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## Luminescence date and archaeological ages: An epistemology of the luminescence dating

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La thermoluminescence utilise le rapport entre la dose accumulée et la dose reçue annuellement.

Il s'agit donc au sens physique d'une méthode de datation absolue, qui contient en elle-même sa propre calibration. Mais, du fait du nombre élevé de paramètres, de leurs natures essentiellement probabilistes, il ne peut y avoir de datation unique. Il n'y a que des représentations que le chronologiste a de la date réelle. Cela nous conduit à réfléchir à la nature exacte d'une datation et sa place vis-à-vis de la date réelle. De manière que les datations puissent être reçues de façon critique par les archéologues et s'intégrer à leurs propres corpus de connaissances.

Thermoluminescence is based on the ratio between the accumulated dose and the annual dose rate. It thus acts with the physical meaning of a method of absolute dating, which contains in itself its own calibration. But, because of the high number of parameters, their probabilistic nature, there cannot be single dating. There are only representations that the chronologist has of the true date. That leads us to think of the exact nature of a dating and its place with respect to the true date. So that the dating can be received in a critical way by the archaeologists and be integrated into their own set of knowledge.

Mots-clefs : datation par luminescence, équation d'âge, incertitude

Keyword: luminescence dating, age equation, uncertainty

## **A. Introduction**

Although the role of the physicist usually stops with the production and the publication of the ages, the ages themselves have a life after. They are an information about artefacts or sites. And then, one expects that they are used as an argument among others in the constitution of the knowledge of the historians (Art historian, historians and archaeologists).

Actually, some times, the age obtained by physical method fully disappears when the site or the object is published by the historians, especially in the communication to the general public, which is very strange when it is obtained after request of the archaeologists and on public funds. In some other cases, it is copied as received without any analysis in review papers, as if they are gospel. A last case, and not the less odd, the historians made a full arbitrary choice between the dates, keeping only the ones in agreement with their own assumptions. In all these cases, the physical age is misused, as it should be used by comparison with other informations to construct the archaeological discourse.

How to present the results (report, papers, list of dates) in view to be useful for the historians? The problem is how have a rigorous (scientific) discourse with the minimum of technical jargon in a such way that the physical result can be transfer to the historical discourse; keeping the possibilities to other colleagues to control and reproduce our work from the published data.

In the present paper, we are interested by the luminescence dating and the nature of the luminescence age.

### **Age equation**

To well understand the luminescence dating methods, we must remind the physical process involved in luminescence (Aitken 1985). The luminescence methods are based on the faculty of

the minerals to accumulate energy from ambient radioactivity.

Actually, the radioactive energy is transferred to electrons. The electrons are transferred, or ionized, to the conductivity band. From here, they can go freely in all the crystal. Most of them come back to their initial level, emitting some luminescence. It is the fluorescence process. But a minority is trapped by defects in the crystal, the so-called traps. Due to the thermal agitation of the crystal, it has some probability that the trapped electrons are evicted from their traps and come back to their initial level emitting some luminescence. It is the phosphorescence process. The time spent in the trap can be more or less long, from some microseconds to several billion years. For a trap with large lifetime compare to historical ages, the eviction can be regarded as zero, and the trap accumulates continuously new electrons. It is possible to accelerate the eviction from the traps by giving to the electron some thermal or optical energy. In the first case, we speak about thermoluminescence (TL), and in the second case, about optical stimulated luminescence (OSL). We must outline that the TL and OSL are irreversible processes: after the heating or the optical bleaching, the traps are empty and the process is stopped. Without a new irradiation, we do not have any new luminescence signal.

Resetting and accumulation are the two characteristics useful to apply the luminescence to dating. Actually, to be able to date, we need to have a clock; that is a physical process, function of the time with an outstanding instant. In our case, the process is the accumulation of electrons in the traps, or indirectly the absorbed energy, i.e. the dose. It is then a dosimetric method. The outstanding, or initial, instant is the last emptying of the traps. It could be the crystallization of the mineral (stalagmite), the heat at a temperature above 500 °C (ceramic, heated stone), the solar bleaching during a daytime (aeolian or water-laid sediment), or even a shock (seismic fault).

