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Deriving spatially resolved beta dose rates in sediment using the Timepix pixelated detector

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HIGHLIGHTS

- Timepix detector used to map beta dose rates in sediment.
- Geant4 simulations used to design measurement procedure.
- Method tested on an artificial micro-stratified sample.
- We show that natural dose rates can be mapped at sub-millimetre scales.

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ABSTRACT

Luminescence dating methods currently allow for the evaluation of the distribution of equivalent dose (D_e) values for individual sand-sized grains of quartz and feldspar from a given sample, but the environmental dose rate is still derived from the bulk sample. Additionally, single-grain optically stimulated luminescence (OSL) dating is performed on disaggregated samples, resulting in the loss of micro-stratigraphic context. To enhance the interpretation of D_e distributions, we aim to estimate the beta dose rate to sub-millimetre regions of intact samples using the Timepix pixelated semiconductor detector. The Timepix contains an array of 256×256 pixels, each $55 \times 55 \mu\text{m}$ in size and with its own preamplifier, discriminator and digital counter. The detector has a total sensitive area of 1.98 cm^2 , and 65,536 independent channels. The output of each measurement is a matrix containing the position and pixel-by-pixel count rate (or deposited energy) of each particle that interacted in the sensitive volume of the detector. The main challenge in using the Timepix detector is low natural sample activity, and the goal of this work is to acquire data with minimal background contribution. With an experimental setup guided by Geant4 simulations, progress has been made to greatly reduce background noise using ad hoc shielding and post-acquisition particle analysis. We have established a Timepix measurement procedure applicable to resin-impregnated sediment samples, including sample preparation, measurement, and data processing and analysis. These steps have been tested on an artificial micro-stratified sample (composed of quartz and biotite grains held together by resin) to derive the corresponding spatially resolved beta dose rates.

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1. Introduction

Optically stimulated luminescence (OSL) dosimetry has been widely used over the past 30 years as a dating method for naturally deposited sediments and implemented successfully on individual sand-sized grains of quartz and feldspar (Roberts et al., 2015). In the single-aliquot regenerative-dose protocol (Murray and Roberts,

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1998; Galbraith et al., 1999; Murray and Wintle, 2000), the equivalent dose (D_e) for a sample can be estimated from the distribution of single-grain D_e values. Single-grain dating enables the OSL behaviour of individual grains to be investigated and unsuitable grains rejected. It also permits an assessment of the extent to which grains were adequately bleached by sunlight before deposition and remained undisturbed thereafter.

The environmental dose rate, in contrast, is represented by an average value obtained from the bulk sample, usually after it has been homogenised in the laboratory. Beta particles can penetrate up to ~3 mm through most sediments and the coefficient of variation of beta dose rates in sediments can be large, owing to the spatially non-uniform distribution of radionuclides in the ^{238}U , ^{235}U , ^{232}Th decay chains and of ^{40}K . At low dose rates, the impact of beta dose heterogeneity will likely be high, such as in the Murray Basin of South Australia where the beta dose rates contributed 55% of the total activity to the dune sands (Lomax et al., 2007). Substantial spatial variations in beta dose rate may also arise in deposits with heterogeneous compositions, such as at Diepkloof Rockshelter in South Africa where the beta component constituted 50–70% of the total dose rate (Jacobs and Roberts, 2015). The use of an average beta dose rate may, therefore, not be appropriate for all single-grain D_e distributions (Jacobs et al., 2008; Galbraith, 2015; Martin et al., 2015; Roberts et al., 2015). Additionally, the extraction of grains for D_e estimation results in the loss of micro-stratigraphic context, so there would be benefits in being able to make measurements of the spatial distribution of beta dose rates on intact samples.

Measuring beta dose rates on intact sediment samples is difficult, because the low levels of radioactivity in nature make signal-to-noise ratios problematic as sample size decreases. For this reason, estimates of dose rate distributions have necessarily come through modelling, most prominently using Monte Carlo transport codes (Nathan et al., 2003; Martin et al., 2014, 2015; Guérin et al., 2015). These models use a simplified geometry of packed grains (usually spheres or ellipsoids), each with a given composition, and the deposited energy is recorded in a selection of ‘dosimeter’ grains.

