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ASSESSING THE POTENTIAL OF THERMOLUMINESCENCE DATING OF PRE-CONQUEST CERAMICS FROM CALIXTLAHUACA, MEXICO

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Abstract — Archaeological dating is very important for Mexico taking into account the abundance of ancient ceramics existing through the country. The results are reported of studying three sherds from the archaeological site in Calixtlahuaca, Mexico, where the Matlazinca culture blossomed before the Spanish arrived in America. The thermoluminescence dating technique used was that of fine grain using grain sizes in the range of 4 to 11 μm. Paleodose and the supralinearity correction factor were determined by means of analysis of the glow curve in the temperature range of 250 to 500°C. The average values of paleodose obtained were 8.46 ± 0.27 Gy and 3.62 ± 0.13 Gy for two of the samples studied; the annual dose rates were 5.55 ± 0.20 mGy and 4.20 ± 0.30 mGy. Taking into account these results, the ages of these two ceramic samples were 1520 ± 90 and 870 ± 80 years which agree well with the historical data for the site. The third sherd could not be dated because it did not show any plateau.

INTRODUCTION

Thermoluminescence (TL), the emission of light when a substance is heated below its incandescence temperature, is a promising technique for testing the authenticity of an archaeological sample by using the TL signal induced by natural radiation through the years of burial.

The TL emitted by the minerals present in a ceramic sample is produced by the prolonged exposure to ionising radiation emitted by the radioactive elements like U, Th (and their daughter products) and ^{40}K embedded in the sample as impurities and present in the surrounding soil. A small contribution to TL is due to cosmic rays.

The accumulated dose which gives rise to the TL is

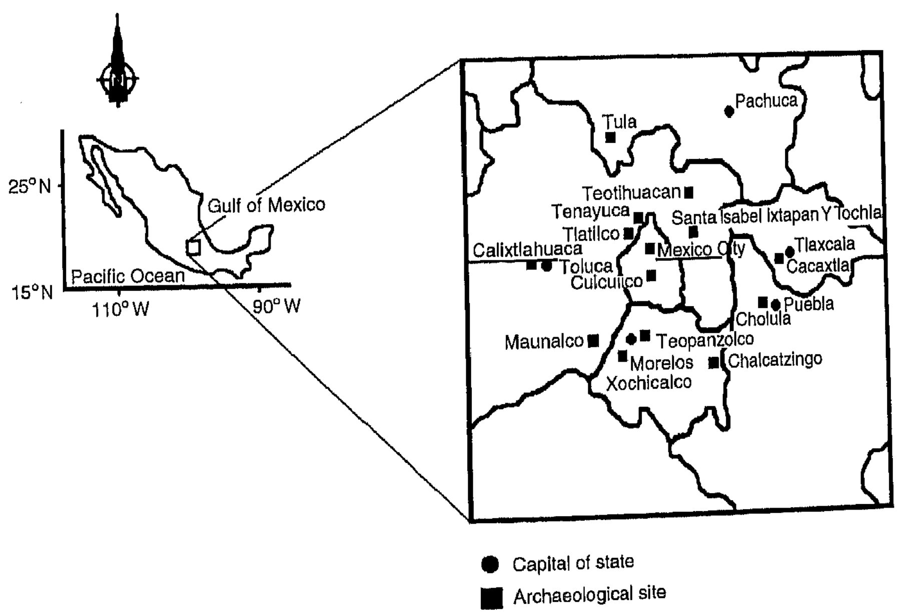

Figure 1. Location of the Calixtlahuaca archaeological site near Mexico City.

named the paleodose (P). Because all the radioisotopes responsible for TL have a very long half-life, their emission of radiation remains effectively constant and the amount of TL induced is proportional to the time that has elapsed since the pottery was fired. This firing removed all the TL acquired by the mineral during geological times and thus sets the clock to zero. The basic form of the age equation is:

$$A = P/D$$

where: A = age (y), P = paleodose (Gy) and D = annual dose ($Gy.y^{-1}$).

