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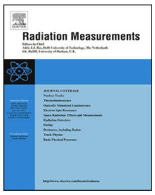

The effect of backscattering on the beta dose absorbed by individual quartz grains

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HIGHLIGHTS

- Modelling and experimental results show that over-dispersion increases with Z of substrate.
- Grain size and shape will contribute significantly to the over-dispersion in measured single grain dose distributions.
- Attenuation of the incoming spectrum can affect the over-dispersion in measured single grain dose distributions.

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ABSTRACT

We describe the effect on dose rates and over-dispersion (OD) of changing the spectrum of energies to which grains of various shapes and volumes are exposed during beta irradiation, either by changing the backscattering medium or attenuating the incident spectrum. Dose rates are found to increase when the atomic number of the backscattering substrate is increased (from 0.038 Gy/s on Al to 0.057 Gy/s on Pb), at the same time the dispersion due to grain shape and volume also increases slightly (9.4% on Al and 12.0% on Pb). By adding attenuators in front of the sample the net spectrum is also altered and the dispersion affected correspondingly. Our model prediction using various grain shapes and volumes are compared with experimental observations using sieved natural grains and the resulting dose rates are in good agreement, although the dispersions cannot be realistically compared in the absence of grain shape information for the natural material. We find from modelling that dose rates (both to grains in single grain discs and to those placed on the backscattering substrates) are sensitive to changes in shape and volume. A relative range across shapes of between 10 and 21% is observed from modelling on backscattering substrates, and of 7.4% from modelling in single grain discs. We conclude that it appears to be desirable to minimise shape and volume variations in grains if over-dispersion is also to be minimised.

1. Introduction

Dose distributions recorded by single sand-sized grains are now widely used in the dating of sediment samples, as a means of identifying and correcting for the effects of incomplete resetting prior to deposition and post-depositional mixing. However, for well-bleached unmixed natural samples the dose dispersion is much larger than that expected from known sources of uncertainty; even grains previously given a uniform dose by γ-irradiation show such over-dispersion (OD) when the dose is measured using a beta source, with the grains supported on some substrate. The dependence of dose rate on the atomic number of the substrate material has been investigated previously using both experiments (Murray and Wintle, 1979; Ingram et al., 2001) and modelling (Greilich et al., 2008); all 3 studies found that the dose rate increased with an increase in substrate atomic number. The dose rate also depended on the size of the sample grains; Wintle and Aitken (1977) observed a 25% lower dose rate at 4–11 μm than at ~100 μm, this was confirmed by Armitage and Bailey (2005) although they found the dose rate difference between that of 4–11 μm and 40 μm grains to be smaller (~12%). They also found no significant difference in dose rates for grain sizes >40 μm, observing a plateau in effective dose rates in the grain size range from 40 to 250 μm. Goedike (2007) reported a sharp decrease in

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dose rate below a grain size of ~40 μm while also observing no significant dependence for grain sizes between 40 and 130 μm. Above 180 μm the observed dose rates begin to decrease as the incident beam is attenuated while passing through the grains. Similar results were reported by Mauz and Lang (2005). Fain et al. (1999) used Monte Carlo modelling to investigate the effect of grain shape on absorbed beta doses; they concluded that for grains of ~200 μm diameter there is no effect.

As part of our investigation into the importance of low energy beta particles in dosimetry, in this study we consider the effect of the backscattering substrate on the dispersion of dose distributions measured using individual grains, by beta irradiating a monolayer of quartz grains on substrates of different atomic numbers (Z) varying from 13 to 82. The doses absorbed by these grains are then measured in an Al substrate (single grain disc) in the usual manner. We also investigate the effect on the observed OD of changing the incident spectrum shape using attenuators. The dependence of average dose and the contribution from backscatter as a function of Z are compared with modelling predictions based on Geant4, and the expected dispersion due to grain shape effects discussed.

2. Backscatter

2.1. Backscatter - modelling

We have used Geant4 (Agostinelli et al., 2003; Allison et al., 2006) to model beta dose absorption in quartz grains. A short summary of the physics involved is given in Supplementary Material Section 1. Similar work has been undertaken by Guérin (2011) and Guérin et al. (2012, 2015). Simulations were run using Geant4 version 10.02 patch 1 on the CentOS 7 distribution, run on a VMware workstation player 12 virtual machine, with the Penelope physics model (Salvat et al., 2001); this is well suited for the simulation of low energy particles and for electromagnetic interactions. A cut-off of 0.5 μm for the production of secondary particles was used (*i.e.* no particle is generated if its range is less than 0.5 μm, and the corresponding energy is instead deposited locally). This value was chosen to be less than 10% of the minimum dimension of our dose absorbers (grains).

