Source calibrations and blind test results from the new Luminescence Dating Laboratory at the Instituto Tecnológico e Nuclear, Sacavém, Portugal

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(Received 10 April 2003; in final form 30 May 2003)

abstract : *Blind test results for the most important materials used in luminescence dating are reported as a measure of the quality of performance of the new luminescence dating laboratory at the Instituto Tecnológico e Nuclear, Sacavém, Portugal. At the same time they serve as a measure of the quality of the source calibrations, which are outlined as well, and an estimation of the required charge particle equilibrium (CPE) distance for gamma irradiation of the calibration material is presented. Several different quartz and measurement techniques are employed for the calibration of alpha and beta sources.*

Introduction

The desire of the Portuguese archaeology for chronometric dating tools in addition to the existing radiocarbon facility led to a protocol between the Instituto Português Arqueologia (IPA) and the Instituto Tecnológico e Nuclear (ITN), in order to set up a luminescence dating facility at the latter. Building work was finished in spring 2001, and the laboratory completed in fall 2002. The task was to set up a basic luminescence dating laboratory, which is able to perform dating application on the following most common minerals/materials used in luminescence dating for archaeology and geology:

- polymineral fine grain sediment (feldspar)
- coarse grained sediment (quartz)
- archaeological heated material (ceramic, pottery, brick, tile, etc.)
- SiO_2-bearing heated archaeological material (silex, chert, etc.)

Additional, but less frequently used, techniques like fine grain quartz or coarse grain feldspar sediment dating can be implemented easily in the future. The primarily function of the laboratory is supposed to be of service.
In order to test the laboratory's performance, a quality insurance program was started by measuring samples, which have already been dated by established luminescence dating laboratories. At the same time, these tests are a check of the calibration of the radioactive sources used at ITN.

The new laboratory

The new laboratory consists of three connected rooms (sample preparation, measurement and irradiation). R10 Encapsulite fluorescent tubes provide the lighting in all rooms. The laboratory is equipped with all the standard chemicals and equipment for the physical and chemical sample preparation employed in luminescence dating. The luminescence reader is a Risø DA-15 with an additional halogen lamp. Two systems were set up for measuring the external gamma dose rates. A portable γ-spectrometer (Target NanoSpec) was calibrated versus the Oxford and the Gif-sur-Yvette blocks, and a system based on α-Al_2O_3:C (TLD-500) dosimeters was developed (Richter & Zink). Furthermore, Neutron Activation Analysis (NAA) is an already well-established tool at ITN (Gouveia et al., 1992).
Grants for upgrading this basic laboratory with equipment like a single-grain luminescence reader and a low level γ-spectrometer are pending.

Beta-source calibration and estimation of Charged Particle Equilibrium (CPE) condition

Two $^{90}Sr/^{90}Y$-beta-sources are employed at the laboratory. One (nominal 40 mCi) is attached to the luminescence reader and the other one is a stand-alone 30-positions irradiator by Daybreak (801E, nominal 200 mCi).

A total of three different quartz samples and three different protocols with TL and B-OSL were used for calibration of these beta-sources.

The Risø calibration quartz, as provided during installation of the system, is sedimentary quartz from Jutland, which was annealed at 500ºC and irradiated with a ^{137}Cs source. After corrections for attenuation

the absorbed dose to the quartz is given as 4.45 ± 0.07 Gy (A.Murray, information sheet for calibration quartz). A B-OSL SAR protocol (Murray and Wintle, 2000) with 3 dose points was used for estimating the dose rates of the ITN beta sources.

The GSF quartz was kindly provided by H.Y. Göksu, and is from the same batch as described in Göksu et al., 1995, but of a different irradiation with a ^{60}Co source. The absorbed dose is given as 0.5 ± 0.015 Gy. Measurements followed as outlined in Göksu et al., 1995. A wide integration range was employed for the blue TL (7-59 and HA-30). A beta dose comparable to the gamma dose was given to aliquots with material identical to the quartz prior to gamma irradiation. All data were normalized by a second glow, and the ratio of integrated beta and gamma irradiated luminescence gave the calibration value. Some B-OSL measurements (see Risø calibration quartz) were performed as well.

