Mediterranean Archaeology and Archaeometry, Vol. 10, No. 4, pp. 9-15

A SURVEY OF SOME NEW APPROACHES IN MAXIMUM AGE LIMIT AND ACCURACY OF LUMINESCENCE APPLICATION TO ARCHAEOLOGICAL CHRONOMETRY

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Received: 12/12/2009
Accepted: 12/05/2010
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INTRODUCTION

In recent years considerable effort has been made to extend the dating limits and accuracies of luminescence dating and its applications to archeological sciences. These include, for example, the use of single grains for dating (Duller 2008: 589-612; Jacobs and Roberts 2007: 210-223), research on athermal fading to explore the use of feldspars (with higher saturation dose) for dating (Huntley 2006: 1359-1365; Lamothe and Auclair 1999: 319-323), the use of red TL for the dating of volcanic ash (Fattahi and Stokes 2003: 647-660; Visocekas and Guérin 2006: 942-947) and direct dating of archeological contexts (Chawla and Singhvi 1989: 416-418; Singhvi *et al.* 1986: 205-207). Use of some of these have provided useful new data on chronometry and have placed luminescence dating on the central stage amongst other chronometric techniques available for the dating archeological sites and several important results on aspects of human dispersal and chronometry have been reported (Anikovich *et al.* 2007: 223-226; Mellars 2006: 796-800; Petraglia *et al.* 2007: 114-116; Roberts 1997: 819-892; Singhvi *et al.* 1998: 23-83).

This contribution outlines some new ideas that offer prospects of developing the luminescence dating technique further and help extend the age range of its applications in archeology. These applications are based on consideration of the manner in which the radiation dose is deposited in natural samples and aim to refine the protocols that are currently used for the estimation of equivalent dose.

KEYWORDS: Red TL, Volcanic ash, Fading, IRSL, Luminescence, Chronometry

DATING THE INTERIOR OF LITHIC IMPLEMENTS

In luminescence dating, quartz has been used widely to provide the burial ages of sediments. The maximum achievable age depends on the dose corresponding to the saturation of the luminescence signal and the lifetime of the stable signal. On the basis of lifetime measurements it has been estimated that the luminescence signal of quartz is stable for 10^8 ka. However, onset of saturation of the luminescence signal at low radiation doses, limits its range of applicability. The saturation of the luminescence signal is typically occurs at ~250Gy (Chawla *et al.* 1998: 53-63). This restricts maximum age obtained from quartz grains to around hundred *ka*, and depends largely upon the absorbed dose rate from the sediment matrix.

In a sediment matrix, the radiation environment comprises alpha, beta and gamma radiations, emitted during the decay of naturally occurring radioactive isotopes of U, Th series, K and ^{87}Rb. The alpha particle range is limited to only few microns, beta has a range of few mm and gamma dose has a range of about 30cm. Under infinite matrix assumption (Aitken 1985), typically for a 100μm grain, the gamma dose is 25-30% of the total dose. In large quartz grains that are generally devoid of internal radioactivity, the distribution of dose inside the grain from the external radiation environment changes with depth due to very different ranges of the alpha, beta and gamma radiations. As a simple approximation, the alpha and beta dose get attenuated with depth and their contribution is negligible at depths few mm. Thus for a quartzite pebble, the interior of the grain receive dose due to only due to gamma radiations. This suggests that in interior region, the dose rate reduces to a third or a fourth of its original value and, in turn implies that the saturation of the luminescence signal in this region would occur over much longer time scales. We explored the feasibility of using the interior of large size quartz grains to extend the age range achievable by luminescence (Chauhan *et al.* 2009: 629-633). In this work, the range of beta particles from different radioisotopes (i.e. ^{40}K, ^{212}Bi, ^{214}Bi and ^{234}Pa) and their relative dose contribution was computed analytically and by using Monte Carlo simulations. Fig. 1 shows the variation of dose inside the quartz grain relative to infinite matrix dose in sediment matrix.

