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L.Stevanato[1], G.Nebbia[2], F.Pino[3], M.M.Mackowiak de Antczak[3], A.Antczak[3], L.Sajo-Bohus[3]*, G.Viesti[1]

[1]Dipartimento di Fisica ed Astronomia dell' Università di Padova, Via Marzolo 8, I-35131Padova, (ITALY)
[2]INFN Sezione di Padova, Via Marzolo 8, I-35131Padova, (ITALY)
[3]Universidad Simon Bolivar, Apartado 89000, 1080A Caracas, (VENEZUELA)
E-mail : sajobohus@gmail.com
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*Corrresponding aurhor's Name & Addess

L.Sajo-Bohus
Universidad Simon Bolivar, Apartado 89000, 1080A Caracas, (VENEZUELA)
E-mail : sajobohus@gmail.com

Neutron-non-destructive assay of archaeological samples from La Tortuga island of Venezuela

Abstract

Archaeological material from the Venezuelan island of La Tortuga is characterized by measuring the ratio of the attenuation of neutron and gamma rays from a ^{252}Cf source. Moreover surface analysis by using X-ray fluorescence has been also performed. The classification using the ^{252}Cf source allows one to obtain the average atomic number of the sample, avoiding problems due to the patina that some time covers the surface of ancient ceramics. The potentiality of the method is discussed also in view of future mobile systems using this technique.

Key Words

Pre-Hispanic culture; Pottery; Non-destructive assay; Neutron and gamma ray transmission; ^{252}Cf source.

INTRODUCTION

Non-destructive assay (NDA) using a ^{252}Cf source has been developed recently at the University of Padova, Italy, for material recognition by using the attenuation of neutrons and gamma-rays. Such technique provides the best results for relatively low-atomic-number material[7,8]. The facility used in this work is optimized for elements and chemical compounds discrimination in small archaeological samples, providing the required accuracy in a short time. A first application of this method is reported here for Amerindian archaeological samples from Latin American sites.

Several years ago, archaeologists at Simón Bolívar University initiated provenance study of pre-Hispanic pottery excavated on the islands located off the Venezuelan mainland that were seasonally occupied by the Amerindians between A.D. 1200 and the European contact period[1,2]. It was suggested that the islands were visited by the bearers of the Valencia style pottery originated at the Lake Valencia Basin. To confirm this hypothesis dozens of human pottery figurines and their fragments were analyzed by different non-destructive techniques including expensive ones such as PGA, INAA and NRCA[3]. Though for many samples, the concentrations of major elements and some trace components have been determined[5,9], it was evident that some kind of preliminary selection of the recollected large number of samples must be made by using less expensive and faster methods, better yet if on site. The other important finding of the study is related to the fact that even if the island figurines are compared by chemical elements to their stylistically related counterparts from the mainland, the analyses require sound chronological control over the samples. Therefore the bulk analysis, based on the non-destructive assay technique with the ^{252}Cf source mentioned above is considered complementary to more precise reactor based techniques, since it provides a fast preliminary classification without damaging the artifact.

ARCHAEOLOGICAL SITE LOCATION AND SAMPLING

The island La Tortuga is the second largest of Venezuela after the island of Margarita.

A set of three groups of ceramic samples was submitted for the analysis:

1) Samples labeled as TRSD come from the eastern edge of La Tortuga Island known as La Salina where extensive archaeological excavations were carried out between 1992 and 2010. More specifically, these samples were recuperated from the archaeological site called Dunas that yielded materials left by the Anglo-American sailors, who exploited salt in the adjacent natural salt pans, between the end of the 17th century and 1781.
2) Samples labeled as TRTRP also come from *La Salina* location, specifically from a site denominated *Terraplén 1* where the remains of a small fortified earthwork existed in association of Dutch salt exploitation between 1622 and 1638.
3) The last of samples labeled as TRH come from *Los Cumaneces* site located two kilometers north from *La Salina.* The systematic excavations carried out during 1996 revealed the existence of a series of pre-Hispanic campsites dated to between A.D. 200 and 900.

The sizes and shapes of the sampled potsherds vary, but in general they have only a few square centimeters of surface, slightly larger than the irradiation window as described in the next section. The interest from an archaeological point of view consists in shedding light on a time frame to which the samples may be assigned, especially because clear stratigraphic, contextual and radiocarbon (except for the TRH sample group) indications are lacking. Preliminary archaeological analysis suggests that the samples labeled as TRSD and TRTRP might have not been related to the pre-Hispanic period in opposition to the samples of the group TRH, for which the contextual associations and radiocarbon dating are congruently pre-Hispanic. There is some possibility that samples identified as TRSD and TRTRP may have the same chronology/origin as they come from sites associated with the colonial activities. If both referred samples come from the colonial times, then there might be a difference of almost a century (i.e. from the 17th to 18 century) between them. Finally, as some tenuous stratigraphic indications seem to suggest, the samples labeled TRTRP may be of pre-Hispanic origin and thus related to the TRH samples (Antczak and Antczak in press). In conclusion, the results of this study may provide valuable chronological/cultural information for the studied samples, contexts and sites. Based on the analysis of its elemental or bulk characteristics, inhomogeneity or homogeneity between samples labeled TRSD, TRTRP and TRH it is conceivable that the results shed light on their chronology. Otherwise from this study we will have some elements to establish the degree of similarity and make some conclusion to guide future research; in any case the analitycal nuclera technique is a novelty in archaeological applications and therefore an original contribution to Interdisciplinary Science.

