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The potential of optically stimulated luminescence for medieval building; A case study at Termez, Uzbekistan

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Abstract

Luminescence techniques thermoluminescence (TL) and optically stimulated luminescence (OSL) are generally used to assess the chronology of the last firing of ceramics. In the field of building archaeology, fired bricks can be dated by these techniques. Nevertheless, these luminescence ages are not exactly related to the construction of the building itself, but to the production of the building materials. In some cases, re-use is possible and this raises problems with the interpretation of the dating results. This led us to employ optically stimulated luminescence in a less conventional way.

Before bricks were sealed in masonry by mortar, they would have been exposed to day light, and, as a result, the optical traps of the crystals on the material surface should have been bleached (zeroed by light). Dating the end of the bleaching period is possible by OSL using blue light for stimulation and by IRSL (infrared stimulated luminescence) using IR stimulation. Thus the OSL or IRSL age for these crystals is directly related to the construction of the architectural structure. Experiments were carried out to determine the suitability of this approach and to solve practical problems of sampling. The results show that the bleaching light penetrates between 0.5 to 1 mm into the bricks, according to their transparency. This depth is sufficient to collect enough quartz and feldspar inclusions that have been affected by light in the past, and thus date the construction of the masonry directly. Attempts at surface dating of bricks collected at the medieval citadel of Termez, Uzbekistan, already dated by TL, were the starting point of this research.

Keywords: Optically stimulated luminescence; Thermoluminescence; Surface dating; Polymineral fine grain technique; Ceramic material

1. Introduction

Usually, dating a structure built of fired bricks by luminescence (thermoluminescence (TL), optically stimulated luminescence (OSL)) means dating the manufacture of the materials. Since bricks can be recycled, it is possible that interpretation problems could arise. This led us to use OSL in a less conventional way to investigate the potential for dating the time of construction of the building.

For Termez medieval citadel, we were interested in applying luminescence dating to the brick laying in the walls. Exposure to light of their surfaces before embedding in a wall makes it possible to empty the OSL traps of the crystals present on the material's surface. Consequently, if we were able to extract those crystals, we could know the sealing time and thus obtain a direct date for the construction of the architectural structures. This is the purpose of this work.

2. The Uzbek citadel of Termez

Termez, an Uzbek city in the centre of Central Asia, was founded during the Hellenic era. Some historians believe that Alexander the Great had been there. Its geographic position along the Amou Darya river, at the crossroads of Iranian, Mesopotamian, Indian and Chinese cultures, allowed it to

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become one of the medieval Tokharestan capitals. The city was destroyed in 1220 by Gengis Khan (Leriche et al., 2001).

Termez archaeological site extends over about 500 hectares, making it one of the largest sites in Central Asia. In addition to the various antique monuments built in adobe bricks and daub, the citadel, about 10 hectares in area, has architectural vestiges built in adobe bricks and strong fortifications built in fired bricks; the latter constitute the predominant material during the Islamic era (7th–14th centuries).

3. TL dating of the fired material

Prior to dating the bleached surface of the material, conventional TL dating of fired bricks was carried out. The corresponding results will provide, in addition to brick manufacturing age, an age control for comparison with the surface age. Indeed, the surface age must be lower than, or equal to, the core age of the brick.

3.1. Protocol and experimental aspects

An additive dose and regeneration procedure has been carried out, following the standard TL-dating process of archaeologically heated materials (Aitken, 1985). Two series of TL measurements are required. The first series is performed on aliquots of the material prepared from the archaeological sample being dated. Prior to the TL experiments (also called first TL reading), some of them are given an additional laboratory dose. After TL measurements, the growth of TL with additive dose above the natural level is then known. Nevertheless, to determine the equivalent dose (D_e), the behaviour of the material at low doses, below the natural level, is also studied. This is the aim of a second series of TL measurements (also called second TL reading). They are performed on aliquots of the material having been previously heated in order to empty the TL traps. After laboratory irradiation and consecutive TL experiments, the growth of the regenerated signals with dose is known, and finally D_e can be determined (see, e.g. Guibert et al., 1996). The aliquots used at this stage of the process are distinct from those used for the first series, but they were derived from the same set of grains extracted from the sample investigated.

