

A cura di **GIOVANNI VISCO**



CMA4CH



*2nd International
Meeting on
Multivariate Analysis
and Chemometry
for Cultural Heritage
and Environment*

Ventotene, (LT) Italy - 1-4 June 2008



Abstract book



Absolute Dating of Cultural Finds. Properties, Problems and Uncertainties of Luminescence Methods

M. I. Dias

Instituto Tecnológico e Nuclear, Estrada Nacional 10, 2686-953 Sacavém, Portugal

Abstract

Luminescence dating is widely used in geology and archaeology to determine the time since material was last exposed to light, or heated, and is based on metastable charge carriers (electrons and holes) trapped at lattice imperfections. Those charges can be released by stimulating the crystal with heat (thermoluminescence) or light (optically stimulated luminescence), enabling them to recombine with opposite charge carriers, giving place to luminescence emission. The intensity of the luminescence is related to the accumulated dose (equivalent dose) and, therefore, to the time interval (age) during which the mineral has been exposed to ionising radiation. The age is then the ratio equivalent dose/dose rate (D_e/D_r). Problems and uncertainties can derive from incomplete bleaching, resetting or zeroing, so accuracy for D_e and D_r determination is a major task.

Introduction

The use of thermoluminescence (TL) and optical stimulated luminescence (OSL) measurements to accurate determination of the equivalent dose for dating [age = equivalent dose / annual dose-rate (D_e/D_r)] has been widely used as a combined dating technique of sediments and cultural finds. Isolating and controlling some major sources of error is need, such as the type of TL-OSL instability, known as anomalous fading, as well as the effects of uncertainty about the degree of zeroing of the luminescence signal in certain depositional environments and ancient artifacts manufacture conditions. A primary condition of existing luminescence dating is that radiation dose delivered naturally and in the laboratory should give rise to the same trapped charge distribution. Without this equivalence, it is not possible to obtain accurate age estimates. Statistical uncertainties on individual estimates of D_e as low as 1% can now be achieved using the single-aliquot regenerative-dose (SAR) method [1]. For both TL-OSL techniques, accuracy has being tested comparing dates with those obtained with other methods. However accuracy for D_e and D_r determination is an ongoing task [2, 3]. A review about main problems and factors that affect the accuracy of the two components, and hence the age, is presented.

Materials & Methods

Types of luminescence dating techniques: 1) photo-transferred (PTTL); 2) thermal (TL); 3) optically stimulated (OSL): (a) green light (GSL) - feldspar and quartz; (b) infrared (IRSL) - K-Spar; (c) blue light (BSL) - quartz; (d) red light (RSL) - volcanic feldspar and quartz.

The age is determined according with the formula: Age (ka) = D_e (Gy) / D_r (Gy/ka) where D_e is equivalent dose in grays and D_r is the average dose rate over time. D_e can be obtained by: (i) additive dose - usually multiple aliquots; (ii) partial bleach - assumes a mix of grains with varying partial bleaches; (iii) total bleach - assumes complete zero; (iv) regeneration - usually single aliquot or more rarely single grain. D_r comes from two main sources: (i) natural radioactivity, alpha, beta and gamma radiation; (ii) cosmic radiation (usually a small contribution), and can be obtained by neutron activation (U, Th, K, Rb) and gamma spectrometry laboratory and in situ field measurements (U, Th, K).

Results / Discussion

OSL dating is a way of establishing sediment deposition chronology of late Quaternary environments by estimating the time since buried sediment grains were last exposed to day light. It makes use of the capability of some crystal structures, like quartz and feldspars, retains charge in traps for a long period of time. TL dating can be applied to all materials whose manufacturing/use requires high temperature heating ($>500^{\circ}\text{C}$), typically including ceramics, bricks, burnt flints, porcelain, cooked hearths and bronze clay-cores. This method implies that it happened an initial "zeroing of the TL signal", generally corresponding to the making of the ceramic, which erases the previous TL signal by emptying all of the electronic traps. To reduce statistical uncertainties, it is useful to collect a large amount of material to be dated. In addition, to minimize uncertainties besides irradiation from the ceramic, also contribution from the environment must be known.

Key assumptions for both methods: 1) materials have uniform dose rates; 2) moisture content of sample and its environment can be determined; 3) depth, altitude, intensity of cosmic rays on site can be calculated or are known; 4) radiation-induced signal has to be thermally or optically reset by the event to be dated. Rate and completeness of resetting can be reliably obtained; 5) TL-OSL must have been stable during time; any spurious fading of TL-OSL can be measured and compensated for in age calculations; 6) TL-OSL growth characteristics follows a mathematical function. However, the closed system assumption can be violated, principally by draining of electrons from traps by agents, such as light (optical bleaching) and heat, and by the anomalous fading, which occurs in some feldspars. Heterogeneous bleaching of the 'primary' (easy to bleach) and/or 'secondary' (less light sensitive) traps and contamination of quartz by feldspar micro-inclusions is a main cause for scatter. Infrared screening detects the presence of feldspars. Moisture attenuates alpha, beta particles and gamma-rays by factors of 1.50, 1.25 and 1.14 respectively [4]. Relationship of commonly-used normalisation procedures with scatter (for precision), and the nature of dose-response curve (for accuracy) have already been discussed [5]. Loss or gain of K, U or Th (or decay products) also violate assumptions, changing D_r . On sampling, trying to avoid road-cuts, river banks and obvious post-depositional alteration, especially iron/manganese deposition, is essential. Mineralogical analysis is suggested for the detection of existence and degree of possible alteration effects.

Conclusions

TL-OSL problems and uncertainties can derive from incomplete bleaching, resetting or zeroing. Resetting is a heterogeneous process, so to improve identification of well-bleached grains the better dose distribution approach is by using small aliquots (~100 grains) or single grains method. Also the number of aliquots must be considered. Accuracy on estimation of D_e (can arise from calibration of laboratory radiation source, luminescence measurement protocols and validity of assumptions of the process) and D_r (affected by calibration procedure on nuclear counting techniques, radioactive disequilibrium, water content) is a major endeavour on TL-OSL dating.

References

- 1) A.S. Murray, A.G. Wintle, Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol, *Radiat. Meas.*, 32(1) (2000) 57-73
- 2) R.B. Galloway, Luminescence dating: limitations to accuracy attainable, *J. Radioanal. Nucl. Chem.*, 247(3) (2001) 679-683
- 3) A.S. Murray, J.M. Olley, Precision and accuracy in the optically stimulated luminescence dating of sedimentary quartz: a status review, *Geochronometria*, 21, (2002) 1-16
- 4) J. Zimmerman, The radiation induced increase in 100°C thermoluminescence sensitivity of fired quartz, *J. Phys. C: Solid St. Phys.*, 4(18) (1971) 3265-3276
- 5) M. Jain, L. Bøtter-Jensen, A.K. Singhvi, Dose evaluation using multiple-aliquot quartz OSL: test of methods and a new protocol for improved accuracy and precision, *Radiat. Meas.*, 37(1) (2003) 67-80