

# Evaluation of SAR procedures for $D_e$ determination using single aliquots of quartz from two archaeological sites in South Africa

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## Abstract

In previous work the conventional SAR procedure has been demonstrated to work well for a large number of samples. Application of the conventional SAR procedure (but with a range of preheat temperatures) to naturally irradiated quartz grains from two different archaeological sites in South Africa showed substantial changes in sensitivity through the measurement cycles; one sample (SIB2) showed no trend in  $D_e$  as a function of preheat temperature, showing that the sensitivity correction was appropriate, whereas for the other sample (ZB4) there was a trend that could be related to the growth curve construction, rather than measurement of the natural signal. However, application of a conventional SAR procedure, with a 260 °C preheat for 10 s and a cut heat at 160 °C, to quartz grains that had been bleached and given a laboratory dose, resulted in underestimation of the dose by  $\geq 10\%$ . The effect of raising the cutheat temperature from 160 °C was investigated using LM-OSL measurements in order to identify OSL components that are present in addition to the fast component for which SAR was developed. The effects of changing the cutheat temperature and adding a high-temperature optical stimulation between cycles in the SAR procedure were measured for dose recovery tests. The effects of these changes on measurement of  $D_e$  values for single aliquots of these two samples are reported. Both modifications improved  $D_e$  recovery, and the study emphasizes the importance of carrying out dose recovery tests.

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## 1. Introduction

Optically stimulated luminescence (OSL) signals from quartz have been used increasingly as the basis of a dating method for sediments, following the first report by [Huntley et al. \(1985\)](#). The single aliquot regenerative (SAR) dose protocol was introduced as a method through which reliable estimates of an unknown dose received by quartz grains in their sedimentary environment could be obtained for a single portion of chemically purified quartz grains

([Murray and Wintle, 2000](#)); the main feature of the SAR protocol is that a correction is made for changes in OSL sensitivity brought about by the thermal treatment used to isolate the appropriate OSL signal, i.e. the preheat. However, there have been a number of reports (e.g. [Stokes et al., 2000](#); [Bailey, 2000](#), [Choi et al., 2003a,b](#)) of problematic behaviour when using the SAR protocol as prescribed by [Murray and Wintle \(2000\)](#), referred to in this paper as the conventional SAR protocol. It thus seems as though some modification to this protocol is required in some instances, e.g. the use of a higher temperature cutheat applied ahead of the OSL measurement used to make the sensitivity correction (e.g. [Bailey, 2000](#); [Choi et al., 2003b](#)) or an additional optical stimulation at high-temperature as advocated by [Murray and Wintle \(2003\)](#).

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In this paper we evaluate the conventional procedure and proposed modifications for two samples of quartz (ZB4 and SIB2) extracted from archaeologically rich occupation layers at two cave sites, Blombos Cave and Sibudu in South Africa. ZB4 from Blombos Cave is of particular importance as it is from the layer in which 39 Middle Stone Age shell beads were found (d'Errico et al., 2005; Henshilwood et al., 2004). Experiments that involved recovering the value of a given laboratory dose are reported using the conventional SAR protocol. Equivalent doses obtained from dating runs made with the conventional SAR protocol (*sensu stricto*, Murray and Wintle, 2000) are presented for different pre-heat temperatures; the changes in OSL sensitivity measured during these runs are reported. Results of dating carried out using both a higher cutheat temperature and an additional optical stimulation at high-temperature are evaluated. The need to modify the procedures for these two samples was investigated by using a linearly increasing stimulation power to look at how the different luminescence components observed were affected by different thermal treatments.

## 2. Single aliquot regenerative (SAR) dose protocol

The SAR protocol involves making repeated OSL measurements ( $L_x$ ) on each aliquot to produce a dose response curve and comparing the natural luminescence intensity ( $L_N$ ) to this curve to determine the equivalent dose ( $D_e$ ), provided that changes in sensitivity are monitored. This SAR protocol uses the OSL response ( $T_x$ ) to a test dose ( $T_D$ ) given after each  $L_x$  measurement relating to the regeneration dose ( $R_x$ ); this monitors for changes in sensitivity. A sensitivity-corrected luminescence response ( $L_N/T_N$  and  $L_x/T_x$ ) is thus determined when the natural ( $L_N$ ) and regenerated ( $L_x$ ) intensities are divided by their respective test dose ( $T_N$  and  $T_x$ ) intensities. The measurement sequence is given in Fig. 1a. In this study, the single aliquots are typically 5 mg of 180–211  $\mu\text{m}$  diameter grains of quartz.

Optical stimulation times to obtain  $L_N$  and  $T_N$  and  $L_x$  and  $T_x$  are typically 40–100 s using blue LEDs at a constant power ( $\sim 6.51$  and  $17 \text{ mW/cm}^2$  for the two Risø readers used for data generation in this paper); this results in continuous wave (CW) OSL signals. The part of the luminescence signals ( $L_x$  and  $T_x$ ) normally used to obtain the  $D_e$  is the initial signal ( $\sim 0.8$  s of the optical decay curve) with a background signal subtracted. The background is a 'late light' background, consisting of the mean of the last 8 s of the respective optical decay curves (either 40 or 100 s), as suggested by Murray and Wintle (2000); the background of the previous  $L_x$  measurement was subtracted from the subsequent  $T_x$  measurement. Fig. 1b shows a typical SAR dose response curve for ZB4, where the corrected natural OSL signal ( $L_N/T_N$ ) is projected onto the regenerated growth curve, for a single aliquot. Construction of such a dose response curve normally requires a range of (or at least 3) regenerative doses ( $R_x$ ) bracketing the expected  $D_e$

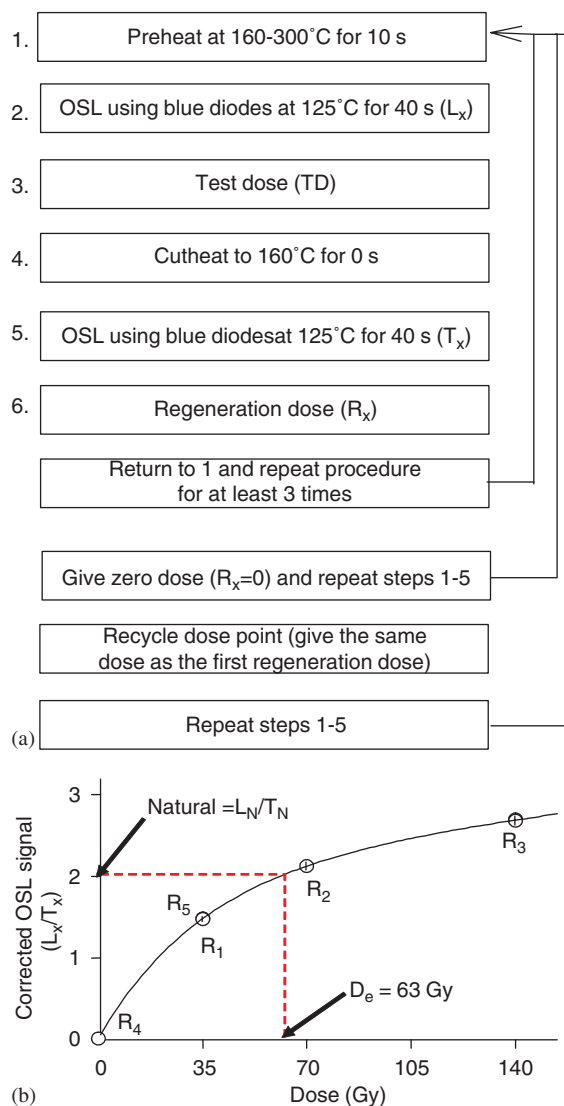


Fig. 1. (a) The conventional SAR protocol (after Murray and Wintle, 2000); (b) a sensitivity-corrected dose response curve for an aliquot of ZB4 obtained using a test dose of 34 Gy and a preheat temperature of 260°C for 10 s.

estimate. It also includes a zero dose ( $R_x=0$ ) point to monitor recuperation and one repeated regenerative dose point at the end of the measurement cycle (e.g.  $R_5 = R_1$ ) to confirm the appropriateness of the sensitivity correction. In addition, an extra measurement was added onto the end of each SAR (dating and dose recovery) run to perform the OSL IR-depletion ratio test of Duller (2003). This procedure was developed to identify the presence of feldspar and is based on the depletion of the OSL signal by prior exposure to infrared illumination. The test dose ( $T_D$ ) is usually a small fraction of the natural dose, though test doses of up to 100%

of the expected  $D_e$  value have been used without affecting the  $D_e$  value (Murray and Wintle, 2000). Experiments on ZB4 showed that changing  $T_D$  had no effect, and a test dose of 34 Gy was used throughout, in order to provide an intensity about half that of the natural dose. For SIB2, a test dose of 6 Gy was used for dating runs and 3.6 Gy for other tests.

