The alpha effectiveness in silt-sized quartz: New data obtained by single and multiple aliquot protocols

B. Mauz, S. Packman and A. Lang

Department of Geography, University of Liverpool, Liverpool L69 7ZQ, UK

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Abstract
The luminescence effectiveness of alpha particles in silt-sized quartz originating from different regions in Europe and North America is presented. Single aliquot regenerative dose (SAR) protocols were used along with the a-value system. The study shows that a-values are independent of a sample's origin, but dependent on the alpha- and beta-doses administered. Sensitivity changes induced by alpha-doses administered during a SAR procedure are not corrected by the beta dose induced test dose OSL. For samples displaying a linear beta-dose response an average a-value of 0.03 was found. For samples exhibiting exponential saturating beta-dose response the a-values determined are higher confirming the dose dependence of the alpha effectiveness in quartz.

Introduction
Quartz is widely used for optical dating. The use of its sand-sized fraction has become the preferred approach in the last few years as this grain size allows the analysis of dose distributions when a small number of grains are used per aliquot and when a single-aliquot regenerative-dose (SAR) protocol is applied. For this grain size, it is assumed that the treatment with hydrofluoric (HF) acid removes the outer rim of the grains which has been affected by alpha radiation in the natural environment. Thus, using sand-sized quartz for optical dating circumvents the need for determining the alpha dose and the alpha dose effectiveness. The silt-sized fraction (4 – 20 μm), however, receives the full alpha dose (α-dose) emitted in the thorium and uranium series and thus, the determination of the alpha dose and its luminescence effectiveness is required.

To determine the alpha effectiveness of a sample, the a-value system (Aitken and Bowman, 1975) is commonly used. Whereas for feldspar silt-sized samples ("polymineral fine grain sample") the a-value has been determined in numerous dating studies (e.g. Lang and Wagner, 1997, Lang et al., 2003), little work has been undertaken to determine the a-value of quartz. Given the relatively low a-values measured for thermoluminescence, typically 0.02 to 0.05 (Aitken, 1985a, p.262), and the mostly insignificant internal radioactivity of natural quartz, there have been few detailed analyses. Rees-Jones (1995) reported a-values of 0.032-0.043 for 4 samples using additive alpha- and beta-dose response curves. Tribolo et al. (2001) for the first time used a SAR protocol to determine the alpha sensitivity of heated quartz. Dating studies (e.g. Stokes et al., 2003a) have used a-values determined on one sample (a = 0.04) and adopted it for the remaining samples or adopted the a-values (e.g. Stokes et al., 2003b) reported by Rees-Jones (1995). It is likely that the luminescence sensitivity of natural quartz for alpha radiation varies little as long as the corresponding alpha- and beta-dose responses are linear. At higher doses, however, less luminescence is created per unit beta-dose; consequently, the contribution of the alpha dose to the total energy deposited increases. The saturation level for beta radiation is grain-dependent (Yoshida et al., 2000), and, thus, the a-value determined using relatively high alpha doses should be sample-dependent and less uniform than the a-value resulting from lower doses.

Our study aimed to test the applicability of a SAR protocol to determine a-values of natural fine-grained quartz particularly with respect to possible sensitivity changes if repeated alpha irradiation is needed. Furthermore, we wished to verify the hypothesis about the dose and sample dependence of the a-value. Silt-sized quartz samples from different areas in Europe and from North America were investigated.

The alpha effectiveness and the a-value system
Alpha particles in the U and Th decay chains have an energy spectrum ranging from 7.8 MeV to zero, where high energy particles travel as far as 47 μm in quartz (Aitken, 1985a, p.253) and affect around 10 mg cm^{-2} (Brennan and Lyons, 1989). The mean absorbed dose fraction for alpha-particles is a function of grain diameter and decreases significantly with grains larger than 20 μm (Bell, 1980). Due to attenuation during travel through matter, the range

