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Spatial variation of dose rate from beta sources as measured using single grains

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Abstract

Dose rates across the centre of a 9.7 mm diameter aluminium disc were measured using a 10 x 10 array of single grains of quartz held in holes drilled with 600 µm separation. The dose rates were obtained by measuring the OSL signals from quartz grains that previously had been given a known gamma dose and comparing them with those measured following increasing doses given by a beta source held 5 mm above the disc surface. The patterns of dose rate were obtained for four $^{90}Sr/^{90}Y$ beta sources. Two were found to produce a non-uniform dose rate at the disc surface, with one showing a factor of two across the 7.6 mm diameter of the area containing the 10 x 10 array. The implications for both single grain and multiple grain measurements are discussed.

Introduction

There has been an increasing number of publications relating to the use of Risø readers that have a special attachment for the measurement of the OSL signals from individual sand-sized grains using a focussed laser for optical stimulation (Duller and Murray, 2000). The main applications have been to the dating of sand related to archaeological sites (e.g. Jacobs et al., 2003), fluvial deposits (e.g. Thomas et al., 2005), glacial deposits (e.g. Glasser et al., 2006) and dosimetry studies of concrete blocks (Thomsen et al., 2003) and mortar (Jain et al., 2004) . The prototype of Duller et al. (1999a; 1999b) used an 8 x 8 array of holes drilled into the surface of a 0.97 cm diameter aluminium disc. More recently a 9 x 9 array (Bøtter-Jensen et al., 2000) and then a 10 x 10 array (Bøtter-Jensen et al., 2003) have been used. The most commonly used discs have holes that are ~ 300 µm in diameter and 300 µm deep with their centres being 600 µm apart; they are designed to receive single grains with diameters ranging from 180 to 250 µm, a common size used in environmental and dosimetric studies.

The reproducibility of measurements made with the single grain system has been reported by Truscott et al. (2000) who used both Al_2O_3:C grains and thermally annealed quartz grains. By making repeated measurements using the same dose, they demonstrated the precision and accuracy of the laser stimulation system. This resulted in a standard deviation of ~3.5% for repeated measurements. Thomsen et al. (2005) found similar results in another study of repeated paired measurements, though they noted that the standard deviation could be decreased by increasing the signal integration time; similar findings were reported by Jacobs et al. (2006).

The experiments described above have established the reproducibility of the measurement procedure. A crucial part of using such a system is calibration of the beta source. This can be undertaken using individual grains that have previously been sensitised and stabilised by heating, and then been given a known gamma dose. The SAR protocol (Murray and Wintle, 2000) can then be used to determine the dose rate. Measurements of such gamma-irradiated quartz, should provide a distribution of doses that has a standard deviation that is similar to that of the grains given repeated beta doses. However, it has been shown that the scatter in dose rate is much larger (Thomsen et al., 2005). Thomsen et al. (2005) also presented evidence that using individual dose rates for each grain position, rather than an average dose rate for the whole disc, caused a reduction in the error term. Their results indicated that for their source, non-uniformity of dose rate did not contribute more than about 5% to the variability observed. However, given the different methods of source construction, it is possible that other sources may be more variable, and that is what this paper explores.

Source	Activity	n	$R_1<R_2<R_3$	R_5/R_1	Recuperation (% of R_1)	Min.	Max.	Average dose rate
5583	74 MBq	600	4	30	4	1.81	3.39	2.66 ± 0.04 mGy/s
5626	24.1 MBq	1600	15	38	6	1.02	1.44	1.236 ± 0.003 mGy/s
6100	1.48 GBq	1200	1	15	1	0.064	0.142	0.103 ± 0.002 Gy/s
6088	1.48 GBq	2000	1	21	1	0.117	0.166	0.147 ± 0.002 Gy/s

Table 1:. *Sources used, source activity, number of grains (n) investigated, percentage of grains that fail a series of quality control tests (dose response curve, recycling ratio and recuperation), average minimum, maximum and average dose rate. R are normalized luminescence values (L/T) for cycles 1 to 5 in the SAR protocol. R5/R1 is the recycling ratio. R1<R2<R3 indicated that the dose response curve grows systematically.*

Although the grains are individually optically simulated with the laser, both irradiation and heating of all 100 grains on a disc is carried out simultaneously. The heating of the grains will be very similar as aluminium is a good conductor. The uniformity of irradiation will depend upon the source-sample distance and the homogeneity of the source. For older versions of the Risø TL/OSL reader, the source-sample distance is 7 mm, as reported by Mauz and Lang (2005). For more recent readers, this distance has been reduced to 5 mm (Bøtter-Jensen et al., 2000). It might be assumed that this geometry would provide uniform irradiation across the 7.6 mm diameter of the area occupied by the holes in the special single grain disc. However, this should not be assumed given recently reported spatial variations in dose rate reported when using a source with a ceramic substrate in a stand-alone irradiator with the sources at distances of 15 to 25 mm from a radioluminescent probe made of CaF_2 (Spooner and Allsop, 2000).

