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PETER SZMUK and SHIGUEO WATANABE

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INSTITUTO DE ENERGIA ATÔMICA
Caixa Postal 11049 (Pinheiros)
CIDADE UNIVERSITÁRIA "ARMANDO DE SALLES OLIVEIRA"
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Divisão de Física do Estado Sólido
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São Paulo - Brasil

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DATING OF BRAZILIAN INDIAN POTTERY BY THERMOLUMINESCENT DOSIMETRY

Peter Szmuk* and Shigueo Watanabe*

ABSTRACT

The thermoluminescent dosimetry method was used to determine ages of funeral urns found in Itapeva, Piraju and Angatuba, São Paulo State, Brazil.

The thermoluminescence induced by radioactivity in the urn and the surrounding earth where the urns were found, is measured in small quartz crystals contained in the urn, after these are separated magnetically from the clay. From this value of TL the total absorbed dose is determined.

The annual β - and γ -dose of the surrounding earth and the urn itself was determined, finding the U, Th and K content by the alpha counting method and chemical method. An annual dose of 240 to 270 mrad was found. At Itapeva 10 Harshaw TLD-100 encapsulated dosimeters were buried for 132 days to confirm the derived γ -dose of about 106 mrad/year.

Dividing the total dose accumulated in the urn by the annual β - plus γ -dose we found ages of approximately 1070 ($\pm$ 130) years for Itapeva urns, 1020 ($\pm$ 120) years for Piraju samples, and 1210 ($\pm$ 145) years for Angatuba potteries. The ^{14}C method applied to human bones contained in the Itapeva urn produced an age of approximately 1000 years.

INTRODUCTION

Pottery dating by thermoluminescent dosimetry (TLD) was first suggested by Daniels (1953) and it was used by many people: Grogler et al.(1960), Fremlin and Srirath (1964), Kennedy and Knopff (1960), Ichikawa (1965), Aitken et al. (1963), Ralph and Michael (1967), and Mazess and Zimmermann (1966). They determined, however, relative ages only, by comparison with age values of other potteries determined by, for instance, ^{14}C method. The error involved amounted to about 20%.

In 1967, Aitken et al.(1967) developed a method to obtain absolute value of the pottery age involving, at first, a large

* Instituto de Energia Atômica and Instituto de Física, University of S.Paulo, S.Paulo, Brazil.

error, but an improvement introduced in the method reduced the error to about 10%.

In the present work we applied the method of Aitken et al. (1967) for dating of urns made by Brazilian indians and found at Itapeva, Piraju and Angatuba, in the State of São Paulo.

EXPERIMENTAL PROCEDURES

A. SEPARATION OF QUARTZ

About 0.5 to 1mm of the surface layer was removed from each piece of urn to clean them and to eliminate the layer that was exposed to alpha- and β-rays from surrounding earth. With this procedure one can be sure that the alpha- and β-rays contributed to the TL of the material are of internal origin.

The piece of urn to be investigated was then pulverized and sieved, keeping only the grains with size between .074mm and .149 mm. These grains were washed in water, HCl and alcohol in order to remove dusts from the surface and to facilitate the magnetic separation of quartz crystals from clay that makes up the pottery. The magnetic separator is Franz Isodynamic model L-1 belonging to Geology Department of the University of S.Paulo. Approximately 1/10 of the original sample is composed of quartz.

B. THERMOLUMINESCENCE OF QUARTZ

The quartz crystals are in most of the cases the only active TL producing agent in the urn. The TL was read out in the Harshaw TL reader model 2000.

To determine the radiation dose that induced TL in the pottery, the TL of one portion of the quartz was read out, the remaining

portion was irradiated to 300 rad ^{60}Co γ-rays and then read out. If we denote by A the height of the 270°C peak in the quartz for samples not irradiated in the lab, and by B that of lab-irradiated ones (300rad), we can find that

$$D = 300A/(B-A)$$

is the dose absorbed in rad during the time the urn was buried in the earth. Therefore, if we know the annual dose, we can calculate

$$\text{Age (in years)} = D/(\text{Dose/year})$$

Since the range of alpha particles is very small, for grains with 75 to 150 micron their effect can be neglected and consider only β - and γ- contribution.