spectrum is approximately rectangular, i.e. there is the same number of alpha particles in each interval of range (Aitken, 1985a, p.253). Unlike beta- and gamma-radiation, the luminescence induced by α-radiation is not proportional to energy deposited in a sample but approximately proportional to the total track length deposited and thus, to the travel range of the alpha particle (Brennan and Lyons, 1989). Since the energy deposited per track increases with track length (as the α-particle slows down) the luminescence signal induced in any particular grain by an α-particle is dependent on track length and not just on alpha energy (Brennan et al., 1991). The track length within a grain depends on the diameter of the grain. Thus, the alpha dose absorbed by a silt-sized quartz grain depends on the alpha particle travel range (grain diameter) and energy, whereas the alpha effectiveness depends on track length. The luminescence effects of an α-particle are limited to a small grain volume along the track (Zimmerman, 1972) and consequently, α-particles induce less luminescence for a given amount of absorbed energy than ß-particles and γ-radiation. At higher α-doses tracks overlap and the spatial energy distribution effect of α-particles should disappear (Zimmerman, 1972). Each alpha particle deposits energy only along a small stretch of its track whereas energy deposition by beta particles is not spatially constrained within the crystal. Therefore, at high doses α- and β-growth curves differ (Fig. 1) with the α-growth curve being linear until higher doses than the β-growth curve (Zimmerman, 1972). At doses close to saturation the energy distribution becomes uniform and the luminescence response to α- and β-doses becomes equal (Fig. 1).

Commonly, the a-value system (Aitken and Bowman, 1975) is used to correct the total α-dose rate calculated from the U- and Th-series for the α-effectiveness. The crucial assumptions of the a-value system are:

1. luminescence per unit track length is the same for all alpha particles independent of their energy
2. luminescence per unit of absorbed energy from beta particles is constant and independent of the energy of the particle

The first assumption allows for using a monoenergetic α-source (e.g. ^{241}Am, 3.7 MeV) to monitor the α-effectiveness of a sample in the laboratory. By comparing the beta (or gamma) dose that would induce the same amount of luminescence as the alpha dose from this monoenergetic α-source,

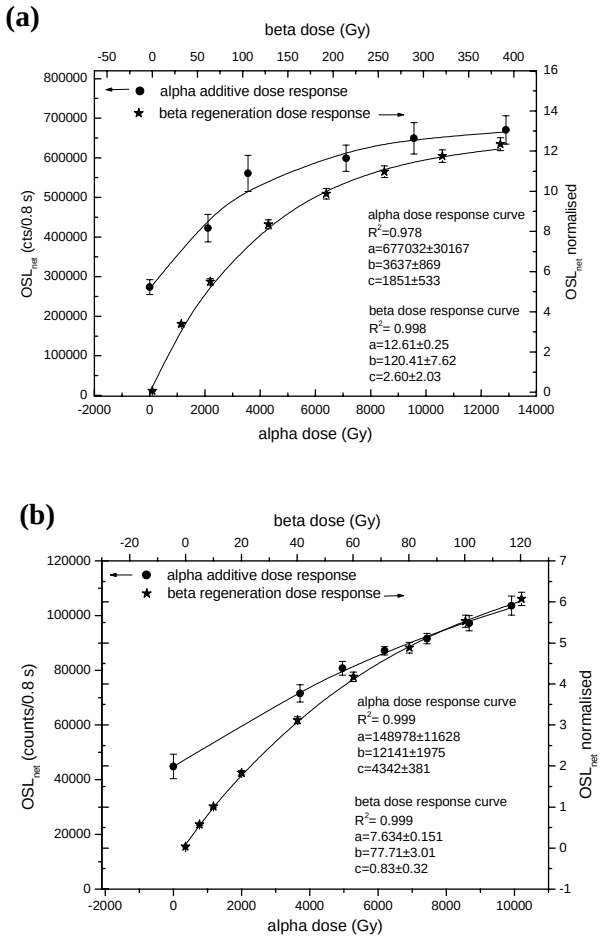

Figure 1: *Alpha- and beta dose response curves of two samples fitted to the equation* $y = a \times (1 - e^{-\frac{x+b}{c}})$. *Each alpha dose point is based on the mean and standard deviation of 3 aliquots after normalisation to the natural OSL. The beta dose points are derived from a SAR measurement of one aliquot. (a) Sample exhibiting a very high natural dose (LV184); (b) sample exhibiting a high natural dose (LV05)*