### 3. Equivalent dose determination using conventional SAR protocol

SAR dating runs were carried out using a range of preheat temperatures from 160 to 300 °C with this temperature being maintained for 10 s, and using a cutheat temperature of 160 °C. Besides being able to obtain plots of  $D_e$  as a function of preheat temperature, it is also possible to investigate how the sensitivity changes with successive 10 s preheats through each dating run. Murray and Wintle (2000) claim that if one can obtain the same  $D_e$  value when using very different preheat treatments which result in very large sensitivity changes, then the correction within the SAR protocol for luminescence sensitivity changes is effective. It is also possible to see how the different preheat temperatures affect both the dose response curves and the values of  $L_N/T_N$ . From these studies possible problems with the simple interpretation of the two standard tests proposed by Murray and Wintle (2000) (namely the preheat plateau test and the recycling ratio test) can be identified.

#### 3.1. Sensitivity change as a function of preheat temperature

The sensitivity changes that occur through the SAR measurement cycles can be investigated by observing changes in  $T_x$  (Fig. 1a, step 5). Temperature dependency of the sensitivity changes has been shown by some investigators (e.g. Armitage et al., 2000; Bailey, 2000; Jacobs et al., 2003). By using a broad range of preheat temperatures, the degree of sensitivity change can be altered. This can be seen when the test dose response ( $T_x$ ) is plotted as a function of measurement cycle during the construction of a dose response curve using different doses. In Fig. 2 the changes in response of the test dose for different preheat temperatures are shown for ZB4 and SIB2. Values are normalised using the value of  $T_N$ , the test dose response after measurement of the natural OSL ( $L_N$ ). The test doses were 34 and 6 Gy for ZB4 and SIB2, respectively. The value of  $T_x/T_N$  at each preheat temperature is the mean of three aliquots. SIB2 (Fig. 2b) shows behaviour similar to that published by Armitage et al. (2000) where data for lower preheats (160–240 °C) show an initial decrease in sensitivity and for the higher preheats (260–300 °C) an increase. For SIB2, the sensitivity change for 280 °C is 3.5 times that for the 160 °C preheat after 10 measurement cycles. ZB4 (Fig. 2a) shows an initial decrease in sensitivity, followed by an increase for all preheat temperatures (except 260 °C). The degree of increase is related to the preheat temperature. The contrasting responses

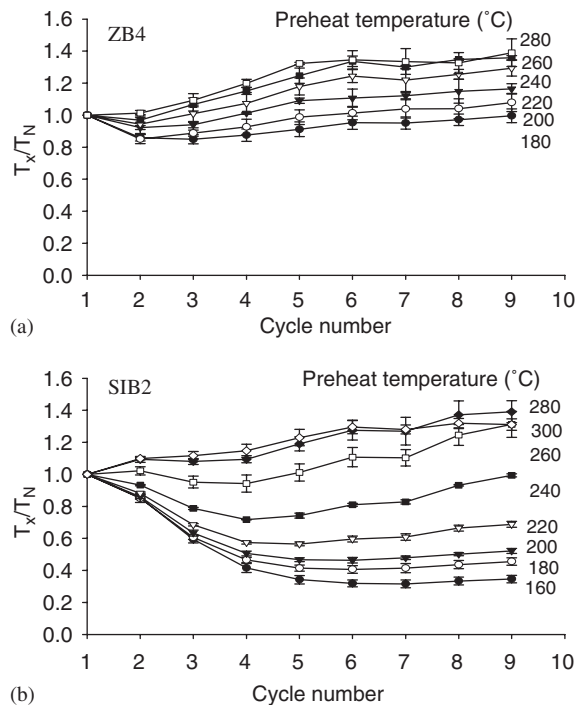


Fig. 2. Ratio of test dose responses ( $T_x/T_N$  as a function of cycle) taken from SAR dating runs on (a) ZB4 (6 preheat temperatures) and (b) SIB2 (8 preheat temperatures).

of these two samples provide excellent scope for testing SAR procedures. If the changes in sensitivity seen in the first five cycles cannot be monitored appropriately, then the dose response curves will be wrongly determined.

#### 3.2. $D_e$ as a function of preheat temperature

Plots of  $D_e$  versus preheat temperature are given in Fig. 3. ZB4 shows large scatter in  $D_e$  for the two lowest preheat temperatures (180 and 200 °C in Fig. 3a). The scatter in the rest of the measurements is considerable ( $\sim 40$  Gy) and it is difficult to assess whether there is any trend in the data. The three aliquots at the 280 °C preheat temperature are tightly grouped and it is worthwhile noting that these aliquots show nearly no sensitivity change at the beginning of the measurement cycle (Fig. 2a). Using the  $L_x/T_x$  measurement of the zero dose point ( $R_4$  in Fig. 1b) gave recuperation doses of less than  $\sim 0.9$  Gy and thus this is not the cause of scatter in the measured  $D_e$  values (Fig. 3a).

The results for SIB2 indicate the effectiveness of the sensitivity correction; the  $D_e$  values of 24 aliquots show no systematic trend with temperature from 160–300 °C, but do show scatter of  $\sim 20$  Gy (Fig. 3b). Measurement of the zero dose point gives recuperation doses of less than  $\sim 0.2$  Gy. SIB2 illustrates that even with large changes in sensitivity (as seen in Fig. 2b) reproducible  $D_e$  values can be obtained

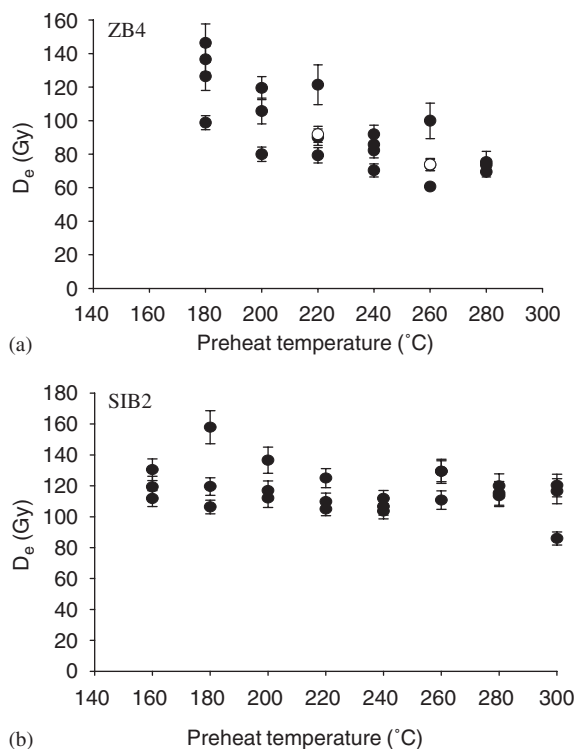


Fig. 3.  $D_e$  values as a function of preheat temperature for (a) ZB4 and (b) SIB2. The mean sensitivity changes at each temperature were given in Fig. 2. For the meaning of the open circles, see text and Fig. 4c.

using the conventional SAR protocol across a wide range of preheat temperatures.

### 3.3. Dose response curves as a function of preheat temperature

Murray and Wintle (2000) demonstrated that the choice of preheat is relatively unimportant provided that changes in sensitivity are corrected for appropriately. When this is the case, Roberts and Duller (2004) have shown that the sensitivity-corrected dose response curves over the preheat range 160–260 °C overlie each other when account is taken of any difference in the test dose; they found this to be the case for single aliquots of coarse-grain quartz from within the same sample, and also between different samples. For preheat temperatures above 260 °C, the normalised OSL signal will drop, due to thermal erosion of trapped charge. The consistency observed at temperatures from 160 to 260 °C led them to propose a standardised growth curve for regenerative doses up to 125 Gy for coarse-grained quartz.

