

Detection of NaCl located in mural painting by stimulated infrared thermography

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Abstract

In this work, we study the possibilities of stimulated Infrared Thermography for NaCl detection in Avignon calcareous stones. We show experimentally that the presence of salts leads to an increase in local thermal effusivity and to a decrease in maximum temperature induced by the photothermal flash analyze. We show then, that measurement of these parameters leads to the NaCl detection.

1. Introduction

Within the framework of the conservation and the restoration of cultural heritage, a lot of laboratories are studying the possibilities of nondestructive testing using passive or stimulated Infrared thermography [1-53]. Among this research teams, we can cite the « Laboratoire de Recherche des Monuments Historiques » (LRMH), the « Centre interdisciplinaire de Conservation et Restauration du Patrimoine » (CICRP) and the « Groupe de Recherche en Sciences Pour l'Ingénieur » (GRESPI) of the Reims University. These three laboratories have worked together for over 10 years. This collaboration has already shown, using stimulated infrared thermography, the possibility to detect delamination located in murals paintings of heritage (painted walls of the church of "Saint Florentin" in "Bonnet", painted ceilings of the Abbey of "Saint Savin sur Gartempe" - World Heritage of UNESCO ...) [40-53]. However, during these about 10 years of research, these teams was mainly interested in structural defects (such as delamination) detection and characterization. But another type of defect is also critical in works of art conservation. It is the presence of hygroscopic salts in the artwork. Indeed, hygroscopic salts, during the crystallization phase, increase in volume and can burst the part of the artwork surrounding the salt location [54-55]. The work presented here aims to detect, using stimulated infrared thermography, the presence of NaCl salt in Avignon calcareous stones. The physical idea is that the presence of NaCl modifies locally the thermal properties of the analyzed stone. Because ultimately, we want to use this method during *in situ* analysis (analysis of thick walls), it was natural to consider the thermal effusivity parameter. Moreover, as it was easy and fast to access, we have also considered the maximum increase of temperature of the studied sample just after the exciting flash. Our presentation is divided into three stages: First, we present the theoretical model developed for measuring the thermal effusivity. We then present the experimental device used for the study. Finally, we show the possibility, using stimulated infrared thermography, to differentiate healthy and contaminated (with NaCl) Avignon calcareous stones.

2. The theoretical model and the thermal effusivity measuring method developed for the study

As previously mentioned, we want to use the stimulated infrared thermography to detect, *in situ*, hygroscopic salts located in murals paintings of the heritage. By construction, they are deposited on thick walls. It is therefore very difficult to thermal perturbation to diffuse through this kind of work of art. The model proposed here is therefore semi-infinite. Furthermore, we want to develop a fast analysis because the access time for a work of art analysis is often very limited. We then chose to implement flash infrared thermography. The theoretical model developed for the study considers then the following assumptions: The studied sample is semi-infinite. It is initially in balance with its environment. The initial temperature is equal to 0 ° C. It is thermally insulated. He is excited in front face by a Dirac pulse. The differential system to solve is then the following:

$$\frac{\partial^2 T}{\partial x^2} - \frac{1}{a} \frac{\partial T}{\partial t} = \frac{-g(x, t)}{\lambda}$$

$$\frac{\partial T}{\partial x} = 0 \quad (x = 0 \text{ and } t > 0)$$

$$T(x, t = 0) = 0 \quad (1)$$

To solve this differential system, we have implemented the Green formalism [57]. It is first to find a general solution to the homogeneous differential system associated with the preceding. The literature [57] indicates that the general solution is given by the following Green function:

$$G(x, t | x', \tau) = \frac{1}{\sqrt{4\pi a \tau}} \exp\left[-\frac{(x-x')^2}{4a(t-\tau)}\right] \quad (2)$$

Then the Green formalism permits to obtain the general expression of the wanted temperature. We obtain:

$$T(0, t) = \frac{a}{\lambda} \int_0^t a \tau \int_{-\infty}^{+\infty} \frac{1}{\sqrt{4\pi a \tau}} \exp\left[-\frac{(x-x')^2}{4a(t-\tau)}\right] 2Q \delta(x', \tau) dx' d\tau \quad (3)$$