independence of instrumental and sample sensitivity is achieved. Thus, Aitken (1985a, p.311) gives:

$$\frac{\xi_s v}{\chi_\beta} = 13a$$

where ξ_sv is luminescence per unit alpha track length per unit volume and χß is luminescence per unit absorbed beta dose. The numerical factor is equal to the energy loss per micron for a 3.7 MeV alpha particle in quartz divided by the mass density (Aitken, 1985a). The factor was later considered to be an arbitrary value (Bowman and Huntley, 1984) derived from the transformation from the k-value,

which preceded the a-value system. Bowman and Huntley (1984) show that the ratio

$$\frac{L \cdot per \cdot unit \cdot alpha \cdot track \cdot length}{L \cdot per \cdot unit \cdot absorbed \cdot beta \cdot energy} \text{ (L=luminescence)}$$

accounts for the beta energy that yields the same amount of luminescence as one unit length of alpha track.

Material and experimental details

The samples used here originate from the German North Sea coast (LV03, 04, 05, 06), the South German loess province (LV95, 96, 97), the English east coast (LV17), the English Lake District (LV104) and North America (LV176, 184). Samples were treated in the laboratory following conventional procedures for fine silt (4-20 µm) sample preparation (Mauz et al., 2002) and etched using 20% and 10% hydrofluoric (HF) acid for various durations to remove the feldspar-derived luminescence component (Mauz and Lang, 2004a). After etching the grains, silt from 4-15 µm was separated by settling in acetone. The aliquots of each sample consisted of 2 mg material pipetted onto 1 cm aluminium discs. All samples were tested using infrared stimulation (IR-OSL depletion ratio, Duller, 2003) which examines the effect of the feldspar OSL component on the normalised OSL (L_x/T_x) and thermoluminescence (TL) measurements, which gives an estimate of the amount of feldspars remaining in the sample after chemical treatment (Mauz and Lang, 2004a, Shen et al., in press). According to these tests, all samples used for further experiments were pure quartz samples.

All measurements were performed using an automated Risø TL/OSL reader equipped with an EMI 9635QA photomultiplier and a 40 mCi $^{90}Sr/^{90}Y$ β source, delivering ~0.09 Gy s^{-1} to fine grain quartz on aluminium discs (Mauz and Lang, 2004b). Alpha irradiation was performed in vacuum (10^{-4} mbar), using six 31.6 µCi ^{241}Am sources delivering 3.7 MeV α-particles. The device was built by Bürgi (1992) following the design of Singhvi and Aitken (1978). The sources were calibrated using TLD 200 dosimeters and cross-calibrated with alpha sources at the MPI Heidelberg (Bürgi, 1992). The six sources gave track lengths in the dosimeter between 0.1873 μm^{-2} min^{-1} and 0.2209 μm^{-2} min^{-1}. With respect to the energy deposition in quartz (Aitken, 1985a, p.135 and p.312) these sources delivered doses of 2.44–2.87 Gy min^{-1} on the calibration day (1.3.1991, Bürgi, 1992). These calibration dates were used here as the half life of ^{241}Am is ~ 450 years. Optical stimulation of samples was performed using blue

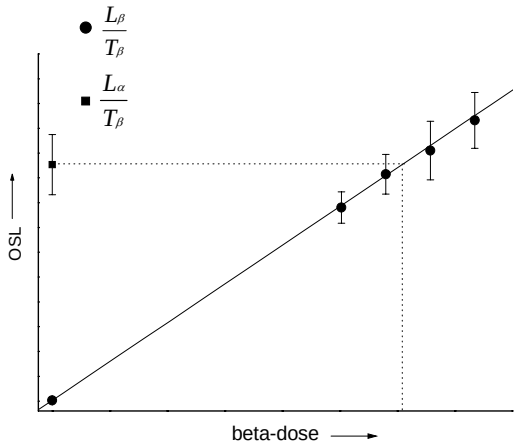

Figure 2: *SAR protocol designed to determine the a-value. The a-value results from the projection of L_α/T_β onto the beta dose response curve obtained from a standard SAR protocol. For details see text.*