C. U, Th AND K CONTENT IN THE URN AND EARTH

If the U, Th and K content in the urn and the earth where urns were found, is known, we can also calculate β- and γ-doses. The U and Th content can be determined by counting alpha particles emitted by members of U and Th series.

The counting system consists of a plexiglass plate on the surface of which a thin film of ZnS:Ag is deposited. The PMT signal is amplified and a discriminator eliminates PMT noises. The pulse is selected in such a way that either each one is registered or a pair of pulses falling within 0.32 sec is registered.

For the analysis of a sample placed on the ZnS plate, the total count of alpha particles emitted by the sample is emasured, and the double pulses of alpha decay of ^{226}Rn and ^{216}Po are counted. The spurious double pulses (those originating from Rn and Po) can be calculated.

The number N_o of scintillations per hour found is related to

the activity S of the sample by the relation

$$S = (N_\alpha/RA\rho) \times 3.008 \times 10^{-14} \text{ Ci/g}$$

where R = alpha particle range, A = area of ZnS plate, and ρ = density of the active medium.

Assuming that the radioactive source is distributed uniformly, the absorbed dose per year in a point P inside the source is given by

$$D_q = 1.158 \times 10^{14} \frac{\bar{d}W\rho XI}{\mu N T_{1/2}} \left[1 - A_1 e^{-\mu R(1+d_1)} - A_2 e^{-\mu R(1+d_2)} \right] \text{rad}$$

where $\bar{d}$ = exposure, $W = D_{air}/D_{medium}$, M = average molecular mass of earth, X = concentration of radioactive centers in ppm, I = γ-ray intensity, A_1, A_2, d_1, d_2 are the coefficients in the expression of Build up factor, and can be found tabulated, Morgan and Turner (1967).

Correspondingly the β-ray dose can be calculated from

$$D_\beta = 6.6 \times 10^9 E_\beta XI/(MT_{1/2}) \quad \text{rad/year}$$

E_β is the average β- ray energy.

Assuming that U and Th series are in secular equilibrium and knowing the U and Th content, the expected β- and γ- dose from the two radioactive series can be calculated. One has to add K contribution. In the present work K concentration was determined by chemical method.

D. ANNUAL γ-DOSE MEASUREMENT USING TLD

A more direct method to measure γ-ray dose absorbed by quartz in the earth, is to bury a TL dosimeter in the earth. Since dose rate is very low the dosimeter must be left in the earth for at least three to four months.

RESULTS

The annual absorbed dose due to U and Th was determined by counting alpha emission from finely pulverized pottery and earth taken from Itapeva, Piraju and Angatuba. In Table 1, Table 2 and Table 3 we find β-, γ- and total dose determined for Itapeva, Piraju and Angatuba samples, respectively.

TABLE 1 - ITAPEVA - ANNUAL DOSE IN mrad/YEAR

Sample No.	γ (earth)	β(urn)	Total
1	115	160	275
2	120	125	245
3	110	125	235
4	95	140	235
5	112	148	260

TABLE 2 - PIRAJU - ANNUAL DOSE IN mrad/YEAR

Sample No.	γ	β	Total
1	150	130	280
2	100	160	260
3	100	140	240

TABLE 3 - ANGATUBA - ANNUAL DOSE IN mrad/YEAR

Sample No.	γ	β	Total
1	130	110	240
2	100	115	215

The average dose determined from TL measurement is listed in Table 4.