EXPERIMENTAL SET-UP

The NDA system used in this study is based on a 10^6 neutron/s ^{252}Cf source collimated by using a Teflon box having an aperture of about 2.5 x 3 cm^2. Neutrons and gamma rays are detected by a 5 cm diameter 5 cm thick EJ-301 liquid scintillator cell coupled with a XP2020 photomultiplier placed at about 1 m from the source. The detector is managed by a CAEN VME electronic front end based on a fast digitizer. The front end makes use of VME mini-crate (4 slots) with a Bridge USB V1718. The mini-crate hosts a HV system (V6533 Programmable HV Power Supply (6 Ch., 4 kV, 3 mA, 9 W) and a V1720 8 Channel 12bit 250 MS/s Digitizer. Inside the V1720, Digital Pulse Processing (DPP) algorithms are implemented by using FPGA, providing the possibility of obtaining on line the Pulse Shape Discrimination (PSD) of neutrons and gamma rays from the liquid scintillator. The measurements consist of sample-out and sample-in runs with the sample placed at the exit of the Teflon collimator so that the neutron and gamma beams from the ^{252}Cf source are attenuated. For each run a PSD 2-D plot is obtained as the one shown in Figure 1 from which the yields of neutron and gamma ray are determined.

As reported in previous works[4,6], material recognition can be obtained from the ratio R of the absorption coefficients for neutrons μ_n and gamma rays μ_γ. The latter being the logarithmic ratio of the measured transmission factors for neutrons ($I_n / I_{n,0}$) and gamma rays ($I_\gamma / I_{\gamma,0}$) respectively:

$$R = \mu_n / \mu_\gamma = \ln (I_n / I_{n,0}) / \ln (I_\gamma / I_{\gamma,0})$$

where I_n andI_γ ($I_{n,0}$ and $I_{\gamma,0}$) are the measured yields for neutrons and gamma rays with (without) the sample. The ratio R characterizes a given material and does not depend on the thickness of the sample. Since in this work we are mainly interested in verifying relative differences between the samples, we have not converted the measured R values in average atomic number $<Z>$.

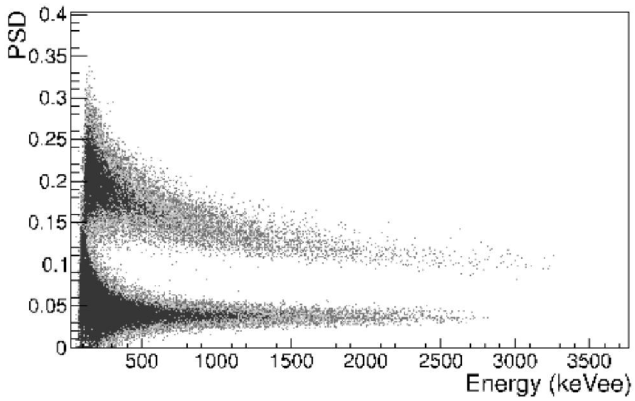

Figure 1 : Neutron-gamma PSD scatter plot. The neutrons are the upper and the gamma ray are the lower distributions, respectively.

As a first step of this work, the reproducibility of the measurements was verified. To this end a Venezuelan handmade brick made in Caracas 30 years ago was cut in 11 samples each with an area of about 3 x 3.5 cm^2 and 2 cm thickness. For each sample the measurement run lasted 15 minutes. The distribution of the measured R values is reported in Figure 2.

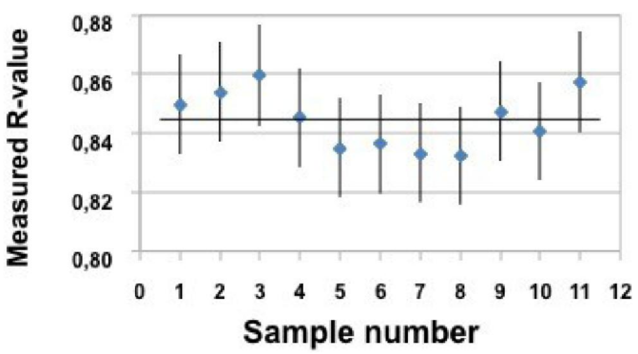

Figure 2 : Calibration with sample of bricks. Distribution of the 11 measured R-values. The horizontal line marks the average value.

