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Quality Assurance for dosimetric measurements of mortar on polymineral fine grain fraction

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***Abstract* – The aim of this work is to evaluate the possibility to use double Single Aliquot Regeneration (dSAR) method on polymineral fine grain fractions extracted from historical mortars for Optically Stimulated Luminescence (OSL) dating. The studied samples were collected from the church of Notre Dame de la Place (Bordeaux, France) and from the Saint-Jean Baptiste chapel (Périgueux, France). The research work was aimed at identifying the best experimental conditions related to the InfraRed stimulation time and the Preheating temperature value for a more accurate and precise dosimetric measurements. The results show the fundamental importance of these dosimetric tests for the Equivalent Dose (ED) evaluation.**

I. INTRODUCTION

In dating field of historical buildings the possibility of dating the mortars represents a very important point as it allows overcoming the problems related to the bricks dating, that can be reused.

The event dated by OSL corresponds to the exposure of the quartz grains to daylight during the manufacturing of mortar and laying. The exposure to light causes the optical zeroing of luminescence signal accumulated by quartz inclusions. When the mortar is layered onto a wall, it is embedded in the structure and hereby the exposure of mortar to light stops. From then on, the mortar is subjected to radiation from its nearest environment and starts accumulating radiation dose that is later used for dating purposes.

In general, mortars are composed of a binder, such as lime, gypsum or cement and an aggregate, such as sand or grit. The aggregate is constituted by quartz that represents the mineral used for luminescence dating. In general, the OSL dating method involves dosimetric measurements on quartz grains with a diameter of 100-250 micron [1-5]. Nevertheless, sometimes only small proportions of quartz grains of these dimensions are found. An alternative for dating is represented by the possibility of use fine grain polymineral fractions constituted by quartz and feldspars. In these cases, it is however necessary to eliminate the luminescent contribution of the feldspars either by etching in HF [6-7], or through InfraRed (IR) stimulation [8-9].

The purpose of this work is to find the best experimental conditions for a more accurate and precise dosimetric measurement on fine grain fractions extracted from historical mortars. The experimental study was performed in order to establish a procedure including different tests useful to eliminate the feldspars contribution to luminescence signals (*IR stimulation test*) and to record only the contribution of the stable traps (*preheating temperature test*).

II. MATERIALS AND METHODS

A. Protocol and experimental aspects

The studied mortars were collected from two French sites: 8 BDX and 16 BDX samples from the church of Notre Dame de la Place (Bordeaux) and 1 BDX from the Saint-Jean Baptiste chapel (Périgueux).

After removal the external 2 mm, each sample was mechanically crushed using an agate mortar. The crushed material was sieved in order to obtain Ø<40 μm granulometric fractions. Then the fractions were etched in 10% HCl for 120 minutes, then in 10% H_2O_2 for 96 hours in order to eliminate, respectively, carbonate phase and organic components. After sedimentation in acetone, according to Stokes' law, polymineral fine grained 4<Ø<11 μm fractions, then named FG, were obtained [6, 10-12].

Artificial luminescence signals were induced by the ^{90}Sr-^{90}Y calibrated beta source integrated in the Risø systems delivering 6 Gy/min.

OSL and IRSL (InfraRed Stimulated Luminescence) for mortars were performed using the semi-automated Risø readers TL-DA-15 with EMI 9235QA photomultipliers. OSL and IRSL signals were obtained using respectively, 41 blue LEDs (470±30 nm) and laser diode (830±10 nm). The stimulation units delivered were ~30mWcm^{-2} for OSL and ~ 240mWcm^{-2} for IRSL at 90% power. Both emissions were detected using a Hoya U340 optical filter.

Equivalent dose values, useful for luminescence dating, were calculated with dSAR protocol [8, 9, 13-14] (Table 1). This protocol, detailed in the Table 1, has been suggested as a method of isolating an optically stimulated luminescence signal dominated by quartz from a polymineralic mixture of grains. In order to eliminate feldspars luminescent contribution, the IR stimulation, before the blue-light one was performed [13, 15-16]. Previous studies have, in fact, shown that IR stimulation at temperatures above 50°C not depletes quartz post-IR OSL signals [17-18]. The degree of feldspars contamination in FG fraction was checked through the R-value coefficient. It is determined by the ratio between the normalized OSL intensity after IR stimulation (post-IR OSL/T2) and the normalized OSL emission (OSL/T1) [7]. OSL/T1 represents the OSL signal normalized by T1 test dose and post-IR OSL/T2 the OSL emission obtained after IR stimulation normalized by T2 test dose. If the R coefficient, calculated on a group of three aliquots for each sample, is close to unity, it indicates that the signal comes from a fraction free of feldspars [7, 8, 11-12, 19].

