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GAMMA RAY SPECTROSCOPY FOR ARTIFICIAL CONTAMINATION EFFECTS EVALUATION IN LUMINISCENCE DATING OF ARTEFACTS FROM LOW DEPTH LAYERS IN SOUTHERN ROMANIA*

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Abstract. Luminescent dating of ancient objects found by archaeologists needs to take into account the gamma-ray dose debit resulting from the natural radioactive series in order to determine the age of the artifacts. In countries like Ukraine, Romania, Poland, Sweden, which have been severely affected by the radioactive contamination resulting from Chernobyl, it seems justified to ask if the artificial contribution does affect the results. Samples from the southern Romanian region were analyzed in order to obtain a straight answer. The first results are presented.

Key words: HPGe detectors, gamma spectra, luminescent dating, artificial radioactivity, coincidence summing effects.

1. INTRODUCTION

Artefacts from the Roman period can be, under certain conditions, dated by luminescent methods, *i.e.* thermoluminescence and optically stimulated luminescence. This type of measurements is performed on fine or coarse grain of silicates that are extracted from the artifact after a certain number of steps in which acid digestion of the original piece of ceramic is performed. It is assumed that the signal selected for dating comes from traps stable in time (the signal has to be carefully selected and the unwanted components have to be removed), trap filling is the same during laboratory and natural irradiation (this assumption is difficult to be tested) [1] and the luminescence per unit of trapped charge is the same during all measurements.

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In order to date the samples one needs to correlate the luminescent signal with the total dose accumulated in the silicates from the moment of their fabrication to the time they are analyzed. This dose is then divided by an annual dose which is specific to the region where the artifact was found in order to determine the age.

The annual dose includes contributions from alpha, beta, gamma and cosmic radiation [2], all affected with specific coefficients, that are converted into dose debit

$$\text{Annual Dose} = \frac{D_{\alpha(U,Th)}}{W_{\alpha}} + \frac{D_{\beta(U,Th,K,Rb)}}{W_{\beta}} + \frac{D_{\gamma(U,Th,K)}}{W_{\gamma}} + D_{\cos}.$$

For further details, the interested reader is referred to the bibliography.

Gamma spectroscopy is a very important method for the dose debit and annual dose evaluation. While analyzing soil samples from the Roman period coming from the Racari site (Southern Romania), we detected presence of ^{137}Cs, which is an artificial radioelement resulting mainly from the Chernobyl accident in the spectra. To our knowledge, there is only one paper [3] linked to this dating topic up to now, but it deals with young sediments instead of archaeological ones. The question that emerged after finding ^{137}Cs in Roman period soil was "has the ^{137}Cs activity a major contribution to the total dose or not?" The aim of this work is to prove that the answer is no.

2. BRIEF DESCRIPTION OF THE DATING METHODS

The basic principle is quite simple, and it consists in dividing the accumulated radiation dose in the luminescent centers (traps) within the quartz crystals from the samples by the dose debit that is characteristic to the environment the sample was found in. This leads us to soil sample analysis.

The crystals are from samples that have their first 2-3 mm removed from all sides, in order to prevent contribution of α and β particles of external origin to the dose, consequently to the luminescent signal. Then, they're broken (or even milled, depending on the size of the grains one needs to obtain), and the grains are treated with HCl and H_2O_2 [4] to remove organic components, and finally with HF (40%), to eliminate α contribution from the inside of the grain. The total dose to be determined could then be considered equal to the gamma contribution and a correction factor for beta activity within the grain and cosmic radiation. Thus, these details are not really necessary for our study, as the present work concerns dose evaluation preliminaries for age evaluation in dating Roman period artifacts from southern Romania. We focus on the gamma spectroscopy contribution to the evaluation of natural and artificial dose, using commercial and in-house software

for spectra analysis. The aim of this study is to show the importance of the artificial component to the luminescent signal generated by TL/OSL stimulations for archaeometry experiments, particularly for the Roman period.