This paper presents the results of dating three ceramic samples from Calixtlahuaca, Mexico, using the TL method. Calixtlahuaca is an archaeological site of the Mexica Culture located at 19°20′ North and 99°41′ West, about 5 km from Mexico City (see Figure 1). The site was occupied by the Matlazinca before the Spanish arrival. However, it seems that there were probably inhabitants in this place for a long period. Ceramic fragments which could belong to the period from the Early Classic (approx. 200–500 A.D.) up to the Early Postclassic (approx. 900–1100 A.D.) have been found at this site.

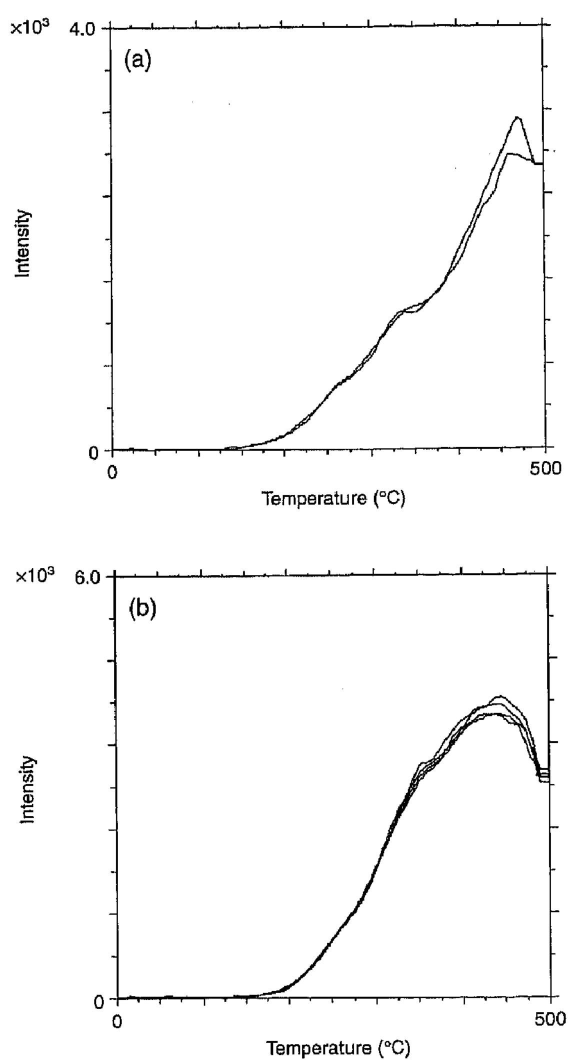

Figure 2. Glow curves of the unirradiated samples (NTL): (a) CAL1; (b) CALIX6.

Direct dating of the ceramics found at this site is important , because the only chronological information available is derived from historical records. These historical data have made it possible to fix four archaeological horizons prior to the Aztec invasion: the first is the period since the site occupation in the Pre-classic (approx. 1500 to 200 B.C.); the second is characterised by ceramics of Teotihuacan influence and ranges from 300 to 600 A.D.; the third period shows a strong Toltec influence (900–1200 A.D.) and the fourth period corresponds totally to the Matlazinca Culture. For this reason, the sherds collected in the site correspond to different periods.

EXPERIMENTAL PROCEDURES

Various fragments of ceramics have been recovered for study; only three samples, labelled CAL1, CALIX6 and CALIX8 have been completely investigated.

The dating technique used was that of the fine grain method[1]. A 2 mm layer was first removed from the surface. The remaining sample was crushed and powdered in an agate pestle and mortar. The grains sized from 4 to 11 μm (fine grains) were treated with H_2O_2 to remove the organic material and with HCl to neutralise carbonate.

More than 50 discs of each sample, each containing 2 mg of the sample, were prepared to check the reproducibility of the procedure . Discs were made of aluminium 0.5 mm thick and 9 mm in diameter. All the sample preparation and analysis was performed under red light illumination.

