Ultrasonic Non-Destructive Testing of Archaeological Ceramics

Addisson SALAZAR, Ramón MIRALLES, Angela PARRA, Luis VERGARA, Begoña CARRASCOSA
Universidad Politécnica de Valencia, Valencia, Spain

Abstract. This paper presents an application of the ultrasounds that pursue to find parameters related with physical characteristics of archaeological ceramic materials and the evaluation of the consolidation materials used in their restoration process. The studied archaeological materials were from the Requena's archaeological museum at the East of Spain and consist of pieces of different ages, i.e., Bronze, Iberian, Roman and Middle age. Pieces were measured in transmission mode at different ultrasound work frequencies. Due to the shape of the pieces, ultrasounds were applied trying with several coupling methods: gel, immersion and using a rubber adaptor. The recorded ultrasonic signals were processed by time-frequency signal processing techniques in order to derive a signature of the material characteristics. Various signal parameters, such as, wave propagation velocity, dominant frequency, signal power, centroid frequency and signal attenuation, were estimated. Differences for the different materials were found by ultrasound non-destructive testing and some parameters seem to be related with the material porosity. The knowledge of the archaeological material porosity is a key factor to choose the proper consolidation product for restoration. A comparative study of the ultrasonic parameters extracted from the signals measured from pieces after impregnation with different consolidation products is included.

1. Introduction

The conservation of archaeological objects is a careful activity that requires the most complete possible information about characteristics of the object to be conserved. In that sense, the application of non-destructive testing (NDT) using ultrasound to archaeological object conservation is relevant, since, unlike others analyses (chemical, and so on) it allows to study the object without damage.

This paper presents the results obtained from the application of NDT by ultrasound to several ceramic fragments or sherds corresponding to different historical periods. The information showed here aims to validate the feasibility of that kind of testing in archaeological ceramics at two levels. At the first level, our goal was to be able to classify and group sherds accordingly to its historical periods and at the second level to test the sensitivity grade of the ultrasonic signals to distinguish archaeological fragments treated with different consolidation products at various concentrations using different penetration methods.

The sherds came from the Arqueological Museum of Requena (Valencia) at the East of Spain. They consisted in pieces made of cooked mud from the Bronze, Iberian, Roman and Middle ages. In the Iberian Peninsula those periods are: the protohistoric period called Bronze age (III millennium BC. - II millennium BC.), the Iberian age (VI BC. to the *romanization*). The Roman age (218 BC. - V AC.), and the Middle age (IV – XV AC.).

This latter has a subperiod featured by the Muslim presence from 711 to 1492 AC. The fragments of the Iberian age were two types: decorated and non-decorated. Roman pieces were three types: *Sigillata,* common, amphora, and *Muslim* pieces were three types: yellow, red, and black paste.

The set of analyzed pieces consisted of 230 sherds: 119 non-consolidated, and 111 consolidated (conserved) pieces, see Table 1. For the conservation of the pieces, the following consolidation products were used: Paraloid (ethyl metacrylato) B-72, Acryl (acryl resin) 33 and Estel (ethyl sylicate) 1000. Each one of the pieces was treated with one of the consolidation products at 5, 10, 20, 40 or 60% concentration using one of two methods for product penetration: impregnation and immersion [1].

Table 1. Sherd Distribution

Age	Non-consolidated	Consolidated				Total
		Consolidation product			Consolidated total	
		Acryl	Paraloid	Sylicate		
Bronze	15	6	5	4	15	30
Iberian	22	10	6	8	24	46
Roman	40	13	10	8	31	71
Middle	42	17	11	13	41	83
Total	119	46	32	33	111	230

Ultrasound has been used in archaeological applications such as, ocean exploration to detect wrecks, imaging of archaeological sites, cleaning archaeological objects [2-4]. There are few references of the application of ultrasonic signal processing to support the conservation/restoration of archaeological ceramics [5].

Ultrasonic signals were analyzed following two approaches: general analysis based on classification by linear discriminant analysis (LDA) and material characterization analysis based on ultrasonic signature using the centroid frequency evolution [6-9]. The following parameters from the ultrasonic signals were extracted by time-frequency techniques: Propagation velocity, principal frequency, principal frequency attenuation, signal power, total signal attenuation, attenuation curve initial value, time-reversibility, centroid frequency, and third order autocovariance [10].

