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# Optimized analysis for nonlinear ultrasonic imaging in complex media : acoustic imaging for cultural heritage

Serge DOS SANTOS<sup>1</sup>, Nathalie POIROT<sup>2</sup>

<sup>1</sup> INSA Centre Val de Loire Inserm U930, Blois, France

<sup>2</sup> GREMAN, IUT de Blois, Blois, France

Contact e-mail: serge.dossantos@insa-cvl.fr

**Abstract.** The interaction between an ultrasonic wave and a complex media has an increase interest for nondestructive testing (NDT) applications. For specific applications using nonlinear ultrasonic imaging, inversion procedure needs specific signal processing techniques and new coding schemes. Nonlinear acoustic testing use the fact that microcracks generate harmonic and/or subharmonic tones of the frequency at which they are excited. Nonlinear signal processing started with the measurements of harmonics. Then came the nonlinear mixing of waves where the nonlinear parameter produced the attachment of the low frequency (LF) message to the high frequency (HF) probe with a mixing of complex physical properties of the medium under study. Today, new optimized excitations are generated thanks to the analysis of symmetry properties of the system such as reciprocity, nonlinear time reversal and other pulse-inverted (PI) techniques. This paper tends to show how symmetry analysis can help us to define new methodologies and new experimental set-up involving modern signal processing tools. Generalized TR (Time Reversal) based NEWS (Nonlinear Elastic Wave Spectroscopy) methods and their associate symmetry skeleton will be taken as an example with some new specific signal processing tools such as Pulse Inversion (PI), ESAM, DORT or SSM modern coding schemes. As an application of mixing properties in a wide frequency range, new broadband techniques are needed in the domain of the preservation of cultural heritage. This is an interdisciplinary challenge which needs the expertise of many scientific disciplines as well as chemistry, physic, optics, etc. The Loire Valley in France has a diversified cultural heritage, including monuments where the mainly construction units is stone. Most castles were built with two French highly porous limestone called Rlichemont and tuffeau stones. The analysis of the composition of the stones is one of the key parameters in the study of aging historic buildings. The use of news multichannel TR-NEWS based analysis (40 MHz) combined with a FTIR-based system (wavenumber range from 2000-800 cm<sup>-1</sup>) has shown a specific property of the tuffeau limestone which is a mix of calcite, SiO<sub>2</sub> and CaSO<sub>4</sub>·2H<sub>2</sub>O whereas other damaged sample is calcite.

## Introduction

The preservation of patrimonial is an interdisciplinary challenge which needs the expertise of many scientific disciplines as well as chemistry, physic, historical, etc.[1]. Cultural protection has the same level of environment protection. In order to generalize the idea of protection, Non Destructive Testing (NDT) is necessary for cultural heritage testing. The destruction of cultural heritage is the destruction of the culture and the memory of a whole civilization. The Loire Valley in France has an impressive amount of historical,



architectural treasure and diversified cultural heritage, including monuments where the mainly construction units is stone. Most castles were built with two French limestone as tuffeau and Richemont stone. The analysis of the composition of the stones is one of the key parameters in the study of aging historic buildings[2]. Throughout the years, because a building may have been subjected to both large and small external forces, including several disasters, its framework structure might have possibilities to be distorted due to aging process. The presence of exogenous pathogens often salts such as gypsum, is considered to be connected to the peeling plates and sheets of tuffeau. Many chemistry and physic analyses are in this work to put in evidence the presence or not of salts and fissuration in two stones that are not part of the same historical building.

Ultrasonic techniques with advanced signal analysis approaches have been developed for characterization of microstructural features in the size range from a few nanometers to few microns in a variety of technologically important materials[3]. Innovative ultrasonic methodologies have been developed for characterisation of microstructures using ultrasonic spectral parameters for applications related to quality assurance, damage assessment and life prediction of engineering components. Based on extensive investigations on various isotropic solid materials, Poisson's ratio is found to decrease with increase in ultrasonic shear wave velocity for all the materials. This new correlation between two independent elastic constants may lead to the reduction in the number of independent material parameters required for isotropic solids. The innovative studies carried out on acoustic wave propagation in complex medium enabled development of ultrasonic devices for measuring linear and nonlinear parameters[4,5]. Using nonlinear ultrasonic measurement facilities, studies have been made on evaluation of damage in a variety of engineering materials. The studies on damage assessment demonstrated that nonlinear ultrasonic measurements enable evaluation of creep-fatigue damage with higher sensitivity, compared to classical ultrasonic measurements. Using advanced ultrasonic signal processing coding schemes and tomographic reconstruction[6], defects could be detected in most of complex materials. Advanced ultrasonic signal processing have also been developed for to overcome several difficulties due to small dimensions of defects and complex geometry of structures, and to meet stringent sensitivity requirements.