Brick samples from Termez showed similar mineral composition, the fine-grain fraction containing both quartz and feldspars. Fine grains (3–12 μm) of these minerals were extracted and chemically etched following a 3-stage procedure carried out on particles with diameter $< 40\,\mu m$: (i) HCl 1 M to dissolve carbonate minerals, (ii) H_2O_2 50 vol. to remove organic material, and (ii) HF 0.5 M + HCl 1 M for 100 min to eliminate clay minerals coating the grains (the presence of HCl in the HF solution avoids precipitation of CaF_2); the 3–12 μm fraction was then separated by sedimentation, and the corresponding grains deposited onto identical brass discs (ca. 1 mg per aliquot). As described above, two separate sets of naturally irradiated aliquots were employed for D_e determination: one for the first TL measurements (additive dose technique), the other set being previously annealed by heating for 1 h at a temperature to be determined, before use for the regeneration measurements. This procedure must be carried out for every sample studied, in order to avoid most changes in luminescence properties, which would distort the shapes of the TL curves, cause curvature of TL growth curves and affect the D_e values and, as a final result, the dates themselves (Roque et al., 2004; Vieillevigne et al., 2004).

TL was recorded with an automated TL reader built in CRP2A (Guibert, 2002), according to the following conditions: heating to 500 °C at 4 °C/s in wet nitrogen atmosphere ($\approx$ 96% N_2, 4% H_2O vapour), preheat at 180 °C for 2 min, spectral window from 380–450 nm (2 BG12 Schott filters, 1 infrared reflecting MTO Ta2 filter, EMI 9813QKA photomultiplier tube).

3.2. TL results

As we used polymineral fine grains (containing both quartz and feldspars), a careful study of possible fading was necessary in order to avoid underestimation of the equivalent dose (D_e) (Sanderson, 1988; Visocekas et al., 1994; Zink, 1996). For this fading study, an additive-dose procedure was carried out on fine grains. A selection of bricks was examined by comparing the TL signals of naturally irradiated aliquots which were given an additional laboratory irradiation (the order of magnitude of which is 3 times the expected natural dose) and then stored at room temperature, with those of freshly irradiated aliquots

Table 1
TL dating results of bricks taken from the eastern tower of the citadel of Termez in Uzbekistan

Sample reference	D_e (Gy)	Annual dose rate (mGy/yr)	TL age (X_i) ± overall error (yr)	Statistical s.d. (σ_i) (yr)	Systematic s.d. (yr)	Dates (AD)	Difference $X_i - X_{m1}/\sigma_i$ (BDX 6372 included)	Difference $X_i - X_{m2}/\sigma_i$ (without BDX 6372)
BDX 6367	2.8 ± 0.2	4.3 ± 0.2	650 ± 40	30	20	1350 ± 40	3.4	1.2
BDX 6369	3.6 ± 0.3	4.3 ± 0.2	830 ± 60	50	30	1170 ± 60	−1.5	−1.5
BDX 6372	4.2 ± 0.2	4.7 ± 0.2	890 ± 40	30	30	1110 ± 40	−4.2	
BDX 6375	3.4 ± 0.3	4.8 ± 0.2	700 ± 50	40	20	1300 ± 50	1.5	−0.3
BDX 6391	3.5 ± 0.4	4.7 ± 0.2	760 ± 60	60	20	1250 ± 60	0.0	−1.2
BDX 6392	3.0 ± 0.3	4.2 ± 0.3	720 ± 60	50	40	1280 ± 60	0.7	−0.6

Mean TL age X_{m1} (yr): 1240 ± 30 (BDX 6372 included).
Mean TL age (X_{m2}) (yr): 1290 ± 30 (without BDX 6372).

for which there was a delay of no more than 4 min between the end of the irradiation and the end of the preheating. For one brick (BDX 6372), we observed a delayed TL signal slightly greater than the immediate TL signal. The signal was increased by 9% after 15 days and a stable level seemed to be reached after 2 days. Another brick (BDX 6392) presented a loss of 4% after 15 days and a stable level seemed to be reached after a further 2 days (Vieillevigne et al., 2004). As a result of this preliminary study, all aliquots (natural plus dosed and regenerated) were then stored for 2 days at room temperature after laboratory irradiation and before TL measurements.