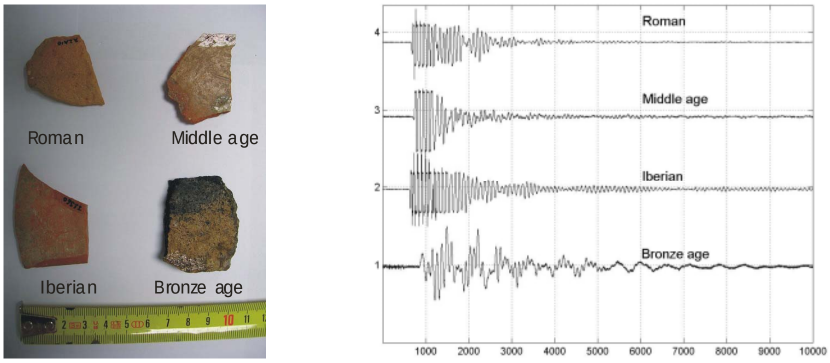
The following sections describe the ultrasound NDT method used for testing the pieces, the parameters extracted from the recorded ultrasonic signal, the classification procedure and results, an ultrasonic signature analysis and findings of material characterization, and finally the conclusions and future work.

2. Ultrasonic Non-Destructive Testing

Ultrasound transducers have a working transmission frequency; the higher transducer frequency the higher capacity to detect small details, but also lower capacity of material penetration. Therefore using high frequencies is possible to detect smaller details but they have to be closer to material surface [11,12]. Several working frequencies were tried, such as 10, 5, and 2.25 MHz, those were the frequencies available for transducers with a suitable size for measuring the small and curved archaeological sherds, see Figure 1. Due to the material physical properties, the ultrasonic reflection could not be measured in pulse-echo technique, thus through-transmission technique was applied. The best results were obtained with 2.25 MHz transducers made to work at a lower frequency (1.050 MHz).

Pieces were measured one time by transmission mode, using a rubber adaptor, see Figure 2. This coupling method was selected due to good ultrasonic transmission and to be innocuous for the pieces, after trying with methods such as immersion and direct contact.

This latter (contact by gel) offers a very good coupling but the time for measuring is very short to avoid gel absorption by the piece.



a. Sherds **b.** Ultrasonic signals

Figure 1. Ultrasonic measuring

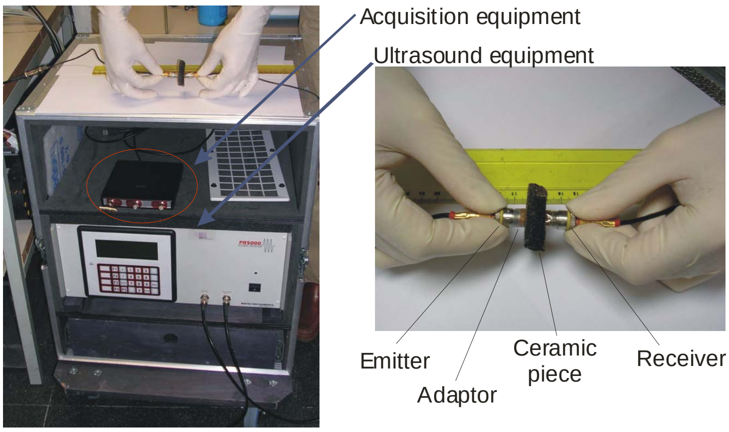


Figure 2. Ultrasonic measuring

2.1 Equipment Setup

The equipment setup used for NDE of the archaeological ceramics is described in Table 2.

Table 2. Equipment Setup

Ultrasound equipment setup		**Acquisition equipment setup**	
Ultrasound equipment	*Matec PR5000*	Acquisition equipment	*Tektronix TDS 3012*
Transducers	*2.25 MHz Krautkramer*	Sampling frequency	*100 MHz*
Pulse width	*4 μs*	Sample number	*10000*
Pulse amplitude	*100%*	Observation time	*1 ms*
Analog filter	*None*	Vertical resolution	*16 bits*
Excitation signal	*Tone burst 1.050 MHz*	Dynamic range	*100 mV/division*
Operation mode	*Through-transmission*	Average	*16 acquisitions*
Amplifier gain	*43 dB*	PC connection	*USB*

2.2 Ultrasonic Parameters for Classification

In order to test the sensitivity of the ultrasonic signals to changes of material physical properties from different ages, various parameters for classifying were calculated from the signals. The ultrasonic parameters -features- and the formulas used for their calculation are in Table 3, where $x(t)$ is the recorded signal and $X(f,t)$ is the Short Time Fourier Transform, $\langle \bullet \rangle$ is the temporal average, σ_x is an estimate of $x(t)$ standard deviation, and instantaneous centroid frequency was calculated at $t_0 = 15\mu s$. The centroid frequency also was used to analyze the signature of the consolidation products.