Finally, as the excitation and analysis are developed on the same side and also as the excitation is very short, this expression simplifies to:

$$T(0, t) = \frac{Q}{b\sqrt{\pi t}} \quad (4)$$

It is therefore theoretically possible to determine the wanted effusivity from the photothermal response of the sample analyzed and knowledge of the deposited energy. Unfortunately, in practice it is often difficult to know precisely the latter parameter. In our study, we have therefore chosen to free us from this knowledge. Instead, we chose to develop an indirect method of estimating the thermal effusivity parameter. We used a comparison method. It consists in analyzing in the same experimental conditions, the unknown sample and a reference sample. Under these conditions and for the same area of analysis, the wanted effusivity is given by the formula (5):

$$b_{inc} = \frac{T_{ref}(t)}{T_{inc}(t)} \cdot b_{ref} \quad (5)$$

3. The samples studied and the experimental device used to study

3.1 The studied samples

Three types of samples were studied in this work (Fig.1.). The first is a reference PVC sample. Its dimensions are a length and a width of 4.1 cm and a thickness of 5 mm. It was characterized thermally with the DICO system [58] of CERTES laboratory (Creteil – France). Its thermal effusivity is equal to 408 J.K⁻¹.m⁻².s^{-1/2}. It is covered with matt black paint to simulate a pictorial layer. It is the reference sample that we use for our thermal effusivity estimates. The second type of considered sample is a series of 8 Avignon calcareous disks. They have a radius equal to 15 mm and a thickness equal to 5 mm. They are covered with the same matt black paint that the reference PVC sample. To obtain crystalline salts, 4 of these samples, before painting, was immersed for 24 hours in a saturated water solution of sodium chloride. They were then left in air for a week. This caused a vaporization of water and the appearance of sodium chloride crystals. These four samples were then covered with the same black matt paint as before. Finally, the last sample is an Avignon calcareous block, containing locally (in the center) sodium chloride salts. (We putted in the center of this sample a few drops of the previous saturated saline solution and was allowed to dry the sample for again one week). Its dimensions are a length of 12.5 cm, a width of 10 cm and finally a thickness of 2.5 cm. Finally, to simulate the paint layer, we have covered this calcareous block with the same black paint than other types of samples.

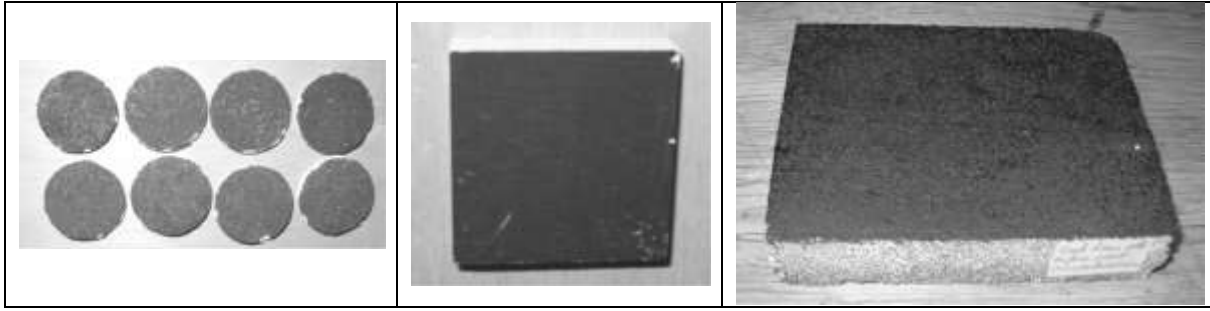


Fig 1. The three kinds of analyzed samples (On the left, the 8 Avignon calcareous disks, in the center, the PVC reference sample, in the right the Avignon calcareous block)

3.2 The experimental device used to study

The experimental device used for the study is the SAMMTHIR system of the GRESPI laboratory. It is first composed of two flash lamps delivering energy of 2×2400 J during 5 ms. It is then composed of a bolometer camera FLIR SC 655 type. It is a "long waves" camera well adapted to the murals paintings analysis [50]. Finally, the device comprises a synchronization electronic and a data acquisition and post processing. (Fig.2.).