The objective of the present study is to make a comprehensive evaluation of modern technologies[2,3,7,8] employed for other NDT applications, and to recommend suitable measurements for the NDT applied to cultural heritage. Even though a number of studies have been carried out on Tuffeau sandstone, a complete and comprehensive in situ assessment using most modern NDT techniques has not been attempted. The advances of instrumentation and signal processing[9,10] related to the chirp-coded TR-NEWS approach have made it possible to have rugged and portable equipment with higher sensitivity and accuracy for exploring the Tuffeau sandstone using a multimodality based on ultrasound, and FTIR structural characterization.

## **2. Non Destructive Testing of Cultural Heritage**

Nondestructive Testing (NDT) of complex media is of increasing importance, not only in aeronautic industry, but also in various areas of industry also including modern biomedical media. A general definition of NDT is a examination, test, or evaluation performed on any type of test object without changing or altering that object in any way, in order to determine the absence or presence of conditions or defects that may have an effect on the usefulness or serviceability of that object. Classical methods for analysing historic materials are often time-consuming and destructive or at least micro destructive. From the structural point of view it has to be pointed out that in complex sandstones, the aging

process give rise to complex stress states, which can become particularly severe in case of vibrations or other causes. Such a severe stress state which can come from environmental conditions, in conjunction with the brittleness of the Tuffeau sandstone, can originate cracks. In this context, the development of restoration treatments and the research on innovative materials for preventive conservation are objects of recent projects[11]. Nondestructive tests are usually conducted in order to measure various characteristics such as size, dimension, configuration or structure, including alloy content, hardness and grain size. NDT, as a technology, has seen significant growth over the past 30 years and is considered today to be one of the fastest growing technologies from the standpoint of uniqueness and innovation.

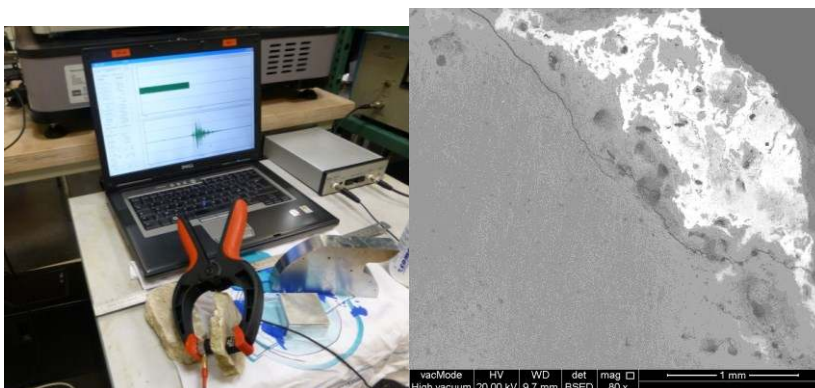
For example, X-ray Computed Tomography can be applied successfully to cultural heritage to obtain morphological and physical information on the inner structure of archaeological samples and works of art[12]. In another domain, NDT of parchment, which has been used as a writing and drawing support since antiquity, are also studied with a great interest. Since, like human skin[3], collagen is its principal component and has a unique triple helix structure, stabilised by hydrogen bonds and Van der Waal's interactions between side chains, the NDT methods to be tested on this sample need to involve chemical aspects of the microstructure observed in the near infrared region [13]. The numerous interactions of these chemical aspects with each other and with environmental components (oxygen, humidity, etc.) in the course of its history make each sample or each structure a unique object. The correct classification and condition monitoring of the composing materials of historical monument is therefore of primary importance when studying aging behaviour and degradation processes. As another example, one can cite the multi-analytical approach adopted for the characterization of the concrete structural elements of the Victory Monument in Bolzano, where a full knowledge on the mechanical and chemical/mineralogical characteristics of the materials was demonstrated[14]. Again, both low frequency ultrasonic and impact echo techniques have been employed for establishing the mode of fabrication of the Delhi Iron Pillar using forge-welding process, and also for gaining insight into characteristics of sounds produced by musical columns of Hampi musical pillars[15].

