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Mortar and surface dating with Optically Stimulated Luminescence (OSL): Innovative techniques for the age determination of buildings

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Summary. — In this work the results of a dating study on bricks and mortars using both Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL) are shown. The samples came from the outside walls of the Certosa di Pavia, located in northern Italy and independently dated (XVII-XVIII century). TL dating, applied to bricks using the fine grain technique, allowed to determine the time of manufacture of the bricks (XII century), that resulted therefore re-used. To circumvent this problem the application of two innovative dating techniques, OSL surface dating and mortar dating, was attempted. The first was applied to the light-shielded surfaces of bricks and allowed to successfully determine the edification of the wall (XVII century). Mortar dating gave instead severe age overestimation: the results obtained on coarse grain quartz with the SAR technique both on multi-grains aliquots and with single-grain analyses were highly dispersed indicating an incomplete bleaching of the quartz grains. The shine-down curves were in fact characterized by the absence of the so-called fast component, as confirmed by Linear Modulated OSL technique.

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1. – Introduction

Delayed luminescence allows to determine the amount of radiation dose absorbed by a material. If this quantity can be correlated with the time elapsed since the starting of irradiation, luminescence can be used as a dating tool [1, 2]. The physical basis of the phenomenon is the presence of defects, both intrinsic and impurity related, in the lattice of insulating materials. These defects act as traps as well as recombination

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centres for the free charge carriers originated by the interaction with ionizing radiation. Thermoluminescence (TL) is the most common technique, and is measured by heating the sample under controlled conditions [1]. More recently Optically Stimulated Luminescence (OSL) [2], by which traps are emptied by optical excitation with appropriate wavelength, has been developed.

In the years 1965-70 it was demonstrated that TL, widely applied in the field of dosimetry, could be fruitfully used also for ceramics dating. The idea was to use the natural quartz and feldspars crystalline inclusions present in the clay and consequently in shards, as dosimeters of the radiation field due to natural radioactivity of both ceramics and surrounding environment. In the following decades specific procedures were developed, allowing to reach precision as good as 5% in determining the age of archaeological shards.

The following equation of the age summarizes what briefly mentioned, and is used in all luminescence dating techniques:

$$\text{Age (years)} = \text{palaeodose (Gy)} \ / \ \text{annual dose rate (Gy/year)}.$$

The palaeodose is the radiation energy deposited within the sample since the last heating for TL or since the last daylight exposure for OSL. The annual dose rate is the rate at which ionizing radiation energy is absorbed.

However, TL could not resolve all the dating problems of archaeologists and architectural historians. This technique in fact gives the time elapsed since the firing of the ceramics in kiln. In the dating of buildings or brick structures, TL is therefore able to determine the period of manufacture of a brick which may not coincide with the construction of the structure itself, because bricks are often recycled. Moreover, all the archaeological remains made with unfired materials (adobe, marble, and stone) cannot be dated by TL.

In order to overcome these limits, in recent years a new approach was attempted: mortar dating. Compared to all other datable materials, mortar has the great advantage of existing in abundance in every stage of construction, and cannot be recycled. Dating mortar would represent a great improvement in historical building studies allowing the identification of different construction phases, interventions and modifications of a structure. Mortar was first recognized as a suitable dosimeter for the reconstruction of doses after accident in nuclear power plants [3]. As mortar is an unfired material, dating is accomplished by using OSL, technique routinely used for dating geological materials such as sediments, sands and loess. This technique, in fact, determines the time elapsed since the last exposure of mineral grains to sunlight, *i.e.* the deposition age [2]. The application of OSL to mortar dating is possible because the quartz crystals contained in the aggregates are exposed to daylight during the mixing and laying of the mortar itself, and could therefore be used as dosimeters, recording the natural dose since the end of the exposure to sunlight, which coincides with the edification.