The TL reader used was a Daybreak 1100 Automated TL system (Nuclear, Guilford, CT, USA). It is equipped with a sample holder which can carry 20 discs and is coupled to a Pentium PC loaded with the software to control the whole reading process. This software permits the analysis of the plateau (region of interest) and uses least squares fitting of the growth curves to give an estimate of the dose equivalent (Q) and the supralinearity correction factor (I).

Artificial irradiation of the samples employed a ^{90}Sr-^{90}Y beta source (Amersham International), with an activity of 3.7 MBq (100 mCi) at a dose rate of 3.6 $Gy.h^{-1}$.

Sixteen readings of each of the unirradiated samples were taken to obtain the natural TL signal (NTL); the remaining samples were irradiated in vacuum at four

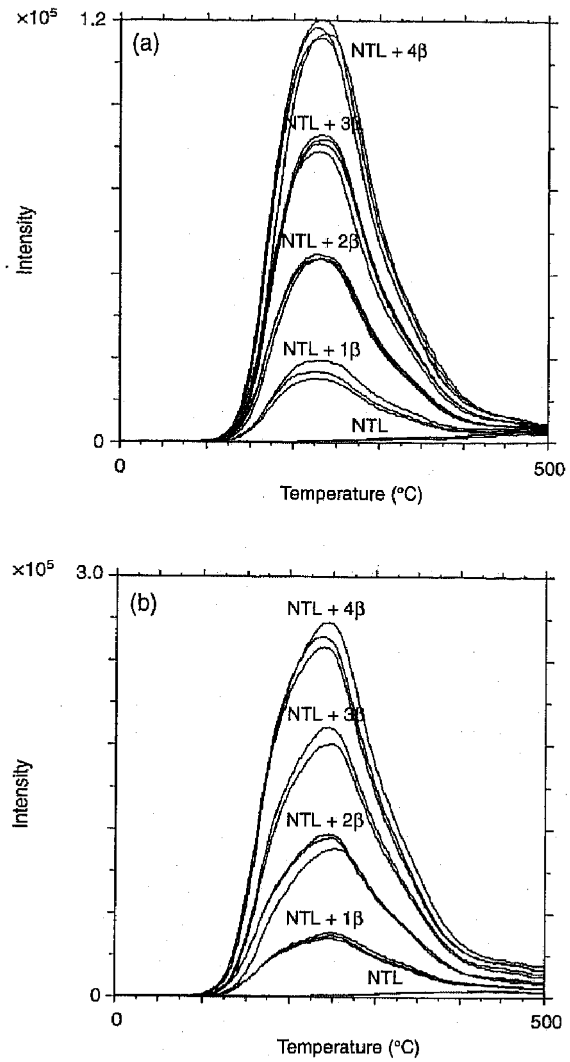

Figure 3. Glow curves of unirradiated samples (NTL) as well as those irradiated with ^{90}Sr beta radiation at four different doses (NTL + ATL). (a) CAL1 irradiated at: 1β = 32 Gy, 2β = 64 Gy, 3β = 96.6 Gy, and 4β = 129 Gy. (b) CALIX6 irradiated at: 1β = 58 Gy, 2β = 130 Gy, 3β = 202 Gy, and 4β = 288 Gy.