Fig. 4a and b shows the standardised dose response curves from one aliquot of ZB4 and SIB2 at each preheat temperature. The sensitivity-corrected luminescence signal ( $L_x/T_x$ ) was adjusted for the size of the test dose to produce the

standardised signal ( $[L_x/T_x]T_D$ ). However, direct comparison as suggested by Roberts and Duller (2004) was not possible because of the use of a high test dose (34 Gy) for ZB4 (i.e. one that is not in the linear region of the dose response curve). The dose response curves in Fig. 4a and b do not show the entire dose ranges that were measured, but only those doses that generate standardised values that are about equal to, and less than, the standardised natural ( $[L_N/T_N]T_D$  shown on the left axis); only two dose points below 100 Gy are shown for sample ZB4. For SIB2 (Fig. 4b), the patterns are very similar to those presented by Roberts and Duller (2004) where samples normally show a decrease in  $[L_x/T_x]T_D$  at higher preheat temperatures due to the thermal erosion of trapped charge. The data for ZB4 in Fig. 4a show an unusual pattern with an increase in  $[L_x/T_x]T_D$  for preheats above 220 °C (Fig. 4a).

### 3.4. Values of $L_N/T_N$

In Fig. 5 the  $L_N/T_N$  ratios for those aliquots of ZB4 whose dose response curves are presented in Fig. 4a are shown. The constancy of  $L_N/T_N$  suggests that there is no thermally unstable signal in the  $L_N$  signal that would be emptied by the 10 s preheat, i.e. from 180 to 280 °C. The lack of an increase in  $L_N/T_N$  with increasing preheats, together with the decrease in  $D_e$  with increase in preheat temperature (Fig. 3a), can be linked to changes in the sensitivity-corrected dose response curves (Fig. 4a).

### 3.5. Assessing the reproducibility of the growth curve at high doses

One of the tests proposed for the SAR protocol by Murray and Wintle (2000) is the recycling ratio test. This involves repeating a dose point within the SAR measurement sequence. The first corrected luminescence ratio ( $L_x/T_x$ ) is obtained at the beginning of the measurement sequence when sensitivity change is most rapid and the second ratio ( $L_y/T_y$ ) is obtained at the end of the measurement sequence to encompass the full range of sensitivities encountered. If sensitivity change has been corrected for appropriately, then the ratios of these corrected intensities (the recycling ratio, RR1) will be consistent with unity.

The recycling ratios for 24 aliquots from ZB4 and SIB2 are shown in Fig. 4c and d as a function of preheat temperature. The recycling ratios for ZB4 show some scatter, except for the aliquots preheated at 280 °C. The recycling ratios for two of the aliquots lie outside of the range 0.90–1.10 proposed by Murray and Wintle (2000); these two appear to have no relationship to the scatter in  $D_e$  values (shown as open circles in Fig. 3a). The recycling ratios for SIB2 are tightly clustered but show some trend with preheat temperature. Although all ratios fall within the range 0.90–1.10, those for the higher and lower temperatures are not consistent with unity within errors.

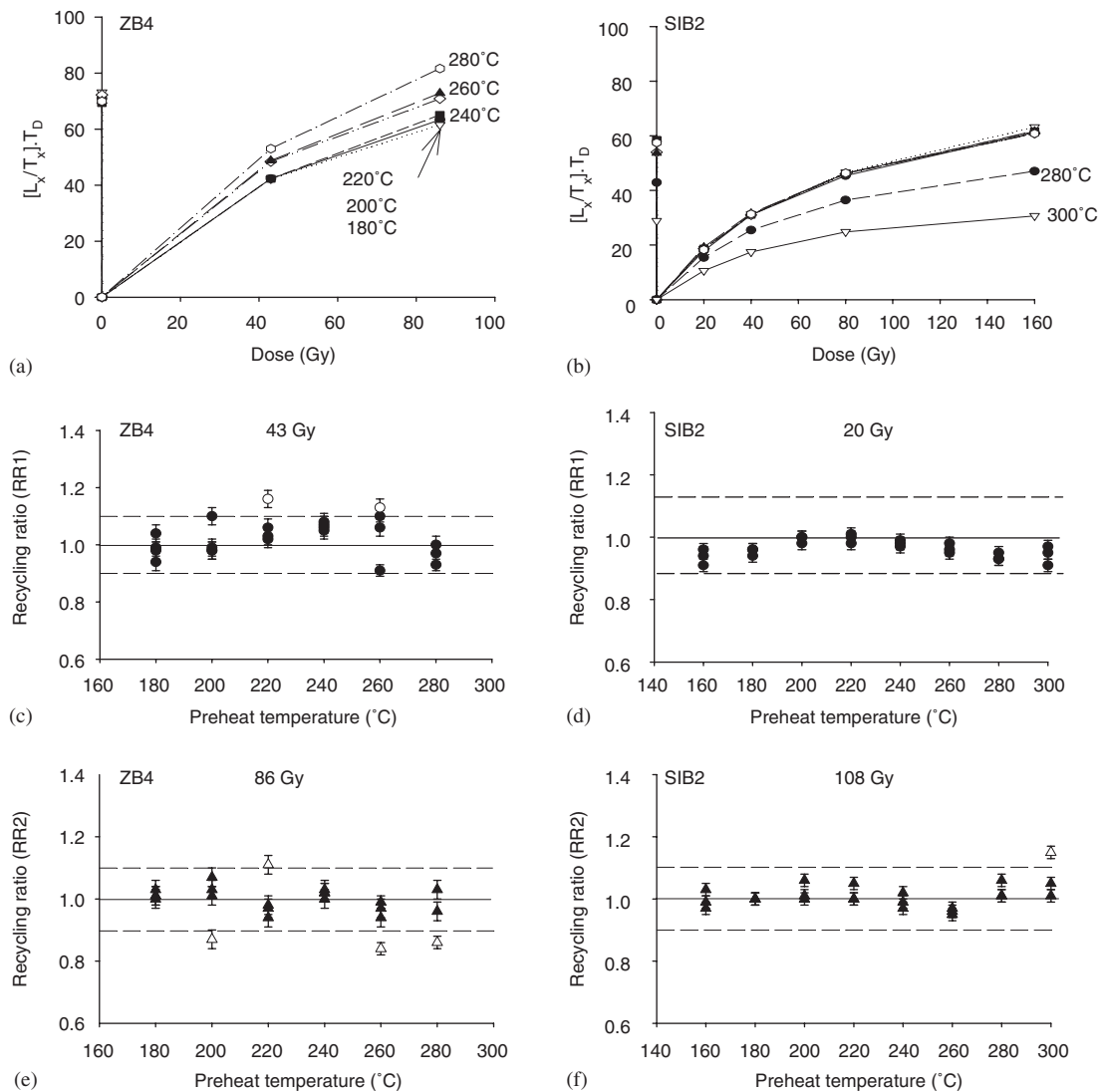


Fig. 4. Standardised dose response curves (one aliquot for each preheat temperature) for (a) ZB4 and (b) SIB2. Recycling ratios (RR1) (c) and (d) and (RR2) (e) and (f) as a function of preheat temperature for all aliquots used to obtain  $D_e$  values in Fig. 3. Aliquots that were deemed to have failed each recycling ratio test are indicated with open symbols.

Since the first measurement will represent the point in time when sensitivity changes are most rapid during the measurement cycle, the first recycling ratio (RR1), as defined above, tests for the accuracy of the sensitivity correction. The drawback of this measurement, in samples whose dose response curves are non-linear at higher doses (Fig. 4a and b), is that this value of RR1 was measured for the lowest regeneration dose (43 Gy for ZB4 and 20 Gy for SIB2) and does not assess the ability of the aliquot to recover a dose near saturation. A second recycling ratio (RR2) was therefore added to the measurement cycle, repeating a high dose point, equal to or higher than the expected  $D_e$  (86 Gy for

ZB4 and 108 Gy for SIB2). As seen in Fig. 2, most sensitivity changes would have taken place already and the second recycling ratio should thus be more consistent with unity. Also, when RR1 is consistent with unity, one would expect that RR2 would show the same value.

