Confirmation of backscattered beta dose enhancement rates based on single aliquot regeneration (SAR) analysis of quartz sand and silt

Sarah Ingram[1], Stephen Stokes and Richard Bailey

Oxford Luminescence Research Group. School of Geography and the Environment
University of Oxford. Mansfield Road
Oxford OX1 3TB
1: Present address: Lamont-Doherty Earth Observatory, Columbia University, Pallisades, New York, USA

(Received 1st August 2001 ; in final form 6 December 2001)

Introduction

In luminescence dating applications numerous factors contribute to systematic uncertainties in age estimation. Many of these parameters are associated with artificial irradiation within the laboratory, necessary for establishing the luminescence sensitivity of the given dosimeter. In dating applications this is now routinely achieved using calibrated 5-100 mCi $^{90}Sr/^{90}Y$ beta radiation sources, typically attached to automated TL/OSL reader devices (Bøtter-Jensen et al., 2000). While accurate cross-calibration of sources, either to known gamma doses or to other beta sources, is readily achievable, such exercises have previously been hampered in some instances by a lack of precision. This lack of precision is derived from a number of factors including (from Murray (1981) with additions):

- Contrasts in attenuating and backscattering of phosphor materials and substrates used.
- Difficulties imposed in off-plate calibrations (including disc orientation and possible source inhomogeneity).
- The size and shape of the phosphor grains.
- Thermal lag effects in thermoluminescence (TL) measurements.
- Inter-aliquot normalization where multiple aliquot approaches are employed.
- For some phosphors (e.g., quartz) problems associated with thermoluminescence analysis include thermal quenching, low temperature TL peak instability, and dose-dependent and other forms of sensitivity change.

By far the majority of the correction factors relating to dose rate contrasts between varying grain sizes and/or backscatter dose enhancements were derived empirically during early phases in the development of luminescence methods, principally employing CaF_2 as a high sensitivity TL phosphor (e.g., Wintle and Aitken, 1977; Murray, 1981; Aitken, 1985). The combination of optical dating and single aliquot regeneration (SAR) approaches provide radically improved means by which high precision estimates can be obtained for many of these important conversion factors. Somewhat surprisingly, these factors have not yet been systematically re-measured using such approaches.

It is the purpose of this note to describe two simple experiments which sought to re-evaluate the substrate dependent backscatter enhancement of beta dose from a $^{90}Sr/^{90}Y$ source. We tested both sand-sized (125-180 μm) and fine silt (4-11 μm) grain sizes; the sand comprising a gamma irradiated, annealed beach sand, and the fine silt consisting of a well mixed natural fine grained quartz sample (Ingram, 2001). Our results confirm the existing Al/stainless steel ratio (0.86) which was initially estimated as part of a larger experiment using 90-125 μm grains of CaF_2, and four contrasting backscattering materials (perspex, Al, stainless steel Pb) of infinite thickness with respect to the incident beta particles (Murray, 1981).

Samples Used and Sample Preparation

Ideally, either a geologically-stable signal from a naturally occurring phosphor, preferably the sample dosimeter used, or an artificially, uniformly irradiated phosphor should be employed to test for contrasts in dose backscattering from differing sample substrates.

In our study we used both the Equivalent Dose (D_e) from fine-grained (4-11 μm) quartz extracted from a deep sea core collected from the Indian Ocean (Ingram, 2001) and an annealed beach sand (125-180

μm) sample which had been artificially gamma irradiated to test for backscatter dose attenuation. The beach sand was collected from the contemporary upper (high tide) shore face of Eastbourne beach, UK and was annealed at 650°C for 50 minutes. The fine-grained aeolian quartz in the deep sea sediment samples is primarily derived, via long-distance aeolian transportation, from the Arabian Peninsula, and is well mixed at deposition by bioturbation processes, occurring in the upper few cm of the sediment column.

The beach sample was collected and underwent standard separation procedures for the refinement of fine sand (90-150 μm) sized quartz (Stokes, 1994). The fine silt sample was collected from a core repository in film canisters in reduced light conditions. The edge of the sample exposed to light during collection was removed in the laboratory. The gamma irradiation took place at the National Physical Laboratory (Teddington, UK) where the prepared quartz fraction was subjected to a total dose of 7.46 Gy using the Hotspot 800 ^{60}Co gamma source. The sample was then deposited as a monolayer on 4 aluminium and 4 stainless steel discs of a standard size (dia. 9.9 mm, thickness 1 mm).