LEDs emitting at 470Δ30 nm (delivering ~30 mW cm^{-2}). The OSL was detected through an optical filter (Hoya U340, 7.5 mm) transmitting 260 to 390 nm wavelengths.

To determine the a-value, a SAR protocol was envisaged (Fig. 2). The ß-dose equivalent to a given α-dose should be determined by projecting the regenerated OSL induced by α-irradiation normalised to the OSL of a test dose induced by β-irradiation (L_α/T_β) onto the sensitivity-corrected regenerated dose response curve generated by ß-irradiation. Three experiments were designed to test the reliability of this SAR protocol. In the first experiment the α-dose was given at the end of a standard SAR protocol. The second experiment adopted principles of the dose recovery test (Murray and Wintle, 2003). Aliquots were optically bleached (to remove the natural OSL), given an α-dose, and this was then treated as an unknown dose in the SAR protocol. For this experiment two naturally low-dosed samples (LV04, LV97) were chosen. The third experiment was conducted on aliquots previously used for $D_{eß}$ determination to investigate sensitivity changes. These were subjected to 7 cycles of alpha irradiation (~90 Gy per cycle, sample LV06), preheating and blue light stimulation. Alpha doses in the first two experiments were chosen to match the linear L_β/T_β growth and aliquots were rejected if the L_α/T_β ratio did not fall in this range.

Samples showing a relatively high natural dose, and therefore requiring an exponential-saturating beta dose response curve (indicated as 'exp' in Table 1)

Sample Code (LV)	Sediment (Origin)	*n*	fit	$D_{e\beta}$ (Gy)	$D_{e\alpha}$ (Gy)	a-value
06	coastal, North Sea (Germany)	14	linear	2.48±0.04	-	0.023±0.002
03	coastal, North Sea (Germany)	10	linear	4.32±0.01	-	0.026±0.002
04	coastal, North Sea (Germany)	6	linear	4.37±0.02	-	0.031±0.002
104	lacustrine, (NW-England)	13	linear	2.5±0.05	-	0.028±0.002
17	coastal, North Sea (Scotland)	7	linear	23.8±0.4	-	0.027±0.002
96	loess derivate, (S-Germany)	12	linear	3.20±0.07	-	0.053±0.004
95	loess derivate, (S-Germany)	10	linear	2.84±0.06	-	0.032±0.003
97	loess derivate, (S-Germany)	6	linear	2.59±0.02	-	0.031±0.002
176	Mississippi river (N-America)	-	exp	248±43	2378±72	0.104±0.018
184	Mississippi river (N-America)	-	exp	203±14	1851±533	0.110±0.032

Table 1: *The a-values of silt-sized quartz samples analysed in this study. n indicates the number of aliquots used in the SAR protocol to calculate the a-value, fit indicates the fitting procedure used to determine the $D_{e\beta}$ of the sample (linear: y=ax+b; exp: $y = a \times (1 - e^{-\frac{x+b}{c}})$; $D_{e\beta}$ is the equivalent dose obtained from a SAR protocol using β-irradiation; $D_{e\alpha}$ is the equivalent dose obtained from a multiple aliquot additive dose (MAAD) protocol using α-irradiation.*

Sample code (LV)	*n*	Given α-dose (Gy)	Recovered ß-dose (Gy)	Recycling ratio	a-value	a-value ratio
04	6	122±8	4.02±0.17	1.03±0.03	0.033±0.002	0.94±0.08
97	6	56.8±3.7	1.76±0.06	0.97±0.02	0.031±0.002	1.00±0.09