Fig. 4e and f presents the RR2 values for ZB4 and SIB2. There are four RR2 values from ZB4 that fall outside the range 0.90–1.10. For SIB2 only a single aliquot with a 300 °C preheat temperature failed the test. For ZB4, rejection of these four aliquots led to a reduction in the amount of scatter in  $D_e$  values at preheat temperatures between 220 and 280 °C; for SIB2, it accounted for the one outlier

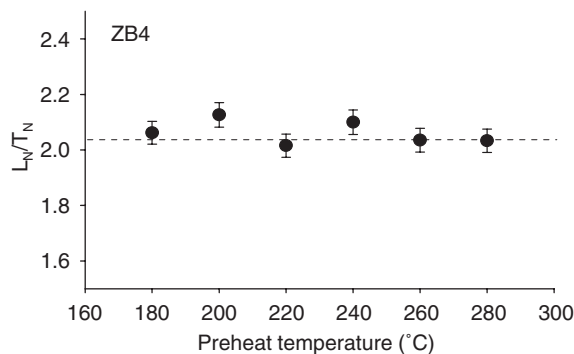


Fig. 5. Mean normalised sensitivity-corrected natural OSL response ( $L_N/T_N$ ) obtained as a function of preheat temperature for aliquots of ZB4 having related data presented in Fig. 2a, 3a, 4c and e.

at 300 °C (Fig. 3b). The application of this recycling ratio (obtained with a high dose, comparable in size with the natural dose) as a rejection criterion for samples with non-linear dose response curves is important. However, even this broader range of tests is insufficient to fully evaluate the appropriateness of the SAR protocol. This is discussed below.

#### 4. Testing the appropriateness of the conventional SAR protocol

The most important method of evaluating whether a laboratory procedure is appropriate is a dose recovery test (e.g. Murray and Roberts, 1998; Roberts et al., 1999). This involves zeroing of the natural OSL signal using a light source, followed by application of a known laboratory dose to the optically zeroed aliquots. The procedure is then applied to see if the value of that dose can be determined. The dose recovery test has been proposed as a useful independent experimental verification procedure for the general behaviour of any dating protocol (Wallinga et al., 2000; Fuchs and Lang, 2001; Murray and Wintle, 2003). If this laboratory experiment does not work, it is unlikely that the measurement of the equivalent dose of a natural sample will be accurate. An accurate result from a dose recovery test should thus be a minimum requirement, since it will display at least the appropriateness of the protocol for the laboratory measurements that make up the sensitivity-corrected dose response curve.

All dose recovery tests in this study used blue light emitting diodes (LEDs) to zero the natural OSL. Murray and Wintle (2003) and Choi et al. (2003a) suggested that two optical stimulations for 1 ks should be performed, both at room temperature, with a 10 ks room storage in between; this is to allow for the thermal decay of any possible charge that may have been transferred into the 110 °C TL trap. A faster option is a 40 s optical stimulation at 125 °C, assuming that this will also prevent charge transfer into the 110 °C

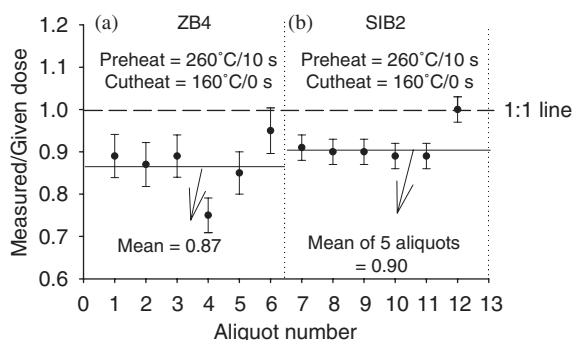


Fig. 6. Measured/given dose ratios obtained for dose recovery experiments on ZB4 and SIB2 using the conventional SAR procedure of Fig. 1a and including data for the lower recycle point (RR1).

TL trap. Dose recovery tests were performed using both types of optical stimulation. The mean recovered doses from six aliquots, obtained using the two different zeroing procedures, were indistinguishable and all further dose recovery tests have been performed using 40 s optical stimulation at 125 °C for bleaching.

Fig. 6 shows the measured/given dose ratios obtained using the conventional SAR protocol for ZB4 and SIB2 for given doses of 68 and 128 Gy, respectively. Data presented include six aliquots for each sample measured with a single preheat temperature (260 °C for 10 s) and a cutheat of 160 °C. An average value of 0.87, i.e. an underestimation of 13%, was found for ZB4 and 0.90 (10%) for SIB2 (one of the six aliquots is consistent with unity ( $1.00 \pm 0.04$ ) and is not included in the ratio of 0.90). The error on each individual dose estimate is < 5%. The underestimation of 10% or more indicates that the conventional SAR protocol is likely to be inappropriate for determining  $D_e$  for these two samples.

#### 5. Use of LM-OSL as a diagnostic tool

A key assumption of the SAR protocol is that the electron traps that are being investigated are the same for all optical stimulations (for test dose and regenerative doses) during a measurement cycle, or where different traps are stimulated that these sensitise in the same way. This holds true for samples used in the development of the SAR protocol; the OSL signals from these samples were dominated by a fast component (Wintle and Murray, 1999). The contributions to the OSL signal from different components in a specific sample can be investigated by observing the linearly modulated OSL (LM-OSL) (e.g. Singarayer and Bailey, 2003; Jain et al., 2003). LM-OSL records the luminescence emission while ramping the power of the light source (Bulur, 1996). The advantage of LM-OSL is that it provides greater discrimination between the various components. The components with different optical cross-sections appear as different peaks in plots of luminescence intensity against

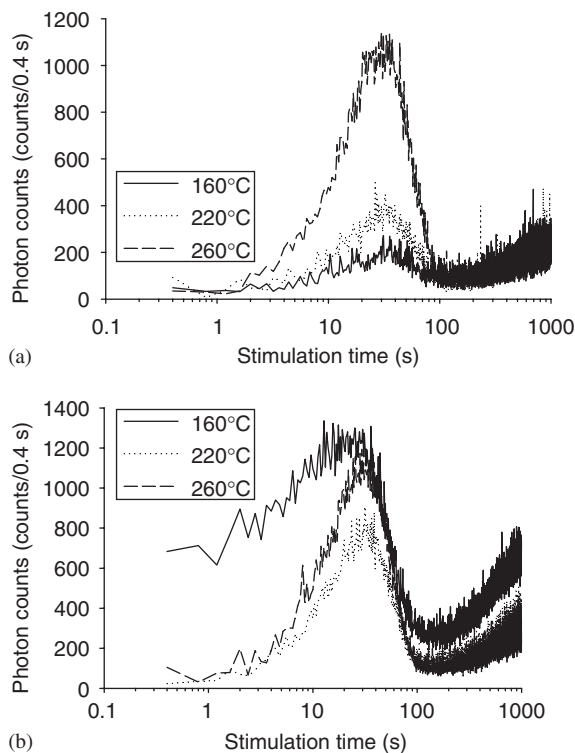


Fig. 7. LM-OSL measured for three aliquots containing (a) their natural dose and pretreated using cutheats of 160 or 220 °C or a preheat for 10 s at 260 °C; (b) same aliquots given 100 Gy and having the same thermal pretreatment as in (a).

stimulation time, when stimulation power is linearly increased.

Singarayer and Bailey (2003) suggest that there are at least five OSL components that commonly appear in quartz. They refer to these components as the fast, medium, slow 1 (S1), slow 2 (S2) and slow 3 (S3) components. Jain et al. (2003) report finding up to seven different OSL components for measurements made at 125 °C. Using LM-OSL Jain et al. (2003) demonstrated that different degrees of sensitivity change in different OSL components could affect the sensitivity correction process and give rise to poor recycling ratios.

### 5.1. LM-OSL measurements on unannealed aliquots

For ZB4, natural aliquots were measured after different thermal treatments had been applied, namely cutheating to 160 and 220 °C and preheating for 10 s at 260 °C; these are the conditions encountered in the SAR protocols used in this study. The LM-OSL was measured at 125 °C, whilst ramping the intensity of the blue LEDs linearly from 0% to 100% power over 1000 s. The datasets for the three different thermal treatments are shown in Fig. 7a. It can be seen that

the signals are dominated by the fast component and the sensitivity after preheating at 260 °C for 10 s was higher than that obtained after cutheating to the lower temperatures.