With the increase of modern signal processing tools observed during the last 20 years, the area of interest of NDT based innovations can be explored in such domain like old or new materials for biomedical, or within the increasing demand of inspection of ancient materials in the domain of cultural heritage. Among these modern signal processing tools, nonlinear signal processing is a new field of research with the aim to optimize the excitation of information coming from nonlinear effects[4,8]. Within the wide range of possibilities of signal processing methods, those using symmetry properties seems to be more efficient for extraction of a specific information. Various nonlinear methods have been investigated in recent years for the detection of faults and fatigue in carbon-fibre reinforced composite materials and structures[5,18]. However, there is no universally-agreed rationale for which technique is best suited to the detection of which type of defect. Furthermore, even if such a rationale were to exist, real-world samples potentially contain a variety of defects (e.g. micro-cracking, delamination and disbonding) induced by various damage mechanisms (stress, impact, heat) such that no single nonlinear testing technique can offer the optimum inspection choice in all circumstances, and particularly for cultural heritage. Various nonlinear methods have been developed in recent years for defect detection in complex materials[5]. The most common include harmonic and overtone generation, inter-modulation product generation and resonant frequency shift, also known as Nonlinear Elastic Wave Spectroscopy (NEWS) methods. Consequently, new optimized excitations are needed and, thanks to the analysis of symmetry properties of the system such as reciprocity, nonlinear time reversal[16] and other pulse-inverted (PI) techniques, a

wide range of innovation can be proposed[8]. Thus, the objective of the present study is to carry out a wide variety of complementary NDT investigations based on nonlinear methods so that comprehensive and statistical information could be obtained related to the aging properties of the material under study. This study aimed to investigate the mechanical properties of tuffeau limestones which was used to build old Loire Valley Castles by measuring the stress wave propagation velocity as well as the internal chemical structure. Such a study could enhance the understanding of ancient stones and also help in better knowledge of our cultural heritage in the Loire Valley.

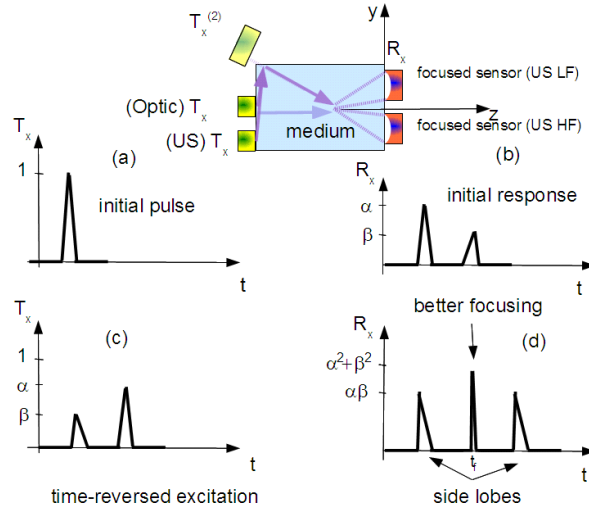
## 2. Materials and methods

The interaction between an acoustic wave and a complex media has an increase interest for both NDT applications and for biomedical ultrasound. For specific applications using nonlinear imaging, inversion procedure needs specific signal processing techniques and new coding schemes. Nonlinear techniques use the fact that microcracks and delaminations generate harmonic and/or subharmonic tones of the frequency at which they are excited. Nonlinear signal processing started with the measurements of harmonics. Then came the nonlinear mixing of waves where the nonlinear parameter produced the attachment of the low frequency (LF) message to the high frequency (HF) probe with a mixing of complex physical properties of the medium under study. Today, new optimized excitations are generated thanks to the analysis of symmetry properties of the system such as reciprocity, nonlinear time reversal and other pulse-inverted (PI) techniques. Generalized TR (Time Reversal) based NEWS (Nonlinear Elastic Wave Spectroscopy) methods and their associate symmetry skeleton constitute an example with some new specific signal processing tools such as Pulse Inversion (PI), ESAM, DORT or SSM modern coding schemes[7,9,16]. Among these family of “pulse coded excitation”, solitonic coding constitutes a new scheme in the sense that solitary waves are the best candidates for pulse propagation in nonlinear and dispersive media[17]. The present investigation was oriented to the assessment of the reliability of the acoustic TR-NEWS method in detecting early stage delamination of the Tuffeau sandstone. The repeatability of the measurement is evaluated through a number of independent repetitions in order to assess the standard deviation of values, helping the successive signal processing. Preliminary tests presented in this report aim at the characterization of the environment (ESM and FTIR analysis), and the characterization of the optimized TR-NEWS set-up configuration. Background noise has been intensively studied, optimizing the transducers and the excitation conditions (figure 4). As shown figure 4a, the sample 1 (noted S1) seems to be tuffeau which is a mix of calcite, SiO<sub>2</sub> and CaSO<sub>4</sub>·2H<sub>2</sub>O whereas the sample 2 (noted S2) is calcite. On figure 4(b), opale and quartz which are the different crystalline phases of SiO<sub>2</sub> can be observed.