In other words we are so far neither interested by the age determination itself, nor the study of the gamma contribution to the total dose, but just to obtain the magnitude of the ratio natural/artificial gamma contribution to this dose, in order to acknowledge if one has to take artificial contributions into account or not in such cases.

3. CONTEXT

The samples which were analyzed during the last period and of which we consequently present the results have their origins in the Racari area, Dolj region, Romania. They were sampled under surveillance of expert archaeologists from the National Museum of Romanian History from a 40 cm deep layer of soil, which corresponds to the Roman period of half 2^{nd} century, when the by that time existing fortress was burnt down. The burning process actually resets the luminescent signal, which is of great interest for dating experiments.

The whole area around Racari is flat, no river makes its way around, and no other factors contribute to soil layers movement. This is the main reason for which the depth at which the roman artifacts are found is so small (typically 40 cm, just as the soil samples of interest to us).

The preparation procedure includes drying the soil, the milling, drying again, sealing it in containers designed for this type of analysis. Then the samples are stored for three weeks in order to obtain the radioactive equilibrium in the Uranium-Radium series.

Samples of about 100 grams each were placed on the top surface of a high efficiency gamma ray detector (Ortec, hyper pure Germanium, GMX series, 47% relative efficiency at 1.33 MeV). Thick lead shielding surrounds the detector, and the vicinity is purged with nitrogen. Acquisition time for the spectra is typically one or two days long, in order to have a good statistic in the peaks. Dead time calculation is performed by default. Blank boxes are also analyzed just as the sample in order to extract the peak areas determined by the blanks and the background for accuracy improvement of the analysis. Even if empty, the blanks are sealed with the same quantity of sealant as the real samples, identically distributed, in order to simulate the exact same conditions.

Monte Carlo computation is achieved using GESPECOR, so that the efficiency curve can be corrected and the geometry effect adapted since the calibration sources were placed in containers of different shape than the samples. GESPECOR is also used for the coincidence summing calculations, as it is frequent to have real coincidence summing effects on detectors of such efficiency [5, 6].

Software used for peak analysis is commercial and also in-house. The commercial one is Gamma Vision 32 v. 6.04 from Ortec. The in-house one is called Gadetool, which is an application designed to analyze peak areas over the half-maximum, adapting the calculus for the apparent efficiency and the resulting activities to this situation. Gadetool is also designed to ask for human intervention in any controversial situation, such as anomalous peak widths or shapes.

We mentioned the apparent efficiency because analyzing peaks over half-height takes only part of the events recorded by the detection chain into account, so a correction needs to be performed. Assuming the Gaussian distribution, numerical integration provides the correction factor: the areas need to be divided by 0.761 in order to get to the full peak value. The reason for which the analysis is performed this way is that the events recorded in the over-half-height corresponding interval have a much greater chance of belonging to the actual peak than any other event in the area. Correct peak integration on a larger energy interval in complex spectra would require too many corrections of the background, as it is heavily influenced by close peaks.

After a quick manual review of the results, we proceed to the activity calculations meant to determine the natural dose debit of gamma radiation in that soil, and to see if the contribution of artificial radionuclides over the last 24 years (Chernobyl origin) is important enough to be taken into account.

This experiment provides a quantitative evaluation of the contribution of human induced radioactivity to the total gamma dose for ancient artifacts in the last decades; further it shall be easy to analyze the magnitude of this contribution and make predictions on the possibility of neglecting their effect, if the case is so. The contribution of artificial radioelements can be predicted by the fact that water brings them to the deeper layers of soil; the question is how much they influence the total dose, as the age equation has to take into account every type of radiation. This means the expression of the total radiation dose includes the contribution of any gamma radiation, whether belonging to the natural series of to events such as the Chernobyl accident.

4. EXPERIMENTAL RESULTS

The ^{137}Cs turns into $^{137}Ba^{m}$ (94.6%) *via* a beta decay; ^{137}Cs has a half life of about 30 years, and $^{137}Ba^{m}$ has one of 2.5 min. This means that statistically, $^{137}Ba^{m}$ emits the 661 keV quanta immediately after the beta decay. In other words, the concentration of ^{137}Cs can be determined by studying the 661 keV peaks resulting from the metastable Barium de-excitation.

