

Determination of the annual beta dose by measuring beta activity using the liquid scintillation technique

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Abstract. This paper presents a method for measuring the gross alpha/beta radioactivity of ancient pottery by using the liquid scintillation technique, Tri - carb2770TR/SL in the alpha/beta discrimination counting mode. The beta radioactivity is converted to the annual dose, which can be applied in dating of pottery by thermoluminescence technique. In comparison with the radiocarbon technique, the preliminary results shown that the liquid scintillation technique may be applied effectively for ancient ceramic dating in Vietnam.

Keywords: Environmental radioactivity, Dosimeter, Thermoluminescence Dating

1. Introduction

Pottery is one of the most important archaeological relics to study on ancient cultures and art histories. Every civilized culture had left some earthen wares featured by their characteristic forms, shapes, designs, materials and other technical (stamps) imprints. Hence, forms and materials are normally used to categorize earthenware by culture and period accordingly. This is particularly important for the archaeological investigation of prehistoric periods, where human culture could not yet be characterized by the written letters remained. The pottery's age is an important term, therefore, if there are more scientifically founded methods to date ancient pottery, the categorization of cultural periods will be more precise. One of the reliable methods to date archaeological pottery is the thermoluminescence method (TL), which is effectively applied in many countries all over the world [1,2].

In the natural environment, the ionizing radiations from the radioactive elements contained in soils causing effects on the material's properties of ancient pottery samples. Natural ionising radiation consist of the alpha, beta and gamma irradiations. These irradiations cause the different effects on the thermoluminescence behaviour of the materials. For example, among all the irradiations, the alpha particles, cause effects only on fine grains due to their short penetration length ($10 \div 40 \mu\text{m}$) although their high ionizing capacity. The beta and gamma irradiation, however, are the main agents to cause thermoluminescence phenomena in large quartz grains due to their very long penetration length, although their lower ionizing capacity. Hence, to date ancient pottery by the quartz inclusion technique, the determination of the beta annual dose plays a determining important role on the precision of method. We have following formula:

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$$A = \frac{P}{0.9D_{\beta} + D_{\gamma} + D_c} \quad (1)$$

where A is thermoluminescence age of pottery (in years -yr); P is the paleodose in sample (Gy); D_{β} is annual beta dose (Gy/yr); D_{γ} is annual gamma dose (Gy/yr); and D_c is annual cosmic dose, which is around 175 $\mu\text{Gy/yr}$ [1-3].

Presently, in the ancient pottery TL dating the following steps for the determination of annual dose rate are: (i) place the thermoluminescence dosimeter at the field; (ii) determine the environmental radioactivity of a representative soil sample for the region concerned and (iii) determine the contents of radioactive elements, such as uranium, potassium and thorium in the soil sample. In general, these measurement procedures shown essential disadvantages, such as: registration time is long, low accuracy and sophisticated equipment is required. Therefore, a new method (procedure), presented in this paper which allows quick and accurate evaluation of the gamma and beta annual dose, will contribute to the significant improvement for the thermoluminescence pottery technique in Vietnam.

In previous research works [5,6], we have presented a procedure to evaluate the environmental gamma dose rate by using the radiometer of the type CPII-68-01 (Figure 1).

The instrument used in this experiment is CPII-68-01 (USSR). A cylinder-shape detector NaJ(Tl) of the size 30mm x 30mm, allowing to measure the energy in range from 50keV to 3000keV. In the practice, such stable and highly sensitive equipment was used to determinate the annual gamma dose in thermoluminescence dating.

In the consistency with the experimental results it was shown that the more accurate the beta dose evaluation is, the more reliable is the technique of thermoluminescence dating [5,6].

