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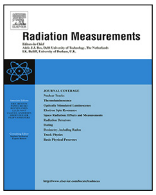

The issue of laboratory bleaching in the infrared-radiofluorescence dating method

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HIGHLIGHTS

- Describes the behavior of K-feldspars during bleaching experiments.
- The bleaching level of an IR-RF signal is dependent on the wavelength and on exposure duration.
- Proposes suitable bleaching parameters adapted to IR-RF measurements.

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ABSTRACT

Measuring the infrared radiofluorescence (IR-RF) signal is methodologically simpler than measuring the IRSL or TL signals. However, unlike classical luminescence measurements, the IR-RF signal is totally resetting when its highest value is reached. Previous studies (Buylaert et al., 2012; Trautmann et al., 2000) reported that the proper bleaching level of an IR-RF signal is difficult to define and seems to be dependent 1) on the wavelength used for bleaching and 2) on exposure duration. The IR-RF signals from K-feldspar samples have been measured with various bleaching wavelengths and exposure time using a *Lexsyg research* luminescence unit. In this study, we propose suitable bleaching parameters adapted to IR-RF measurements.

1. Introduction

Introduced for the first time in 1999, the infrared-radiofluorescence (IR-RF) protocol seemed promising for dating feldspars exposed to light before burial. The method allows evaluating directly the density of charges that have been accumulated in electronic traps over time, without involving any process of liberation and recombination as in the case of the measurement of the infrared stimulated luminescence (IRSL) signal. As a consequence, measuring the IR-RF signal should be methodologically simpler. Moreover, the dating of feldspars by IRSL and thermoluminescence (TL) has long been plagued by anomalous fading, a laboratory observation invoked as the cause of systematic age underestimations when dating feldspar (Aitken, 1998). A few studies have previously reported that IR-RF signal might yield accurate equivalent doses for K-feldspar grains. The absence of fading was deducted from the fact that IR-RF ages were consistent with expected ages (Degering et al., 2007; Erfurt, 2003; Novothny et al., 2010; Wagner et al., 2010), and because Krbetschek et al. (2000) mentioned that this signal seems to be stable over a period of storage of several months (without showing experimental evidences). Despite these promises, there were very few applications of the IR-RF technique in the context of dating, chiefly due to technical constraints. This method might improve our understanding of the physical mechanisms giving rise to luminescence within feldspars.

In IR-RF dating, regeneration of the IR-RF "decay curve" is achieved following laboratory bleaching and a pause. This bleaching step in the dating protocol is presumed representing the IR-RF level at time of deposition. However, it has been shown that the proper bleaching level of an IR-RF signal is difficult to define (Buylaert et al., 2012). In conventional IRSL and TL methods, the zero signal can be reached following exposure to light or heat, as it becomes distinguishable from instrumental background. In contrast, the IR-RF signal increases to a maximum intensity, during light exposure.

Properly defining the IR-RF "zero level" is of paramount

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importance and constitutes one of the pillars of any dating protocol. We show here that this level seems to be dependent 1) on the wavelength used for bleaching and 2) on the exposure duration, whatever are the origins and natural doses of the samples.

2. Materials and measurement conditions

The samples have been selected according to these criteria:

(i) they are expected to have been well exposed to daylight before burial,
(ii) they could be associated with an independent age control (radiocarbon dating, U/Th…),
(iii) in the absence of independent age control, reliable luminescence ages are available,
(iv) they have different natural accumulated doses (from low to saturated dose).

The information about the geological setting of the samples and their age control are reported in Table 1. They carry a large spectrum of accumulated doses, from zero to more than 3000 Gy.

The coarse grains fraction (180–250 μm) of the six sediment samples were extracted and treated with hydrochloric acid (10% HCl) followed by hydrogen peroxide (30% H_2O_2) for 24 h in order to remove carbonates and organic matter, respectively. The coarse K-feldspar-rich fraction was then isolated using a heavy liquid solution (LST) at the density of $\rho < 2.58$ g cm^{-3} (up to three successive cycles). No HF acid etching was used with the feldspar preparation (Duller, 1992), to avoid splitting or disintegrating grains. Minerals are then passed through a Frantz magnetic separator (Mauz and Lang, 2004), in order to remove iron oxides and other minerals with low magnetic susceptibility.