Table 2: *The a-values of two samples determined using a dose recovery SAR protocol. A given α-dose is recovered by a SAR protocol based on ß-irradiation. The a-value ratio is the ratio between the a-value determined with the alpha irradiation at the beginning of the SAR protocol and the a-value determined with the alpha irradiation at the end of the SAR protocol.*

were measured with a multiple aliquot additive dose (MAAD) protocol. Between three and five different additive alpha doses were administered and a $D_{e\alpha}$ was determined by extrapolating the dose response curve to the dose axis. The a-value of a sample then results from the comparison of $D_{e\beta}$ and $D_{e\alpha}$.

Preheat temperatures were chosen on the basis of results from a preheat test and were 200°C, 220°C and 230°C for 10 s for low dose samples (indicated as 'linear' in Table 1) and 260°C for 10 s for high dose samples (indicated as 'exp' in Table 1). The cut heat was always 200°C, the test dose size was ~ 10% of the expected natural dose and the OSL was stimulated for 40 s at 125°C. All a-values obtained from a SAR protocol were determined as arithmetic mean and standard deviation of n values per sample (for n see Table 1 and Table 2). Assuming a normal and log-normal distribution of doses, a small number of aliquots was investigated per sample.

Results and discussion

The results of all measurements are listed in Table 1 and 2. From the first experiment, a-values ranging from 0.023 to 0.053 were determined (Table 1). Samples with linear beta dose responses gave an average a-value of 0.029±0.003 (LV96 has been excluded from this average a-value). The relative standard deviation (RSD) of 11% of this average does not originate from differences between regions, but seems to be sample-dependent as shown by LV96. The a-values of the second experiment are 0.033 and 0.041 (Table 2) and, thus, the a-value of samples with low doses seems to be independent of the measurement protocol. If the OSL-signal induced by a beta dose ($T_ß$) corrects L_α for sensitivity changes, the relationship between $T_ß$ and L_α should be proportional and the regression line should pass through the origin.

Figure 3(a) shows that the relationship is proportional for some aliquots only and that the regression line

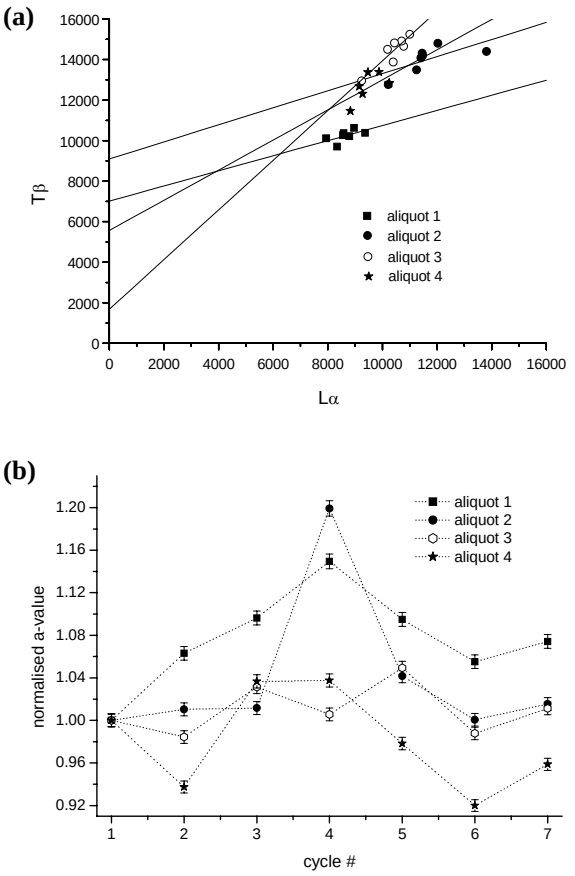

Figure 3: *Results from the sensitivity change test shown by sample LV06. Data were obtained using the protocol described in Figure 2, but using repeated alpha doses. An alpha regeneration dose of ~ 90 Gy was administered for 7 cycles (beta test dose was ~ 4 Gy). (a) Plot of $T_ß$ versus $L_α$; linear fitting was applied to the data set of each aliquot. (b) the a-values of the 4 aliquots of each alpha regeneration cycle normalised to the a-value of the first cycle.*