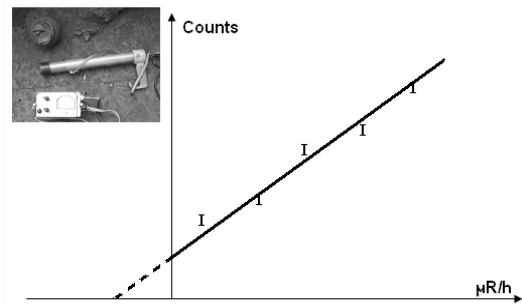


Fig. 1. the measurement of the gamma dose rate using the radiometer CPII-68-01.

This paper presents a new method to evaluate the annual beta dose (D_{β}) by measuring the gross beta activity using the liquid scintillation analyzer Tri-carb 2770TR/SL.

2. Experimentals, results and discussion

The sample preparation procedure for measuring by liquid scintillation technique is presented in Figure 2

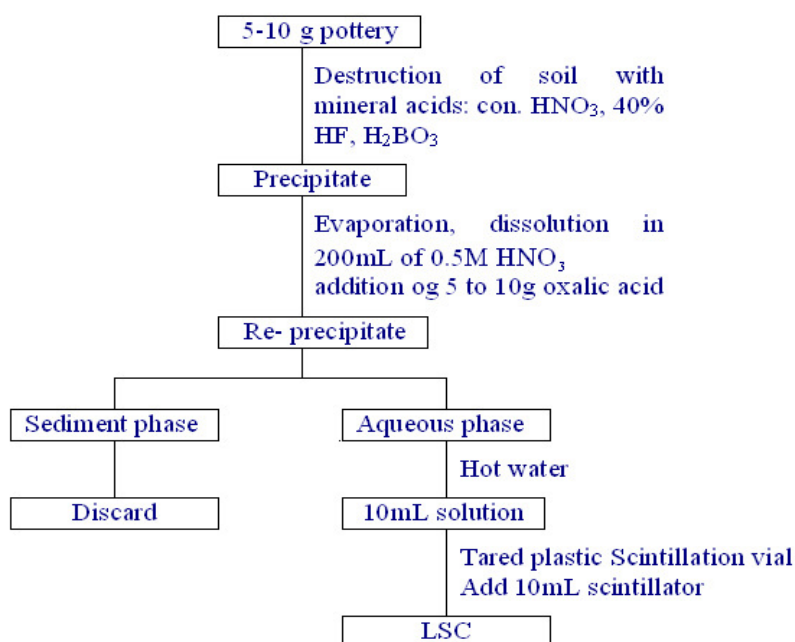


Fig. 2. The scheme of sampling preparation.

The scintillation measurement was carried out on a liquid scintillation analyzer, Packard Tri-carb 2770TR/SL (USA), which is shown in Figure 3.

The liquid scintillator used in the experiment was of the Ultima Gold AB series of DIN-based cocktails. This cocktail was designed specifically, for alpha/beta separation, by the Packard Instrument Company with an aqueous concentration of approximately 25% and can effectively detect both of alpha and beta particles at the same time [3].



Fig. 3. Measurement the total beta radioactivity by LSA Tri-carb 2770TR/SL.

The total alpha/beta activity in ancient pottery were measured with the Tri carb 2770TR/SL in the alpha/beta discrimination counting mode. The high percentage of triplet states produced by alpha particles were translated into electronic pulses at the PMT anode which may be as much as 30 nsec longer than those derived from beta particles and gamma photons produced in the sample cocktail. This is illustrated graphically in Figure 4.

In order to optimize alpha/beta separation performance, it is necessary to determine the correct PDD (Pulse Decay Discriminator) setting. The optimum setting is the setting where there is equal and minimum spill of alpha pulses into the beta MCA and beta pulses into the alpha MCA. In the Parkard Tri-carb 2770TR/SL the spillover percentage or misclassification percentage of Am-241 and Cl-36 samples is less than 0.5%.