The laboratory procedures to isolate fine-grained quartz from the bulk samples for OSL dating were adapted from standard practice as outlined by Aitken (1998). The bulk samples were sequentially treated with dilute hydrochloric acid to remove carbonates, hydrogen peroxide to remove organic matter and a dilute sodium oxalate solution to deflocculate the grains. To remove unwanted feldspars the samples were treated with fluorosilicic acid (Rees-Jones, 1995). The 4 –11 μm grain size fraction was isolated from the quartz fraction by settling in acetone according to Stokes' Law of settling velocities. The fine-silt suspension was then deposited, in acetone, onto aluminium and stainless steel discs of a standard size.

The purity of both samples was tested by infra-red stimulation – no significant IRSL above background was observed.

Single Aliquot Regeneration (SAR) Procedure

The Equivalent Dose (D_e) of the refined quartz was analysed using the single aliquot regeneration (SAR) protocol (Murray and Wintle, 2000). Measurements were made using a RISO TL/OSL DA-15 reader fitted with a blue (λ= 470 nm) diode array, and a calibrated ^{90}Sr/^{90}Y beta radioactive source and Electron Tubes type 9235QA photomultiplier. The natural signal was measured as initial (0-1s) luminescence intensity from which background levels (measured after 11-15 seconds exposure) was subtracted. The same aliquot was irradiated with successively increasing laboratory radiation doses and the regenerated OSL signals measured as for the natural to generate a 'growth curve'. A pre-heat of 260C for 10 seconds was employed prior to all measurements. Sensitivity changes induced by multiple irradiations were monitored and corrected via the measurement of OSL resulting from a test dose (10 Gy) and pre-heat (quick heat to 200C, no hold) in the SAR analysis. The D_e required to produce the natural OSL signal was interpolated from the growth curve. The robustness of the SAR analysis was tested in all cases via the re-dosing and measurement of the initial regeneration dose at the end of the experiment, producing a so-called 'recycling ratio'(Murray and Wintle, 2000).

Results

Our analysis has confirmed the efficiency and precision of the SAR approach (Tables 1 & 2, Figure 1). In all cases the recycling rations fell within 10% of 1.0. Our study confirms the great potential of the SAR procedure for reducing uncertainties associated with some conversion factors and ratios crucial to accurate luminescence ages. Grouping D_e estimates into averages for both aluminium and stainless steel, and sand-sized (Table 1) and fine silt-sized (Table 2) dosimeter fractions and calculating standard errors for those mean values results in substrate backscattering ratios which are known to better than 3%.

The resulting stainless steel to aluminium D_e ratios are 0.82±0.01 and 0.84±0.03 for sand and fine silt size fractions. These respectively correspond to backscattering ratios of 1.22±0.01 and 1.19±0.04, which are the same within errors.

The result for the sand size fraction is identical to the value calculated by Murray (1981) using thermoluminescence emissions from calcium fluorite, though in that study no uncertainty was quoted for the ratio.

The ratio for the fine silt sized fraction is statistically the same as previous estimates for fine grains using thermoluminescence (Doreen Stoneham, unpublished data).

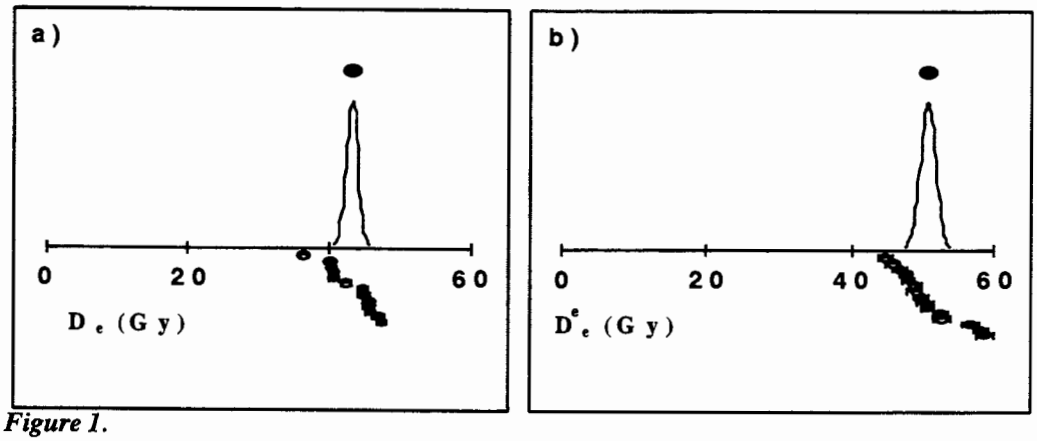

Figure 1.
Examples of SAR D_e populations for deep sea core samples 70KL 85-88
a) stainless steel substrate, b) aluminium substrate.