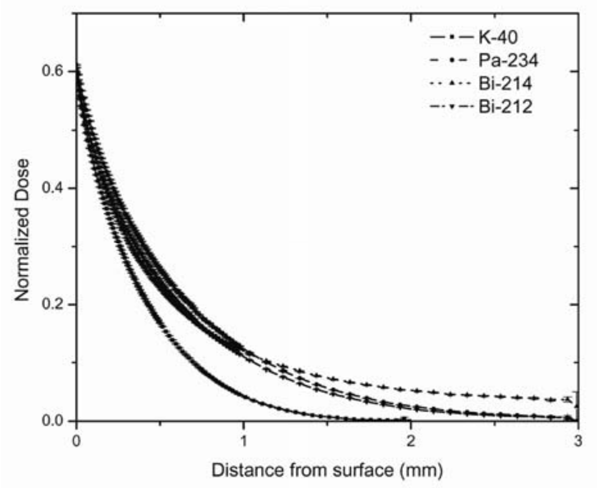

Fig. 1 Attenuation of beta dose inside a quartz grain due to various naturally occurring radionuclides. The radioactivity is considered be external to the grain. Notice that at depths of 2mm most of the beta dose gets attenuated.

These computations comprised,

1. estimation of beta dose profile inside a quartz grain with depth normal to surface using Monte Carlo simulation,
2. development of analytical methodology to compute the annual radiation dose and compare with the simulation results,
3. estimation of the depth at which beta dose inside quartz grain is reduced to a negligible value, and
4. examination of the possible causes that may inhibit the applicability of the new methods.

Result suggest that in large cm size quartz grains, the net beta dose (from all the possible beta emitters) is confined to outer ~2 mm skin. The interior of quartz grain receives radiation dose only due to gamma rays. This is about 30% of the total dose rate which implies that by analyzing the interior of a large size quartz grain, dating of a sample which is three to four times older should be possible.

Based on this, we suggest the possibility of directly dating heated or sun exposed, archaeological artifacts. Microliths and small lithic implements are good candidates to be used for dating by this approach, considering that often these were heated to a high temperature to make it easy to shape them. Further, at times some of the implements are translucent and this fact further ensures that their geological luminescence can he photobleached by daylight during their manufacture, use, disposal and eventual burial. Initial experiments with large (~ cm sized) pieces of quartz indicated that their interior regions were also well bleached partly due to the presence of multiple cleave plains and fractures, that ensured multiple reflections of light in the lattice and hence bleaching. The extraction of the interior of the grains is non-trivial and two possible approaches are, 1) Hydrofluoric acid etching for extended periods, or 2) the imaging of a slice (taken normal to the grain surface) towards the interior and using imaging techniques to measure the spatial distribution of dose through the sample matrix. This can be done using the spatially resolved luminescence measurement system analogous to that initiated by Greilich *et al.* (2005: 645-665). A system for spatially resolved luminescence is being constructed at our laboratory and such cases of the interior of lithic implements will then be examined via the paleodose gradient in a slice through its interior. Also, it should in principle be possible to dates lithic implements with finite internal radioactivity due to the fact that only a part of the beta dose arising from the internal radioactivity is trapped inside the volume considered. In this context we also refer to an early works by Liritzis (1980: 242-251) and Huntley and Prescott (2001: 687-699), who exploited low dose environments for dating over an extended time range.

SENSITIVITY CHANGES DURING NATURAL LUMINESCENCE MEASUREMENTS-THE IMPROVED SAR PROTOCOL

Generally, the Single Aliquot Regeneration (SAR) protocol (Murray and Wintle 2000: 57-73) is used to estimate the dose deposited in a sediment matrix. In this protocol, the sensitivity change during measurement is monitored by a test dose (TD) signal. The natural and regeneration luminescence signals are normalized by the TD luminescence signal. This procedure therefore corrects for sensitivity change during a dose measurement cycle. However, it has often been observed that the intensity of a natural aliquot lies way above the intensity of the regenerated curve and often the regenerated intensity even saturates well below the natural intensity. Such a situation can only arise when the sensitivities (luminescence intensity per unit mass per unit dose) are different, that is, the natural luminescence and the regenerated luminescence signals are recorded with different sensitivities. It was therefore pointed out that a finite change in sensitivity can occur during the pre-heating measurement of natural OSL and consequently the SAR protocol as used now does not fully correct for the sensitivity changes. Laboratory studies indicated that such sensitivity changes during OSL readout and preheat cycles can be significant (up to 30% or more) and can therefore cause significant systematic errors or even make a sample unsuitable for analysis (e.g. the case of natural signal exceeding the saturation limit of the regeneration signal) as shown in Fig. 2.