Radioluminescence stimulation was achieved in a *Lexsyg* Research system from Freiberg Instruments (Richter et al., 2013). The aliquots placed on the storage wheel are physically isolated from the aliquot residing inside the measurement chamber, thus preventing any cross-talk effects. For measurements, the aliquot is mechanically transported from the sample wheel to the measurement chamber where it is placed on a heater plate, attached to a rotating arm. The heated plate consists of a metal plated dielectric heater, which can raise the temperature up to 700 °C.

The device is equipped with a Sr-90 beta irradiation unit delivering a dose rate of 0.06 ± 0.003 Gy s^{-1}. It was specifically designed as a ring source to achieve optimum spatial uniformity across the aliquot and also to allow RF measurements during beta irradiation (Richter et al., 2012). To measure the emission for IR-RF during irradiation, a Hamamatsu H7421-50 photomultiplier was employed, along with a Chroma D850/40 interference filter, allowing detection between 810 and 890 nm.

An internal solar simulator included within the measurement chamber was used for bleaching experiments. It is equipped with six LEDs: UV (365 nm), blue (462 nm), green (525 nm), amber (590 nm), red (625 nm) and IR (850 nm). Their optical power is regulated via a feedback loop.

All measurements were performed in a nitrogen atmosphere. Erfurt and Krbetschek (2003) had previously observed an increase in dispersion in their measurements, whenever they had to handle the discs out of the system. They hypothesized that it might be due to a change in the measurement geometry, either by having placed the disc in a different orientation or by grain movements. Here, our K-feldspar grains were most of the time dispensed on nickel cups and we took great care to prevent the movement of grains whenever handling was required.

3. Signal resetting

In any luminescence dating method, the signal must be susceptible to being reset by natural sunlight and the level reached during this process needs to be reproduced with laboratory procedures (Trautmann et al., 1999). For the IRSL signal, the zero level is reached after a reasonably short exposure, and the residual level becomes quickly indiscernible of the instrumental background. On the contrary, the IR-RF signal increases during optical bleaching since this signal reflects the probability to trap charges in the IR-sensitive traps (Krbetschek et al., 2000; Trautmann et al., 1999). The highest probability is then reached when all the traps are empty. Thus, by measuring the IR-RF signal as a function of the bleaching time, the signal intensity tends to a single value. For natural sunlight, this plateau is reached after 2–5 h (Trautmann et al., 1999), but wavelengths lower than 500 nm produce the same effect (Trautmann et al. (2000; their Fig. 2). Krbetschek et al. (2000) used a solar simulator to bleach their samples and achieved complete bleaching within a few minutes. Buylaert et al. (2012) used a UV bleaching unit (395 nm UV LED; 1 W optical power) and reached a bleaching plateau after a few minutes of exposure. However, these authors showed that the zero levels were different, whether it was achieved with a UV LED or with a solar simulator.

In our first observation, we investigated the resetting behavior of the IR-RF signal for different wavelengths and bleach durations. The following single aliquot protocol was designed for this task: the bleaching experiment was divided in steps and at each bleaching step, we monitored the IR-RF signal by inducing a short, (5 s i.e. ~0.3 Gy), beta exposure (hereafter named short IR-RF signal, or simply “sIR-RF”). The natural signal was probed using this sIR-RF for each successive bleaching time, from 10 s to ~90 000 s (~25 h). Each bleached sIR-RF signal was normalized by the first sIR-RF bleached signal (e.g. the one obtained after 10 s).

Erfurt and Krbetschek (2003) pointed out that samples exhibit strong phosphorescence immediately after bleaching. Due to the possible superposition of the phosphorescence and the radiofluorescence, these authors recommend to wait at least 1 h after bleaching. Recently, Huot et al. (2015) shows that phosphorescence is thermally dependent: as the temperature increases during the bleaching step, so does the waiting time to cool the sample to room temperature. Thus, in this study we decided to measure the bleached sIR-RF signal after a pause from few minutes to 1 h, in function of the bleaching time.

Table 1
Summary of geological details and previous dating results on studied samples.

Sample	Location	Site	Origin	IRSL		Age control		
				De$_{fc}$ (Gy)	Age (ka)	Method	Age (ka)	Reference
TH0	Morocco	Sebka Tah	Aeolian	0.18 ± 0.01	~0	*Stratigraphy*	*Modern*	Bouab, 2001
TH8	Morocco	Sebka Tah	Aeolian	76 ± 9	50 ± 4	*n.d.*	*n.d.*	Bouab, 2001
C5	Peru	San Mateo Paleogulf	Marine sand	104 ± 11	76 ± 9	U/Th	85 ± 2	Pedoja et al., 2006
TML1	Canada	–	Fluvial	>3000		*Stratigraphy*		