Each aliquot was then given a dose of 100 Gy, given the same preheat and the LM-OSL was measured under the same conditions (Fig. 7b). From these curves it can be seen that the sensitivities of each aliquot have been increased by the combination of their previous thermal treatment and by holding at 125 °C for 1000 s during the LM-OSL measurement. In addition, for the lower temperature cutheat (160 °C), there is an additional signal in the first 20 s of measurement which results in the LM-OSL signal not starting with zero counts. Singarayer (2002) observed a similar non-zero origin of the LM-OSL curve for one of her samples (SL203) when stimulated at a temperature of 200 °C. Choi et al. (2003b) also noted the presence of such a component. Singarayer (2002) proposed that it could be due to a luminescence component that was still present after the low preheating temperature and that decayed over a relevant time period during the LM-OSL measurement. Such a source of light is the decay of thermally stimulated luminescence (TL) as a function of time at a constant temperature.

### 5.2. Choosing the cutheat temperature

Having established the behaviour following natural irradiation (Fig. 7a) and a subsequent irradiation of 100 Gy for ZB4, further experiments were carried out on aliquots of both ZB4 and SIB2 that had been heated to 500 °C in order to increase the sensitivity of the response and thus improve reproducibility. The aim of these measurements was to investigate potential reasons why the dose recovery experiment was unsuccessful for ZB4 and SIB2. Heating to 500 °C had the additional advantage of reducing all OSL components to undetectable levels (Singarayer, 2002). Following heating to 500 °C, all aliquots were given a laboratory dose (37 Gy), followed by a preheat temperature of 260 °C for 10 s. Following the preheat, and after allowing the sample to cool down, the LM-OSL signal was measured at 125 °C, whilst ramping the intensity of the blue LEDs linearly from 0% to 100% power over 1000 s. These signals are representative of the  $L_x$  measurement in the SAR protocol (labelled 260 °C in Fig. 8a and b). These signals are very similar to those published previously for quartz (e.g. Bøtter-Jensen et al., 2000; Bulur et al., 2000; Choi et al., 2003a; Kuhns et al., 2000; Larsen et al., 2000). There is a well-defined peak at 23 s and this is believed to be the same as component A of Bulur et al. (2000), better known as the fast component (Bailey et al., 1997; Singarayer and Bailey, 2003). It is concluded that the fast component dominates the signals for these samples.

To obtain signals representative of the  $T_x$  measurement in the SAR protocol, the aliquots were again heated to 500 °C and irradiated (37 Gy). This time a cutheat temperature of 160 °C for 0 s was used and the LM-OSL signal measured as before for 1000 s (labelled as 160 °C in Fig. 8a and b).

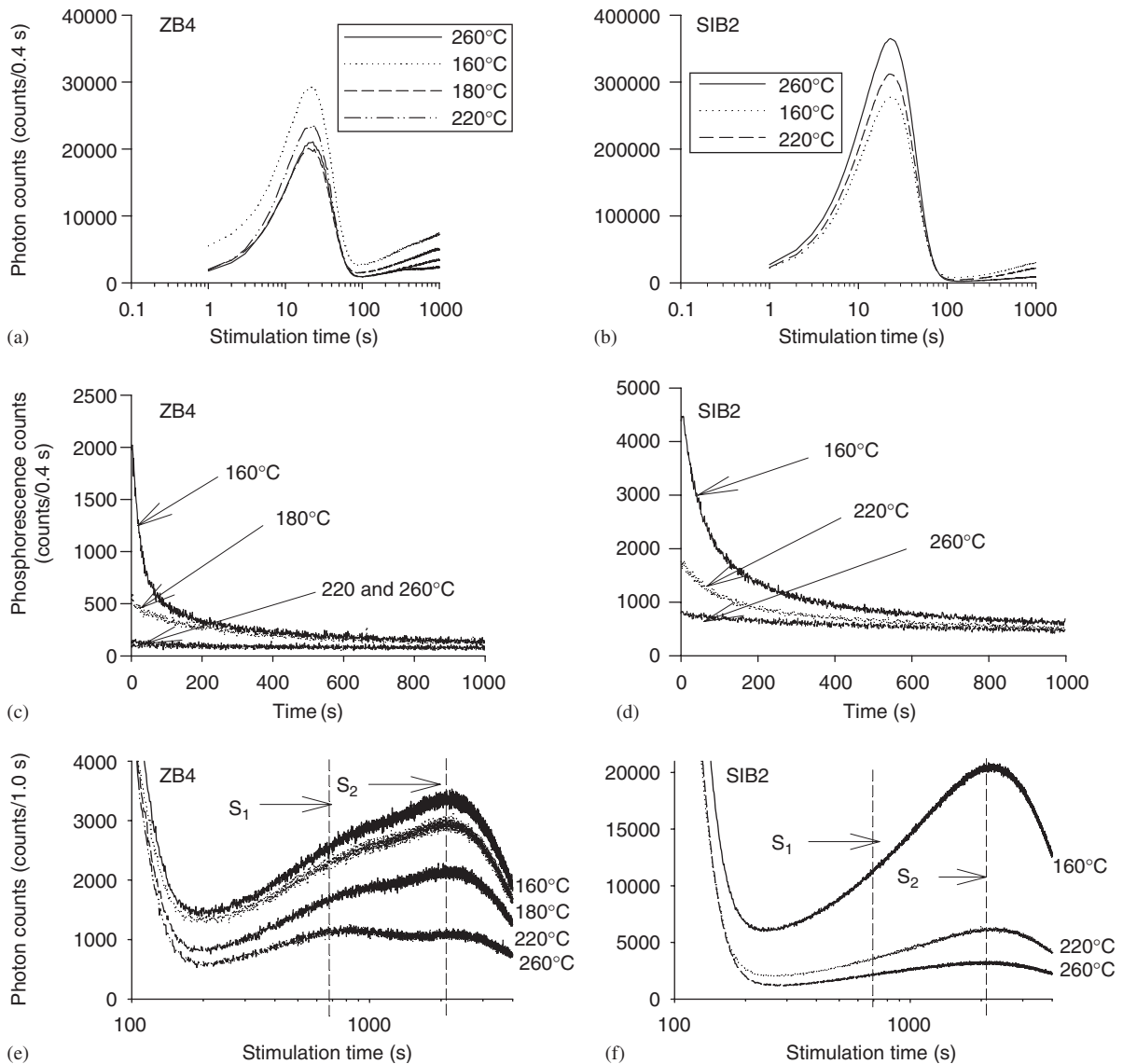


Fig. 8. LM-OSL curves at 125 °C (a) for ZB4 and (b) for SIB2 following different thermal treatments; in each figure, 260 °C refers to a preheat for 10 s and the other temperatures are cutheat temperatures. (c) and (d) are the phosphorescence decay curves obtained when the sample is held at 125 °C for the same length of time, but without optical stimulation. (e) and (f) are LM-OSL curves at 125 °C following the same thermal treatments as for (a) and (b), but for a longer stimulation time to enable the slow components S1 and S2 to be identified.

For ZB4, the curves representative of the  $L_x$  and  $T_x$  measurements in the SAR protocol (labelled as 260 and 160 °C in Fig. 8a) appear to be different, and once again the initial signal intensity (at  $t = 1$  s) for  $T_x$  for the 160 °C cutheat is substantially above background. For ZB4, two additional LM-OSL measurements were made following application of cutheats at 180 and 220 °C. For SIB2 only one additional measurement was made following a cutheat at 220 °C. These curves are also plotted in Fig. 8a and b. The LM-OSL measurements following the 180 and 220 °C cutheats are

more similar to the measurements that followed the preheat of 260 °C for 10 s. These data support the use of cutheat temperatures above that originally proposed by Murray and Wintle (2000), though still not so high as to cause additional sensitivity change.

When luminescence is observed at 125 °C (Fig. 8c and d) after the same thermal treatment as in Fig. 8a and b, but without optical stimulation, a relatively large signal is seen after the 160 °C cutheat. This isothermal signal may interfere in the SAR measurement protocol for derivation of  $D_e$  if



the fast component is not dominating the signal. However, it is possible that variability in this isothermal signal may explain the scatter in  $D_e$  observed for ZB4 when a cutheat temperature of 160 °C was used in combination with a low preheat temperature (e.g. 180–200 °C) (see Fig. 3a). For this reason, a higher cutheat temperature should be used when dating this sample.