In the last few years another novel innovative luminescence methodology was developed: the so-called surface dating. It allows dating events resulting in the permanent covering of surfaces of buildings previously exposed to the light, like construction, destruction or land-slides covering. The first few published studies on surface dating focussed on TL measurements of calcitic samples such as marble [4, 5]. This approach results in a low precision preventing a probably widespread application. Recently an alternative strategy using OSL to date the last time a stone surface was exposed to daylight, has been investigated [6]. If such a surface is buried or permanently covered, this approach should provide a direct method for dating the time of construction. Rocks contain the same minerals that are used for dating archaeological and geological materials, and many rock surfaces are exposed to daylight for very long periods of time before being exploited and covered. Thus it is likely that, in many cases, all the mineral grains in at least some of the surfaces of stones used by ancient peoples would have been sufficiently exposed to daylight to have been completely zeroed, and thus provide an accurate chronometer.

In this work TL and OSL were applied to bricks and mortars belonging to a portion of the perimeter wall of the Certosa di Pavia, fallen down in 1995. The Certosa di Pavia is a monastery located 8 km north of Pavia dated XIV-XV century, while the perimeter wall is dated by historic sources to a later period (XVII-XVIII century).

The collapse of the wall allowed to sample "sandwiches" of bricks still stick together with the original mortar and other with clean surfaces exposed to light since the collapse.

According to sample's typology and technique, we should therefore obtain different results:

– the manufacture of bricks by TL dating

– the building of the wall either by OSL dating of mortar or by surface dating of the light-shielded surfaces of bricks

2. – Samples and experimental

TL dating was attempted on three bricks (B2, B5 and B23), still presenting a thick layer of mortar on one of the major surfaces. The fine-grain technique [7] was used. The samples were prepared under dim red light, using the standard procedure and the polymineral fine grain (4–11 μm) fraction was deposited on aluminum discs. To evaluate the dose absorbed since manufacturing (palaeodose) the multi-aliquot additive technique (MAAD) was used [1].

For surface dating, only one sample (B23) was selected. It presented one surface completely covered by a very thick layer of mortar (about 3 cm) and another surface completely clean, unshielded and exposed to light since the collapse. In red light, cores (10 mm in diameter and 40 mm long) were drilled from the two surfaces using a low speed drill. Mortar was mechanically removed and residual contamination was eliminated leaving the whole cores in 5% HCl solution for 2.5 h. The cores were then carefully washed, dried and gently filed in steps of 0.5 mm from the surface, to obtain material from nine different depths. The same 4–11 μm polymineral fraction used for TL dating was selected, but this time the evaluation of palaeodose was made with the Single Aliquot Regenerative dose OSL protocol (SAR) [8].

For OSL dating of mortars, samples of the mortar associated to bricks B2 and B5 were taken. They are referred to as M2 and M5, respectively. The 180–250 μm quartz fraction was selected using the standard procedure [9] and fixed on stainless steel discs (10 mm diameter) with a heat resistant resin (Dow-Corning 805). The evaluation of palaeodose was made even in this case with the SAR OSL protocol.

TL measurements were performed using a homemade system based on the photon-counting technique with a photomultiplier tube (EMI 9235QB) coupled to a blue filter (Corning BG12). The samples were heated from RT to 480 °C at 15 °C/s. Artificial irradiations were carried out by a 1.85 GBq ^{90}Sr $-$ ^{90}Y beta source (dose rate: 4.21 Gy/min), a 37 MBq ^{241}Am alpha source (dose rate: 14.8 Gy/min).