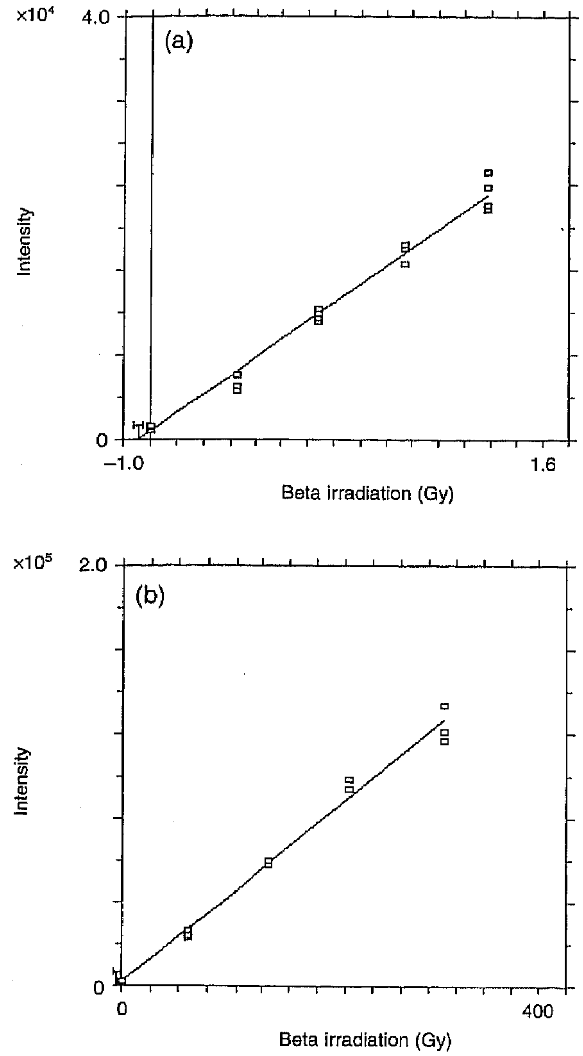

Figure 4. Dose–response curves of the samples obtained by the additive method. (a) CAL1; (b) CALIX6.

Table 1. Concentration of K, U, and Th, as well as the α dose and β dose (mGy.y^{-1}) produced by each radioactive material.

Sample	Element	Concentration	α dose (mGy.y^{-1})	β dose (mGy.y^{-1})
CAL1	K	0.86 ± 2.58%	—	0.71 ± 0.02
	U	3.12 ± 0.22 ppm	8.66 ± 0.62	0.45 ± 0.03
	Th	5.06 ± 0.78 ppm	3.73 ± 0.58	0.14 ± 0.02
CALIX6	K	0.82 ± 3.11%	—	0.68 ± 0.02
	U	1.33 ± 0.59 ppm	3.69 ± 1.64	0.19 ± 0.08
	Th	4.17 ± 0.70 ppm	3.08 ± 0.52	0.12 ± 0.02

different doses: 32, 64, 96.6, and 129 Gy for CAL1 and 58, 130, 202 and 288 Gy for CALIX6 and CALIX8, which were labelled as 1β, 2β, 3β, and 4β respectively in both cases. The samples were stored in the dark prior to measurement.

Dose equivalent (Q) was determined by the additive dose method; determination of the I factor was carried out by means of the regeneration method[1,2], using those samples from which NTL had been previously erased.

To calculate the annual dose rate due to alpha and beta radiation the concentrations of K, U, and Th in the ceramics and in the surrounding soil were determined. Determination of K concentration was performed by means of the microanalysis technique using a sweeping electron microscope (Philips XL30). The determination of U and Th contents was carried out by the neutron activation analysis technique in a Triga Mark III reactor.

The gamma radiation dose rate was measured using locally made $CaSO_4$:Dy TL dosemeters[3].

To test for anomalous fading five discs of each sample were beta irradiated at a dose of 30 Gy and stored in the dark for 30 days at room temperature.