In the model, primary particles are released individually and the release of the next particle does not occur until tracking of the previous primary and all generated secondary particles is complete. Tracking of a particle ceases when it reaches the boundaries of the world volume (at which point its energy is considered lost from the experiment) or its step size becomes shorter than the 0.5 μm cut-off. The release of primaries from the active source layer occurs isotropically. Each simulation is run for 10^8 primary electrons, corresponding to approximately 1/15 of a second of irradiation using a 1.5 GBq ^{90}Sr/^{90}Y source, this number of primaries provides us with good counting statistics (events per grain) as well as realistic computation times.

Individual geometrical components (structural, shielding and sample) are placed inside a predefined world volume otherwise filled with air and are given material properties based on user-defined elements and alloys. The irradiation geometry is constructed using the beta-source manufacturer's description (taken from Greilich et al., 2008), the characteristics of the irradiator and source flange (Dalsgaard, 2017 private communication), and a given grain geometry and substrate. The characteristic emission spectrum of the ^{90}Sr/^{90}Y active layer is defined as a cumulative probability distribution function (CDF) and given as input to the simulation. Beta particles were emitted with energies randomly sampled according to the Fermi model for beta spectra, modified by Behrens and Szybisz (1976). This spectrum is then modified by the source construction, the 0.125 mm thick Be window, and scattering by the flange, before arriving at the sample position. This part of the simulation is substantially the same as that undertaken by Greilich et al. (2008), and details of the irradiation geometry are given in Supplementary Material (Fig. S2).

The backscattering substrates used in the model are made of materials in the range $0 \leq Z \leq 82$. One hundred quartz grains are placed on a 10 × 10 grid, spaced with 600 μm between adjacent centres (this is similar to the layout of grains in a standard single-grain measurement disc; Bøtter-Jensen et al. (2003) except that in our case the grains are placed on the substrate surface rather than in holes in the substrate). Each quartz grain is uniquely labelled to allow the tracking of events within individual grains. The simulations record both the total energy deposited in each grain and the energy of any electron which enters any grain, the latter with a resolution of 1 keV.

To test the dependence of dose deposition on grain shape and volume, simulations were run for 7 different sizes and shapes of quartz grains: 100 μm or 200 μm diameter spheres, cylinders with a base radius of 150 μm and a height of 5 μm or 300 μm, and a cone of base radius and height of 150 μm. For comparing shape changes, a cylinder with base radius 50 μm and a height of 66 μm, as well as a cone of base radius 50 μm and a height of 200 μm were added. The latter two are chosen to have the same volume as the 100 μm diameter sphere; any differences in dose absorbed by these three grains types will thus arise because of shape and not volume. In contrast, dose absorption in the two spheres will be affected only by volume. In the two cylinders dose absorption will be affected by both shape and volume. Given that the backscattered spectrum should be softer than the incident spectrum, we expect to see a higher backscatter contribution to the absorbed dose in the short cylinder compared to the tall cylinder (See Supplementary Material Section 1, Fig. S1). Note that all grains fit completely within the grain holes of standard single grain discs.

The simulation records the spectra as follows: the energy of an electron is recorded (1 keV bin width) only if it (a) leaves a volume which is not the backscattering material and then (b) immediately enters the backscattering substrate, i.e. when an electron originating outside the substrate takes a first step in the substrate; secondary electrons generated within the substrate are not recorded. In the case of the incident spectrum (Fig. 1), there is no absorber (quartz grains) sitting on the substrate, and the substrate is vacuum; this gives the incident spectrum with no backscattered contribution. Backscattered spectra are recorded in a similar manner, except that a separate 200 μm thick air volume is placed directly on the substrate, covering the entire sample surface; all electrons leaving the substrate and then entering the air volume are recorded (Fig. 1).

The energy distribution of the backscattered electron spectra (Fig. 1a; for completeness, corresponding backscattered photon spectra and total photon and electron spectra are shown in Figs. S3a and b, respectively) does not change substantially from substrate to substrate. In contrast the intensity does change substantially, and at low energies and high substrate Z the backscattered intensity is comparable to that of the incident spectrum. As expected, the backscattered spectrum is softer than the incident spectrum, i.e. it contains a relatively larger low energy component; this can be seen more clearly in the normalized cumulative spectra shown in Fig. 1b (calculated by progressively summing all lower energy bins and then normalized by dividing by the total sum of all bins). It is clear that the backscattered spectrum tends towards the incident spectrum when the substrate Z increases.

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