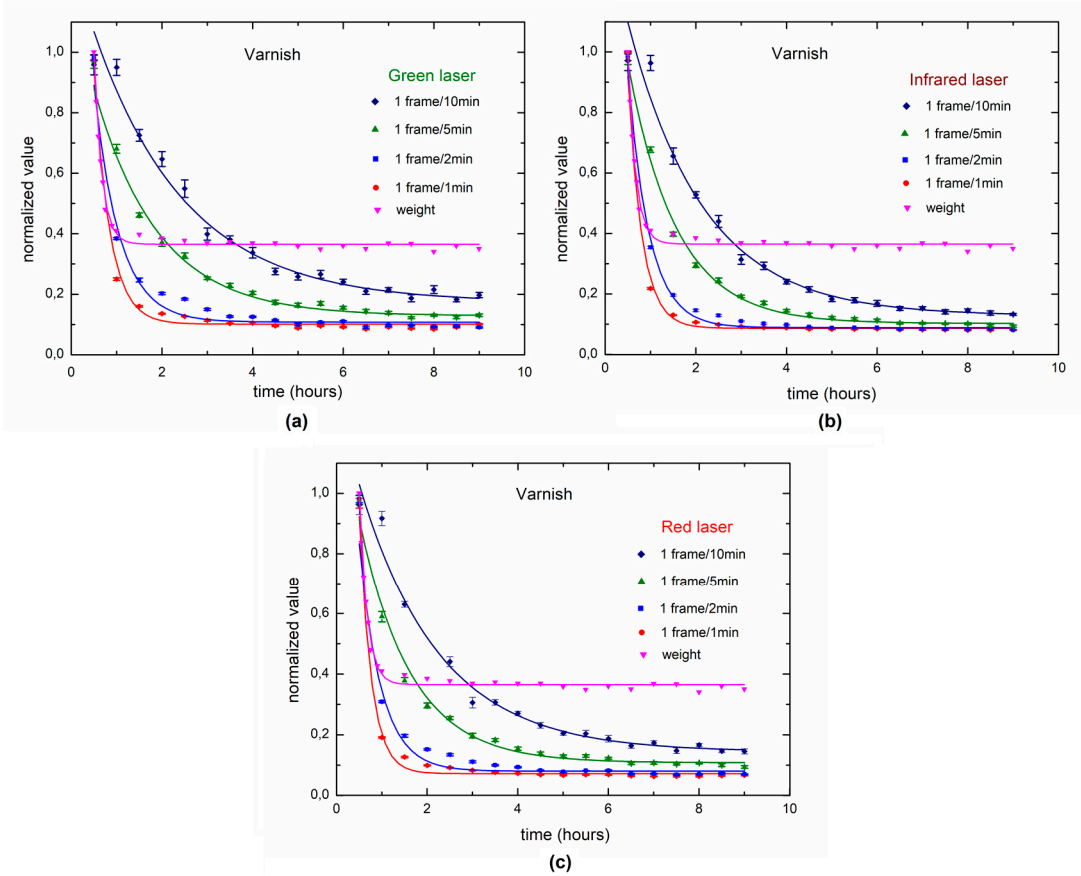
**Figure 9.** Dynamic laser speckle in fast dynamics of drying colloids using three wavelengths submitted to each painting treatments (a) Varnish (b) Cyclododecane.

In Figure 10, the DLS and weighing values are presented, and related to the three wavelength lasers. Varnish and Cyclododecane were monitored during 20 minutes, and both analysed with the fast dynamics, i.e. 10 fps. The fast dynamics presented best ability to follow the fastest drying process presented by the Varnish, but the DLS could not sense the Cyclododecane that presents the slowest drying dynamic.