Aliquot	Substrate	
	Aluminium	Stainless Steel
1	8.78 ±0.01	7.26 ±0.03
2	8.78 ±0.02	7.22 ±0.04
3	8.80 ±0.01	7.37 ±0.02
4	9.21 ±0.03	7.38 ±0.02
Mean	8.89 ±0.11	7.31 ±0.04
Ratio	SS/Al	0.82 ±0.01

Table 1. *Equivalent Dose (Gy†) and summary ratio data for gamma irradiated annealed sand-sized quartz.*

Aliquot	Substrate	
	Aluminium	Stainless steel
1	58.8 ±1.3	44.8 ±0.7
2	66.4 ±1.7	46.9 ±0.8
3	46.7 ±1.1	47.2 ±0.7
4	52.6 ±1.2	36.5 ±0.6
5	50.3 ±1.2	48.6 ±1.0
6	52.5 ±1.0	40.0 ±0.5
7	48.9 ±1.1	44.5 ±0.7
8	50.5 ±0.8	45.5 ±0.7
9	48.0 ±0.7	42.2 ±0.7
10	49.8 ±1.0	40.5 ±0.6
11	45.8 ±0.8	45.6 ±0.7
12	47.7 ±0.8	40.4 ±0.5
13	44.7 ±0.9	40.2 ±0.5
14	56.7 ±1.1	45.4 ±0.7
15	48.8 ±0.8	
16	58.2 ±1.2	
Mean	51.7 ±1.4	43.5 ±0.9
Ratio	SS/Al	0.84 ±0.03

Table 2. *Equivalent Dose (Gy†) and summary ratio data for silt-sized quartz from deep sea sediments.*

Summary

Minimizing systematic uncertainties in optical and thermoluminescence dating is essential to make full use of the potential for high precision measurements now available to dating practitioners via single aliquot and single grain equivalent dose determination procedures. These new approaches can also usefully be exploited in reducing errors associated with absolute or relative alpha and beta source calibration, and laboratory irradiation related dose rate and backscattering conversion factors. This study has confirmed the previous backscattering efficiency ratios for coarse grains and confirmed the equivalence of the ratio for fine silt size dating fractions. It has additionally demonstrated that errors in such parameters can easily be reduced considerably (to better than 2-4%). Given that many other important ratios (e.g., coarse/fine dose rate) are likewise based on old techniques and technologies, it would be sensible for the community to systematically re-evaluate all such parameters, with known and low levels of uncertainty.

Acknowledgments

We are grateful to the review comments of Michel Lamothe and the patience of Didier Miallier.

References

Aitken, M.J. (1985). *Thermoluminescence Dating.* Academic Press, 359pp.

Bøtter-Jensen,L., Bulur,E., Duller, G.A.T., Murray, A.S. (2000) Advances in luminescence systems. *Radiation Measurements*, 32, 523-528.

Ingram, S. (2001). *Optical dating of deep sea sediments from the Arabian Sea.* Unpublished BA dissertation, University of Oxford.

Murray (1981). *Environmental radioactivity studies relevant to thermoluminescence dating.* Unpublished DPhil thesis, University of Oxford.

Murray, A.S. and Wintle, A,G, (2000). Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol. *Radiation Measurements*, 32 (1): 57-73.

Rees-Jones, J. (1995). *Optical dating of selected British archaeological sediments*. Unpublished DPhil thesis, University of Oxford.

Stokes, S. (1994). *Optical dating of selected Aeolian sediments from the Southern High Plains, USA*. Unpublished DPhil thesis, University of Oxford.

Wintle, A.G. and Aitken, M.J. (1977). Absorbed dose from a beta source as shown by thermoluminescence dosimetry. *International Journal of Applied Radiation and Isotopes*, 28: 625-627.

Reviewer
Michel Lamothe

Comments

The manuscript fully deserves to be published in Ancient TL. It is a nice and brief demonstration that the development of the SAR approach in luminescence opens a whole range of investigations. Some of this will hopefully concern the early work revolving around calibration procedures. This note might revive our need to initiate a true inter-calibration program between laboratories of radioactive sources, or to check for luminescence efficiency between the different types of irradiation and s.o.