Table 3. Ultrasonic parameters

Ultrasonic parameter	**Formula**	**Ultrasonic parameter**	**Formula**
Propagation velocity	$v = \dfrac{piece\ thickness}{ultrasound\ time\ of\ flight}$	Attenuation curve initial value (dB)	$x(t) = Ae^{-\beta t}$ $Po = 10\log(A)$
Principal frequency	$f_{max} = \underbrace{max}_{f} \lvert X(f,t) \rvert$	Time-reversibility	$\dfrac{1}{\sigma_x^3}\left\langle \left(\dfrac{dx(t)}{dt}\right)^3 \right\rangle$
Principal frequency attenuation (dB)	$x_i(f,t) =$ $TF^{-1}\{X(f,t) \cdot Notch_{filter,f_i}(f)\}$ $x_i(f,t) = Ae^{-t\beta_i}$ $\Rightarrow At\_F\,Reson = \beta_i$	Centroid frequency	$fc(t) = \dfrac{\int_{f_1}^{f_2} f \cdot \lvert X(f,t) \rvert df}{\int_{f_1}^{f_2} \lvert X(f,t) \rvert df}$
Signal power	$P\_Total = \dfrac{\int_0^T \lvert x(t) \rvert^2 dt}{T}$	Third order autocovariance	$\langle x(t) \cdot x(t-1) \cdot x(t-2) \rangle$
Total signal attenuation	$x(t) = Ae^{-\beta t}$ $Aten\_Total = \beta$	Instantaneous centroid frequency	$fc(t = t_0)$

3. Discrimination Analysis of the Archaeological Sherds

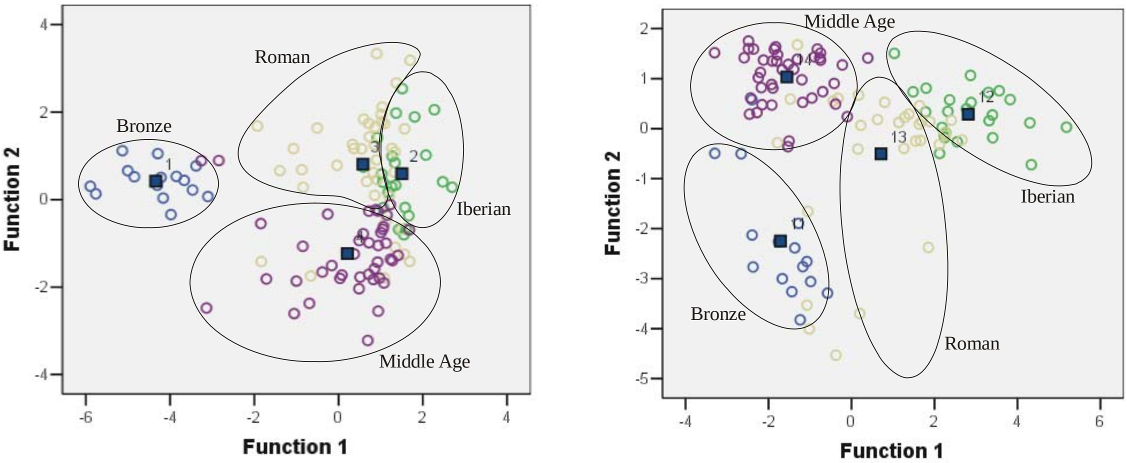
Classification procedure followed a supervised scheme with 5 steps: i.) Label the database cases with the known archaeological period, ii.) Select a case of the database, iii.) Estimate an archaeological period for the selected case by LDA algorithm using the remaining cases as training data, iv.) Repeat steps ii and iii until the end of the cases, and v.) Calculate the percentage of success for classification results. Basically the classifier works calculating distances between the cases (pieces) and separating them in disjoint classification subspaces, delimited by discriminant functions calculated as linear combinations of the original variables (ultrasonic parameters).