In order to ensure the laboratories independence, commercial Merck quartz (107536 quartz, washed and calcined, charge TA 1077836 204, 0.2 - 0.8 mm) was purchased, prepared and irradiated with a ^{60}Co source at ITN (Oliveira, et al., 2000). Some of this quartz was further crushed with an agate mill prior irradiation. Tests revealed that the sensitivity could be increased and sensitivity changes decreased by a repetition of rapid heating to 700ºC and fast cooling, followed by HCl-washing after the last cycle. Long heating was found to decrease the sensitivity and a further sensitization by beta irradiation in between heating cycles to be unnecessary. The gamma irradiation was performed on a dense mix of grain sizes between 0.001 mm and about 1.0 mm, in order to allow the irradiation of a dense material with the least cavities. It is thus assumed that any corrections for interactions at repeated air-grain boundaries are not necessary. The required container thickness to obtain charged particle equilibrium (CPE) for ^{60}Co-irradiation was estimated for fused (pyrex) quartz of 2.23 g cm^{-3} density, using mass stopping power values provided by the ESTAR (NIST) database (ESTAR). It is assumed that CPE is reached in matter at a distance equivalent to the maximum electron pathlength. As we were unable to find sufficient data for secondary electrons, the upper limit estimate of the range of penetration was calculated assuming electron energy to be equal to the total photon energy, which is certainly a slight overestimation. Additionally, the average range of penetration is smaller than the average pathlength because the trajectory of electrons in solids is zigzak-like. We further assume, that for energetic photons (above 200 keV) the photoelectric electrons originate mainly as result of interaction of secondary (Compton) photons with the mater, and any effect of Bremsstrahlung is negligible. The obtained value of 3.6 mm as the required thickness to reach CPE is in good agreement with empirical data, indicating 3 mm to 4 mm (pers. comm.. H.Y.Göksu).

Employing the mass absorption coefficients of the XAAMDI (NIST) database and from Aitken, 1985, we obtain for ^{60}Co a ratio of 0.996 (0.0266/.0267, the latter interpolated for 1.17 and 1.33 MeV), in order to convert for Air Kerma to gray (Gy) in quartz.
The self-attenuation of a sample can be calculated in a simplified approach by the following formula:

(1) $$D = D_O * (1 - mu*r/3),$$

where D is the attenuated dose of gamma radiation, D_O is the dose delivered by the source, mu is the linear attenuation coefficient of the sample, r is the inner radius of the container (or half of the length of the base in the case of quadratic base containers)

This formula (1) was derived for homogenous and isotropic radiation around a container of overall small dimensions, as compared to the mean free path of gamma photons ($mu*r << 1$). For ^{60}Co irradiation we found the self-attenuation factor to be 0.985.

Specially build fused quartz containers of quadratic base with 3.6 mm wall thickness and 8.0 mm inner base-length were employed for ^{60}Co (nominal 300 Ci) irradiation equivalent from all four sides. Any attenuation due to the thickness of the "sample" for such irradiation is assumed to be similar as resulting from homogenous and isotropic radiation field. Applying the above, the total absorbed dose in our case is 1.96 ± 0.018 Gy.
After irradiation the 90-160 μm fraction was obtained by dry-sieving and the 4-11 μm fraction was extracted by settlement in acetone. TL measurements were performed using a blue detection window (7-59 + HA-30). The peak-integrated TL from the gamma irradiated material was matched to the linear regression of 3 dose points from beta irradiated material. Additionally, a portion of the zeroed material was given a ^{137}Cs dose of 2.0 Gy at the Gif Luminescence Laboratory (LSCE at the CNRS-CEA), and measured in the same way.

Some measurements were performed with and without silicon oil as a fixing agent. Only an insignificant tendency to slightly lower results with silicon was observed. All the results are corrected for a common date of 12/10/2002 (Table 1).