Experimental validation of such models has required the artificial raising of the dose rate, so that the dose received by mineral grains can be detected (Nathan et al., 2003; Cunningham et al., 2012). Nathan et al. (2003) prepared sand boxes with uranium ore as the radioactive mineral, and allowed many months for the dose to build in the dosimeter grains; Cunningham et al. (2012) sped up the experimental process by using neutron-activated NaOH grains as the radioactive source. Unfortunately, these experimental enhancements are not possible for natural samples. A Monte Carlo model of the dose rate distribution is unlikely to capture the complexity of natural sediments, where mineral inhomogeneities and micro-stratification are common occurrences (e.g., Roberts et al., 2015; Fig. 5). Furthermore, since we are dealing with dose rates only slightly higher than background levels, it is essential to use a detector that has high sensitivity and is stable over long measurement periods (7–40 days).

Here we investigate the use of a measurement device, the Timepix, which has the potential to resolve this problem. The Timepix hybrid-pixel detector, developed by the Medipix2 collaboration, consists of a pixelated semiconductor detector sensor coupled to the Timepix readout chip (Llopert et al., 2007; Jakúbek, 2009). The sensor is made of 300 μm -thick high resistivity silicon, which consists of 256×256 pixels, each with dimensions $55 \times 55 \mu\text{m}$. Sensor pixels are individually Flip Chip[®] (Lau, 1996) bump-bonded to corresponding pixels in the Timepix readout chip that lies underneath. Each readout pixel has its own preamplifier, threshold discriminator and digital counter. The complete assembly has a total sensitive area of 1.98 cm^2 and 65,536 independent

readout channels (Fig. 1). With sub-millimetre spatial resolution, the detector has the potential to register emissions from individual mineral grains, as well as the sediment matrix in which they are embedded. Additionally, the detector is able to discriminate between particle types (alpha, beta, gamma and cosmic radiation), based on the morphology and energy deposition of the resulting clusters (Bouchami et al., 2011). This means that sources of background radiation (mostly from the gamma- and cosmic-ray components) can be suppressed, and the distribution of beta-like hits can be separated for analysis.

In this paper, we develop a procedure for using the Timepix to estimate the beta dose rate distribution in natural sediments. The proposed method for achieving spatially resolved dose rates is tested using an artificial micro-stratified sample prepared from grains of quartz and biotite (a dark mica mineral). To quantify the beta dose rate from the emitter grains, a calibration is performed using a sample prepared using an international standard of known ^{40}K radioactivity concentration distributed uniformly within the sample.

2. Materials and methods

2.1. Materials: the Timepix detector and simulation platform

2.1.1. Timepix measurement

The Timepix contains a matrix of semiconductor detectors – that is, pixels – and each pixel is able to measure the energy deposition in the “time-over-threshold” mode, where the charge collected in each pixel is recorded while the preamplifier output is over the threshold (Llopert et al., 2007); or each pixel can function simply as an event counter. Measurements are stored as a matrix containing information on the x-y location and energy deposition in each pixel (when an energy calibration is provided). The Timepix chip used in this experiment is part of the Jablotron MX-10 unit, which is factory-calibrated and comes with calibration parameters that can be imported into the acquisition software (Pixelman: Turecek et al., 2011), so that particle energy can be registered. Prior to data acquisition, a threshold equalisation procedure (Llopert et al., 2007) is carried out to compensate for pixel-to-pixel variations, and dead pixels are masked to avoid over-response.

Particle types are recognised based on cluster morphology (Holy et al., 2008). Beta particles usually have a cluster of pixel hits that appears as a long, curly track, or as single, double, triple and quadruple hits (Fig. 2). The shape of the cluster depends on the charge, velocity and mass of the incoming particle, as well as its angle of incidence. Clusters produced by gamma rays are similar to those of beta particles, as they mainly deposit energy by producing electrons (Teyssier et al., 2011).

Due to the typically low natural activity of environmental samples, background radiation contributes significantly to the total measured dose. We used several techniques to minimise incident radioactivity outside of the sample of interest, including shielding and cluster analysis. In this study, the Timepix detector was embedded in a 3 cm-thick lead castle for measurement acquisition, which stops most of the background radiation from reaching the detector (Fig. 1). A post-measurement cluster analysis was performed, based on the Medipix Analysis Framework (MAFada: Bouchami et al., 2011; Idarraga, 2012), to ensure that only beta particle tracks were counted in the dose rate evaluation. Hits registered above a specific threshold (which was determined in the equalisation process) were grouped into clusters. The clusters were then characterised based on cluster length in the x-y directions, energy deposition, and energy-weighted centres of mass. Beta particles were classified as ‘light tracks’ consisting of non-straight line morphology, as opposed to the straight tracks of minimum

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