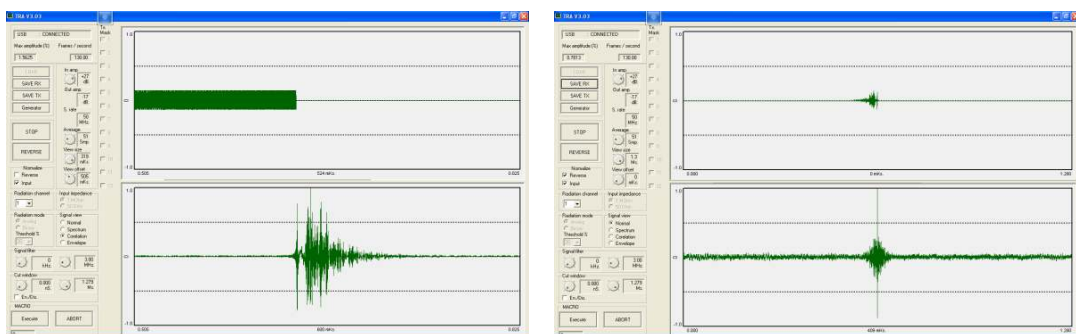
**Fig. 1.** : Experimental set-up (left) and ESM image of the tuffeau standstone (sample S1)

The test equipment (Figure 1) consisted of the TR-NEWS device Juvitek TRA-02 (0.02-40 MHz) connected to a computer, an Amplifier ENI model A150 (55 dB at 0.3-35 MHz), shear wave transducer Technisonic ABFP-0202-70 (2.25 MHz), longitudinal wave transducer Panametrics V155 (5 MHz).



**Fig. 2.** : The four elementary steps of the multi-modal TR-NEWS method : (a) after the excitation of a broadband initial excitation (a short pulse here), the recorded initial response (b) is averaged, filtered, time reversed (c) and rebroadcasted as a time-reversed excitation with the same experimental conditions. The optimized excitation based on this TR-NEWS approach is used for extraction of the local acoustic signature located in the time domain with respect to the maximum  $T=t_f$  and the side lobes of the crosscorrelation function  $T_h(T-t, T)$  given by the focused signal (d). The multimodal property come from the fact that the focusing is increase thanks to the use of a broadband multi-physics modality (US LF and HF, optics, Terahertz, etc.).

The chirp-coded TR-NEWS method uses TR for the focusing of the broadband acoustic chirp-coded excitation. The method consist in the successive steps (Figure 2) : emission of a linear frequency sweep signal (the chirp-coded excitation); recording of the response to the emitted signal (the chirp-coded coda), computation of the pseudo-impulse response which is the correlation between the chirp-coded excitation and its response, recording of the response to the time-revered emitted pseudo-impulse excitation (chirp-coded TR-NEWS coda). The sensitivity improvement of chirp-coded signal processing has been validated in various domains and is also validated here within cultural heritage applications (figure 5). The sensitivity improvement is difficult to conduct due to the fact that broadband excitation is conducted. SNR evaluation of chirp-coded can be found in literature and has been intensively studied in our specific case of Tuffeau sandstone (Figure 3).