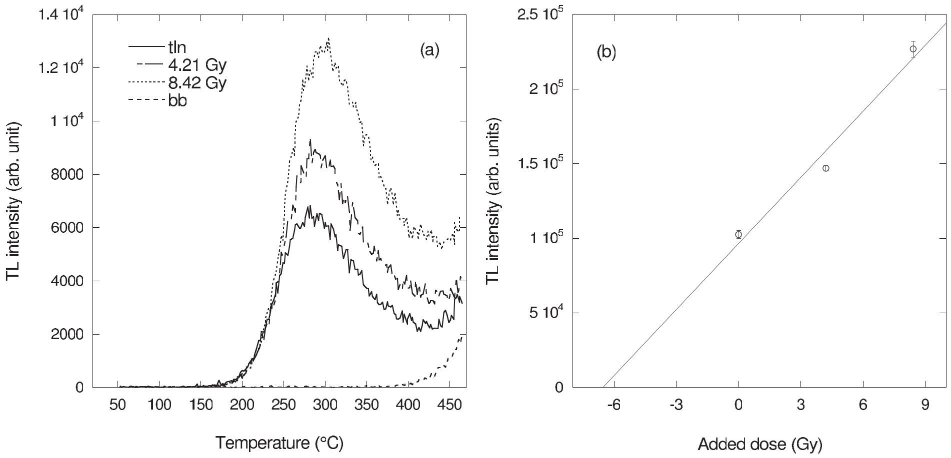

Fig. 1. – a) TL glow curves and b) TL growth curve with dose obtained on brick sample B23 applying the MAAD protocol.

OSL/IRSL measurements were undertaken using a Risø TL-DA-20 equipped with a ^{90}Sr/^{90}Y beta source delivering 0.135 ± 0.013 Gy/s to quartz coarse grains (180–250 μm) and 0.23 ± 0.02 Gy/s to fine grains (4–11 μm). The samples were stimulated by an array of blue LEDs (470 ± 30 nm) with a constant stimulation power of 54 mW/cm^2 or with IR LEDS (830 ± 10 nm) with a constant stimulation power of 360 mW/cm^2. Photons were detected by a bialkali photomultiplier tube (EMI 9235QB) coupled to a 7.5 mm Hoya U-340 filter (280-380 nm) for quartz or to a blue filter pack (Corning 7-59 plus Schott BG-39; 300-500 nm) for feldspars.

Single-grain measurements were made using a single-grain laser attachment of the Risø system. The stimulation source was a 10 mW Nd:YVO4 solid-state diode-pumped laser emitting at 532 nm, which can be focused sequentially on to each of 100 grains mounted on a special aluminum sample disc [10].

The annual dose rate was indirectly derived from the measurement of the radioactivity of the samples. The U and Th concentrations were obtained by total alpha counting using ZnS scintillator discs assuming a Th/U concentration ratio equal to 3.16 [1]. The contribution due to ^{40}K content was deduced from the total concentration of K obtained by flame photometry. Because water absorbed part of the radiation that would otherwise reach the sample, the saturation water content was evaluate and a water content equal to $(80 \pm 10)\%$ of this value was assumed for all samples, taking into account the available information of humidity of the area. Attenuation of the beta dose for coarse-grain quartz was taken into account [11], as well as the cosmic ray contribution to the final dose rate [12].

3. – Results

3˙1. *TL dating of bricks.* – As mentioned before, the TL palaeodose was measured with MAAD technique. Four aliquots of the same sample were used to get the natural signal, others were given different artificial doses (4.21 and 8.42 Gy) superimposed to the natural one and preheated (200 °C for 10 s) to eliminate the unstable component of the TL signal. After identifying the thermally stable portion of the curves (plateau

TABLE I. – *Assumed average water contents, measured radioactivity concentrations, annual dose rates, palaeodose and dating results obtained on brick samples.*

Sample ID	H_2O % (±10%)	U ppm (±5%)	Th ppm (±5%)	K_2O % (±3%)	a factor
B2	11	2.64	8.34	2.84	0.12
B5	7	2.71	8.56	2.81	0.25
B23	9	3.76	11.56	2.83	0.17

Sample ID	Annual dose (mGy/a)	Palaeodose (Gy)	TL data	Surface Palaeodose (Gy)	OSL surface data
B2	5.0 ± 0.2	4.3 ± 0.2	1150 ± 50 AD		
B5	6.9 ± 0.3	5.7 ± 0.3	1190 ± 50 AD		
B23	7.2 ± 0.5	6.3 ± 0.3	1140 ± 50 AD	2.7 ± 0.5	1640 ± 70 AD

test) [1], the growth curve of TL as a function of dose allowed to obtain by extrapolation the values of palaeodose, as shown in fig. 1 for sample B23. Palaeodoses, radioactivity concentrations, assumed average water contents, annual dose rates and dating results are reported in table I.