The first TL study concerned a tower, located in the east of Termez citadel. Six bricks were taken from the building and two of them (BDX 6369 and 6372) dated from the 12th century, although the ages of the four others are younger (Table 1). We supposed that the two bricks BDX 6369 and 6372 may have been re-used but a χ^2 test confirmed that only one (BDX 6372) provided anomalous results with respect to the assumption of contemporaneity (Table 1). Indeed, χ^2 value is 27.8 when we include BDX 6372 (χ^2 range: 1.6–9.2 with 80% of probability). The weighted average date for the five other samples is 1290 ± 30 AD with a χ^2 equal to 3.4 (χ^2 range: 1.1–7.8) (Vieillevigne et al., 2004). Moreover, brick BDX 6372 could have been taken from the ruins of the east corner of the citadel after the destruction by Gengis Khan in 1220 (Vieillevigne, 2005).

4. Towards the dating of a brick surface

We describe here a feasibility study for dating the surface of a brick to provide an age that is related to the construction of the building. A three-step procedure has been performed. First we determined the penetration depth of light into a brick sample. Then, if this depth is sufficient to collect enough bleached material, we have to find a mechanical process to collect the material of interest below the mortar layer. Finally, D_e of the anciently bleached material has to be determined.

4.1. Experimental aspects

The same mineral fraction and chemical etching procedures were used to prepare the grains. The OSL was recorded with an automated OSL reader built in CRP2A, according to the following conditions: preheat at an appropriate temperature (around 200 °C) for 2 min (Vartanian, 1999; Guibert et al., 2001) and OSL measurements at room temperature. The stimulation source is a halogen–tungsten lamp (Schott KL 1500, emission spectrum 370–830 nm) with 3 filters (MTO J400, to avoid UV-emission of the lamp) coupled to a monochromator (Jobin–Yvon H20) and a light guide (Schott). The spectral window detection range is 250–400 nm (combination of the spectral transmittance of optical filters, 2 Schott DUG11, and the spectral efficiency of photomultiplier tube EMI 9813QKA). For this study, the stimulation wavelength was fixed at 450 ± 10 nm.

The IR stimulation is provided on the same reader by 12 As Ga Al diodes (SEP 8075) emitting at 880 ± 80 nm. Two other filters were employed in front of the photomultiplier tube (one BG39 Schott filter in order to totally absorb the infrared stimulation and one KG5).

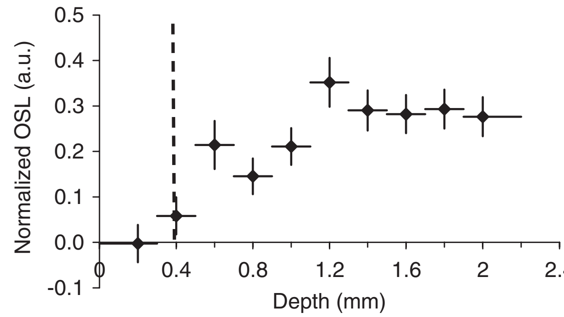

Fig. 1. Brick BDX 6392. Normalised OSL intensity (L/T) from polymineral fine grains as a function of depth in 0.2 mm steps. L is the natural signal measured after the brick surface has been exposed to daylight for 2 weeks. T is the OSL signal in response to a subsequent test dose of 8 Gy. This graph shows that the bleached depth of this brick is around 0.4 mm.