**Fig. 3.** : Chirp coded excitation (left) and TR-NEWS focusing of the tuffeau limestone

In order to describe chirp-coded TR-NEWS process in terms of a time-invariant system, we consider a chirp excitation  $c(t)$  send in a medium with impulse response  $h_{21}(t)$ . As described schematically for a simple medium (Figure 2), side lobes could be interpreted with cross-correlations functions and present a symmetry property with respect to the focusing time  $t_f$ . The chirp-coded TR-NEWS method uses TR for the focusing of the broadband acoustic chirp-coded excitation. In order to describe chirp-coded TR-NEWS process in terms of a time-invariant system, we consider a chirp excitation  $c(t) = A \cos(2\pi f(t)t)$  where  $f(t) = At + f_0$  send in a medium with impulse response  $h_{21}(t)$ . The chirp-coded coda response  $y(t, T)$  recorded for a finite time duration  $T$  is given by  $y(t, T) = h_{21}(t) * c(t)$  where  $h(t-t', T)$  is an approximation of the (linear) Green's function that satisfy the linear wave equation. The correlation  $\Gamma(t)$ , computed during  $\Delta t$ , is called the pseudo-impulse response (figure 5).  $\Gamma(t)$  is also proportional to the impulse response  $h(t)$  if  $\Gamma_c(t) = c(t) * c(-t) = \delta(t)$ . Under these hypothesis,  $\Gamma(t)$  can be considered proportional to the impulse response (referred as the coda) of the medium and used for enhancing the TR-NEWS focusing. If  $\Gamma(t)$  is time reversed and used as a new excitation, the response  $y_{TR}(t)$  of the medium (called chirp-coded TR-NEWS coda) is then given by  $y_{TR}(t, T) = \Gamma(T-t) * h_{21}(t) = \Gamma_h(T-t, T)$ , and provides the linear crosscorrelation of the system which peaks at  $t=T$  and induces a spatial focusing at the receiver. All this theory is valid under linear behaviour of the medium represented by its impulse response  $h_{21}(t)$ , and will be applied specifically to the Tuffeau sandstone. Any source of nonlinearity (or aging) in the system will result to a perturbation of this method, and will induce additional terms.

### 3. The physical meaning of the cross correlation function

The systemic approach of TR-NEWS has also an important feature related to the physical meaning of TR-NEWS signals. All these signal processing steps (chirp broadcasting, reception, correlation, time reversal and rebroadcasting, reception) help us to understand the physical meaning of the crosscorrelation function  $\Gamma_h(T-t, T)$  of a (simple or) complex medium, like the Tuffeau sandstone. The classical properties of the crosscorrelation function  $\Gamma_h(T-t, T)$ , such as single maximum at  $t = T$ , and such as the symmetry properties (parity, invariance with respect to linear pulse inversion, etc.) gives to this function a real physical interest connected to the symmetries of the excitation. The Curie's principle described in 1894 can be also applied to the cross-correlation function extracted experimentally on the sample with TR-NEWS methods. When certain causes (amplitude of excitation, but also external chemical or environmental reactions) produce nonlinear effects, aging or long time behaviour, the elements of symmetry of the causes must be found in the effects produced in the TR-NEWS cross-correlation signal  $\Gamma_h(T-t, T)$ . And consequently, the symmetry breaking of TR-NEWS signal coming from nonlinear effect would be measured (in the time domain thanks to the temporal representation of the cross-correlation function  $\Gamma_h(T-t, T)$ ) by the "loss of symmetries" that should, of course, be calibrated also, and connected to excitations that break the linear behaviour of the complex system. As described in [3], TR-NEWS experimental set-up which provide a physical meaning of the crosscorrelation function  $\Gamma_h(T-t, T)$  of a complex medium can be considered as an experimental test of the Curie Symmetry Principle.