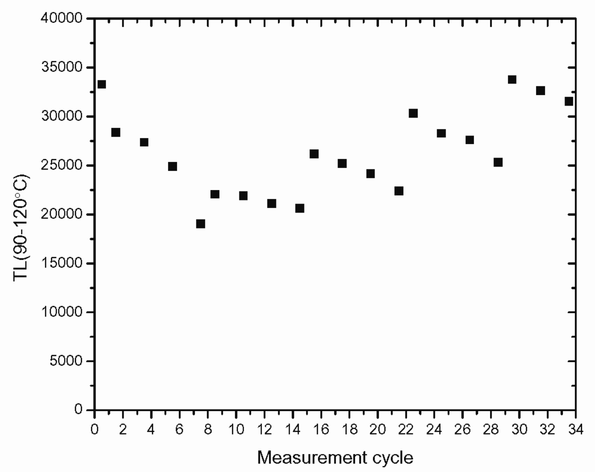

Fig. 2Changes in the sensitivity of 110°C peak during the measurement cycles. The large steps of changes are due to preheats. The SAR protocol corrects for these effectively but the initial change is not included. The area under the peak from 90°C to 120°C was integrated to obtain the intensity of the110°C peak

With this proviso, we recommend additional measurement steps for the analysis of a sample using SAR protocol. This correction procedure assumes that the 110°C quartz OSL peak (Murray and Roberts 1998: 503-515) can be used as a surrogate for the OSL sensitivity of the sample (a correlation that ideally should be established for each sample). In this protocol the ratio of the 110°C TL peak response for TD, recorded before and after the measurement of natural OSL and this ratio is used to correct for the sensitivity change during OSL measurements (Fig. 3).

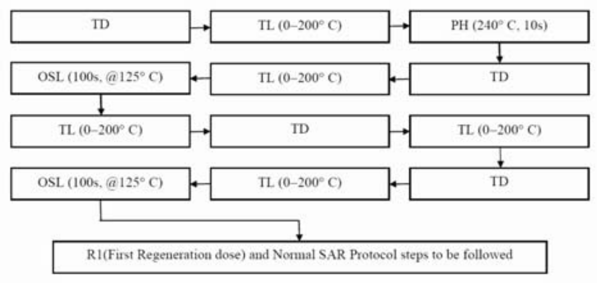

Fig. 3 The protocol for sensitivity correction during the preheat and readout of the natural OSL. The intensities of steps 2 and 5 provide the correction factor NCF.

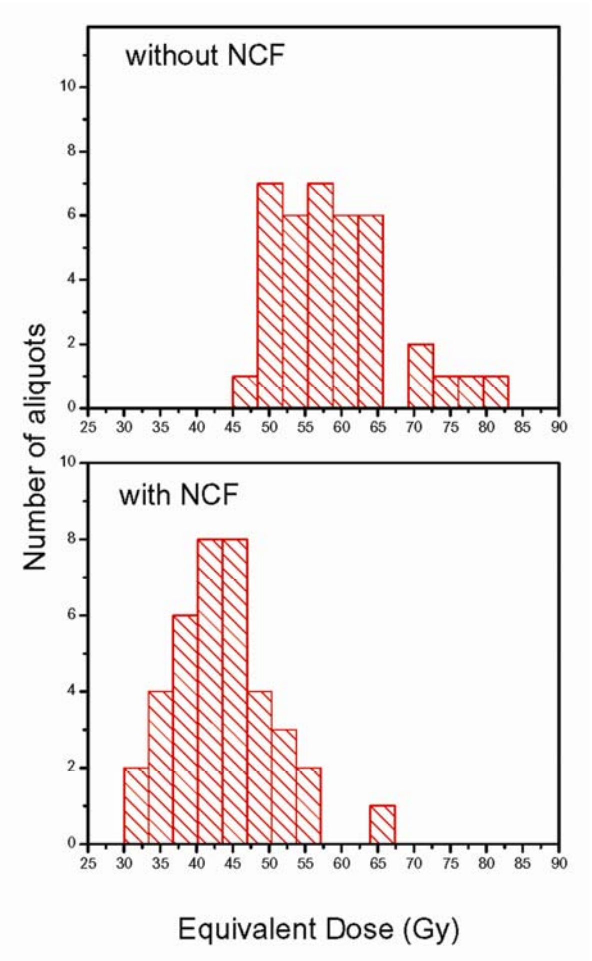

Fig. 4 Changes in the distribution of SAR paleodoses with and without NCF correction. Notice the reduction in the spread and the shift in the mean value.