The space for ceramic age classification was composed by 10 features (Table 3) and the cases were 230, for a total of 2300 data. The classes were defined as the ceramic archaeological periods, i.e., Bronze, Iberian, Roman and Middle age. The space for consolidation product classification was composed by the same 10 features and the cases were 111, for a total of 1100 data. The classes were defined as the consolidation products, i.e., acryl, paraloid, sylicate.

3.1 Ceramic Age Classification

Figures 3a and 3b are scatterplot diagrams of the ceramic pieces set using two of the three discriminant functions, obtained by the classification procedure, for non-consolidated and consolidated pieces respectively. The cases (sherds) are represented as circles with belong to a centroid group that represents the gravity center of each one of the classes (Bronze, Iberian, Roman, and Middle age). There are some curves added to the diagrams to visualize the delimited region of each class (where can be found more cases of the class). Due to the classification was not perfect, i.e. 100% of success assigning each case to its known class, some cases were located in wrong regions.

Figure 3a shows the centroids corresponding to Roman and Iberian very close, and many cases near to those centroids, so the classifier fails assigning the right class to the Roman and Iberan cases. Besides, it is not possible to depict a symmetric axis region that borders the most of Roman cases projected in the subspace delimited by the estimated LDA hyperplanes of the principal discriminant functions 1 and 2. Figure 3b shows a better result than Figure 3a, with centroids more separated and symmetric classification regions depicted.



3a. Non-consolidated ceramics **3b.** Consolidated ceramics

Figure 3. Classification by age of the archaeological ceramics

Figure 4a shows general quantitative results of the classification by age of the ceramics pieces. General average percentages of success are good, 72.3 and 80.2% respectively for non-consolidated and consolidated pieces. The higher percentages of success are Bronze (non-consolidated) and Middle age (consolidated); and the lower percentages of success are Roman (non-consolidated) and Roman (consolidated).

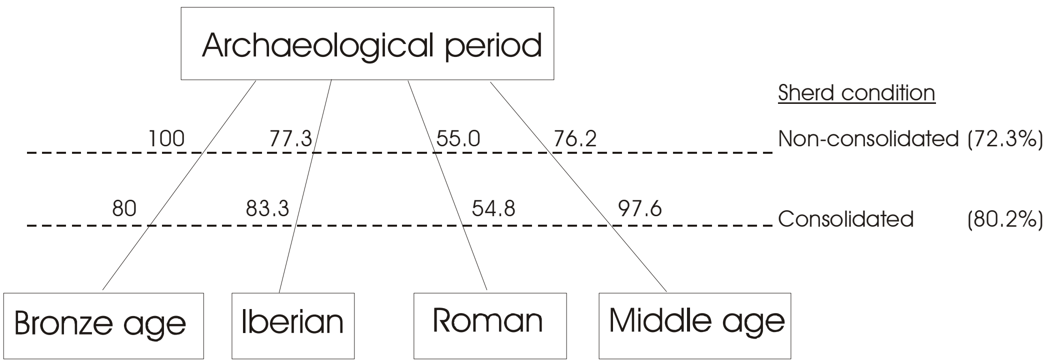


Figure 4. Percentage of success in age classification

The variables employed by the classifier for non-consolidated pieces were: attenuation curve initial value, centroid frequency, third order autocovariance, and principal frequency: The variables entered for consolidated pieces classification were: time-reversibility, total signal attenuation, attenuation curve initial value, and instantaneous centroid frequency. It has to be noted that better parameters selected for the classifier for consolidated pieces are related with non-linearity degree measure (time-reversibility) and non-stationarity of the signals (instantaneous centroid frequency) that could reflects a phenomenon of inhomogeneity of the pieces due to superficial penetration of the consolidation product. In contrast better parameters for non-consolidated pieces assume signal stationarity. In addition, ultrasound propagation velocity that was a very good discriminant in a previous work [5] was not included in classification. This latter could be due to difficulties in measuring piece thickness because aspects such as, curved geometry, irregular shape, plaits, and calcareous and soil formations.

The resulting groups obtained in both age classifications (consolidated and non-consolidated pieces) were highly correlated with *a priori* knowledge on the kind of ceramic porosity: Bronze and Iberian - high porosity -, Middle age - medium porosity -, and Roman - low porosity -.

