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# Alpha sensitivity determination in quartzite using an OSL single aliquot procedure

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## Introduction

In the process of dating burnt stone artefacts, the determination of the alpha sensitivity is of paramount importance especially when the radioisotopic contents of the pieces under study are high (typically > 1 ppm), since in such cases the contribution of the alpha dose rate to the total dose rate is not negligible. This parameter is also known to vary from sample to sample, as in the case of burnt flints dated by TL (see for instance Mercier et al., 1995), and its value is needed for each sample.

Up to now, the OSL technique has been rarely used for measuring this parameter and in cases where it has been done, it concerned mainly quartz grains extracted from sediments (Rees-Jones, 1995). Here we report alpha sensitivity measurements, using TL and OSL, made on burnt quartzite pebbles, which we wish to date by OSL. It will also be shown that an OSL single aliquot method can be used in order to determine this parameter for each disc, after its equivalent dose (ED) has been measured.

## Alpha sensitivity : S- $\alpha$

The alpha sensitivity under consideration (S- $\alpha$ ) (Valladas and Valladas, 1982) is defined as the ratio of the relative alpha and beta sensitivities :

$$S-\alpha = \frac{L\alpha \cdot D\beta}{\phi\alpha \cdot L\beta}$$

where  $L\alpha$  and  $L\beta$  are the luminescence intensities induced by the artificial alpha integrated flux  $\phi\alpha$  (number of  $\alpha/cm^2$ ) and the artificial beta dose  $D\beta$  (Gy), respectively. The equivalent beta dose rate  $D\alpha_{\text{equ.}\beta}$  to be considered in the age equation is the product of the S- $\alpha$  value and the alpha flux rate, which depends on the specific U and Th alpha fluxes (19400 and 5377 respectively; Valladas, 1988) and on the radioisotopic contents of the sample<sup>1</sup>.

<sup>1</sup> Thus, S- $\alpha$  can also be expressed to a first approximation as:

$$S-\alpha = k \cdot \frac{[U] \cdot d_{\text{SU}} + [\text{Th}] \cdot d_{\text{STh}}}{[U] \cdot \phi_{\text{SU}} + [\text{Th}] \cdot \phi_{\text{STh}}}$$

where k is the usual k-value (Zimmerman, 1971) and  $d_{\text{SU}}$  and  $d_{\text{STh}}$  are the specific alpha doses for U and Th, respectively.

$D\alpha_{\text{equ.}\beta} = S-\alpha \cdot \{ \eta_U \cdot [U] \cdot \phi_{\text{SU}} + \eta_{\text{Th}} \cdot [\text{Th}] \cdot \phi_{\text{STh}} \}$   
where units are :

$$S-\alpha : \text{Gy} / \alpha/cm^2$$

$$\phi_{\text{SU}}, \phi_{\text{STh}} : \text{number of } \alpha/cm^2 / \text{p.p.m.} / \text{y}$$