As it is, we can use the luminescence methods to date facts. But, as the most of the clock, it

depends of the environment. And they can be used only as a relative method to date objects from the same environment. The dating is not limited to the dosimetry of the sample. Actually, to obtain an absolute dating method, we need to calibrate this clock by a term reflecting the environment. It is made by measuring the dose-rate, or annual dose, using analytical or radiometric methods and by dividing the accumulated dose by the dose-rate. Hence, one define the age equation as:

$$\text{Equation 1} \quad \text{Age} = \text{accumulated dose} / \text{annual dose}$$

We must outline that the dimensional analysis associated to this equation: a quantity is divide by its own rate, shows that the result is, physically, a time. It means that we obtain always a calendar age. In comparison, the radiocarbon dating gives a so-called C14-age. To have the corresponding calendar age, we need to use external calibration, generally another dating method as the dendrochronology. The age equation can be split in two terms: the numerator corresponds to the luminescence clock; the denominator plays the role of a correction factor of the error due to the environment. From a physical point of view, it displays what we know about the environment of the sample: the luminescence clock going more or less quickly following the ambient radioactivity. Hence, a granitic soil has a higher radioactivity compared to a loessic environment. The dose rate with its uncertainties should reflect our knowledge on the environment (radioelements, wetness...). In this case, the final result is fully correct as a representation of our knowledge on the age.

### ***Uncertainties***

Usually, the uncertainties in luminescence dating are based on the works of Aitken and Allred, summarize in Aitken (1985).

In this work, the uncertainties are separated in two groups: the uncertainties arising from random

effects and the uncertainties arising from systematic effects. We prefer here to speak about uncertainty than error, as the term 'error' must be reserved to the difference between the measured value and the true value (VIM). Several sources of uncertainties are identified regrouped into seven components. Two components are related to random effect, they concern the measurement: one for the accumulated dose measurement, and the other for the annual dose measurements. The other components are related to systematic effects and deal with the parameters or the calibration of the equipment. Their mathematical expressions for usual thermoluminescence can be found in Appendix B of Aitken (1985). Following the used methods (TL or OSL, fine grains or coarse inclusions, single aliquot or multi-aliquot), they can be adapted. In some cases, other sources of uncertainties can be added (eg. Anomalous fading, external dose rate for museum objects...).

We must keep of mind that in luminescence dating the uncertainty  $U_c$  on the age is quoted with a coverage factor of one, or one standard uncertainty. That means that the age  $A$  of the sample is believed to lie between  $a - U_c$  and  $a + U_c$  with a level of confidence of about 2 out to 3 chances (ca. 68 percent), which is commonly written  $A = a \pm U_c$ . On other terms, if we have measured three ages, for two of them the true age is expected to be inside the confidence interval. But for one of them, it should be outside. The test with samples of known ages gives informations on the evaluation of the uncertainty. In the case where all the true ages are inside of the confidence interval, the uncertainty are overestimate. Opposite, if less than two out of three true ages are inside the confidence interval, the uncertainty is underestimated (Zink et al. 2005).

Following the recommendation of the International Committee for Weights and Measures CIPM (ISO, 1995; Taylor and Kuyatt, 1994), the scientific community of the metrological science prefers separated the uncertainties in two groups:

A. those which are evaluated by statistical methods,

B. those which are evaluated by other means.

rather than used the classification of uncertainty components as 'random' and 'systematic'. The nature of the uncertainty as 'systematic' or 'random' is relative depending of the treatment of the measurement.

The table 1 shows an example of uncertainty budget in the case of a Hallstatt site in East of France, with their classification in type A or type B evaluation. As you can see, there is not a simple correspondence between 'systematic'/'random' and 'type A'/'type B' classifications. The wetness is classified by Aitken as an uncertainty arising from a systematic effect, but in the present case is based on several measurements in laboratory, and then a statistical treatment.

Actually, only the uncertainties on luminescence measurements come from random effects. The uncertainties on the dose rate and the wetness, although are evaluated by statistic methods, are related to systematic error. As say before, the dose-rate (including the wetness) are use to correct the error due to the environment, which is a systematic error for all the samples with the same context.

### ***Law of propagation***

Actually, the distinction between random and systematic error given by Aitken is best understood as a distinction between type A and type B evaluated uncertainties. But their treatment for several measurements in the same context, their propagation, is best understood with the notion of correlation or uncorrelation between samples as suggested by Aitken (1985) to identify 'systematic' and 'random' effects.

For the components of the uncertainty uncorrelated between samples, Aitken suggests to use the usual method for combining standard deviations, the “root-sum-of-squares” (square root of the sum-of-the squares). And for the component of the uncertainty correlated between samples, he

suggests to take the weighting mean of the individual uncertainties (Aitken 1985).

We can illustrate the difference between correlated and uncorrelated uncertainties between samples in the case of the dating of a stratigraphic unit. A stratigraphic unit is the less archaeological unit which define an age based on chronological links (Harris 1979). Then it is associated to an unique age (the timespan of the age can be very short or very large). We have measured several samples from the same stratigraphic unit and we want to combine them to obtain the age of the unit.