This ratio is termed the Natural sensitivity Correction Factor (NCF) and is used to ensure that the natural OSL and the regenerated OSL are compared with the same sensitivity. The remainder of the protocol remains similar as for the SAR protocol. Finally the equivalent dose is obtained by subtracting the TD value (given prior to natural OSL measurement) from the obtained dose value. Fig. 4 gives the paleodose distribution of a sample and it is clear that a noteworthy reduction in the spread of paleodoses occurs, due possibly to hitherto unaccounted for changes in the sensitivity in the conventional SAR protocols.

This study suggests, a revisit of the ages using the SAR protocol and a simple test of a comparison of the sensitivity of 110°C glow peak to a test dose before and after the OSL readout, is recommended. Recent experiments with dose distribution from a variety of environments show similar trends. A decrease in scatter in the paleodoses using this protocol was seen in all the samples tested and this suggested that in many situations, well bleached samples may be misunderstood as a poorly bleached sample. This aspect has been dealt with elsewhere (Chauhan and Singhvi 2010: in process).

DATING OF VOLCANIC ASH

In Indian archeology, the presence of archeological implements in central –western India and the dating of associated volcanic ash layer have been extensively debated on methodological grounds. Thus for example, Korisettar et al. (1989: 564-566) first dated the Bori ash by K-Ar method to 1.4 Ma. This was later revised to 600ka using Ar –Ar ages. On the other hand, based on geochemical similarity, this layer was considered to be the Youngest Toba tuff (YTT) dated to 73±4 ka by Chesner *et al.* (1991: 200-203). Recently, Petraglia*, et al.* (2007: 114-116) dated a pre and post Toba archeological sample, in Jwalapuram, Andhra Pradesh, by luminescence dating technique as 77±6 and 74±7 ka respectively. The age bracketing shows that the age of this ash is ~74 Ka. Direct luminescence dating of Toba ash was first attempted by Horn *et al.* (1993: 326-329). The thermoluminescence age of the glasses, derived from Bori Ash, was 23.4±2.4 ka and was interpreted to date the secondary reworking and consequent photobleaching of light-sensitive TL signals in the tuff, post deposition. An alternative explanation was that this ash suffered athermal fading. Not many ash beds have been dated in the Indian contexts.

To resolve some of the basic issues of dating volcanic ash in India, we initiated a systematic evaluation of the ash sample using a variety of approaches. Towards this, a volcanic ash layer sample from different stratigraphic contexts was taken such that the age of the ash bed could be constrained with SAR age on quartz grains from horizons that sandwich the ash layer. Based on field evidences, one of the ash layers was also considered as the Toba ash. Dating of sediments, above and below the ash layer, using SAR protocol for quartz, gave ages of ~52 and ~76 ka suggesting the age of the ash layer to be within the age range of 52-76 ka. These ages however did not permit a clear resolution on the age except that the difference between this and ages on horizons above and below. Direct dating of the ash using TL- Multiple Aliquot Additive Dose method, in blue emission window (430±30 nm) yielded an age of 47±8 ka. This age is twice that obtained by Horn, *et al.* (1993: 326-329), suggesting that their inference of ash bed being reworked/redeposited (and hence not in primary context) was possibly correct. Using a correction procedure reported by Someshwararao (1996), the sensitivity correction procedure for athermal fading indicated a correction factor of 1.6 for the underestimated ages. The protocol is outlined in Fig. 5.