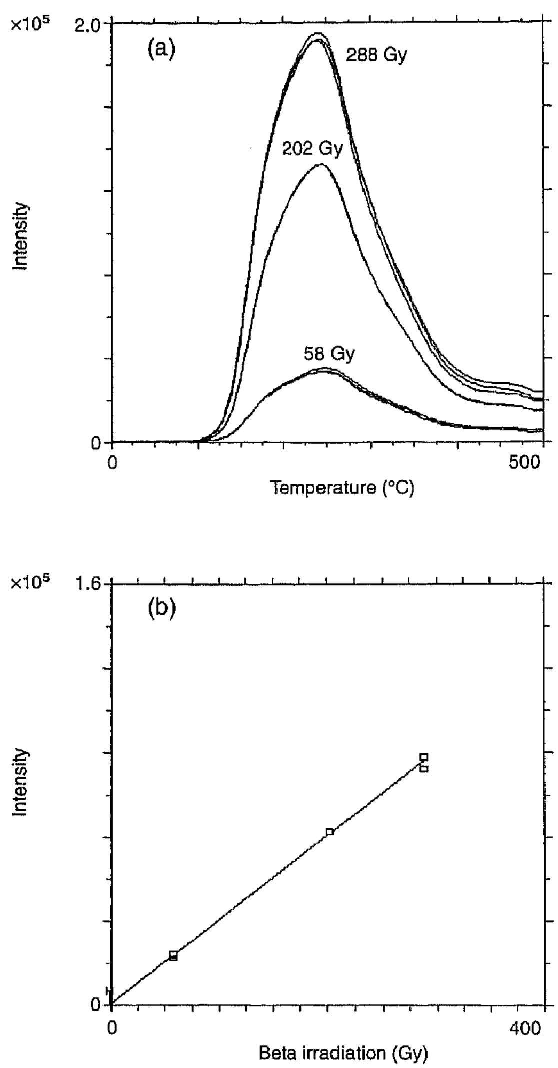

Figure 5. (a) Glow curves of CAL1 and CALIX6 samples beta irradiated at 58, 202 and 288 Gy. (b) Regenerated growth curve of the CALIX6 sample obtained from the glow curves.

RESULTS

The data for the sample CALIX8 are not reported here because it was not possible to date it because its growth curve did not show any plateau.

Table 1 shows the K, U, and Th concentrations and the annual dose rate due to alpha and beta radiation respectively for the two samples studied (CAL1 and CALIX6).

No signal loss was detected (to within 4%) compared with samples which were dosed and measured immediately. It is concluded that anomalous fading could not be detected in either sample.

Gamma radiation dose rate as measured by $CaSO_4$:Dy TL dosemeters was 2.77 ± 0.15 $mGy.y^{-1}$. Taking an α efficiency factor of 0.15[2] and a Th/U ratio of 3:1[1], annual dose rates in dry conditions of 5.55 ± 0.20 and 4.20 ± 0.30 $mGy.y^{-1}$ were obtained for CAL1 and CALIX6 respectively.

Figure 2 shows the natural TL (NTL) intensity signal of the ceramic samples CAL1 and CALIX6 as a function of temperature. As can be seen, these curves do not exhibit a well defined peak but present two very small peaks in the temperature range from 250 to 360°C.

In Figure 3 we can observe the glow curves of unirradiated samples (NTL) as well as those irradiated with ^{90}Sr beta radiation at four different doses (NTL+ATL). The dose response curves obtained by the additive dose method[4], for the two samples, are shown in Figure 4. Interpolation of these curves to zero TL intensity (TL=0) gives the dose equivalent (Q); this was determined at each 10°C temperature interval in the

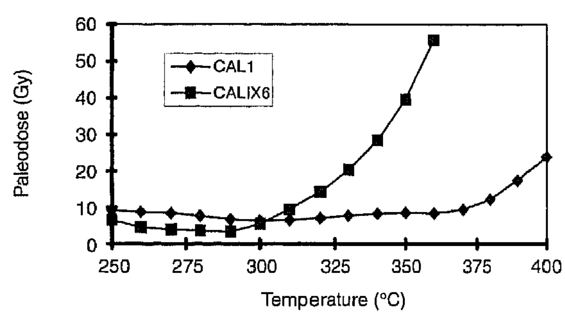

Figure 6. Paleodose plotted as a function of temperature in the range of 340 to 360°C.

range from 250 up to 400°C for both samples. The correction factor due to supralinearity was determined by means of the regeneration method at the same 10°C temperature intervals. These results are shown in Table 2 together with the paleodose values for the two samples. Figure 5(a) shows the glow curves of the CALIX6 sample beta irradiated at 58, 202 and 288 Gy. The regenerated growth curve shown in Figure 5(b) was obtained from these glow curves to show the measurement of the Y factor for the CALIX6 sample.

Using the data of Table 2, the paleodose as a function of temperature was plotted for the two samples, as shown in Figure 6. From this plot, the average paleodose in the temperature range 340 to 360°C was determined for both samples. According to the method proposed by Mejdahl[5], mean values of paleodose determined in this temperature range were 8.46 ± 0.27 Gy and 3.62 ± 0.13 Gy for CAL1 and CALIX6 samples respectively.