TABLE 4 - ABSORBED DOSE IN rad

Sample No.	Itapeva	Piraju	Angatuba
1	253	280	240
2	270	260	215
3	262	240	
4	280		
5	264		

Encapsulated TLD-100 (Harshaw) was buried in the spot where the urns of Itapeva were found. The TL read out after 132 days period resulted in about 106 mrad/year. This value is comparable to the average value of 110 mrad/year found from the previous method.

In Table 5 we have the age values found for urns we investigated.

TABLE 5 - AGE OF URNS IN YEARS

Sample No.	Itapeva	Piraju	Angatuba
1	970	1150	1450
2	1100	960	980
3	1110	955	
4	1190		
5	1010		
Average	1076	1020	1210

Considering a maximum 5% error involved in the irradiation of pulverized sample, approximately 3% error in the TL reading and ~5% error in the determination of annual dose rate, we estimate an overall error between 10% and 15%.

Bones contained in Itapeva urns were analised by ^{14}C method (elsewhere) yielding a value of about 1000 years, in reasonable agreement with our result.

The dating of each piece took about 4 to 5 days.

The fact that our measurement, as far as the pottery age is concerned, is in relatively good agreement with that of ^{14}C method, and the fact that buried TL dosimeter registered a γ-dose very close to that found by alpha counting method, give support to the method of Aitken et al.(1967).

RESUMO

O método da dosimetria termoluminescente foi utilizado para determinar a idade de urnas funerárias encontradas em Itapeva, Piraju e Angatuba, no Estado de S.Paulo, Brazil.

A termoluminescência induzida pela radioatividade da urna e da terra vizinha nas urnas foi medida separando os pequenos cristais de quartzo contidos na urna, depois de tê-los separados magneticamente da argila. Determina-se a dose total absorvida a partir dêste valor de TL.

A dose gama e beta anual do solo vizinho e da urna foi determinada pela medida do teor de U, Th e K pelo método da contagem alfa e pelo método químico. Uma dose anual de 240 a 270 mrad foi encontrada. Em Itapeva foram enterradas 10 cápsulas dosimétricas de TLD-100 (LiF) da Harshaw Chemical Co. durante 132 dias, para confirmar a dose gama calaculada de aproximadamente 106 mrad/ano.

Dividindo-se a dose total acumulada na urna pela dose total de raios beta e gama, obtivemos uma idade de aproximadamente 1070 (± 130) anos para as urnas de Itapeva, 1020 (± 120) anos para as amostras de Piraju e 1210 (± 145) anos para as de Angatuba.

O método de ^{14}C aplicado aos ossos humanos contidos na urna de Itapeva deu uma idade de cêrca de 1000 anos.

RÉSUMÉ

La méthode de dosimetrie thermoluminescente a été utiliseé pour déterminer l'âge d'urnes funéraires découvertes à Itapeva, Piraju et Angatuba, dans l'Etat de São Paulo, Brésil.

La thermoluminescence produite par radioactivité dans l'urne et le terrain environnant dans lequel les urnes ont été découvertes est mesureé dans de petits cristaux de quartz contenus dans l'urne, aprés les avoir magnetiquement separés de l'argile. On détermine la dose totale absorbeé a partir de cette valeur de TL.

La dose annuelle gama et beta du sol environnant et de l'urne elle même a été déterminée par mesure de la quantité de U, Th et K par la méthode de contage alpha et la méthode chimique. Une dose annuelle de 240 à 270 mrad a été trouveé. A Itapeva, on a enterré 10 capsules dosimetriques Harshaw TLD-100, pendant 132 jours, pour confirmer la dose gama calculeé de approximativement 106 mrad/an.

Divisant la dose totale accumuleé dans l'urne par le rayonnement beta annuel plus la dose gama nous avons trouvé des âges d'approximativement 1070 (± 130) ans pour les urnes de Itapeva, 1020 (± 120) ans pour les échantillons de Piraju et 1210 (± 145) ans pour les poteries de Angatuba. La méthode ^{14}C appliqueé aux os humains contenus dans l'urne de Itapeva a donné un âge d'approximativement 1000 ans.

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