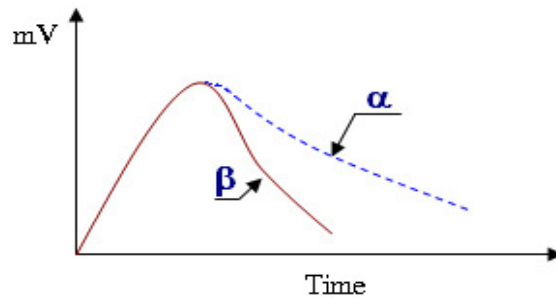


Fig. 4. Characteristic light pulse shapes of alpha and beta pulse in a liquid scintillator.

In theory, the alpha events should be sent to the alpha MCA and the beta events to a beta MCA, negating the requirement for spectral overlap calculations.

In this experiment, nine pottery samples collected from three archaeological sites at three different provinces were evaluated for the annual beta dose as the following: GoThap located in Tan Kieu commune (Thap Muoi district Dong Thap province); BaiBen located in Hien Hao commune (Cat Hai district, Hai Phong City); ThapBa located in Nha Trang town (Khanh Hoa province). At each site samples were chosen with the same identification, i.e. with the coarse material consisting of sand, and their ages have been determined by the radiocarbon method [5].

- GoThap two values were obtained: 2150 ± 130 and 1990 ± 115 yrBP
- BaiBen two values were obtained: 3450 ± 120 and 3540 ± 140 yrBP
- ThapBa two values were obtained: 1450 ± 125 and 1340 ± 115 yrBP

Besides, for all pottery samples collected at each of the above locations, the regional gamma radioactivity from surrounding terrestrial environment and cosmic irradiation was predetermined using the portable radiometer CPII-68-01. The results as obtained are shown in the Table 1. The total specific beta activity in pottery was calculated based on the standard ThS sample and calculated data are also included in Table 1.

Table 1. The measured results of the beta and gamma radioactivity

Sites	Beta specify activity* (C_β) (Bq/kg)	Gamma dose rate (I_γ) (μ R/h)
GoThap	1320 ± 50	17.0 ± 0.5
BaiBen	1084 ± 52	10.5 ± 0.5
ThapBa	1380 ± 48	17.0 ± 0.5

^{*)} averaged in three counting.

The annual dose of beta (mGy/yr) was evaluated from: $D_\beta = 2,22 \cdot 10^{-3} \cdot C_\beta$ and that of gamma (mGy/yr) from: $D_\gamma = 0.1752 \cdot I_\gamma$, where C_β is the specify activity of beta (Bq/kg), and (I_γ) is gamma dose rate (μ R/h).

Water in the pore of pottery or in the soil absorbs some parts of the radiation that would otherwise reach the thermoluminescent grains. Alternatively, on the infinite matrix basis, one can

think of the water as decreasing the radioactivity per unit mass compared to the dry situation. Consequently, if the dose-rates have been evaluated from measurement on the dry material, the corrected values to be used in the age-equation are given by [1,2]:

$$D_{\beta} = D_{\beta, \text{dry}} / (1 + 1.25 WF) \quad (2)$$

$$D_{\gamma} = D_{\gamma, \text{dry}} / (1 + 1.14 W_1 F) \quad (3)$$

where, $D_{\beta, \text{dry}}$ and $D_{\gamma, \text{dry}}$ are the dose-rates to be evaluated for dry materials by beta or gamma respectively, W and W_1 are the saturation water contents expressed as (weight of water/dry weight) for pottery or soil, respectively; F is the fraction of saturation to which the assumed average water content corresponds.

With the values given as: $W = 0.2$; $W_1 = 0.2$ and $F = 0.8$, the corrected values were calculated for the annual gamma and beta dose of pottery in TL dating by quartz inclusion technique. The obtained results are presented in Table 2.