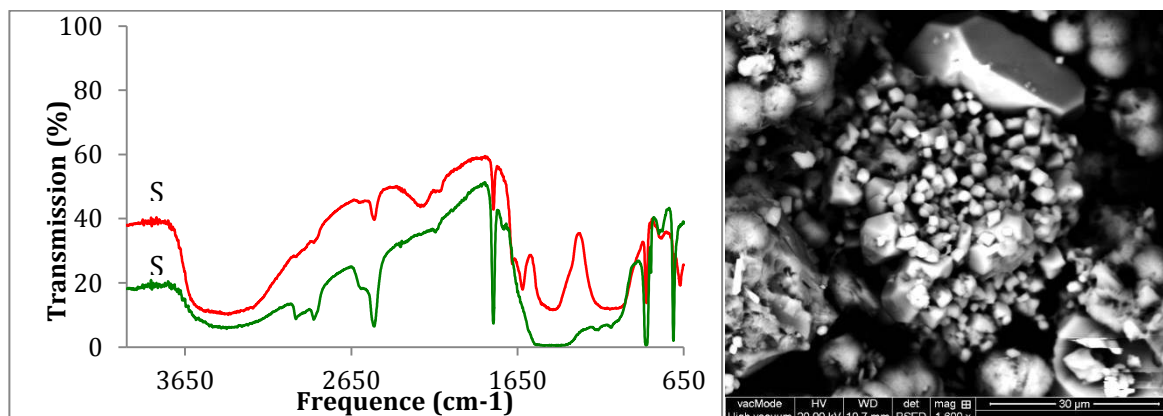


Fig. 4. : (a) FTIR spectra obtained from S1 and S2 samples. (b) : ESM image obtained from S1.

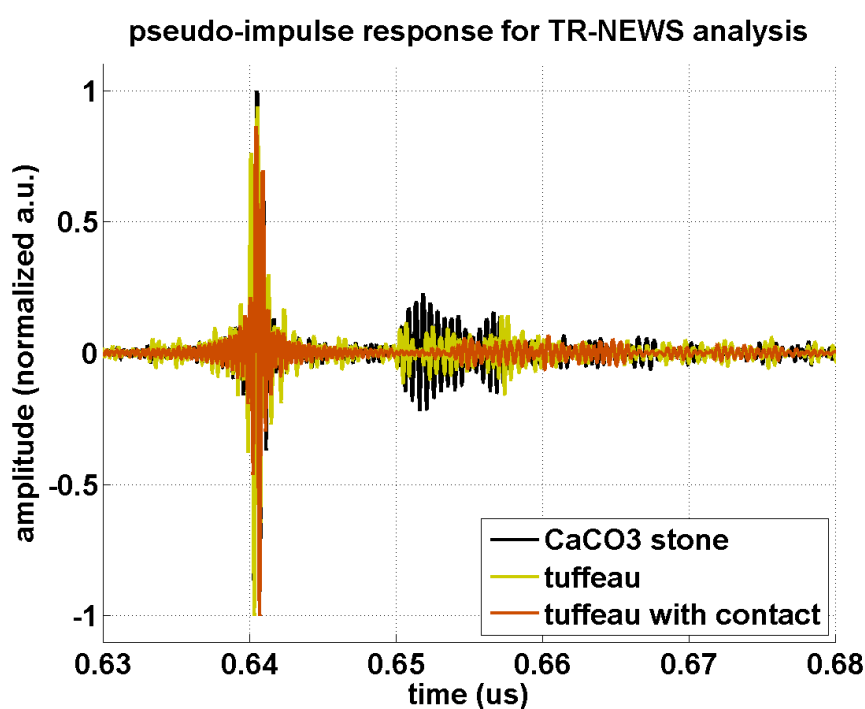


Fig. 5. : TR-NEWS analysis applied to CaCO<sub>3</sub> stone and tuffeau sandstone

#### 4. Conclusion

In this work a database of tuffeau limestones has been tested with TR-NEWS methods coupled with FTIR analysis. The coupling between ultrasonic characterization conducted at 50 MHz sampling frequency, and an FTIR-based system (wavenumber range from 2000-800 cm<sup>-1</sup>) has shown a specific property of the tuffeau limestone which is a mix of calcite, SiO<sub>2</sub> and CaSO<sub>4</sub>·2H<sub>2</sub>O whereas other damaged sample is calcite. The aging of these specific materials are classified into groups, according to their chemical (and therefore spectroscopic) properties. The recorded spectra establish clear distinctions allowing a fast and non-destructive identification of the degradation observed in the micro-sample. Its high sensitivity, highly noticeable for an ultrasonic based method make this technique a very important tool in standard testing procedures for in situ monitoring of the degradation processes of historical monuments. Properly integrated in an extensive and preventive investigation, TR-NEWS methods can provide important indications on the presence and size of subsurface cavities, preventing the loss of structural integrity of the monument



under study. The degradation and non-destructive characterisation of a small set of historic samples was studied and a significant interest of using TR-NEWS method associated to FTIR was demonstrated. The method could be used for rapid condition assessment of historic in situ objects.

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