3.2 Consolidation Product Classification

It was confirmed that the classification of the pieces considering the consolidation products as classes has no sense, because the physical properties of the pieces are different for the several ages and they do not change significantly with the consolidation procedure. Then the classification of consolidation products was made by historical age. Results of classification by age-consolidation product were not good, except for the Iberian pieces. General percentages of success were: Bronze (53.3%), Iberian (70.8%), Roman (51.6%), and Middle age (53.7%). Figure 5 shows the scatterplot diagram for Iberian consolidated pieces, corresponding to the following percentage of success: Acryl (80%), Paraloid (66.7%), and Sylicate (62.5%). For this group of pieces, the variables used for the classifier were: attenuation curve initial value, third order autocovariance, instantaneous centroid frequency, and principal frequency.

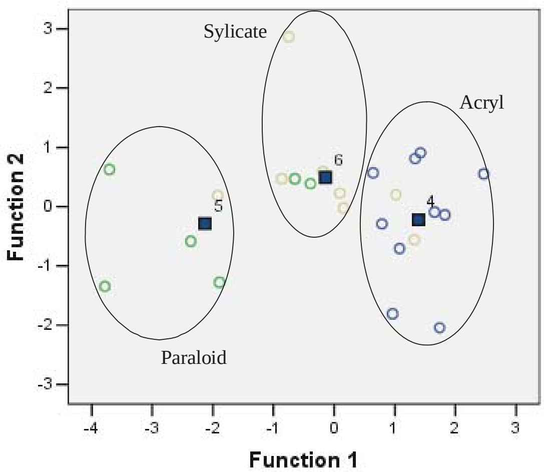


Figure 5. Classification by age of the archaeological ceramics

The causes of misclassification by consolidation product could be related with the variables, consolidation product concentration and consolidation method that were no considered in the analysis. Taking into account the number of consolidated pieces by age, if those variables had been considered, the number of cases by class had not been enough for LDA. However, the results are not quite short and they reveal possibilities for distinguishing the consolidation products. The second approach applied in this work, could be more suitable with this kind of small quantities of samples. It is explained in next section.

4. Ultrasonic Signature Analysis of the Consolidation Products

The ultrasonic signature consists in the evolution curve of the central frequency of the injected ultrasonic pulse while it propagates inside the material. The centroid frequency evolution is related to the way the different frequency components of the pulse spectrum progressively are attenuated. In general, the frequencies attenuate in a different way causing spectrum variations and consequently the central frequency of the pulse varies. These variations are inherent to the physical properties of the material and can enable its characterization. The technique to obtain the ultrasonic signature is made by a non-stationary analysis of the grain noise at different penetration depths of the pulse and it has two steps: estimation of the pulse spectrum and estimation of the central frequency of the spectrum.

Grain noise is generated by the superposition of multiple echoes caused by the material microstructure, when an ultrasonic pulse with a suitable frequency is injected in the material. Grain noise can be modelled stochastically and spectral techniques can be applied to the model to extract parameters and correlate them with material physical properties, allowing its characterization. A classic method to analyze the dependence of the attenuation with the frequency is measuring the significant frequency through the grain noise spectrum. The instantaneous centroid frequency, the one corresponding to the maximum energy, and the resonance frequency feature certain variation with depth due to the dependence with the frequency attenuation [8,9].

Figure 6 shows the evolution of the centroid frequency in the signal record time for the Iberian age ceramics consolidated by the immersion method. This 5-piece set features a dissimilar ultrasonic signature for each case, i.e., the signature of the consolidation product (acryl, paraloid, sylicate) used in the ceramic can be discriminated. This result of high correlation between the ultrasonic signature and the material properties was obtained in some subsets of the analyzed pieces. Thickness of the consolidated pieces shows a variance that did not allow comparing the ultrasonic signature of all the pieces of an age.

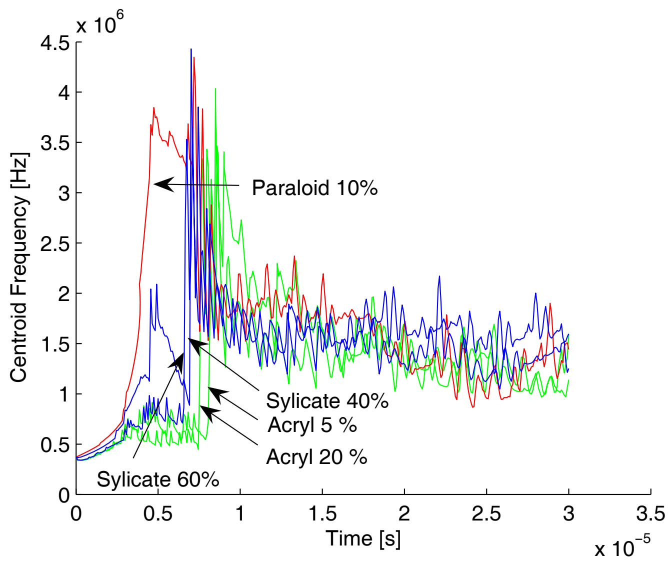


Figure 6. Ultrasonic signature by centroid frequency evolution in Iberian Age pieces for the different consolidation products (thickness 5 - 6.5 mm.)