It is particularly important to take account of all laboratory-derived sources of error in the measurement of single grains before obtaining dose distributions for naturally-irradiated sand grains. Thus it is important to investigate the dose rate for each position across a single grain disc. In this study, we investigate the uniformity of the dose rate across a sample disc for discs irradiated in a Risø TL/OSL reader

Equipment

The Risø TL/OSL DA-15 reader employed in the study was purchased in 2002 and has a source-sample distance of 5 mm (Bøtter-Jensen et al., 2000). The carousel used to carry the sample discs has 48 positions. Single grain discs were placed on the carousel of the reader with alternate positions left empty in order to avoid cross-talk during irradiation (Markey et al., 1997; Bøtter-Jensen et al., 2000) and optical stimulation (Bray et al., 2002). The discs were carefully aligned so that they were identically oriented.

Four $^{90}Sr/^{90}Y$ sources are assessed, one original SIP silver plaque type source and three, more recent, SIF ceramic-substrate type sources manufactured by AEA Technology (Germany). The relative merits of these different types of source have been discussed by Aitken (1985) and by Spooner and Allsop (2000). In particular, it was reported that the active area of the SIP source has a diameter of 12 mm, whereas the equivalent diameter for the SIF source is only 5 mm (Spooner and Allsop, 2000). The sources were moved to the single grain reader. Each source was mounted in turn in the rotating aluminium wheel, which is built into a lead castle to provide shielding (Markey et al., 1997). The laboratory code and nominal activity for each source is given in Table 1.

Experimental procedure

The sources were calibrated using two batches of quartz (grain size of 180-212 µm) that had received doses of 5.00 and 3.18 Gy, respectively, using a ^{137}Cs γ-source at the Risø National Laboratory. This quartz has been heat treated in the Risø National Laboratory and is used by them for beta source calibrations. For all the quartz grains used in this study an OSL signal could be measured. For source 5626, quartz with a calibration dose of 3.18 Gy was used; for the other sources, quartz with 5.00 Gy was used. A SAR protocol was applied using a 10 s preheat at 240°C prior to the OSL measurement that was made for 1 s at 125°C, and a cut heat to 220°C after the delivery of the test dose. L_i and T_i are derived from the initial OSL signal (0.1 s) minus a background estimated from the last part of the stimulation curve (0.2 s). The SAR protocol used three regenerative beta doses to build up the dose-response curve, with R = L/T.

The reliability of the protocol within a measurement was assessed through three checks. First, the dose-response curves were tested for consistency; i.e. that

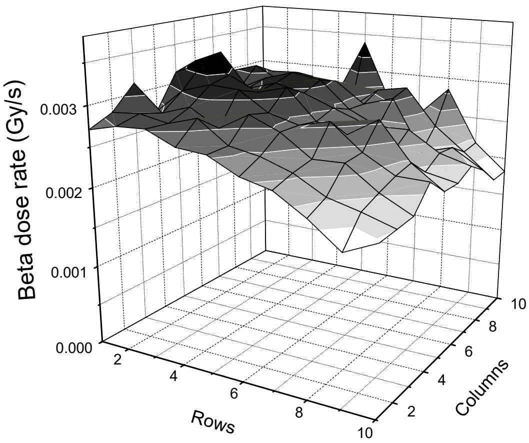

Figure 1: *3-D plot of dose rate (z axis) as a function of grain position (x, y) on ten-by-ten grid position for source 5583 (74 MBq SIP silver plaque source).*