	Risø-β		Daybreak-β	
	B-OSL	TL	B-OSL	TL
GSF	5.68*; 5.56	5.78	n.a.	12.69#
Risø	5.69*; 5.55	n.a.	Failed	n.a.
ITN-Merck	n.a.	5.54	n.a.	12.82
CNRS-Merck	n.a.	n.a.	n.a.	12.49
ITN-Merck fine	n.a.	4.52	n.a.	10.18

Table 1.
*Summary table of calibration results. All values in Gy min^{-1}, corrected for a common date of 12/10/2002. For clarity the error estimates are omitted. * not fixed with silicone oil. # average of 4 measurements with 10 gamma and 10 beta irradiated cups.*

Alpha source calibration

The Littlemore 6-seater alpha irradiator (type 721A, AMM3, nominal 182 mCi) with ^{241}Am-foils was calibrated by the manufacturer with a low level alpha counter under vacuum. This calibration was verified by the blind tests and additional measurements of a polymineral fine-grained sample (HDS-243; loess), which exhibits a linear alpha-dose growth and no fading (kindly provided by the the Forschungsstelle Archäometrie am MPI für Kernphysik, Heidelberg). Natural material (4-11 μm) was irradiated at Heidelberg with an identical alpha irradiator of a different activity, which was calibrated in 1978 by the manufacturer. Some of the natural material was irradiated (on disc) with five different alpha doses at ITN, with dose points set to be lower, about equal and higher than the Heidelberg irradiation, while other discs were left unirradiated (natural). The IRSL was measured after storage at 70ºC for one week, following the Heidelberg protocol as closely as possible. The Heidelberg irradiated material could not be normalized by a short shine because it had to be scraped off the discs after irradiation and re-deposited at ITN. Therefore a normalization by a beta dose after the first readout was employed. Subsequently a D_E-alpha was calculated using only the natural and ITN irradiated material and compared to the result obtained in Heidelberg (Table 2). The result of the measurement of the Heidelberg irradiated material was then plotted against values obtained for ITN irradiated discs. This gave the dose rate of the ITN source relative to the Heidelberg source by matching the luminescence of the former versus the linear fit of the latter (Table 2).

Littlemore calibration (μm^{-2} min^{-1}) *	0.191
Calibration versus Heidelberg (μm^{-2} min^{-1})º #	0.215
D_E-α Heidelberg (μm^{-2})º	67.88 ± 7.48
D_E-α ITN (μm^{-2}) #	79.55 ± 8.1

Table 2.
*Results obtained for fine grain material of sample HDS-243, as verification of alpha source calibration. * average of five out of six positions, as one position is significantly different then the others and is thus not used. º corrected for decay since calibration. # Based on the integrated luminescence of the first 20 s (last 10 s background subtraction) of 60 s room temperature stimulation by IRSL at 35% to adjust for the higher power of the DA-15 versus DA-12 system (30 % roughly equals, pers. comm. S. Lindauer), using a blue detection window (set of BG39, BG3, GG400, BG3), after a preheat of 220ºC for 2 min.*

The results cannot be regarded as independent, as, presumably, the manufacturer used the same method for calibrating both sources. Nevertheless, giving the long time span between these calibrations, the parameters (equipment, etc.) were certainly not identical. The result of the calibration versus the Heidelberg source of 0.215 μm^{-2}min^{-1} shows good agreement with the manufacturers average value of 0.191 μm^{-2}min^{-1}, as do the D_E-α determinations of 67.88 ± 7.48 μm^{-2} at Heidelberg and 79.55 ± 8.1 μm^{-2} at ITN, which both employ the calibration values given by the manufacturer. This gives confidence in the determination of the alpha sensitivity at ITN.

The blind test samples

The purpose of the exercise was to test the quality of luminescence dating at the new laboratory at ITN, with the protocols established there. Therefore only the palaeodoses and the alpha sensitivities (where applicable) are reported here. The dosimetric tools for determining the dose-rates are either already evaluated (Gouveia et al., 1992) or will be presented separate by Richter & Zink, 2003.