3·2. *OSL surface dating of brick.* – The OSL surface dating technique was applied to two surfaces of brick B23, one covered by mortar and one exposed to light. First of all, the bleaching efficiency of solar light and the bleaching extent were investigated by measuring the normalized blue and IR stimulated OSL signal as a function of depth from cores drilled from a surface exposed to sunlight for 24 hours. Samples were taken from surface down to 4.5 mm depth at steps of 0.5 mm. After measuring the natural OSL signal (L_n), the samples were irradiated with a normalizing test dose, preheated at 200 °C and measured (T_n). The mean normalized signals (L_n/T_n), obtained integrating the shine-down curves in the first 0.8 s and subtracting the background (average OSL signal of the last 10 s of each measurement) are reported in fig. 2 as a function of depth. It can be seen that, for both OSL and IRSL, the same trend is observed: L_n/T_n rapidly increases with depth in the first 1.5 mm, then reaching an almost constant value. It is also evident that the signal from the most superficial layer (*i.e.* the first 0.5 mm) is not fully bleached. The extent of incomplete bleaching was quantified by evaluating the residual dose remained after the exposure: a value 1.50 ± 0.05 Gy was obtained. This value will be therefore subtracted to any measured palaeodose.

Aliquots were then prepared using the powder obtained from the first 0.5 mm of any core drilled from the surface of Bricks23 covered by mortar. After the natural luminescence signal of the sample is registered, the very same aliquot is irradiated in laboratory with increasing doses. To overcome the problem of sensitivity changes due to the repetition of irradiation and heating, a normalizing test-dose measurement is performed after each measure. Details on SAR protocol are reported in table II. To account for thermal transfer, a zero dose was included in the SAR protocol (recuperation test); the correction

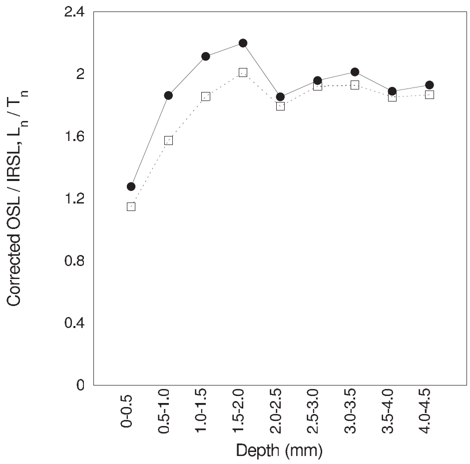

Fig. 2. – OSL (filled circles) and IRSL (empty squares) intensity as a function of depth in brick B23. Each point is obtained as the mean of four measurements on different aliquots of sample.

of the test dose sensitivity procedure was also checked with a repeat dose point (recycling ratio test). For all aliquots the recuperation never exceeded 5% and the recycling ratio test always yielded a value near to unity [13]. This allowed the construction of a sensitivity-corrected dose response curve for each aliquot (see typical decay curves and dose response curves in fig. 3). The palaeodose is obtained by interpolation of the natural signal, corrected for sensitivity changes. For the mortar-shielded surface, the net mean value of palaeodose and date are reported in table I.