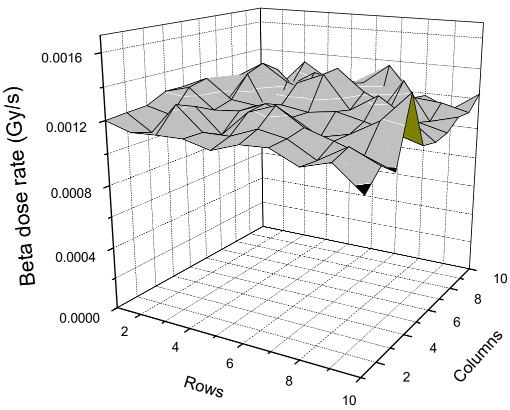

Figure 2: *3-D plot of dose rate (z axis) as a function of grain position (x, y) on ten-by-ten grid position for source 5626 (24.1 MBq SIF ceramic source).*

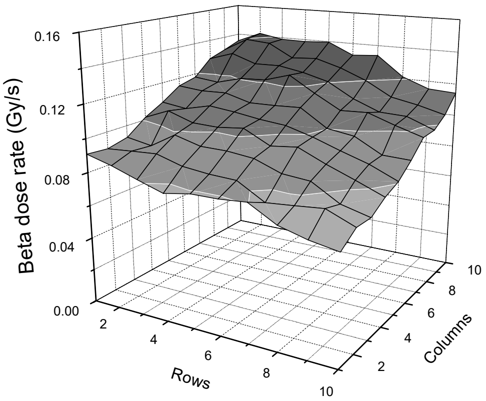

Figure 3: *3-D plot of dose rate (z axis) as a function of grain position (x, y) on ten-by-ten grid position for source 6100 (1.48 GBq SIF ceramic source).*

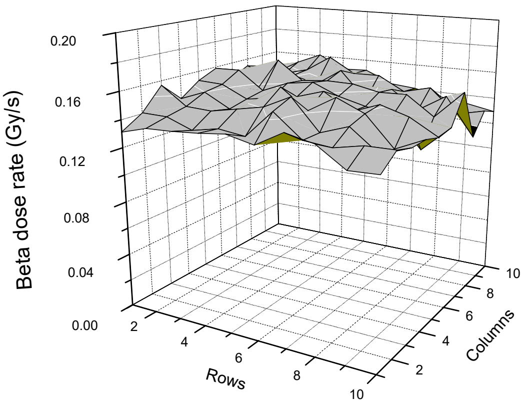

Figure 4: *3-D plot of dose rate (z axis) as a function of grain position (x, y) on ten-by-ten grid position for source 6088 (1.48 GBq SIF ceramic source).*

larger doses gave larger OSL signals (R1 < R2 < R3). Second, the ratio (R5/R1) between the two sensitivity-corrected OSL responses generated from the same regenerative dose (recycling ratio) is within 10% of unity. Third, the OSL response when a zero regenerative dose is administered, expressed as a percentage of the corrected natural OSL signal L_n/T_n, is small (recuperation test of Murray and Wintle, 2000). The percentage of grains that failed the above checks, and thus were rejected, are listed in Table 1.

Results

The results of the experiment are plotted in 3-D graphs where on the x- and y-axis is the ten-by-ten position-grid, and on the z -axis is the estimated dose rate. The spatial distributions of dose rates obtained using single-grain discs are shown in Figures 1-4. All four data sets have been rotated by about 90 degrees; this rotation was chosen for the best display of the plots for sources 5583 (Figure 1) and 6100 (Figure 3). The data used to obtain the plot shown in Figure 3 are given in Table 2. For this source (6100), 12 discs, each carrying 100 grains, were used. Each point in the table is obtained by calculating the mean and standard error (not shown) from the individual measurements. Table 3 gives the individual measurements (and the mean and standard error) for two grain positions (1, 10) and (10,2); these positions

	1	2	3	4	5	6	7	8	9	10
1	0.090	0.087	0.081	0.076	0.078	0.080	0.078	0.071	0.067	**0.064**
2	0.090	0.091	0.092	0.087	0.087	0.084	0.075	0.076	0.078	0.073
3	0.091	0.089	0.098	0.095	0.094	0.091	0.086	0.084	0.076	0.071
4	0.102	0.108	0.102	0.101	0.103	0.095	0.094	0.089	0.083	0.076
5	0.108	0.106	0.108	0.108	0.105	0.103	0.099	0.093	0.088	0.080
6	0.116	0.114	0.114	0.110	0.110	0.111	0.101	0.098	0.089	0.088
7	0.118	0.116	0.120	0.121	0.123	0.118	0.108	0.108	0.102	0.098
8	0.128	0.133	0.128	0.127	0.125	0.120	0.110	0.105	0.104	0.097
9	0.131	0.140	0.138	0.131	0.132	0.126	0.121	0.112	0.105	0.104
10	0.136	**0.142**	0.139	0.137	0.140	0.133	0.134	0.121	0.115	0.111