Table 2. The annual dose in pottery for dating by quartz inclusion technique

sites	Annual beta dose rate (D_{β}) (mGy/yr)	Annual gamma dose rate (D_{γ}) (mGy/yr)
GoThap	2.344 ± 0.089	2.283 ± 0.073
BaiBen	1.925 ± 0.092	1.284 ± 0.073
ThapBa	2.451 ± 0.085	2.283 ± 0.073

The equivalent dose (Q) was evaluated applying the additive dose method: The thermoluminescence intensities of the samples were measured after the exposure to the radiation from radioisotope cobalt-60 source at known dosages as: 1Gy, 2Gy and 3Gy; 5Gy, 10Gy and 20Gy. Results are plotted in Figure 5.

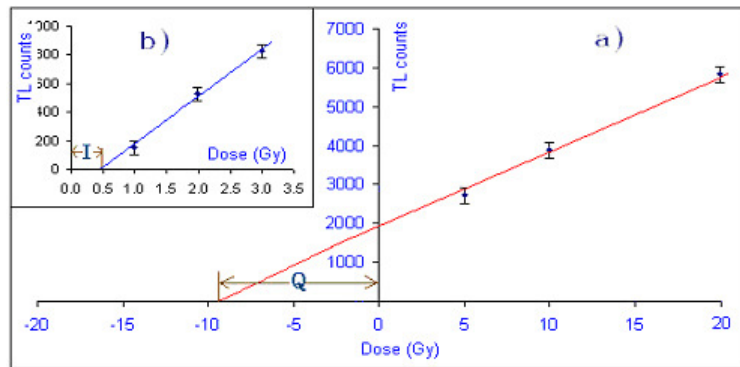


Fig. 5. The additive dose method for evaluation of the paleodose.

The paleodose P that the sample has received during antiquity is usually greater than Q because of the pre-dose effects [1,2]. The evaluation of this correction is presented in Figure 5b. Thus, the paleodose is calculated as $P = Q + I$; The results are shown in Table 3.

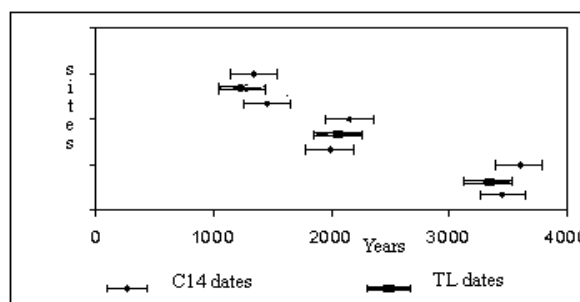
From the value of cosmic irradiation ($2\mu\text{R/h}$) and the annual dose-rates presented in Table 2, the thermoluminescence dating of pottery are calculated (see Table 3).

Table 3. The paleodoses and thermoluminescence ages

Sites	Paleodose (Gy)	TL age (year)
GoThap	9.03 ± 0.26	2056 ± 109
BaiBen	10.02 ± 0.29	3322 ± 216
ThapBa	5.56 ± 0.15	1239 ± 69

A comparison between the obtained thermoluminescence ages and the radiocarbon ages is presented on the Figure 6.

The results in Figure 6 indicate that, the TL ages of ancient pottery determined by the quartz inclusion technique were a good agreement with the radiocarbon ages as well as the approximate dates in archaeology, which were the early bronze period (BaiBen), and some AD begining centuries (GoThap), and from 8th -11th AD centuries for ThapBa [7].

Fig. 6. Comparison the TL ages and ¹⁴C ages.

3. Conclusions

The procedure of sampling preparation and measurement for the total alpha/beta radioactivity in ancient pottery using the liquid scintillation technique using Tri-carb 2770TR/SL with cocktail Utima Gold AB is presented. Based on the results obtained from the measurement the annual beta dose rates were evaluated and applied in thermoluminescence dating of ancient pottery samples collected from three archaeological sites. The TL ages obtained are compared with the radiocarbon ones. Preliminary results illustrate that the determination of the annual beta dose using the liquid scintillation technique Tri-carb 2770TR/SL can be considered as a high sensible and accurate method in archaeological sample dating. Only a low-mass of sample is required. This method is quite suitable for thermoluminescence dating of the ancient pottery in Viet Nam.

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