3˙3. *Mortar dating*. – Mortar dating follows the same procedures which are also used for dating sediments. Two samples of mortar (M2 and M5) were dated, associated to

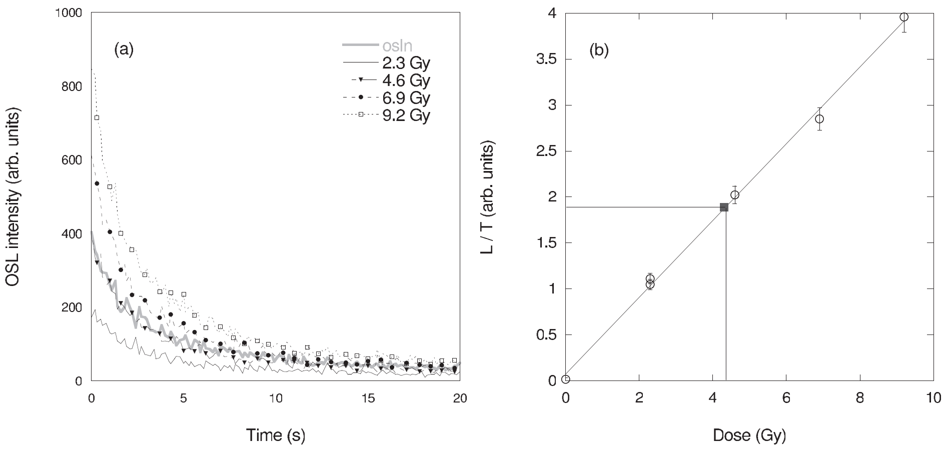

Fig. 3. – a) OSL glow curves and b) OSL growth curve with dose obtained on light-shielded surface of brick B23 applying the SAR protocol. The full square corresponds to the natural signal while the empty circles to regenerated signals.

TABLE II. – *Single Aliquot Regeneration (SAR) sequence used for surface dating of brick B23.*

Step	Treatment	Observed[c]
1	Given dose, $D_i^{(a)}$	-
2	Preheat at 200 °C for 10 s	-
3	Stimulate for 40 s at 125 °C	L_i
4	Given test dose, $D_t^{(b)}$	-
5	Heat to 180 °C	-
6	Stimulate for 40 s at 125 °C	T_i
7	Return to 1	-

(a) For the natural sample $i = 0$ and $D_0 = 0$ Gy. Four regenerative dose (2.3, 4.6, 6.9, 9.2 Gy) plus a zero dose measurement and a repeat dose (2.3 Gy) were used.
(b) The test dose was 1.3 Gy.
(c) L_i and T_i were derived from the decay curve, integrating the first 0.8 s minus a background estimated from the last 10 s.

bricks B2 and B5, respectively. The quartz extracted was checked for the absence of feldspar contamination using IR stimulation on laboratory irradiated samples [14]. The already discussed SAR procedure was used; details are reported in table III. Example of shine-down curves and related growth curve are shown in fig. 4 for sample M5. Sixty aliquots for each sample were measured and only those with a recuperation lower than 5% and a recycling ratio within ±10% of unity were accepted for palaeodose calculation. Forty-nine aliquots of sample M2 and fifty-six of sample M5 were accepted, passing all tests. The palaeodoses weighted mean, the results of radioactivity measurements and the related dose rates are reported in table IV. The distributions of palaeodoses are