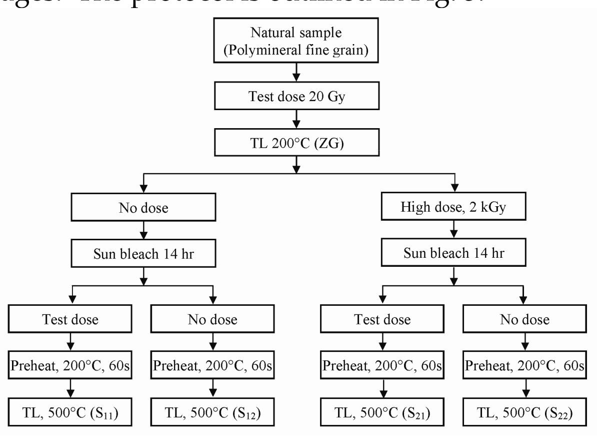

Fig. 5 Correction procedure for fading using a sample-based analysis following Someshwarararao, 1996.

The correction factor is the ratio of sensitivity at the time of burial (S_2=S_{21}-S_{22}) to that as-received sample (S_1 = S_{11}-S_{12}). This resulted in a value of 75±10 ka and is closer to its being the YTT ash. We then attempted the Post IR IRSL SAR protocol, as introduced by Buylaert *et al.* (2009: 560-565) (Fig 6) and the preliminary results, gave an age ~70 ka.

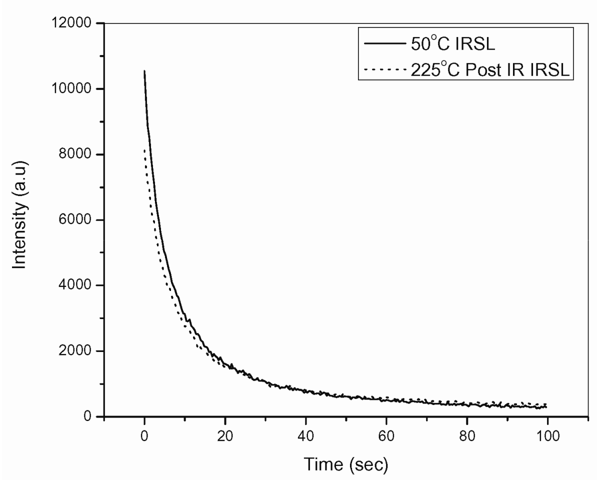

Fig. 6 Post IR IRSL SAR protocol for the dating of volcanic ash (following Bayleaurt et al. 2009).

It thus seems that the both the Sensitivity correction and the post IR IRSL SAR protocol work well. Fig. 7 provides typical optical decay curves obtained using IRSL and post IR IRSL elevated temperature measurements. The question remains that as to why they provide similar results and the physics that leads to this convergence needs further elucidation.

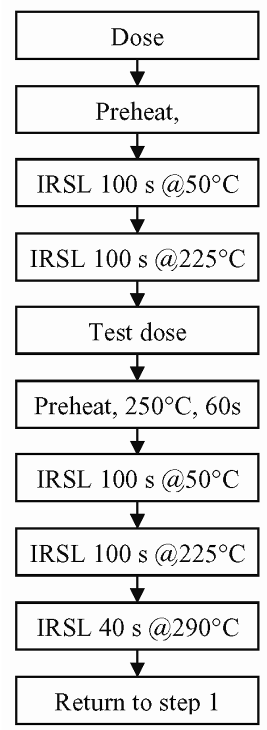

Fig. 7 A typical IRSL and post IR IRSL optical decay curve for a fine grain sample of volcanic ash.

CONCLUSIONS

The conceptual formalism of three methodological aspects of luminescence dating has been presented. Feasibility studies on these,

1. show the possibility of increasing the dating range, three fold,
2. indicate the potential for decreasing the systematic errors that can occur in SAR protocol in OSL, and
3. provide the prospects of improving the accuracy of ages in the case of volcanic ash beds. We sincerely hope that this report will induce other groups to test their efficacy.

ACKNOWLEDGEMENTS

AKS thanks S. Stokes for early collaboration on the sensitivity correction procedures in SAR protocol. We thank Dr. R. Raj and Dr. N. Juyal for help with sample collections and Mr. P. Adhayaru for his sustained help with the instrumentation. We thank the referees for their painstaking comments and for pointing out language flaws. But for them, the manuscript would not have been as palatable.

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