For the assessment of the dosimetric accuracy in equivalent dose measurements, in dating processes field, the dose recovery test is used. This test consists on the application of dSAR protocol on fined grain aliquots giving a laboratory dose and followed by optical bleaching [20]. In such test, the ratio of the measured to given dose should be closer to unity.

Table 1. Steps for ED evaluation with dSAR protocol. Li and Ti are derived from the decay curves, taking the first 0.8 minus a background estimated from the last 3.5 s integral of the OSL signal. Li [post- IR] OSL, Ti is the sensitivity-corrected [post-IR] OSL intensity.

Step	Treatment	Observed
1	Give dose (0Gy for natural signals) D_i	-
2	Preheat X°C for 10s	-
3	Stimulate with IR light for X s at 40°C	-
4	Stimulate with blue light for 40 s at 125°C	Li
5	Give test dose ~20% natural dose	
6	Cut heat at X°C(preheating temperature-40°C)	
7	Stimulate with IR for X s at 40°C	
8	Stimulate with blue light for 40 s at 125°C	Ti
9	Repeat steps 1-8 for 5 times for regeneration doses in the appropriate range dose	

B. Choice of IR stimulation time

In dSAR protocol, in order to reduce as much as possible the feldspars signal, IR stimulation, before the reading of the optically stimulated luminescence, is performed.

In the test, different times at 100s, 300s and 500s have been used. The variation of the sample sensitivity and the dispersion of the data were considered in order to determine the optimal value of this parameter.

C. Choice of Preheating temperature

Preheating (*Ph*) is useful in order to empty unstable traps, before the reading of the optically stimulated luminescence. The signals generated from these traps, in fact, could cause an underestimation of the age of samples. In dating of samples as mortars, preheating temperature is normally set between 180°C and 240°C.

High temperatures could cause the heating transfer of charge from deep energy levels to OSL traps and could lead to an overestimation of the age of the sample. Considering the expected ages of the samples values, the preheating temperature was determined for each sample observing the results at 180°C, 200°C, 220°C and 240°C. The test to choose the preheating temperature is conducted at IR time value of 100 s.

III. RESULTS

The best dosimetric conditions for each sample have been determined considering R-value and data dispersion. The Relative Standard Deviation (RSD) associated to each R-value was defined as:

$$\mathrm{RSD}(\%) = \frac{\sigma}{\bar{x}} \cdot 100 \qquad (1)$$

where σ is the standard deviation and $\bar{x}$ the mean value. An example of Recovery values vs IR time and vs Ph temperature for 8 BDX is reported, respectively, in Figures 1 and 2.

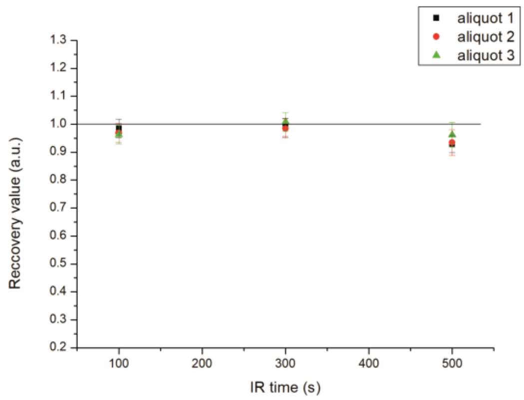

Fig. 1. Recovery values vs IR time for 8 BDX sample.

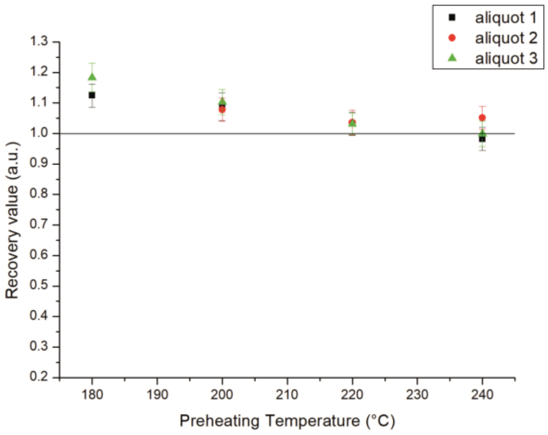

Fig. 2. Recovery values vs Ph temperature for 8 BDX sample.

Tables 2 and 3 show, respectively, the R-value and the RSD (%) results for the different IR stimulation times and for various Ph times for all samples.

Table 2. R-value results and RSD (%) for each IR time for all samples.

Sample	IR time [s]	R	RSD (%)
8 BDX	100	0.97	1
	300	0.99	1
	500	0.94	2
16 BDX	100	1.00	10
	300	1.00	14
	500	0.96	5
1 BDX	100	1.10	15
	300	0.98	8
	500	1.00	6

Table 3. R-value results and RSD (%) for each Ph time for all samples.