Fig 2. The experimental device used for study of the 8 disks studied (left) and of the calcareous block of Avignon (right)

4. The experimental results obtained

4.1 Study of 8 painted Avignon calcareous disks

Initially, to test the sensitivity of the photothermal signal with the presence of salt, we studied simultaneously 8 Avignon calcareous disks. All have been bonded with a double sided tape on a wooden support. The four salted samples were placed on the same horizontal. The four healthy samples were placed just below them, again on the same horizontal. This support was placed in front of the camera at a distance of about 50 cm. The two flash lamps were then placed on either side of the camera, again at a distance of about 50 cm from the sample. These 8 samples were then simultaneously excited for about 5 ms and then analyzed by infrared thermography for 2 seconds. An example of obtained results is presented in Figure 3. It is the photothermal response of the thermal scene at the end of the excitation. It shows less important photothermal signals in place of salt samples (upper band). This result is very interesting because it suggests that the photothermal method is sensitive to the presence of NaCl salts type in mural paintings.

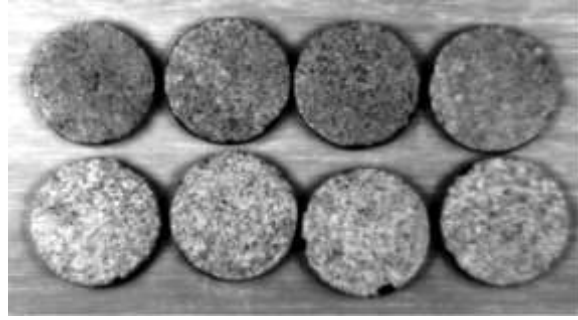


Fig.3. Raw photothermal responses of the 8 studied Avignon calcareous disks

To quantify these different photothermal responses, we then estimated two characteristics thermophysical parameters. This is the maximum increase of temperature during the analysis and the thermal effusivity. Table 1 shows the values obtained. It shows that in presence of salt, the maximum increase of temperature is lower than in the absence of salts. Indeed, the average maximum value of increase of temperature is equal to 21.3 °C in the case of healthy samples. It is equal to 17.8 °C in the case of contaminated samples. This variation confirms on one hand the possibility to detect NaCl in Avignon calcareous sample. In an over hand, this temperature variation is quite significant. It gives hope that the method can be sensitive enough to a salts concentration analysis.

<i>healthy samples</i>		<i>Contaminated samples</i>	
	Tmax (°C)		Tmax (°C)
Sample 1	22,9	Sample 1'	18,2
Sample 2	21,1	Sample 2'	18,1
Sample 3	20,8	Sample 3'	16,6
Sample 4	20,3	Sample 4'	18,4
Average	21,3	Average	17,8

Table 1. The highest temperature elevations obtained during the photothermal analysis of the 8 Avignon calcareous disks

In Table 2, we presented the thermal effusivity obtained during the photothermal analysis of the 8 Avignon calcareous disks. It shows that the presence of NaCl leads to an increase of the estimated average effusivity. This parameter is equal to an average value of 1067 J.K⁻¹.m⁻².s^{-1/2} for healthy samples and about 1139 J.K⁻¹.m⁻².s^{-1/2} for contaminated samples. Again the result obtained is encouraging. Indeed, it shows on the one hand, the possibility to detect NaCl infiltration using thermal effusivity estimation. It shows in another hand that the change in thermal effusivity is important, which can lead to NaCl concentration estimation by stimulated infrared thermography.

<i>healthy samples</i>		<i>Contaminated samples</i>	
	Effusivity (J.K ⁻¹ .m ⁻² .s ^{-1/2})		Effusivity (J.K ⁻¹ .m ⁻² .s ^{-1/2})
Sample 1	1055	Sample 1'	1118
Sample 2	1080	Sample 2'	1157
Sample 3	1066	Sample 3'	1134
Sample 4	1067	Sample 4'	1146
Average	1067	Average	1139

Table 2. The thermal effusivity values obtained during the photothermal analysis of 8 Avignon calcareous disks

4.2 Study of an Avignon calcareous block locally contaminated with NaCl

Following these encouraging results, we wanted to approach a real case. We have then studied an Avignon calcareous block locally contaminated with NaCl. As for the previous study, the sample was placed in front of the SAMMTHIR system, approximately 50 cm of the latter. It was then illuminated for about 5 ms using light sources. It was then filmed for 2 seconds with a frequency of 50 Hz by the infrared camera of thermography. An example of result obtained is presented in Figure 4. It is the photothermal response obtained at the end of the excitation flash. It clearly shows a lower photothermal signal at the place of contamination with NaCl. This confirms on the one hand the possibility of detecting the local presence of NaCl in calcareous by stimulated infrared thermography. This also

confirms the presence of sodium chloride in calcareous causes a decrease in the maximum temperature rise induced by the photothermal analysis.