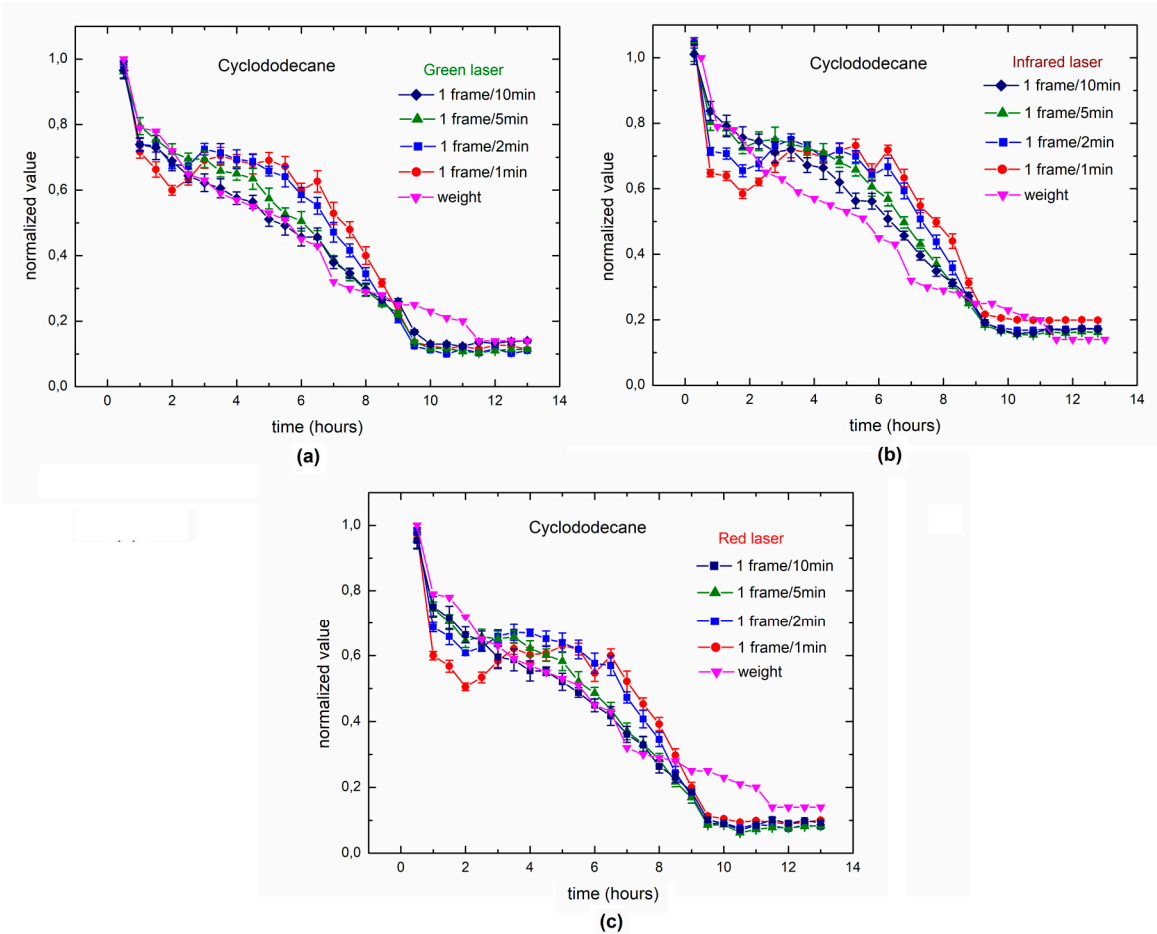
**Figure 10.** Dynamic laser speckle index for fast dynamics (10 fps) and the weighing to each painting treatments (a) Varnish (b) Cyclododecane.

The slow (long-term) dynamics is presented in Figure 11, to Varnish drying, and it is also compared to the weighing and the three lasers (wavelengths). Four time-rates were used to evaluate the answer of long-term process. The slower the time-rate, higher is difference to the weighing process. In this case, the acquiring of 1 frame at each 10 minutes provided the best ability to sense changes in long-term monitoring. Otherwise, the weighing output did not have the ability to follow the drying process after one hour.



**Figure 11.** Dynamic laser speckle in low dynamics of drying Varnish using three wavelengths submitted (a) Green laser (b) IR laser (c) Red laser, observed in four different time-rates and compared to the weighing.