does not always pass through the origin (sample: LV06). This inter-aliquot discrepancy is reflected by the ~ 8% relative standard deviation of the SAR-derived a-values (Table 1). Moreover, Figure 3(b) shows that the a-value determined from constant alpha regeneration doses is affected by these sensitivity changes. Details plotted for one aliquot of the same sample indicate that the $T_ß$-OSL and the $L_α$-OSL behave differently with each cycle (Figure 4). Sensitivity changes induced by alpha irradiation are not sufficiently monitored by the $T_ß$-OSL and, consequently, aliquots to be used for a-value determination should be irradiated only once.

The a-value of samples showing a relatively high natural dose was determined by comparing $D_{eβ}$ and

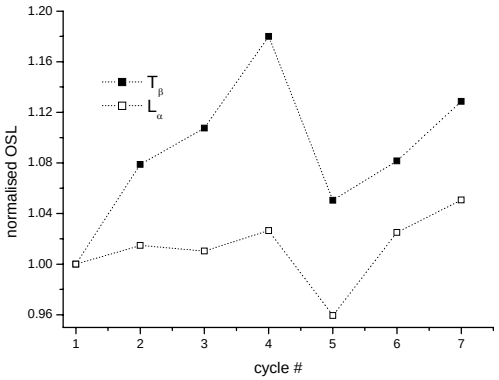

Figure 4: *Detail of the sensitivity test illustrated in Figure 3(a). $L_α$ and $T_ß$ of aliquot 1 (LV06), normalised to the first cycle are plotted separately against cycle number.*

$D_{eα}$. This comparison resulted in a-values of around 0.1 (Table 1). This value, however, is poorly constrained as it is based on the results from only two samples out of 10 samples analysed. The 8 other samples (not shown here) showed scattered dose response curves which could not be extrapolated. We believe that this is largely derived from large errors of individual dose points due to inappropriate experimental conditions (alpha source is not mounted on the luminescence reader but external). Nevertheless, the a-value of ~ 0.1 indicates that the a-value is dose-dependent. However, this result was obtained from adding charge induced by mono-energetic alpha particles to charge induced by alpha- and beta-particles and gamma photons. The different spatial distribution of charge resulting from alpha dose compared to beta and gamma radiation results in different dose response curves and, thus, this additive dose protocol is actually not accurate if the natural β-dose was relatively high. Aitken (1984) showed that the D_e is likely to be overestimated if a significant alpha dose is added to a beta dose which induced a non-linear OSL behaviour.

Conclusion

The a-values determined in this study range from 0.023 to 0.053 for samples with linear dose response. These are in agreement with a-values determined by additive dose protocols (Rees-Jones, 1995). Given the low α-effectiveness and apparent independence of sample origin, we conclude that a-values of 0.03 can be assumed for samples displaying a linear beta dose response curve allowing for an uncertainty of around 10%. With respect to the difference how alpha- and beta-particles deposit energy in silicates, comparison between regenerated alpha- and beta-doses could always be performed for low doses. We

further found in this study that sensitivity changes induced by alpha dose are not corrected by the beta dose induced test dose OSL and thus, aliquots should not be regenerated with alpha-doses but irradiated only once. The comparison between $D_{e\beta}$ and $D_{e\alpha}$ indicates that the alpha effectiveness of quartz increases with increasing dose.

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Reviewer

Norbert Mercier

Reviewer’s Comment:

During the past two decades, only a few studies aimed at understanding how alpha particles produce luminescence in silicates, especially in quartz. This paper is a useful contribution as it confirms some previous observations and justifies the use of a mean "standard" a-value for quartz. Moreover, it shows that the effects of alpha particles are still not well understood and, consequently, the paper is an incentive for further studies.