### 5.3. The slower components

The signals observed for stimulation times from 100 to 1000 s (Fig. 8a and b) may be associated with the slow components of Singarayer and Bailey (2003) and Jain et al. (2003); in this paper the notation S1 and S2 of Singarayer and Bailey (2003) is used. For the selection of preheat and cutheat temperatures, it is important to assess the presence, and extent, of the thermally unstable S2 component. The S1 component has been shown to be the most thermally stable of the OSL components and it has dose saturation values similar to that of the fast component (Singarayer and Bailey, 2003). Presence of the S1 component is therefore not considered to be problematic when the conventional SAR measurement protocol is used; also, the late light background subtraction of the previous  $L_x$  measurement for both  $L_x$  and  $T_x$  in a measurement cycle reduces the effect of the S1 component. However, Singarayer (2002) has demonstrated that the S2 component appears to correspond to a trap that would give rise to TL at 280 °C. Jain et al. (2003) calculated that only 50% of its initial intensity is left after heating to 260 °C and it can be completely removed by heating to 340 °C. The S2 component is not sufficiently stable to be used for dating on Quaternary timescales and, if not accounted for, the presence of this component may lead to dose underestimation.

In order to assess the presence and relative magnitude of the S2 component, the same aliquots as shown in Fig. 8a and b were remeasured, but this time for 4000 s ramping the blue LEDs from 0% to 100% power. The results are shown in Fig. 8e and f where the  $x$ -axes are reduced to focus on only the later part of the measurements (100–4000 s) to show in more detail that part where Singarayer and Bailey (2003) found their slow components, S1 and S2. The approximate peak positions of the S1 and S2 components are indicated on the graphs with broken lines. Comparing measurements made following different cutheats, as the cutheat temperature increases both samples show a preferential reduction in the S2 component compared to S1. This is consistent with the findings of Singarayer and Bailey (2003) that the S2 signal is more thermally unstable than the other signals.

## 6. Dose recovery tests for changed SAR procedures

### 6.1. Application of an increased cutheat temperature

Based on the results presented in Section 5.2, a higher cutheat temperature of 220 °C was applied to six aliquots

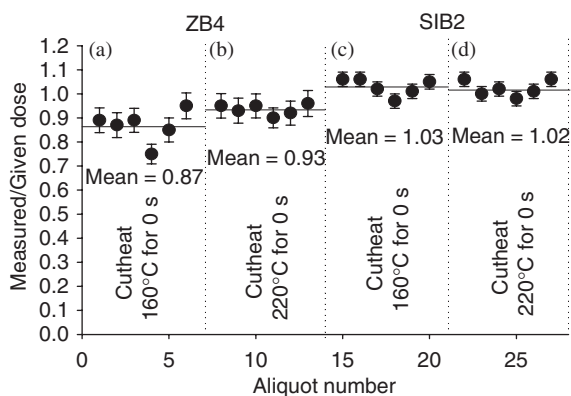


Fig. 9. Measured/given dose ratios obtained for dose recovery experiments on ZB4 (a and b) and SIB2 (c and d) using a 10 s preheat at 260 °C. The dose response curves were fitted including both the first (RR1) and the second (RR2) repeat points. Measurement conditions are given in the figure, together with the mean ratio for each set of six aliquots.

for both ZB4 and SIB2; a similar cutheat was recommended by Bailey (2000) and Choi et al. (2003b) for their samples. Fig. 9b shows the dose recovery results for ZB4 using the 220 °C cutheat, and including the higher repeated dose point value to fit the dose response curve. The results can be compared with those obtained using a 160 °C cutheat (Fig. 9a) and show an improvement in the measured/given ratio from  $0.87 \pm 0.07$  to  $0.93 \pm 0.02$ . Also, the scatter observed when applying the lower cutheat is reduced when using the 220 °C cutheat. The ratios for SIB2 remained about the same,  $1.02 \pm 0.04$  (Fig. 9d) as compared to  $1.03 \pm 0.04$  (Fig. 9c). The higher cutheat is therefore improving the dose recovery results for ZB4, but the measured dose is still underestimated.

### 6.2. Application of a high-temperature optical stimulation

Murray and Wintle (2003) suggested an additional optical stimulation at a temperature close to, or higher than the preheat temperature, directly after the measurement of every test dose ( $T_x$ ) during the measurement procedure. This extra thermal and optical treatment was designed to reduce recuperation occurring between repeated SAR cycles. The SAR protocol of Fig. 1a was modified by adding a 40 s optical stimulation with blue LEDs at 280 °C and by using a higher cutheat temperature (220 °C). The dose response curves were fitted using all the  $L_x/T_x$  measurements including the two repeated dose points that are being used to obtain the recycling ratios (RR1 and RR2). Both sets of recycling ratios were consistent with unity and incorporation of this data improved the precision with which the growth curve was defined. The results for ZB4 are presented in Fig. 10 c and d when the high-temperature optical stimulation is

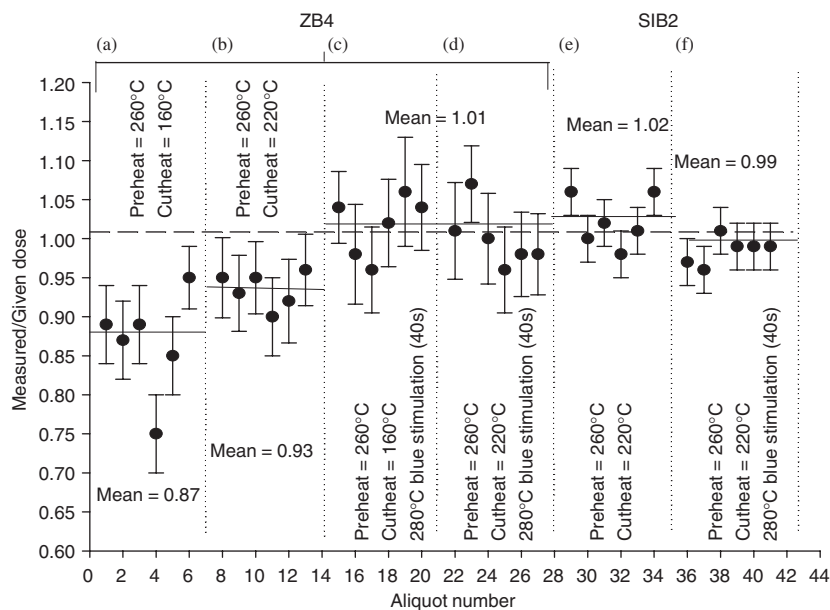


Fig. 10. Measured/given dose ratios obtained for dose recovery experiments on ZB4 and SIB2. The dose response curves were fitted including both the first (RR1) and the second (RR2) repeat points. Measurement conditions are given in the figure, together with the mean ratio for each set of six aliquots. Note expanded scale compared with Fig. 9.

employed after every test dose measurement, for both the conventional 160 °C cutheat and the higher 220 °C cutheat. For comparison, the equivalent data set from Fig. 9a and b is shown as Fig. 10a and b. The datasets shown in Fig. 10c and d show an improvement in the ability to recover a known dose, regardless of the cutheat employed, with a mean ratio of  $1.01 \pm 0.04$ . For SIB2, the dose recovery ratio was  $0.99 \pm 0.05$  (Fig. 10f) when using the high-temperature optical stimulation of Murray and Wintle (2003); this is similar to the ratio of  $1.02 \pm 0.04$  using the 220 °C cutheat alone (Fig. 10e). From the dose recovery data, it appears that the modifications to the conventional SAR protocol suggested by Murray and Wintle (2003) are successfully correcting for problems that may be contributing to the inappropriateness of the conventional SAR protocol for sample ZB4 from Blombos Cave; the modifications did not affect sample SIB2 from Sibudu.

### 6.3. Testing the modified SAR on other samples from Blombos Cave

The appropriateness of the modified SAR measurement procedure was also tested for a further six samples taken from the archaeologically rich layers at Blombos Cave. Dose recovery tests were carried out, using doses similar to those obtained in the preliminary dating runs. The results are presented in Fig. 11 and demonstrate that the modified measurement procedure is appropriate for all the samples from archaeological occupation levels at Blombos Cave.