5. Conclusions

Ultrasound NDT applied to archaeological ceramics has shown good results in general age classification for a set of ceramic fragments from the East of Spain. The resulting clusters were highly correlated with a priori knowledge on the kind of ceramic paste porosity. More detailed studies could demonstrate the feasibility of such technique as an economic alternative for age analysis.

Results of ultrasonic signature are promising to distinguish among consolidation products for ceramic conservation/restoration, nevertheless it is necessary a more detailed study to confirm preliminary results.

It has to be noted that parameters employed by the classifier are different for non-consolidated and consolidated pieces. For the latter pieces, parameters that exploit the non-stationarity of the signal (instantaneous centroid frequency) and parameters that measure the non-linearility degree (time-reversibility, third order autocovariance) are selected as better discriminants. In contrast, parameters that assume stationarity of the signal become better discriminants for non-consolidated pieces. Those results could be explained by the knowledge that consolidation techniques only consolidate the more superficial area of the ceramic pieces, thus highlighting the non-stationarity signal behaviour.

Measuring of archaeological ceramics involves some difficulties in thickness measuring due to sherd characteristics such as, curved geometry, irregular shape, plaits, and calcareous and soil formations. The manual coupling method using a rubber adaptor has to be standardized in order to obtain a standard press during measuring and to access opposite piece sides properly. Measuring standardization will produce more reliable measurements of the archaeological ceramics. It is especially significant for parameters such as ultrasound propagation velocity.

Acknowledgements

This work has been supported by Spanish Administration and the FEDER Programme of the European Community under grant TEC 2005-01820, and *Universidad Politécnica de Valencia* under interdisciplinary grant 2004-0900.

References

[1] B. Carrascosa, and M. Lastras, “Socarrats”, Restoration & Rehabilitation, América Ibérica, n. 100, 2005.
[2] A. Murray, M. F. Mecklenburg, C. M. Fortenko, and R. E. Green, "Detection of Delaminations in Art Objects Using Air-Coupled Ultrasound", Proceedings of Materials Issues in Art and Archaeology III, pp. 371-378, San Francisco CA, 1992.
[3] R. Cribbs, and F. Saleh, “An Ultrasonic Based System Used for Non-destructive Imaging of Archaeological Sites”, in: Informatica ed Egittologia all’inizio degli anni ‘90. A cura di Francesco Tiradritti, Roma, Bulzoni Editore, pp. 97-108, 1996.
[4] W.I. Sellers, “Ultrasonic Cave Mapping”, J. of Archaeological Science, Elsevier, n. 25, pp. 867-873, 1998.
[5] B. Carrascosa, R. Miralles, A. Salazar, M. Lastras, A. Parra, “Aplicación de los ensayos no-destructivos por ultrasonidos en cerámicas arqueológicas”, II Conference of GEIIC - Spanish IIC, Barcelona, 2005.
[6] R. Duda, P.E. Hart, and D.G. Stork, Pattern Classification, Wiley-Interscience, 2nd edition, 2000.
[7] B.D. Ripley, Recognition and Neural Networks, Cambridge University Press, 1997.
[8] L. Vergara, J. Gosalbez, R. Miralles, and I. Bosch, “On Estimating the Center Frequency of Ultrasonic Pulses”, Ultrasonics, Elsevier, v. 42, pp. 813-818, 2004.
[9] L. Vergara, et al., “NDE Ultrasonic Methods to Characterize the Porosity of Mortar”, NDT&E International, Elsevier, v. 34 n. 8, pp. 557-562, 2001.
[10] L. Cohen, Time-frequency analysis, Englewood Cliffs, Prentice Hall, 1995.
[11] J. Krautkrämer, Ultrasonic Testing of Materials, Springer, 4th edition, Berlin, 1990.
[12] J.D. Cheeke, Fundamentals and Applications of Ultrasonic Waves, CRC Press LLC, USA, 2002.