Spectra acquisition typically lasts from 24 to 48 h, in order to obtain a decent statistic for the regions of interest.

In order to evaluate the contribution of each isotope, Gadetool analyzed the following peaks:

– 1460.8 keV for ^{40}K

– 295.3 keV (^{214}Pb) and 609.3 keV (^{214}Bi) for ^{238}U

– 338.3 keV for ^{232}Th

- 661.6 keV for ^{137}Cs

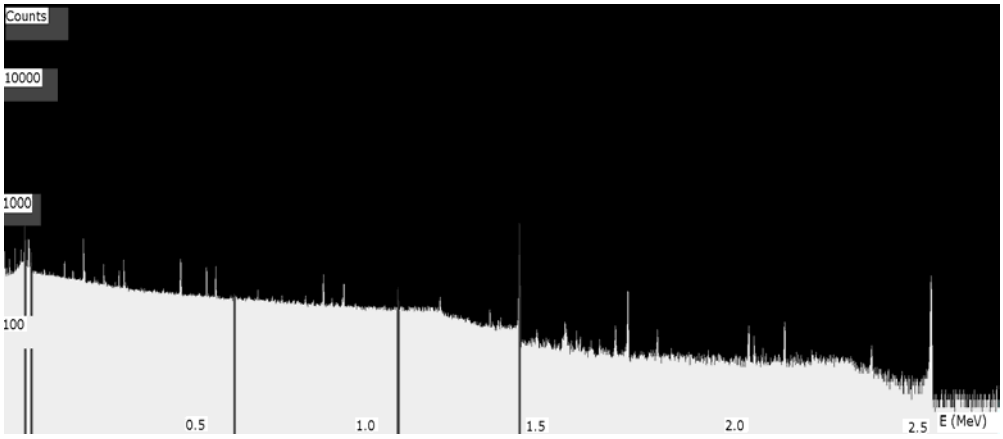

Fig. 1 – Typical soil sample spectra.

Correction factors were applied for the true coincidence summing, the geometry effects and the statistic effect of above FWHM analysis, as the entire Gaussian shape in not taken into account by Gadetool. Monte Carlo computation was performed for complementing experimental calibration and also coincidence summing corrections. Use of the response function ensured a better simulation. [7] Blank sample peak area subtraction ensured that we eliminated the background effects, although very small.

The efficiency calibration was completed with a reference material sample from the IAEA. The procedure is quite simple: first, one uses a calibration source of well-known activity. Then one makes the efficiency calculations for the energy range available. Then follows the identification of peaks from radionuclides within the reference material that provide at least one gamma ray energy in the calibrated range. Finally, by identifying peaks from the same element at higher energies that those available from the calibration source and taking into account the ratio of the gamma emission intensities, one can obtain efficiency values for those higher energies.

The activities resulting from the analysis we performed indicate the following:

Table 1

Radionuclide activities [Bq/kg]

Nuclide/Sample	1	2	3	4	Average
U-238	46.66	47.03	44.97	45.42	46.02
Th-232	99.91	99.16	97.93	95.84	98.21
K-40	682.42	697.51	722.47	707.85	702.56
Cs-137	4.98	5.31	5.59	5.81	5.42

5. CONCLUSIONS

Although almost a half life of ^{137}Cs has passed since the Chernobyl accident and the diffusion process in that soil is not well known, we can conclude that Cesium activity contributed at most with 1% of the total annual gamma dose. Knowing the contribution to the archaeological dose has been at least 2 orders of magnitude smaller and timing was 24 years out of almost 2000, and also the total dose is greater than the Gamma dose, it is obvious that the ^{137}Cs contribution to the total dose is less that 0.01%.

Now, knowing that the uncertainty of TL/OSL measurements which are performed in order to obtain the age of ancient artifacts is typically about 10%, we can easily decide not to take Cesium contributions into account for the concerned region.

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