Each point reported in Figure 2 is affected by statistical uncertainty of about 2% whereas the deviation from the average R-value (R=0.84) is between 1-2 %. This demonstrates that with a 15 minutes run results are already acceptable and highly reproducible when the sample thickness is about 2 cm.

RESULTS

The results relative to the samples from La Tortuga Island are reported in TABLE 1.

The measured R-values are generally larger compared to the calibration brick indicating a higher average atomic number $<Z>$. Moreover, the statistical uncertainty is fluctuating about 6% depending on the sample thickness. Comparing the measured R as a function of the different sites at La Tortuga Island, it appears that measured values are around R=1 for all locations with the exception of some TRSD samples that are well below this number.

TABLE 1 : Group of samples analyzed

Sample	Measured R-value
TRSD1	0.89 $\pm$ 0.04
TRSD2	0.93 $\pm$ 0.06
TRSD4	1.06 $\pm$ 0.09
TRSD5	1.06 $\pm$ 0.07
TRH441	1.08 $\pm$ 0.05
TRH443	0.95 $\pm$ 0.05
TRH321	1.03 $\pm$ 0.06
TRH322	0.95 $\pm$ 0.05
TRTRP11	1.02 $\pm$ 0.06
TRTRP14	1.07 $\pm$ 0.05
TRTRP15	0.99 $\pm$ 0.05

At first sight it would seem that the group TRTRP has dispersion similar to the TRH group, suggesting similar pre-Hispanic origin of these samples. The third group, TRSD, shows a larger dispersion value compared to the other two groups, suggesting that it may be related to a different time frame, possibly the colonial times. We are aware that the similar time frame does not warrant a common cultural provenance of related samples. Therefore, the above hypotheses need to be substantiated by increasing the number of samples for each group and at the same time lowering the uncertainty in the measured R-value to the value of about 2% as the one in the measurements on the (thicker) modern brick. This will leave us with a smaller uncertainty in the difference between sample groups.

Finally, samples were analyzed by X-ray fluorescence looking at the correlation of those results with measured R-value, i.e. the correlation between the surface chemical composition and the bulk average atomic number, as reported in Figure 3. Optical inspection reveals indeed that some samples are covered by a patina that will interfere with sample bulk composition during X-ray measurements. It seems from Figure 3 that a linear correlation exists between the two measurements with the inclusion of the modern brick data and with the exception of the samples in which the patina is covering the surface (points outside

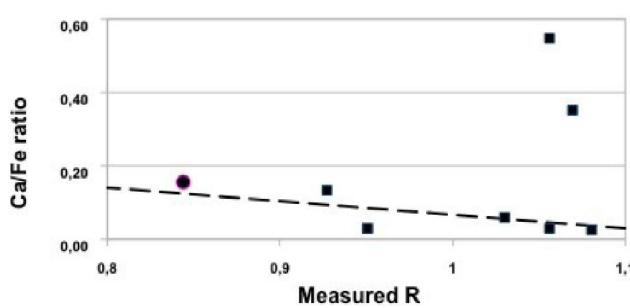

Figure 3 : Correlation between the measured R-value and XRF results. The black dot refers to the calibration bricks.

the expected values far away from the data trend). It is interesting to note that for the TRSD4 sample (R = 1.06) the reported measure of both surfaces shows the well-known dependence of the XRF from the patina.

DISCUSSION AND CONCLUSIONS

The set of samples analyzed by using the attenuation of neutrons and gamma-rays technique show some interesting results on two aspects being the first a new archaeological application, the other a good correlation existing with the XRF measurements. The presence of calcium on the sample surface may be related to the local environment soil rich in shell sand; however the most important parameter is its ratio with iron concentration. On general term results given in TABLE 1, indicate that material recognition for La Tortuga Island sample set, related to different archaeological sites, is useful for a preliminary selection and gives important information on its bulk composition. In this study case, we observe that two groups of samples have similar response to bulk analysis. On the other hand, a difference exists between *Los Cumaneces* samples of pre-Hispanic origin, taken here as reference, and those from *La Salina* site of *Dunas*. Furthermore, we learn that difficulty exists in the definitive interpretation mainly due to the fact that too few specimens for each group of samples were available at our disposal and therefore limited statistical data could guide us to determine precisely their belonging to a given chronological group.

The potentiality of the method is therefore evidenced in this preliminary study also in view of the development of a transportable instrument that might be employed to obtain on-site analysis of sample. The technology development that allows the use of this technique to support archaeological studies is at hand and in the near future a portable device will be available.

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