## 7. Impact of the modified SAR protocol on the dating of ZB4 and SIB2

Following the successful dose recovery with the modified SAR protocol, another set of 24 aliquots from ZB4 and SIB2 were measured using this approach in order to obtain  $D_e$ . A single preheat temperature of 260 °C for 10 s was employed and a higher cutheat of 220 °C was chosen because of the illustrated improvement it had on the LM-OSL curves from ZB4, removing both an isothermal signal and the thermally unstable S2 component. The additional high-temperature optical stimulation was for 40 s with blue LEDs whilst the sample was held at a temperature of 280 °C. Recycling ratios (RR1) were all within 5% of unity (Fig. 12a and b) and are comparable with the 4 data points for the 260 °C for 10 s preheat shown in Fig. 4c. The RR2 ratios, that made a large difference to the dose recovered in the SIB2 samples, are now all consistent within 3% of unity.

The OSL measured when an aliquot is given no dose, but after different thermal treatments, is shown in Fig. 13a and b. It indicates that the high-temperature optical stimulation has evicted all optically sensitive charge for ZB4 and the majority for SIB2 and that there is no thermal transfer of charge. It can be seen that the signal after every  $L_x/T_x$  measurement cycle is erased completely, when the high-temperature optical stimulation is used. Fig. 14a and b shows the seven decay curves obtained during the extra stimulation with blue LEDs at 280 °C after each  $T_x$  measurement in a dating run. The decay curves from ZB4 (Fig. 14a) imply that there is

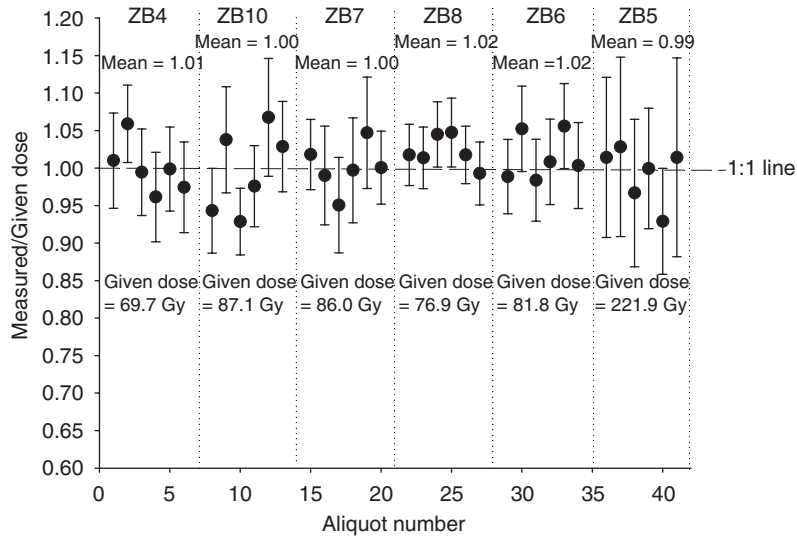
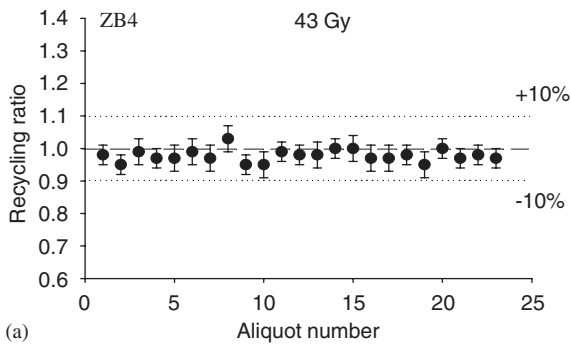
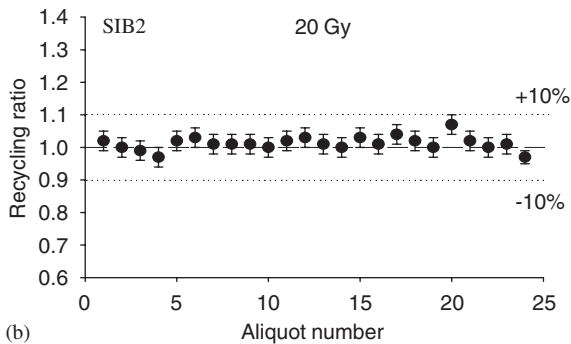


Fig. 11. Measured/given dose ratios obtained for dose recovery experiments for aliquots from a number of different samples from the Middle Stone Age archaeological layers in Blombos Cave.

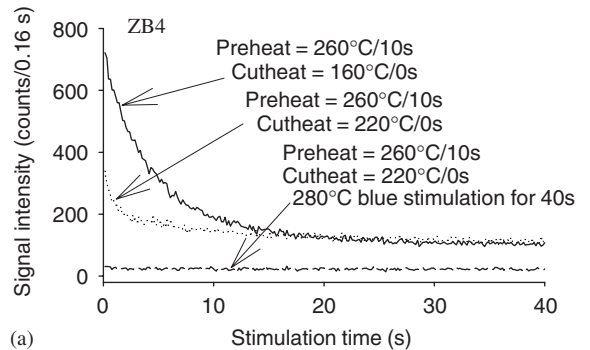


(a)

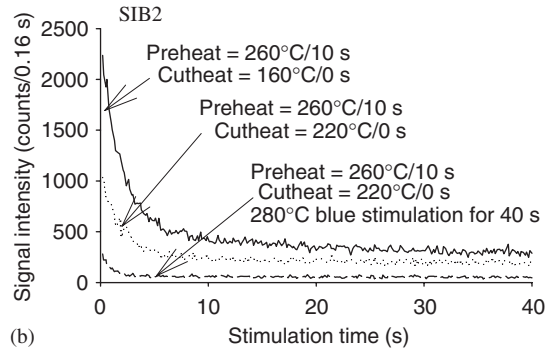


(b)

Fig. 12. Recycling ratios (RR1) obtained for the specified doses in dating runs for (a) ZB4 and (b) SIB2 using a preheat temperature of 260°C, cutheat temperature of 220°C in each cycle and an additional 40 s optical stimulation at 280°C between each cycle.



(a)



(b)

Fig. 13. OSL decay curves obtained for (a) ZB4 and (b) SIB2 for stimulation at 125°C when no laboratory dose had been given, but different cutheats had been applied (160 or 220°C), and also after a 280°C blue light stimulation was applied at the end of the previous cycle.

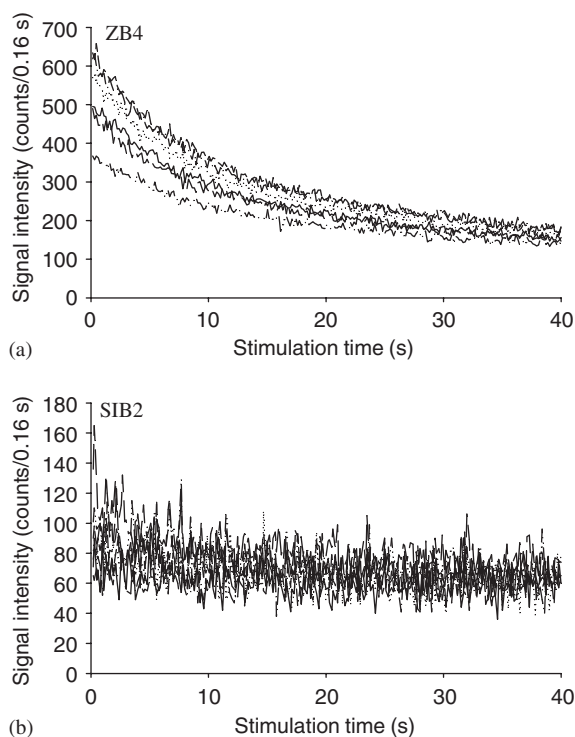


Fig. 14. OSL decay curves obtained for (a) ZB4 and (b) SIB2 during optical stimulation with blue LEDs at 280 °C following each measurement of  $T_x$  in a SAR dating run.

still a significant amount of optically stimulatable charge left after stimulation at 125 °C to obtain the measurement of  $T_x$ . Given the thermal stability of this signal, it may be the same as the thermally stable slow component (S1) of Singarayer and Bailey (2003). The OSL decay curves for SIB2 shown in Fig. 14b are smaller than those from ZB4 (Fig. 14a), in spite of the greater sensitivity of SIB2.