The luminescence parameters for the following four different samples, representing the most important materials used in luminescence dating, were determined:

- loess from Nussloch (Germany), provided by the Geography Department, University Bonn
- heated flint from the Upper Palaeolithic site Geißenklösterle (Germany), provided by the Forschungsstelle Archäometrie, Heidelberg

- dune quartz from the Southern United Arab Emirates, provided by the School of Geography and the Environment, Oxford
- brick from St. Denis (France), provided by the C2RMF Louvre, Paris

Unprepared portions were received for all four samples and the full preparation cycle was performed at ITN. With the exception of the heated flint, the tests were true blind tests.

The fine grain sediment sample is loess originating from the long sequence at Nussloch in Germany. It is used in an intercomparison of German luminescence dating laboratories (Mauz et al., 2003), and soon will become an internationally recognized standard for γ-spectrometry. Results from two participating laboratories are published by Mauz et al., 2003, who also summarize the standard sample preparation procedures. Five different methods were used in order to establish the parameters of equivalent dose and alpha sensitivity. Additive dose TL and IRSL-methods were applied, as well as SAR-IRSL and post IR B-OSL-SAR.

The heated flint was previously measured at the Forschungsstelle Archäometrie am MPI für Kernphysik, Heidelberg. The results are published as sample GK8 in Richter et al. 2000, thus this sample cannot be regarded as a true blind test. Sample preparation followed as outline by Aitken, 1985 and Richter et al., 2000. The palaeodose was determined by the additive dose and regeneration methods, and the additive method for the alpha sensitivity.

The School of Geography and the Environment, Oxford provided a coarse grain quartz sample. It originates from 22 m depth of a core (LIWA2) of a dune in the Southern United Arab Emirates. The Oxford laboratory employed a B-OSL SAR protocol on the coarse grain quartz fraction, using preheats of 260ºC for 10s for 0, 3, 6, 3, 0 Gy regeneration doses, with a 1 Gy test dose for 190 aliquots. An almost identical protocol was employed at ITN as well, with a variation of the regeneration doses.

The brick sample was provided by the Centre de Recherche et Restauration des Musées de France, Louvre, Paris (C2RMF). The preparation of the polymineral fine grain sample was performed according to Aitken 1985. All measurements reported here are TL, using the additive and the regeneration methods. The alpha sensitivity was determined by the additive dose technique.

Blind test results

The results (Table 3) are reported, grouped by sample and technique, together with the values obtained by the other laboratories. The true ages of all samples are unknown. Therefore, only the difference between the determined results on the identical (or almost identical) samples versus the average of all results can be employed as a measure of the quality of the ITN laboratory performance (Table 5). This is applicable despite the fact, that the number of determinations exceed two in only one case.

The values in Table 5 show no systematic difference, neither towards over- nor under-estimation. The mean deviation from the average for the palaeodose determination is 3.64 % and 20.71 % for the a-value determination. The large value for the latter is not surprising, as it combines the differences in alpha and beta source calibration and their associated errors between the laboratories.

Discussion

The differences in calibration values obtained for all samples used are small, and agree very well to each other. As the grain sizes are similar, the average values are employed for each source in routine dating at ITN. It is anticipated to perform more measurements on other fine grain material in order to strengthen the calibration of the $^{90}Sr/^{90}Y$-beta-sources at ITN.

The discussion of the blind tests will be limited to the comparison of only the methods where results from other laboratories are available, and separated for the two parameters determined (where applicable the palaeodose is $D_E + D_I$) (Table 4).

Conclusions

The values obtained for beta source calibrations from the different materials and techniques exhibit a small spread, which allows the calculation of an average value as estimate of the source dose-rates. It also indicates the validity of all gamma irradiations with regards to the irradiation setup and calculation of the absorbed dose (e.g. estimation of distance needed for CPE).

The calibrations of the alpha sources are in good agreement, and the calibration by the manufacturer is preferred, as it is of primary significance.