4.2. Measurement of the solar bleaching depth

Solar bleaching affects a volume of material rather than only the very superficial crystals. To date this event, a sufficient quantity of bleached material is required. Thus we measured the depth of brick material below the surface that is affected by the exposure to daylight by using freshly cut pieces of the archaeological bricks. The exposed face has then been eroded by steps of 0.2 mm with a low-speed disc saw. The corresponding fine grain material cut from the brick was used as OSL or IRSL subsample and etched by hydrochloric acid (1 M) only. The remaining natural OSL or IRSL of every subsample obtained was measured, a laboratory test dose was then given to each aliquot and the subsequent luminescence signal measured in order to normalise the first OSL or IRSL measurements.

Experiments were carried out on two brick samples using blue stimulation (BDX 6372 and BDX 6392) and infrared stimulation for the other (BDX 6387). The use of IRSL, instead of OSL, for the third sample was motivated by the low OSL sensitivity of the Termez brick material. Fig. 1 shows the penetration of sunlight into brick BDX 6392 observed using the OSL signal and Fig. 2 shows it using the IRSL signal for BDX 6387. The results show that sunlight penetrates the brick to a depth that varies from sample to sample: from 0.4 mm in 2 week to 1 mm in 1 week, respectively, for bricks BDX 6392 and BDX 6387. Such effects of daylight on the interior of other materials have been observed for granites or gabbro (Haberman et al., 2000; Greilich, 2004) and for quartzite (Tribolo, 2003). It is thus possible to sample anciently bleached material in the surface layer (up to 1 mm) of bricks.

4.3. Collection of the superficial layer

Once we had established that the light penetrated to a depth sufficient to collect material for dating, we decided to continue this feasibility experiment on sample BDX 6372. As the surface of bricks is commonly irregular and covered by mortar, a par-

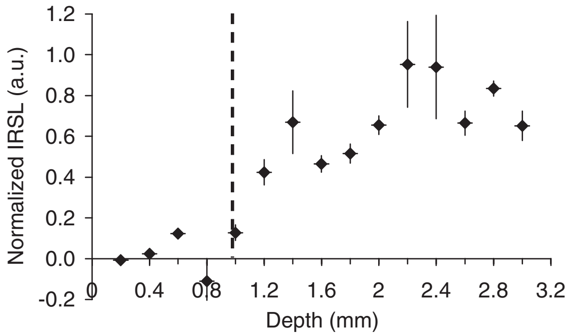

Fig. 2. Brick BDX 6387. Normalised IRSL intensity (L/T) from polymineral fine grains after daylight exposure for 1 week, and with a test dose of 5 Gy. We can observe that the bleached depth obtained by IRSL is around 1 mm.

ticularly careful sampling was performed. First, the mortar is removed by a mechanical and manual method using a scalpel, taking care not to remove fragments from the brick surface. Then because of the irregular topography, regular sampling is carried out every 5 mm (order of magnitude of the blade dimensions) at the brick surface. The thickness of each point sample is controlled by a micrometer. We cannot hide the fact that it is a very long, time consuming operation.

4.4. *A first attempt at surface dating by OSL*

Once the material of interest has been extracted and chemically treated (fine grain), D_e was measured by OSL (stimulation at 450 ± 10 nm). Unfortunately, the OSL signals were very weak and we only obtained an upper limit for the estimation of the palaeodose: less than 4.7 Gy at the 2σ level.

According to the corresponding annual dose (4.8 ± 0.2 mGy/yr) including full alpha dose from brick, beta dose from brick (50%) and mortar (50%) and environmental dose rate, we found that the surface age should be less than 990 yr, a result that is not contradictory to the firing age (890 ± 40 yr, Table 1).

5. Conclusion

The methodology and the practical aspects of surface dating have been established. This feasibility experiment encourages us to continue with more OSL-sensitive materials, from other sites. Surface dating by OSL can provide archaeologists with direct dating information for the construction of architectural structures. It is necessary to provide age control by TL in order to compare it with the surface age.

This work permitted the observation of some interesting optical properties at the brick's surface. Indeed, it is possible to detect a bleached depth able to be sampled, i.e. a depth characteristic of each brick. Sampling this bleached depth is not very easy.

The first result of surface dating by OSL was obtained on one brick sampled at Termez in Uzbekistan: it is not very accurate because of the low sensitivity of the material.

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