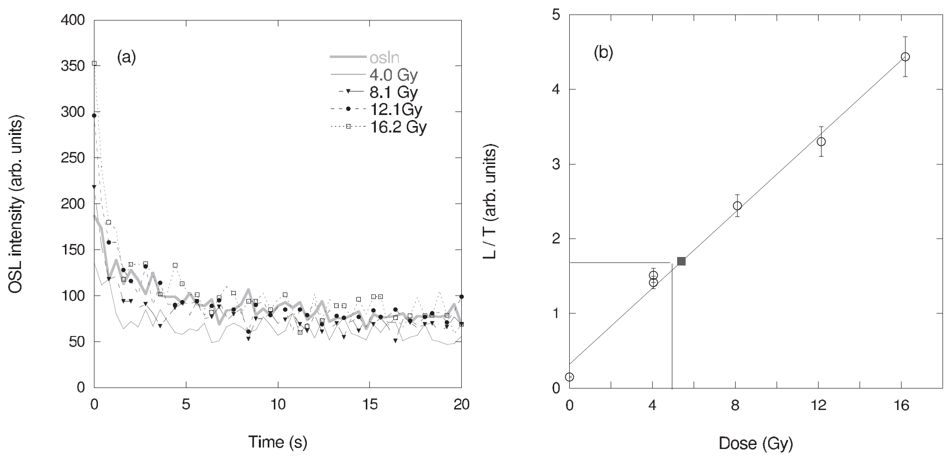

Fig. 4. – a) OSL glow curves and b) OSL growth curve with dose obtained on mortar M5 applying the SAR protocol. The full square corresponds to the natural signal while the empty circles to regenerated signals.

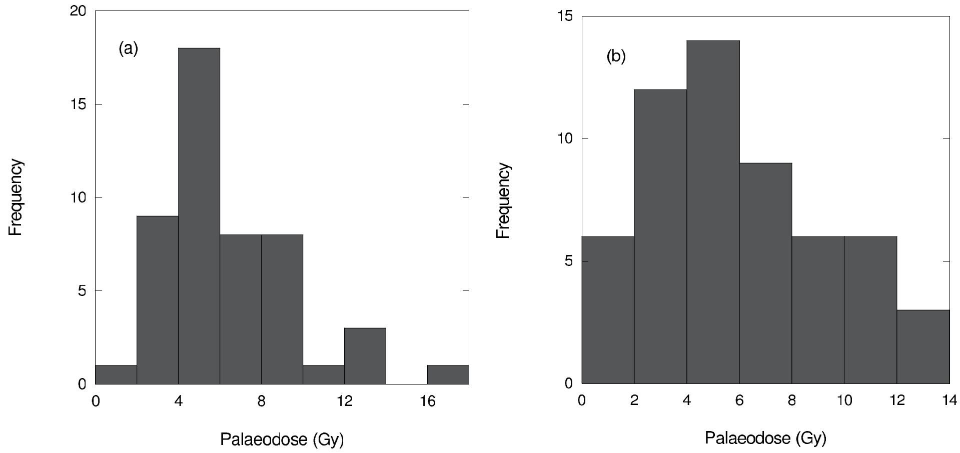

Fig. 5. – Distribution of palaeodoses from multi grains quartz aliquots extracted from mortar M2 (a) and M5 (b).

reported in fig. 5 in which the histograms show a set of values not symmetrically clustering around a mean value. This non-Gaussian distribution suggests that the samples were not completely bleached, and the use of such palaeodoses should result in a severe age overestimation (table IV).

It is often reported that mortar shows adversary properties such as partial bleaching, low signal and slow decay [15]. In contrast to the application of OSL dating in geology, the zero-setting event, *i.e.* the event to be dated, is not the bleaching during deposition or during aeolian transport, but the end of a production process starting from the retrieval of sand in the gravel pit, ending when lime slurry covers the surface of the quartz grains. As a result, the degree of bleaching of the geologically accumulated signal is uncertain. Quartz grains extracted from mortar may therefore show a wide distribution of doses, with possibly only some completely bleached grains. The challenge in using such a material as dosimeters is in identifying these well bleached grains. Measurements of incompletely bleached samples using multi-grain aliquots should normally result in an overestimation of the dose accumulated in the sample because it is the result of a mixed population of bleached and unbleached sample. The single-grain dating technique [16] can be useful in identifying the well-bleached grains of quartz.

Single-grain OSL technique was therefore applied to M2 samples. A total of 2400 grains were submitted to the SAR protocol (table V), 105 grains giving a detectable light signal (*i.e.* three times higher than the standard deviation of the background signal). Figure 6 displays a histogram of the results obtained with single-grain techniques: doses range from 0.20 to 32 Gy and the average weighted dose was 3.87 ± 0.12 Gy, lower than that obtained with multi-grain aliquots. The dose distribution clearly shows that the sample was poorly bleached, making the results unreliable (table IV).