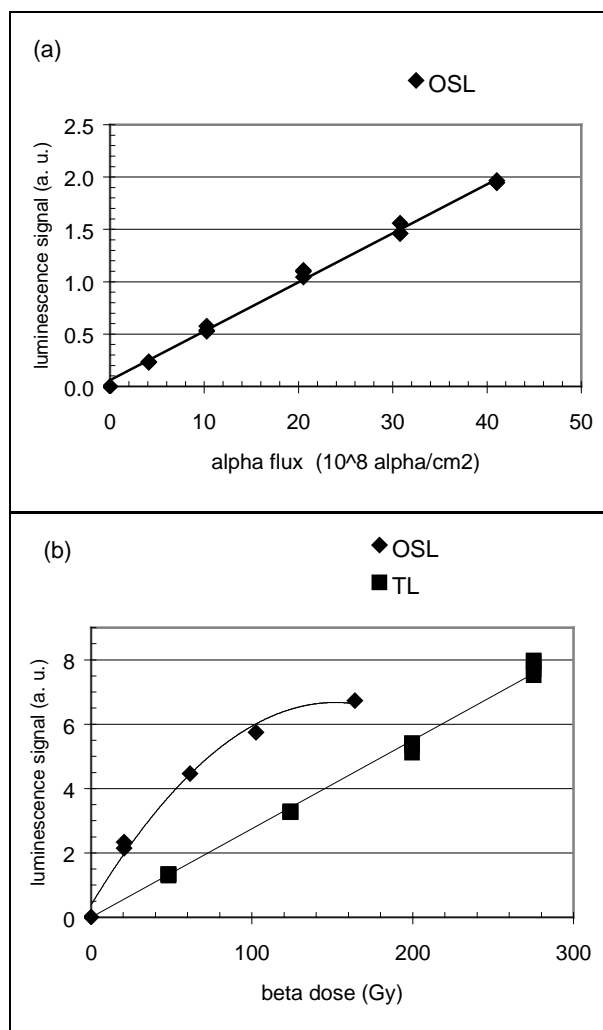
$$[U], [\text{Th}] : \text{p.p.m.}$$

and where  $\eta_U$  and  $\eta_{\text{Th}}$  are two correcting factors that are close to unity, the exact values depending on the alpha particle energies inside the sample under irradiation.

This definition assumes that the luminescence signals vary linearly with the beta dose and the alpha integrated flux. For all the samples we studied, this last assumption was true for OSL signals induced by alpha integrated fluxes up to  $40 \cdot 10^8 \alpha/cm^2$  (see sample CAR 7 in Fig.1a as an example), a value which is generally higher than the natural alpha integrated flux received by the sample since it was last heated. However, the OSL beta growth curve can exhibit a strong saturation behaviour as in the case of sample CAR 7, which contrasts with the beta induced TL signals which increase linearly up to 250 Gy (Fig. 1 b). Consequently, a correction of the OSL S- $\alpha$  value is necessary for samples exhibiting this behaviour in order to evaluate the mean S- $\alpha$  value they experienced during the burial time.

## Samples and equipment

The samples used were archaeological burnt lithics (quartzite pebbles) from 3 Upper Palaeolithic sites in the Côa valley (Portugal): 4 from Cardina (CAR4, CAR5, CAR7, CAR8), 3 from Olga Grande Sul (OGS1, OGS2, OGS5) and 1 from Quinta da Barqua (QBS2). Their TL S- $\alpha$  values had been measured during a prior dating study (Valladas et al., 2001; we note here that the units for S- $\alpha$  in Table 1 of that paper should have been shown as  $\mu\text{Gy} / 10,000 \alpha / \text{cm}^2$ ), in which fine grains previously heated at 350°C for 90 minutes were deposited on stainless steel discs, then irradiated either with a  $^{90}\text{Sr}/^{90}\text{Y}$  beta source delivering 0.134 Gy/sec or with a  $^{238}\text{Pu}$  source having a flux of  $2.279 \times 10^6 \alpha/cm^2/sec$ . The TL signals were measured with a home-made automatic reader (Valladas et al., 1994) equipped with a blue-UV filter



**Figure 1.**

(a) Sample CAR7: the linearity of the growth curve as a function of the integrated alpha flux is typical of what is observed when the luminescence signal is measured using the OSL and TL techniques.

(b) TL and OSL beta regenerated growth curves for the same sample. The TL and OSL growth curves behave differently, the TL one showing greater linearity.

(MTO 380 nm) and normalised by irradiating all discs with a fixed beta dose.

The OSL measurements reported here were performed on a Risø TL/OSL-DA-15 apparatus. The stimulation was done with a broad-band blue and green light (420-550 nm) filtered from a halogen lamp (Bøtter-Jensen and Duller, 1992). Luminescence was measured by a Thorn-EMI 9235QA photomultiplier tube using 3 Hoya U-340 filters. Beta irradiation came from a <sup>90</sup>Sr/<sup>90</sup>Y source delivering 0.165 Gy/sec (attached to the Risø reader)

and alpha irradiation from the <sup>238</sup>Pu source mentioned above.

#### Sample preparation and measurement procedure

Two sets of fine grains were prepared to evaluate the influence of pre-treatment on the OSL  $S-\alpha$  values: one containing recently heated grains (set H) and another bleached grains (set B).

The preparation of set H was very similar to the one used in TL: the pebbles were crushed and sieved in order to obtain a < 100  $\mu$ m granulometric powder which was heated at 350°C for 90 minutes in a furnace, then washed with hydrochloric acid, pure water, ethanol and acetone. Fine grains were deposited in acetone on stainless steel discs according to Zimmerman's protocol (1971). Five discs were prepared for each sample.