In Figure 12, the result of a long-term monitoring of Cyclododecane is presented in comparison to the weighing and to three different wavelength lasers. It was also observed the ability of each time-rate to follow the process. The characteristic of the Cyclododecane is drying in the external layer first, then to continue the drying process in the inner layers. In this case, the weighing process could follow the slow drying during hours before the stabilization, which also happened with the DSL outputs using low time-rates.



**Figure 12.** Dynamic laser speckle in low dynamics of drying Cyclododecane using three wavelengths submitted (a) Green laser (b) IR laser (c) Red laser, observed in four different time-rates and compared to the weighing.

## 5. Discussion

### 5.1 Validation of portable device to monitor drying processes in paint restauration

Beyond the ability of the proposed device to follow the drying process, it is relevant to highlight its ability to sense it in a smoother way than the traditional setup. One can note that the explanation comes from the different cameras used in both cases; whilst in the proposed device the size of the pixel in the CCD camera was of 2.2  $\mu\text{m}$  the size of the system in the optical laboratory was of 4.65  $\mu\text{m}$ . The smoothness of the exponential curve from the proposed device allow us to follow the drying process for larger time if compared to the camera used in the optical laboratory. So much so, the size of the pixel matters and it can improve the sensitivity of our sensor [31].

### 5.2 Comparison of back and forward scattering using the portable equipment

The ability of the backscattering to sense the drying process of Cyclododecane in a larger evolution, from 1 to approximately 0.1 in normalized values of AVD index, than the ability of the forward scattering, was sensed from 1 to approximately 0.3 in normalized values of AVD index, can be attributed to the less sensitivity of the transmission in the dynamic laser speckle outcomes [32]. That leads us to adopt the reflection, backscattering, as prior when possible.

### 5.3 Third experiment: Changes in time rate using different lasers compared to weight

The long-term monitoring by means of DLS presented best ability to follow the drying process even if compared to the weighting process. This means that the DLS in the long-term monitored as proposed is a better sensor than the traditional DLS with high time-rate (10 fps) and also better than the weighing process that could be considered the Gold Standard. The monitoring of drying paint using the traditional fps are usually restricted to the first minutes [20,23,33].

The different dynamics of drying presented by Varnish and Cyclododecane were better followed by the long-term methodology proposed, where in the Cyclododecane the presence of two phases of drying can explain the relation of the time-rate with the fast and with the slow drying, each one linked to the surface and to the inner layer of the sample.

Varying the laser wavelength can be interesting in fast dynamics, a lower wavelength is more sensitive to small changes. In the Varnish, the lower the wavelength is, the longer the process can be sensed (Figure 10). The composition of the colloid and the fast drying in the surface can explain that ability of the lower wavelengths. In long-term dynamics varying the wavelength do not show a difference in sensitiveness, but can be useful to make the image capture adjustments more independent from the colour of the studied surface.

The portable equipment presented reliable results, thus offering the facility to be used *in situ*. And with the association to the IR laser (the independence of the external light) makes the equipment more robust. The commercial equipment using DLS in drying ink also uses IR [24], but only limited to fast dynamics of drying, restricting its use in long-term monitoring.

## 6. Conclusions

The portable equipment to monitor the activity of painting treatments during the restauration of paint *in situ* presented reliable outcomes, compatible to the equipment used in an optical laboratory. The modified image processing biased to a long-term measurement presented best results if compared to the traditional method driven by the fast dynamics data acquisition and analysis. The proposed long-term methodology presented ability to sense different drying dynamics. The laser wavelengths testing proved that more accurate measurements can be obtained in fast dynamics and in general some improvements can be obtained regarding the sample and light interaction. The weight monitoring proved to be less sensitive to long-term changes in colloids with slow drying process, as the case of the Varnish. On the other hand, the weight monitoring setup could not be possible in real painting restauration, or even if it is possible, could be more complicated than the proposed DSL method.

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### Author Contributions:

Alberto J. Pérez Rolando J. González-Peña and Roberto Braga. Conceived, designed the experiments, performed the experiments, analyzed the data and wrote the paper.  
Ángel Perles Analyzed the data and wrote the paper.  
Eva Pérez -Marín and Fernando J. Garcia-Diego contributed reagents/materials and wrote the paper

**Conflicts of Interest:** The authors declare no conflict of interest.

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