For samples thought to consist of some grains that were fully bleached before burial and other grains that were incompletely bleached when buried, the "Minimum Age Model" (MAM) [17] may be an appropriate mean of estimating the palaeodose specific to the population of well bleached grains. The palaeodose for M2 calculated with the 3-parameter MAM is 2.5 ± 0.6 Gy (table IV).

TABLE III. – *Single Aliquot Regeneration (SAR) sequence used for mortar dating of samples M2 and M5.*

Step	Treatment	Observed[c]
1	Give dose, $D_i^{(a)}$	-
2	Preheat at 180 °C for 10 s	-
3	Stimulate for 100 s at 125 °C	L_i
4	Give test dose, $D_t^{(b)}$	-
5	Heat to 160 °C	-
6	Stimulate for 100 s at 125 °C	T_i
7	Stimulate for 40 s at 290 °C	
8	Return to 1	-

(a) For the natural sample $i = 0$ and $D_0 = 0$ Gy. Four regenerative doses (4.0, 8.1, 12.1, and 16.2) plus a zero dose measurement and a repeat dose (4.0 Gy) were used.
(b) The test dose was 1.3 Gy.
(c) L_i and T_i were derived from the decay curve, integrating the first 2 s minus a background estimated from the last 20 s.

Fast bleaching of the OSL signal is a necessary pre-requisite for OSL dating. OSL from quartz is thought to arise from different traps, each having a different photoionisation cross-section [18, 19]. Under constant power stimulation, the continuous-wave OSL (CW-OSL) signal can be approximated by three exponential components designated fast,

TABLE IV. – *Assumed average water contents, measured radioactivity concentrations, annual dose rates, palaeodose and dating results obtained on mortar samples.*

Sample ID	H_2O % (± 10%)	U ppm (± 5%)	Th ppm (± 5%)	K_2O % (+ 3%)	Annual dose (mGy/a)	Palaeodose[a] (Gy)
M2	14	1.15	3.63	1.56	2.20 ± 0.06	4.60 ± 0.09
M5	12	1.71	5.40	1.73	2.43 ± 0.07	2.11 ± 0.08

Sample ID	Data[b]	Palaeodose[c] (Gy)	Data[d]	Palaeodose[e] (Gy)	Data[f]	
M2	80 ± 70 BC	3.87 ± 0.12	250 ± 70 AD	2.5 ± 0.6	880 ± 260 BC	
M5	1140 ± 40 AD					

(a) Palaeodose and
(b) data obtained on multi-grains quartz aliquots.
(c) Weighted mean palaeodose and
(d) relative data obtained on single quartz grains.
(e) Palaeodose calculated with MAM and
(f) relative data obtained on single quartz grains.

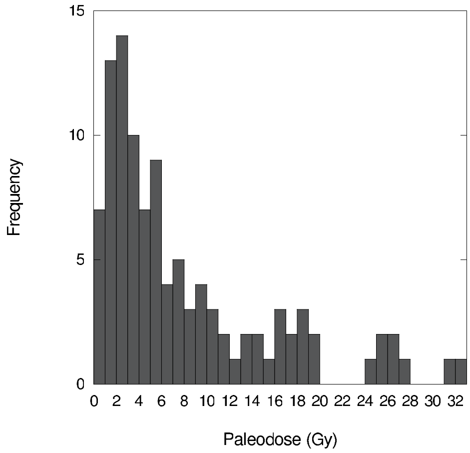

Fig. 6. – Distribution of palaeodoses obtained on quartz single grains of sample mortar M2.