The same procedure was applied to set B, except that unheated fine grains were bleached for 100 sec at 125°C in the Risø reader.

Both sets were exposed to the alpha source at the integrated fluxes reported in column 1 of table 1. For comparison, integrated fluxes experienced by the samples during their period of burial (estimated from the TL ages and radioisotopic contents) are given in column 2.

| sample | (1)   | (2)   |
|--------|-------|-------|
| CAR4   | 41.02 | 26.20 |
| CAR5   | 41.02 | 5.11  |
| CAR7   | 41.02 | 28.21 |
| OGS1   | 41.02 | 4.48  |
| OGS2   | 41.02 | 3.98  |
| OGS5   | 54.70 | 15.84 |
| QBS2   | 30.08 | 5.99  |

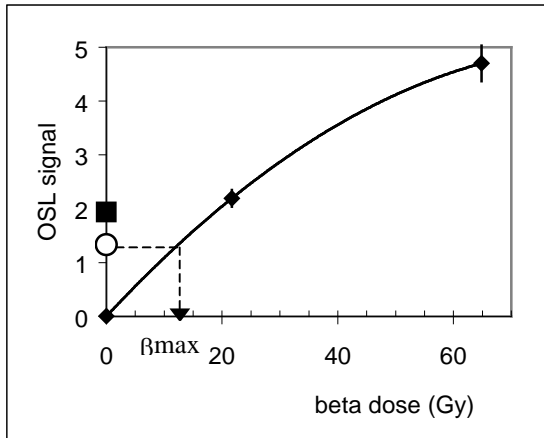
**Table 1.**

(1) : integrated alpha flux (10<sup>8</sup> alpha/cm<sup>2</sup>) used for  $S-\alpha$  determination

(2) : integrated alpha flux (10<sup>8</sup> alpha/cm<sup>2</sup>) experienced by the sample during burial calculated from age estimation and U, Th contents

We used a standard SAR protocol (Murray and Wintle, 2000) to determine the  $S-\alpha$  value. For each aliquot, 7 cycles were measured, each one consisting of :  $\beta$  dose (= 0 for cycles n°1 and n°2), preheat at 220°C for 10 sec, OSL measurement for 100 sec at 125°C, test dose, cut-heat at 160°C and OSL for 100 sec at 125°C. The percentage of thermal transfer was estimated during cycle n°2. Increasing regenerative doses allowed us to build the beta growth curve; the dose of cycle n°7 was identical to the one of cycle n°3 to check the efficiency of the sensitivity correction. Using set B of sample CAR7 as an

example (Fig. 2), the beta doses for the successive cycles were 0, 0, 22, 65, 108, 173 and 22 Gy and the test dose was 7 Gy.



**Figure 2.**

SAR protocol applied to sample CAR7 (set B). The plot shows the region up to 65 Gy. The square represents the alpha induced OSL signal and the diamonds the beta regenerated signals.

The open circle represents the signal that would have been observed if the integrated alpha flux had been equal to the estimated archaeological alpha flux ( $\phi_{arch.}$ ). As defined,  $\beta_{max}$  is the equivalent beta dose corresponding to this flux, giving a maximum value of  $S-\alpha$ :

$S-\alpha_{max} = \beta_{max} / \phi_{arch.}$ . The mean value  $S-\alpha_{corr.}$  corrected for non-linearity is then obtained by integrating or, simply, by averaging:

$$S - \alpha_{corr.} = (1/100) \sum_{n=1}^{n=100} \{ \beta_n / (n \cdot \Phi_{arch.} / 100) \}$$

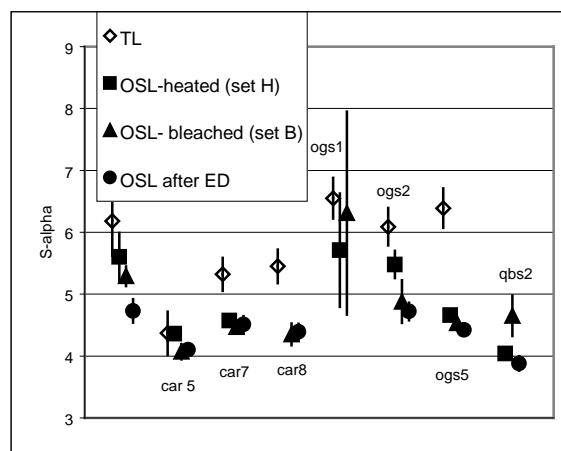
where  $\beta_n$  is the dose corresponding to an integrated flux of:  $n \cdot \phi_{arch.} / 100$