Table 2: *Source 6100 - Average individual single-grain dose-rate estimates (Gy/s) calculated for each position on the disc (100 grains are measured on each of 12 discs). In bold are the minimum and maximum dose rate values.*

Position	Individual dose rate estimates (Gy/s)								Mean and s.e.
(1,10)	0.136	0.147	0.142	0.132	0.149	0.150	0.137	0.142	**0.142±0.002**
(10,2)	0.065	0.060	0.071	0.060	0.054	0.060	0.063	0.085	**0.064±0.003**

Table 3: *Source 6100 - Dose rate measurements for 8 discs for two positions*

correspond to the positions giving the lowest and highest dose rates, respectively. In each case, the dose rate was measured on 8 grains (out of the possible 12 prior to the grain rejection criteria being applied).

For determination of the dose rate for one grain on one disc, e.g. that at position (1, 1) on the first disc, it is necessary to be sure that the OSL signal is measured reliably. The reproducibility of the OSL signal measurement through a SAR run can be assessed by looking at the recorded position of the laser beam during successive measurements. An example of this is shown in Figure 5, where for the first grain the position of the measurement of the gamma dose is shown at the centre of a circle drawn to represent the size of a 300 µm diameter hole. The co-ordinate centres for each subsequent beta dose measurement (irradiation with source 6100) are shown. There is a slight movement (<20 µm) for the second measurements and the remaining six are clustered at ~100 µm from the initial position. This shows that no further relative measurement has occurred as a result of the movement of the disc whilst in the reader.

The fact that the disc is not moving far, with respect to the coordinate system, implies that it is not moving far relative to the beta source either. This will allow for the dose response curve for each grain in that position to be well defined. However, for it to be meaningful to calculate the mean dose rate for a grain position, the discs should be placed in a similar position relative to the source. That this has been accomplished in this study, can be seen by plotting the recorded co-ordinates for four consecutively measured discs. In Figure 6 these positions are shown for both the first measurement (i.e. related to the gamma dose) and the last measurement (i.e. related to the final beta dose). From this plot it is inferred that the dose rate is being measured at positions relative to the source that are within 500 µm.

Discussion

A source's homogeneity can be assessed by visual inspection of the 3-D plots. Flat surfaces indicate spatial homogeneity of irradiation and such a surface is found for sources 5626 and 6088. If a non-flat surface is observed, this indicates that irradiation does not occur uniformly, and these are found for sources 5583 and 6100. The results shown in Figures 1-4 indicate that two of the sources result in a steep gradient in the dose rate across the 10 x 10 array of grains. In Figure 1, showing the data obtained using the SIP type source (5583), the dose rate varies from 3.39 mGy/s to 1.81 mGy/s. In Figure 3, showing the

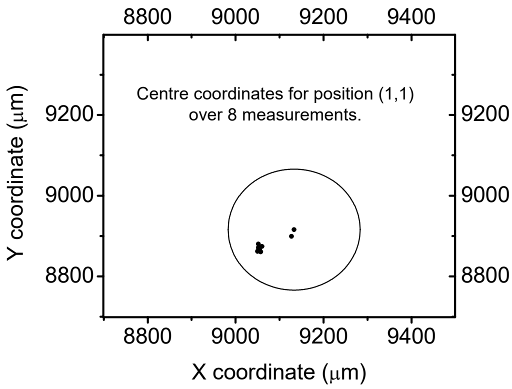

Figure 5: *Plot of centre co-ordinates for position (1, 1) as obtained over 8 OSL measurements made in a SAR cycle. Circle drawn to show size of hole with its centre at the first measurement position.*