Total uncertainty was estimated taking into account the random fitting errors on the growth curves produced by the propagation of the uncertainties in the values of Q and I given in Table 2. The uncertainties obtained in the radionuclides analysis shown in Table 1 were also taken into account to yield uncertainties of 4.7% and 7.4% in the paleodose and in the annual dose rate of CAL1 and CALIX6 respectively. The assumed average water content of the samples, on which the corrections were based, was 10% for CAL1 and 13% for CALIX6. The effect of radon escape on the determination of the dose rate was neglected since that determination of U and Th was made by means of neutron activation analysis.

Finally, according to Aitken[2] and taking into account moisture corrections, by substituting the above values in the age equation, the ages of 1520 ± 90 years and 870 ± 80 years for CAL1 and CALIX6 respectively were determined.

CONCLUSIONS

From the results obtained, one can have confidence that archaeological dating by means of TL is reliable, despite the unusual Q/I ratio, because of the agreement of the results with the historical data. This encourages us to continue this research since that our country is rich in archaeological sites which need to be dated.

Table 2. Values of dose equivalent (Q), supralinearity correction factor (I), and paleodose (P) determined for each 10°C interval in the temperature range 250 to 400°C for the samples CAL1 and CALIX6 from Calixtlahuaca, Mexico.

	CAL 1			CALIX 6		
Temperature (°c)	Q(Gy)	I(Gy)	P(Gy)	Q(Gy)	I(Gy)	P(Gy)
250	2.341 ± 0.034	6.808 ± 0.185	9.149 ± 0.188	0.814 ± 0.019	5.697 ± 0.105	6.512 ± 0.107
260	2.194 ± 0.035	6.575 ± 0.208	8.768 ± 0.211	0.057 ± 0.001	4.509 ± 0.097	4.567 ± 0.097
270	1.949 ± 0.032	6.499 ± 0.205	8.448 ± 0.207	0.733 ± 0.020	3.145 ± 0.073	3.878 ± 0.076
280	1.475 ± 0.024	6.203 ± 0.196	7.678 ± 0.197	1.671 ± 0.048	1.982 ± 0.050	3.654 ± 0.069
290	0.824 ± 0.012	6.923 ± 0.166	6.747 ± 0.166	2.739 ± 0.079	0.590 ± 0.016	3.330 ± 0.081
300	0.155 ± 0.002	6.204 ± 0.141	6.359 ± 0.141	4.060 ± 0.123	1.474 ± 0.042	5.534 ± 0.129
310	0.594 ± 0.009	5.955 ± 0.143	6.549 ± 0.143	5.627 ± 0.175	3.901 ± 0.129	9.529 ± 0.217
320	1.483 ± 0.021	5.654 ± 0.134	7.137 ± 0.138	7.371 ± 0.229	6.962 ± 0.295	14.334 ± 0.373
330	2.328 ± 0.034	5.443 ± 0.142	7.772 ± 0.146	9.321 ± 0.304	11.100 ± 0.647	20.421 ± 0.715
340	3.148 ± 0.049	5.214 ± 0.175	8.362 ± 0.182	11.500 ± 0.405	17.000 ± 1.425	28.500 ± 1.481
350	4.333 ± 0.073	4.237 ± 0.137	8.570 ± 0.156	14.100 ± 0.537	25.600 ± 3.379	39.700 ± 3.421
360	6.074 ± 0.112	2.374 ± 0.058	8.448 ± 0.126	17.300 ± 0.696	38.400 ± 8.102	55.700 ± 8.332
370	8.409 ± 0.162	1.070 ± 0.028	9.478 ± 0.164			
380	11.800 ± 0.220	0.422 ± 0.008	12.222 ± 0.220			
390	16.700 ± 0.300	0.733 ± 0.007	17.433 ± 0.300			
400	22.500 ± 0.397	1.366 ± 0.016	23.866 ± 0.397			

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