The overall agreement of the blind test results from the ITN luminescence dating laboratory with the values obtained independently by other laboratories is fully satisfactory. Indirectly it indicates the validity of the source calibrations as well. It was shown that the new luminescence dating laboratory at ITN can provide satisfactory dating results for the four most important materials (fine grain polymineral sediment, quartz from sediment, ceramics and flint) employed in luminescence dating.

SAMPLE	METHOD	PARAMETER	OTHER LAB1	OTHER LAB2	ITN LAB
Loess	TL additive	D_E (Gy)	-	-	63.7 ± 5.7
	TL additive	a-value	-	-	0.024 ± 0.01
	IRSL additive	D_E (Gy)	59.8 ± 14.6	62.4 ± 5.4	57.1 ± 1.6
	IRSL additive	a-value	-	0.09 ± 0.01	0.05 ± 0.01
	SAR-IRSL	D_E (Gy)	-	-	56.9 ± 7.4
	SAR-post IR (B-OSL)	D_E (Gy)	-	-	61.8 ± 6.8
Silex	TL additive	D_E (Gy)	-	8.17 ± 0.06	8.37 ± 0.35
	TL regeneration	D_I (Gy)	-	2.87 ± 0.40	2.86 ± 0.30
	TL additive	a-value	-	0.23 ± 0.03	0.15
Quartz	B-OSL	D_E (Gy)	-	4.64 ± 0.45	4.01 ± 0.20
Brick	TL additive	D_E (Gy)	-	2.6 ± 0.3	2.7 ± 0.08
	TL regeneration	D_I (Gy)	-	-0.8 ± 0.6	0.0 ± 0.08
	TL additive	a-value	-	0.09	0.07

Table 3. *Results of luminescence measurements at ITN and other laboratories of the four blind test samples. D_E is the β-equivalent dose and D_I is the supralinearity correction.*

SAMPLE	PARAMETER	OTHER LAB1	OTHER LAB2	ITN LAB
Loess	D_E (Gy)	59.8 ± 14.6	62.4 ± 5.4	57.1 ± 1.6
	a-value	-	0.09 ± 0.01	0.05 ± 0.01
Silex	Palaeodose (Gy)	-	11.04 ± 0.46	11.23 ± 0.46
	a-value	-	0.23 ± 0.03	0.15 ± 0.05
Quartz	D_E (Gy)	-	4.64 ± 0.45	4.01 ± 0.20
Brick	Palaeodose (Gy)	-	2.6 ± 0.3	2.7 ± 0.08
	a-value	-	0.09 ± 0.01	0.07 ± 0.01

Table 4. *Luminescence results obtained at ITN versus results of other laboratories.*

SAMPLE	PARAMETER	AVERAGE	ITN DEVIATION	ITN DEVIATION (%)
loess	Palaeodose (Gy)	59.77 ± 2.65	-2.67	4.47
	a-value	0.07 ± 0.03	-0.02	28.57
Silex	Palaeodose (Gy)	11.14 ± 0.13	0.09	0.81
	a-value	0.19 ± 0.06	-0.04	21.05
Quartz	Palaeodose (Gy)	4.33 ± 0.45	-0.32	7.39
Brick	Palaeodose (Gy)	2.65 ± 0.07	0.05	1.89
	a-value	0.08 ± 0.01	0.01	12.5

Table 5
Average results for the blind test samples. The deviation of ITN determination from these averages expressed in absolute and relative values.

Acknowledgements
We owe a very special thank to S.Lindauer, BSc.(Forschungsstelle Archäometrie, Heidelberg), for helping in questions of sample preparation, and especially for providing sample material and assistance in the α-source calibration. We would like to thank all the laboratories and persons who provided the blind test samples and made their data available, namely Dr. H.Bray (Oxford Luminescence Research Group),Dr. J.R.Gaborit (Louvre Museum, Paris), Dr. J.Castaing (C2RMF, Louvre), Dr. B.Mauz (University Bonn). Additionally we would like to thank Dr. A.Ferro de Carvalho (LMRIR at ITN), and Dr. N.Mercier (LSCE at the CNRS-CEA) who performed the gamma irradiations.

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Reviewer
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