Sample	Ph temp [°C]	R	RSD (%)
8 BDX	180	1.00	8
	200	0.99	8
	220	1.01	5
	240	1.05	4
16 BDX	180	1.07	1
	200	0.96	2
	220	1.02	0.5
	240	1.03	0.2
1 BDX	180	1.00	8
	200	0.99	8
	220	1.01	5
	240	1.05	4

Based on the obtained results (Tabs. 2 and 3), Table 4 shows the parameters chosen for the equivalent dose measurements useful for dating. The ED was evaluated by radial plots.

Table 4. Parameters chosen for the equivalent doses measurement

Sample	IR time [s]	Ph temp [°C]
8 BDX	300	240
16 BDX	500	200
1 BDX	500	200

In Figures 3-5 the radial plots, respectively for 8-BDX, 16-BDX and 1-BDX, were shown. In these graphs, the x-axis reports the relative error (σ/t) expressed as percentage while the y-axis the precision t/σ.

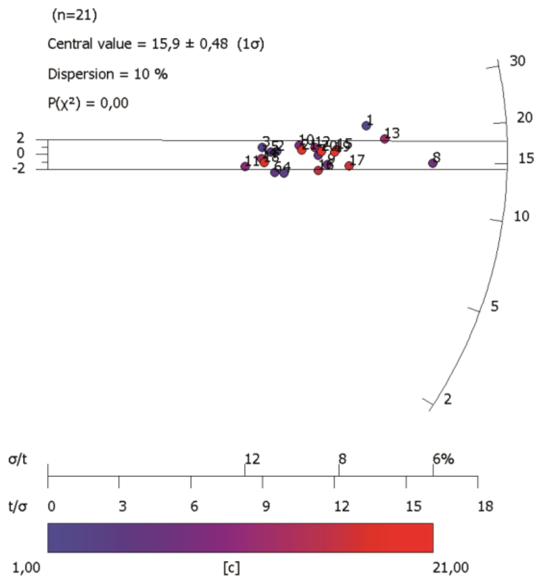

Fig.3. 8-BDX: Radial plot

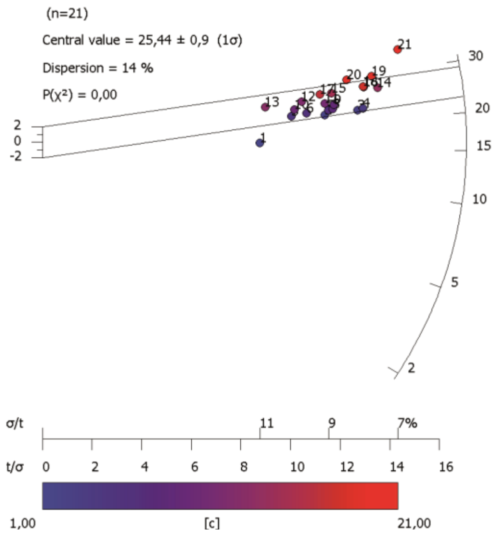

Fig. 4. 16-BDX radial plot

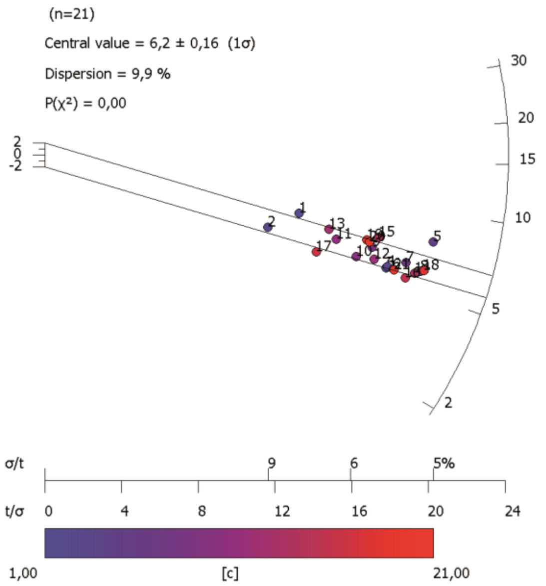

Fig.5. 1-BDX Radial plot

IV. CONCLUSIONS

The results show that the dosimetric tests performed on the samples are of fundamental importance for a more accurate estimate of equivalent dose. Considering the feldspar contamination found for the samples analyzed, the dSAR protocol was used for the dating of mortars. Through the recovery test, the best dosimetric conditions for IR time and preheating temperature were identified. For the selection of the optimal value, the accuracy and precision of the measurements made on the parameters used for the tests were taken into account (three for each sample). The optimal value identified for the three parameters represents a compromise between these two characteristics. The accuracy and precision of the measurement are given respectively by the value of R and the value of RSD.

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