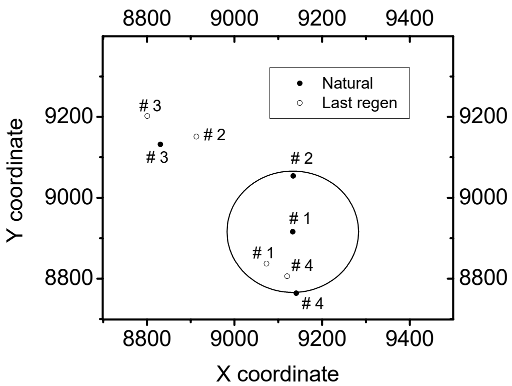

Figure 6: *Plot of centre co-ordinates for first and last measurements of SAR run for position (1, 1) on each of four discs (#1, #2, #3 and #4). Circle drawn to show size of hole.*

data obtained using the SIF type source 6100, the dose rate varies from 0.142 Gy/s to 0.064 Gy/s (Table 1). For the other two SIF type sources, 5626 (Figure 2) and 6088 (Figure 4), the dose rate is much more uniform, and the maximum and minimum values for each source are given in Table 1. The ratio of the maximum to minimum values for the latter two sources is 1.41.

Non-uniform irradiation could be caused by non-uniform distribution of radioactive material on the source face. The method of manufacture of both types of sources employs the dropping of a liquid containing ^{90}Sr onto a surface. In the case of SIP sources, the liquid is evaporated from the silver plate that forms the front of the source (Aitken, 1985). In the manufacture of SIF sources, the liquid is dropped onto a ceramic surface, into which it can penetrate; this may result in it not being uniformly distributed prior to evaporation. There appears to be no immediate solution to this problem with source construction, though it has been suggested that a mini X-ray generator may provide an alternative (but uniform) irradiation source (Andersen et al., 2003).

An alternative interpretation of the results is that the steep gradient in dose rate for sources 5583 and 6100 is caused by a mis-alignment of the source and the aperture through which the electrons pass. This possibility is however not likely seeing the different orientations of the dose-rate gradients for the two sources. The experiment was not repeated with the sources in different orientations.

For single grain measurements, it is possible to obtain individual dose rates for each hole in a single grain disc and, indeed, it is essential to do this to avoid incorrect dose evaluation. However, it should be pointed out that there is likely to be a problem if these poorly-performing sources are used for measurement of aliquots made up of several thousand grains. It would still be possible to measure an average dose rate for a non-uniform source, using the uniformly bright calibration quartz spread over a 9 mm diameter area in the centre of the disc. However, in the case of un-sensitised sand-sized quartz grains, it has been demonstrated that only a small percentage of grains produce almost all the OSL signal (e.g. Duller et al., 2000; Jacobs et al., 2003). These bright grains would be randomly distributed amongst the thousand or so grains on a sample disc. The value of the equivalent dose that would be calculated would depend upon exactly where the bright grains were situated. For a non-uniform source such as 6100, this would result in a previously unconsidered source of scatter in the distribution of the values of the equivalent dose. In contrast to the case for single grains, it is not possible to obtain an appropriate calibration and this will result in meaningless D_e distributions or, at the very least, prevent using overdispersion measurements to obtain information on bleaching history. For laboratories without access to a single grain reader, or equipment of the type used by Spooner and Allsop (2000), a simple test can be applied. A quartz grain can be placed several mm from the centre of a regular sample disc and given a dose. The disc should then be rotated by 180° and the SAR protocol applied. If the number of seconds of irradiation required to match the first irradiation time is identical, then the disc may be considered to be uniformly irradiated.

Conclusions

Using OSL signals from highly-sensitised quartz grains that had been given a laboratory gamma dose, we have demonstrated that two of the four $^{90}Sr/^{90}Y$ sources in the Netherlands Centre for Luminescence Dating in Delft give non-uniform dose rates across the inner area of a standard 9.7 mm diameter sample disc. The measurement used single grains mounted in a 10 x 10 array within an area of 7.6 mm diameter.

Of the two sources that resulted in a uniform dose rate, both were of the SIF type. Of the two sources that showed poor uniformity, one was of the SIP type and one of the SIF type. This study has shown that it is essential to make a calibration for each hole position when using the single grain facility. The gradient across the 7.6 mm diameter area would make it inappropriate for use in regular dosimetry measurements in which grains of variable sensitivity are randomly distributed across a 9 mm diameter area of the 9.7 mm diameter sample disc. Effects of the non-uniform dose rate on regular dosimetry measurements may be mitigated by mounting the sample only on the central area of the disc.

It is not clear whether the strong non-uniformity of the measured dose rate is the result of non-uniformity of the radioactive material on the source face or the mis-alignment of the source and the aperture, but the former is considered more likely.

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Reviewer

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