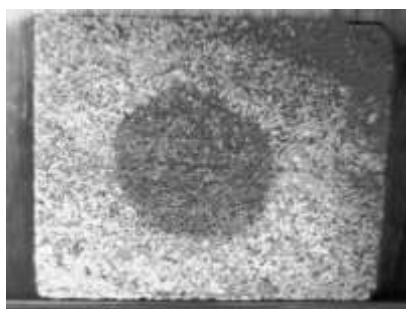


Fig. 4. Photothermal response obtained at the end of the excitation light

To quantify this effect, we have plotted the maximum temperature reached during the photothermal analysis, along a straight line intersecting the signature of the defect approximately at its center. We then considered eight measurement points. 4 of these points have been taken out of the salty area. The other 4 were taken in this salty area. The result obtained is presented in Figure 5. It shows that the presence of salts causes an average decrease in temperature of about 1.8 ° C. This change was much higher than the equivalent signal to noise of our instrumentation (about 40 mK). This confirms the potential quantitative possibilities of the analysis method regarding salts located in murals paintings.

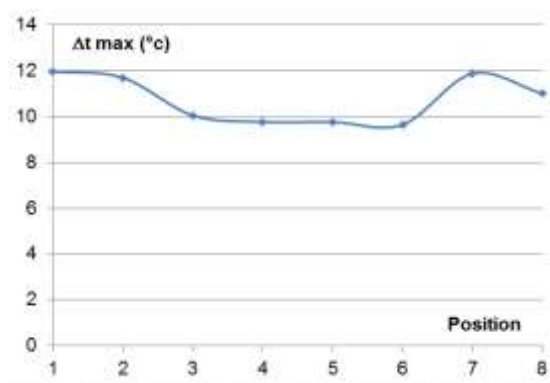


Fig 5. Profile of maximum increase of temperature, drawing along a line perpendicular to the photothermal signature of the contaminated area

In a second step, we estimated the thermal effusivity parameter. We considered the same line and the same measurement points. The results obtained are presented in Table 3. It shows, as previously, that the presence of salt causes an increase in the thermal effusivity parameter. It shows then the possibility of detecting this salts using stimulated infrared thermography. Finally, it shows that the variation of thermal effusivity is also important, which should allow quantitative analyzes of salt concentration.

<i>healthy samples</i>		<i>Contaminated samples</i>	
	Effusivity (J.K ⁻¹ .m ⁻² .s ^{-1/2})		Effusivity (J.K ⁻¹ .m ⁻² .s ^{-1/2})
Sample 1	1038	Sample 1'	1246
Sample 2	1047	Sample 2'	1230
Sample 3	1246	Sample 3'	1263
Sample 4	1230	Sample 4'	1336
Average	1130	Average	1269

Table 3. The thermal effusivity values obtained during photothermal analysis of an Avignon calcareous block contaminated locally with NaCl

5. Conclusion

In this work, we approached the possibilities of stimulated infrared thermography for sodium chloride located in Avignon calcareous detection. In a first step, we presented the theoretical model and the thermal effusivity estimation method developed for the study. In a second step, we have studied 8 Avignon calcareous disks, 4 healthy and 4 contaminated with sodium chloride. We have shown on one hand that the presence of salts induces a decrease of the maximum temperature increase reached during the photothermal analysis. We have shown on the other hand, that this presence also led to an increase of the thermal effusivity parameter. Finally, in a last step, to approach a real case, we showed during the photothermal study of an Avignon calcareous block contaminated locally with NaCl that the two parameters allow as previously a good detection of the salt. The results obtained in this study are encouraging. Indeed they seem to open the way to the salt located in work of art detection using stimulated infrared thermography. They have now to be confirmed during in situ analysis. They also have to be extended to the detection of other types of salts and other types of stones. Studies in this direction are in progress.

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