medium and slow, according to their rate of decay under optical excitation [18, 20]. The palaeodose estimation procedure such as SAR uses the initial part of the OSL signal minus a background based on the signal level at the end of the stimulation period. This net initial signal includes contributions from any fast component as well as medium and/or slow components. For many samples, the fast component dominates the initial OSL signal, but in some cases the contribution from other less light-sensitive signals can be significant [21]. These medium and slow components have different thermal stabilities and dose response characteristics as compared to the fast component and they can therefore adversely affect the dose estimation. So it is desirable to have a dose-estimation protocol based on a well-separated fast component OSL signal, which is the most readily

TABLE V. – *Single Aliquot Regeneration (SAR) sequence used for mortar dating of samples M2 with the single-grain technique.*

Step	Treatment	Observed$^{(c)}$
1	Give dose, $D_i^{(a)}$	-
2	Preheat at 160 °C for 10 s	-
3	Stimulate for 1 s at 125 °C	L_i
4	Give test dose, $D_t^{(b)}$	-
5	Preheat at 160 °C for 10 s	-
6	Stimulate for 1 s at 125 °C	T_i
7	Return to 1	-

(a) For the natural sample $i = 0$ and $D_0 = 0$ Gy. Four regenerative doses (1.3, 2.6, 3.9, and 5.2 Gy) plus a zero dose measurement and a repeat dose (1.3 Gy) were used.
(b) The test dose was 1.3 Gy.
(c) L_i and T_i were derived from the decay curve, integrating the first 0.2 s minus a background estimated from the last 0.2 s.

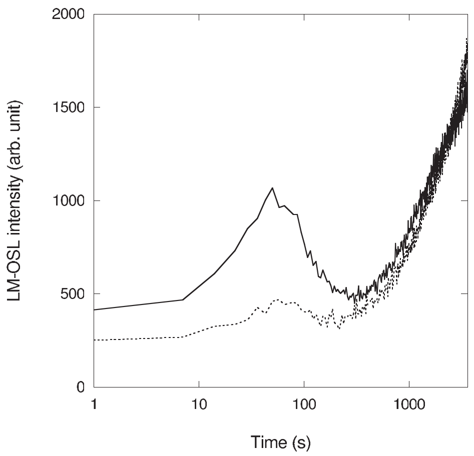

Fig. 7. – LM-OSL curves measured for quartz extracted from mortar samples M2 (continuous line) and M5 (dotted line).

bleached. In order to separate the overlapping OSL components, the Linear Modulation OSL (LM-OSL) [19] was applied on the quartz extracted from samples M2 and M5. This technique consists in linearly increasing the intensity of the stimulation light from 0 to 100%. The LM-OSL from a single trap initially increases and then decays after reaching a maximum. The time at which the OSL reaches its maximum value depends on the photoionisation cross-section. If more than one trap contributes to the OSL decay curve then more than one peak will be seen in the LM-OSL plots. The first peak in the LM-OSL curve was attributed to the fast bleaching component [19]. If this first peak is equal to or smaller than the second, then the fast component is not predominant. This is the case of sample M2, whose LM-OSL emission is shown in fig. 7. Because the fast component is the most bleachable signal, its low contribution can be the cause of the overestimation obtained on mortars.

4. – Conclusions

The three bricks from the wall of Certosa di Pavia were dated by TL to the XII century, instead to the expected XVII-XVIII century. Because the period of manufacture of the bricks did not coincide with the time of the construction of the wall, it was evident that they were re-used. In order to circumvent this problem, surface dating of brick and mortar dating were attempted.

The surface dating technique, applied here for the first time to bricks, permitted to date the construction of the wall to the XVII century, in very good agreement with the historical sources.

OSL dating of the mortar using multi-grain aliquots gave instead a severe overestimation of the age, due to the problem of incomplete bleaching that often affects mortar. In order to circumvent this problem, the single-grain OSL approach was attempted, but with unsuccessful results. The cause of the failure of mortar dating is that the most bleachable component of the OSL signal, *i.e.* the fast component, is not the predominant component in our sample. So the mortars were not completely bleached during transport, mixing and laying.

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