As suggested by Millard (2006), we use a hierarchy between the components of the uncertainty (fig.1). At a first level, we have the constant parameters and the calibrations of the equipments. Assuming that only a laboratory produces the date, the uncertainties are fully correlated. At a second level, we have the gamma and cosmic dose-rates. These radiations have large length path and their contribution are generally homogeneous in a single stratigraphic unit. The gamma dose-rate comes usually from various measurements and then is type A evaluation. We can use an average associated with an uncertainty based on the "root-sum-of-squares" for the stratigraphic unit. For the cosmic dose rate, we take usually reference data (type B evaluation).

We should now consider the alpha and beta dose-rate. They are absorbed quickly in the matter. And we can not assume the homogeneity of their contribution on the entire unit. They are measured from sub-samples different from the sub-samples used for luminescence measurements, even if they come from a same sample in the stratigraphic unit. Due to their short path range (some micron for the alpha particles), and the a-priori inhomogeneity of the radioelement contents at micro-scale (micro-dosimetry), we can not assume, a-priori, that the alpha and beta dose-rates measured for a sub-sample correspond to the dose-rate for an other sub-sample. Then, as for the gamma dose-rate, but for opposite reasons, we should take the average value of the

individual measurements of the alpha and beta dose-rate associated with uncertainties based on the “root-sum-of-squares” for the stratigraphic unit. In this case, we disagree with Millard (2006) who put the alpha and beta dose-rate at lower levels than the stratigraphic unit.

At the level of the sample, we group the parameters calculated by luminescence measurements, and especially the accumulated dose. They arise from the same sub-samples (we neglect the differences between aliquots) and characterise each sub-sample. A priori, it has no reason that the sub-samples are similar. We can say that the age (true value), corrected by the environment (correction factor from a systematic effect) is transform into a particular accumulated dose depending on the nature of the mineral (the nature of the mineral is a-priori independent -uncorrelated- from the other minerals in the vicinity). Then we cannot take the average of this parameters on the stratigraphic unit and it combined “root-sum-of-squares” uncertainty. We must first calculate the age for each sub-samples using the measured value of the accumulated dose and the average for the dose-rates. The uncertainty of the age of the stratigraphic unit is obtain by combining the uncertainties of the average value of the correlated parameters, including the dose-rate and the “root-sum-of-squares” of the components of the uncertainty due to uncorrelated parameters, including the accumulated dose.

The efficiency alpha is a special case. It is a correction parameter of the alpha dose-rate (systematic effect) measured by luminescence (random effect). We see here the conceptual difficulty of the notion of systematic and random effect. In the present case, we consider that the components of the uncertainty due to the efficiency alpha is an uncorrelated between samples, and is treated as the component of the uncertainty due to the accumulated dose..

## ***Conclusion***

The luminescence clock is the accumulated dose by a mineral during some time. By dividing the

accumulated dose by the annual dose, we convert the luminescence clock in time clock. The annual dose or dose rate is a calibration factor used to reduce the systematic effect due to the environment. Then the result is accurate, i.e. very close to the true age, even though it has a large uncertainty. The luminescence age gives an absolute information to the archaeological age, which is based on temporal relations (e.g. stratigraphical unit). The luminescence dating is a metrology science, giving a measurement in the time dimension. As it is, it has no particular archaeological meaning. It reflects just what the physicist believes about the age from his own expertise in luminescence dating. It is only when this measurement is understood as an information for a specific problematic investigated by the archaeologist or the Art historian, that one can speak about a archaeometrical analysis.

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Table 1 example of uncertainty budget for luminescence dating in the case of the archaeological site of Marsal – Pransieu (Moselle, France) modified from Zink and Porto (2006). The subscript associated to the uncertainties refers to the Appendix B from Aitken (1985), except  $\sigma_8$  and  $\sigma_{10}$  which are adapted to the present case.

source of uncertainties	Relative uncertainties for a sample ( <i>resp.</i> for a stratigraphic unit)	
	Type A	Type B
luminescence measurements (s1)	4-5% (1%)	
dose-rate measurements (s2)	7-9% (3 %)	
calibrations (sources, alpha counters) (s4)		1%
parameters (s5)		3%
uranium/thorium ratio (s6)		2%
wetness (s7)	5-8 % (3%)	
calibration (gamma spectrometer) (s8)		5%
fading (s10)	2-8 % (1-3%)	
Combined relative standard uncertainty	13 % (9 %)	

figure 1 hierarchy of the parameters used in luminescence dating.  $\Theta$  is the age,  $D_i$  with  $i=\alpha,\beta,\gamma,\text{cosmic}$  corresponds to the dose-rate associated to the radiation  $\alpha$ ,  $\beta$ ,  $\gamma$  and cosmic.