The effect of using the modified SAR protocol can be observed by comparing the new set of  $D_e$  data (Fig. 15c and d for ZB4 and Fig. 16c and d for SIB2) with that obtained originally using different preheats (Fig. 15a and b for ZB4 and Fig. 16a and b for SIB2). The  $D_e$  values measured using the conventional SAR protocol (the same data as in Fig. 3a) are shown as a sequentially measured data set as well as on a radial plot with the central value set at the calculated mean  $D_e$ . For ZB4 this mean  $D_e$  ( $N = 6$ ) is represented by the line indicated on the plot for those aliquots that received a preheat of 260 °C and 280 °C for 10 s that is similar to the 260 °C preheat used for all aliquots in the new set of measurements. The mean  $D_e$  for the data obtained using the conventional SAR is  $70 \pm 5$  Gy, whereas that obtained using the modified SAR is  $84 \pm 2$  Gy; thus an underestimation of approximately 17% occurred when the conventional SAR protocol was employed. This underestimation is similar in magnitude to the 13% underestimation obtained with the

dose recovery test when the conventional SAR protocol was used. For the aliquots from SIB2 (Fig. 16), the mean  $D_e$  for the data obtained by conventional SAR is  $117 \pm 3$  Gy, whereas that for the modified SAR is  $110 \pm 3$  Gy; these values are consistent within errors.

For ZB4 an age of  $75.7 \pm 3.3$  ka was calculated from the single aliquot  $D_e$  value of  $84 \pm 2$  Gy. This result provides the age for the layer containing the shell beads at Blombos Cave (d'Errico et al., 2005; Henshilwood et al., 2004). The age for SIB2 was  $53.8 \pm 1.6$  ka, based on the single aliquot  $D_e$  value of  $110 \pm 3$  Gy.

Given the difference in  $D_e$  values found for ZB4 when the modified SAR protocol was applied, it is appropriate to consider the OSL ages based on single aliquot measurements made using the conventional SAR protocol for dune sands at Blombos Cave (Henshilwood et al., 2002; Jacobs et al., 2003). An age of  $69.2 \pm 3.9$  ka was obtained for sample ZB15 taken from a sterile sand unit inside the cave, overlying archaeological units that included ZB4. This age was based on a value of  $D_e$  of  $50.3 \pm 1.7$  Gy obtained using the central age model and overdispersion of 16.4% was calculated for this sample (Jacobs et al., 2003). The conventional SAR procedure was found to work well, passing the standard tests; the recycling ratios were found to be consistent with unity over the preheat temperature range (160–300 °C) and values of  $D_e$  were independent of temperature, in spite of sensitivity changes of 30% being found during the dating run (Jacobs et al., 2003). Following the development of the modified procedure described in this paper, dose recovery tests were also carried out on ZB15 using both the high-temperature optical stimulation between cycles and the higher cutheat temperature. The ratio of measured to given dose was 0.99, compared to 0.98 obtained when no high-temperature optical stimulation was employed. Since no problem was encountered, it was not deemed necessary to redat sample ZB15.

## 8. Conclusions

The behaviour of representative samples from two archaeological sites has been shown to be inappropriate for application of the conventional SAR protocol using a 160 °C cutheat. SIB2 exhibited a preheat plateau and passed the recycling test, but a given dose could not be recovered. For ZB4 no preheat plateau could be obtained because the dose response curves from this sample showed atypical behaviour.

The effect of applying a higher cutheat and an optical stimulation at 280 °C for 40 s after each  $T_x$  measurement were investigated. Regardless of the cutheat, the use of optical stimulation at 280 °C made it possible to recover a known dose for both samples (Fig. 10). Increasing the cutheat temperature from 160 to 220 °C reduced the phosphorescence from ZB4 and SIB2 (Fig. 8c and d) and also reduced the relative contribution of S2, the thermally unstable OSL component (Fig. 8e and f).

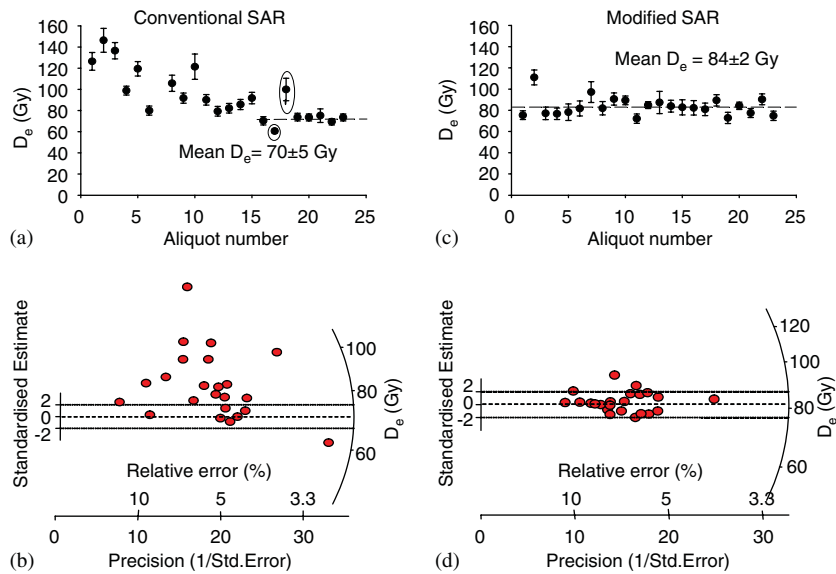


Fig. 15.  $D_e$  values for ZB4 (a) and (b) obtained using the conventional SAR protocol and for a range of preheat temperatures (plotted as a function of preheat temperature in Fig. 3a). (c) and (d) obtained using a preheat of 260 °C for 10 s, a 220 °C cutheat, and an optical stimulation at 280 °C between each cycle.

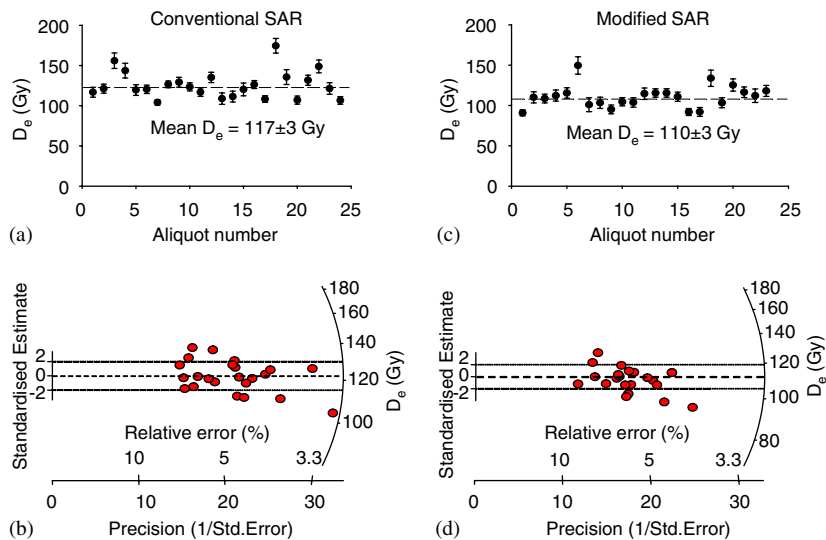


Fig. 16. (a)–(d) as for Fig. 15, but for SIB2.

From this study it appears that the standard tests of recycling a low dose point and measuring  $D_e$  as a function of preheat temperature as suggested by Murray and Wintle (2000) may not be sufficient to tell whether the protocol is appropriate for that sample. There are several other tests that could be performed that may provide additional diagnostic information. These include:

1. A dose recovery test, demonstrating the ability to accurately recover a dose;
2. Measurement of a second recycling ratio point using a higher radiation dose, especially for older samples that are approaching saturation;
3. Analysis of standardised growth curves to investigate any preheat dependency;
4. Making additional LM-OSL measurements, specifically for a signal following the chosen cutheat temperature to look at the relative contributions of the different components to the signal region used for dating.

Where problems exist, these may be circumvented by the application of

1. A higher cutheat e.g.  $\sim 20\text{--}40^\circ\text{C}$  below the preheat temperature
2. A high-temperature optical stimulation, such as  $280^\circ\text{C}$  with blue LEDs for 40 s, between each cycle of the SAR protocol.

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