To correct for the non linearity of the regenerative curve, we first calculated by proportion the OSL signal which we would have observed if the integrated alpha flux had been equal to the estimated archaeological alpha flux ( $\phi_{arch.}$ ) and deduced the corresponding beta dose ( $\beta_{max}$ ) by using the regenerative beta growth curve. In the last step of the calculation, the  $S-\alpha$  value was computed by averaging the values calculated between 0 and  $\beta_{max}$ . The obtained  $S-\alpha$  value is then slightly dependent on the estimated age and an iterative procedure should sometimes be employed.

For the present sample under consideration (set B of CAR7, Fig. 2),  $\phi_{arch.} = 28.2 \times 10^8 \alpha/cm^2$  and  $\beta_{max} = 12.6$  Gy. The corrected value  $S-\alpha_{corr.}$  is then =  $4.47 \mu Gy / 10^3 \alpha/cm^2$ .

**Results of comparisons**

The mean OSL  $S-\alpha$  values obtained after bleaching (set B) or after heating (set H), and the TL  $S-\alpha$  values, are plotted in figure 3. For all the samples under study, in view of the associated errors, the two OSL  $S-\alpha$  values are indistinguishable. However, the TL values tend to be higher than the OSL ones (20% higher for CAR4, CAR7, CAR8 and OGS2, 40% higher for OGS5 and 190% higher for QBS2). The reason for this difference is not clear. It may be due to the fact that traps and luminescent centres involved during the two kinds of measurements are not strictly identical.



**Figure 3.**

TL or OSL  $S-\alpha$  values (in  $\mu Gy / 1000 \alpha/cm^2$ ) obtained after different sample pre-treatments. In view of the uncertainties, the OSL values measured for each sample are all very similar. On the other hand, the TL values tend to be higher: they are almost 20% higher for CAR4, CAR7, CAR8 and OGS2, 40% higher for OGS5 and 190% higher for QBS2. The TL  $S-\alpha$  value for QBS2 ( $12.1 \pm 1.2 \mu Gy / 1000 \alpha/cm^2$ ) is not plotted for clarity.

We have already seen that the  $S-\alpha$  measurements make possible the determination of the alpha sensitivity from a single aliquot, which is an appreciable advantage over protocols using k or a-values requiring several aliquots. Moreover, the good agreement between the two sets of OSL  $S-\alpha$  values indicates that the alpha sensitivity is independent of the past history of the sample. Consequently, it should be possible to use the same disc to determine first the ED and then the  $S-\alpha$  value.

To test this hypothesis, another set of 5 fine grain discs were prepared for all the archaeological samples and their EDs were measured using the SAR protocol. Subsequently, each disc was alpha irradiated and its OSL  $S-\alpha$  measured with the same SAR protocol. For each sample, the mean OSL  $S-\alpha$

value is reported in fig. 3. As expected, it does not appear to be significantly different from the values of sets H and B: all measurements can thus be performed on a single aliquot.

If we consider sample CAR7 once again, the OSL  $S-\alpha$  values measured for 5 discs after ED determination were 4.68, 4.53, 4.55, 4.42 and 4.39, giving a mean of  $4.51 \mu\text{Gy}/10^3\alpha/\text{cm}^2$ . Considering the U and Th concentrations of this sample (1.475 p.p.m. and 12.086 p.p.m., respectively) and the  $\eta_U$  and  $\eta_{Th}$  coefficients specific to the alpha source used, the beta-equivalent alpha dose rate ( $D\alpha_{\text{equ},\beta}$ ) is then :

$$4.51 \times 10^{-3} \times \{ 0.87 \times 1.475 \times 19400 + 0.91 \times 12.086 \times 5377 \} = 379 \mu\text{Gy}/y .$$

### Conclusion

For the quartzite pebbles under study, the OSL  $S-\alpha$  sensitivity seems to be independent of the pre-treatment of the sample. To our knowledge, this is the first time that an experimental procedure allows for the ED and alpha sensitivity determinations to be made on a single aliquot. This might be particularly interesting for the dating of both sediments and small samples like pottery sherds.

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### Reviewer

Richard Bert Roberts