



ASPECTS OF INFRARED RADIOLUMINESCENCE DOSIMETRY IN K-FELDSPAR

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Abstract: Infrared radioluminescence (IRRL) of K-feldspar, detected at peak wavelength of 865 nm, is emerging as a potential geochronometric tool. The present study explores and attempts to optimize the IRRL dating protocols and proposes a revised protocol for estimation of palaeodose. UV light (395 nm; 700 mW/cm²) bleach of 800 s was optimum to remove the trapped charges responsible for IRRL and, reduced the interference of radio-phosphorescence due to prior irradiations. Validation of the proposed protocol was carried out by dose recovery tests on mineral and sediment K-feldspar samples of different provenances. An overestimation in dose recovery was observed and was attributed to difference in sensitivity of natural IRRL and regenerated IRRL. The sensitivity changes were significant and systematic and were documented by repeating bleach–IRRL cycles. Corrections for sensitivity changes between natural and regenerated IRRL, gave reliable results and, have now been included in the proposed dating protocol.

Keywords: infrared radioluminescence, radio-phosphorescence, bleaching of IRRL, sensitivity correction, K-feldspar, geochronology.

1. INTRODUCTION

Natural ubiquitous minerals like quartz and feldspar have been used for the dosimetry of natural radiation environment for geochronometric applications. These applications use the fact that luminescence signal increases with radiation dose in a cumulative manner and that, as a first approximation, annual radiation dose arising from the decay of natural radioactivity is constant over time-scales of interest. Similar to thermoluminescence (TL) and optically stimulated luminescence (OSL), radioluminescence (RL) is dose dependent. The stored radiation dose (termed equivalent dose or palaeodose) measured using RL when divided by the annual dose rate provides the age. Trautmann *et al.* (1998; 1999) reported that RL

signal in infrared (IR) region helps to monitor trapping of electrons and the recombination of electrons can be monitored using RL signal in ultraviolet-visible (UV-VIS) region. RL in both spectral windows is dose dependent and has been developed as a dating/dosimetric tool. Present study examined RL in IR region for its use as a dating technique.

Infrared radioluminescence (IRRL) is the luminescence emission in IR region due to radiative trapping of electrons stimulated by irradiation from ionizing radiations (radiation dose). IRRL in K-feldspar (microcline and orthoclase; Trautmann *et al.*, 1999) has a peak emission at ~865 nm (Trautmann *et al.*, 1999; 2000; Trautmann, 2000; Krbetschek *et al.*, 2000; Erfurt and Krbetschek, 2003). IRRL in feldspars has a higher onset of saturation dose (~1500 Gy) as compared to few hundred Gy for TL and OSL signals of natural ubiquitous

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minerals like quartz and feldspar, and this makes IRRL of K-feldspar a candidate to extend the dating range of luminescence methods (Erfurt and Krbetschek, 2003).

Natural samples accumulate radiation dose in the environment. Thus during an IRRL measurement of the natural sample in laboratory, the stimulating irradiation becomes an added dose to the natural one. The trapping probability of charges is highest if the traps are empty and lowest when the traps are full. Consequently the IRRL intensity decreases with increasing dose, leading to a decay curve for the sample being observed and this provides a means to convert the intensity to radiation dose. The maximum intensity in decay curve corresponds to zero dose and minimum intensity corresponds to saturation dose in the sample (Fig. 1).

Erfurt and Krbetschek, (2003) developed a protocol for dating sedimentary K-feldspar using IRRL. The measurement sequence was; natural (additive) IRRL + 30 min bleach under solar conditions + 1 h pause (waiting) for the decay of radio-phosphorescence + regenerated IRRL. A stretched single exponential function was fitted to regenerated IRRL and natural IRRL was then interpolated over the fitted curve to compute the equivalent dose (D_e) in the sample. This protocol was refined by Buylaert *et al.* (2012) as: Natural (additive) IRRL + Bleach 1500 s (UV light – 395 nm, 700 mW/cm²) + 1 hr Pause + Regenerated IRRL.

In this study, D_e was estimated by simply displacing a part of natural IRRL curve along time (or dose) direction (x-axis) towards the regenerated curve (Fig. 1) and deviation between their intensities was estimated. The dis-

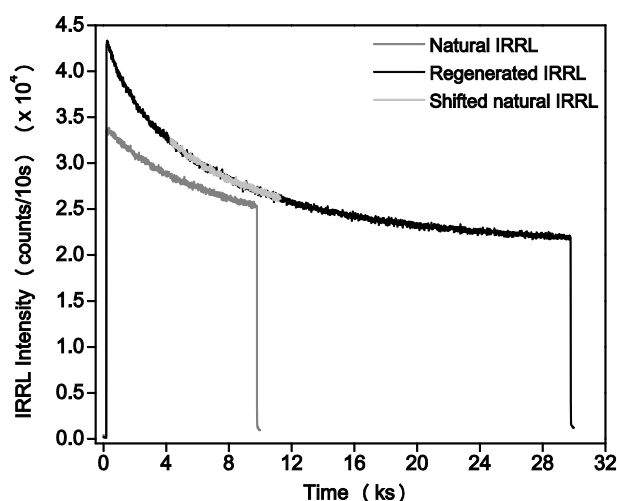


Fig. 1. Decay curve of IRRL for mineral K-feldspar. Natural is measured after dose of 200 Gy and hence has lower intensity as compared to regenerated IRRL which is measured after complete bleach of sample. Natural is then displaced along x-axis or time to minimize its deviation from regenerated. This displaced curve is seen to be overlapping on regenerative IRRL curve. The analysis is done using MATLAB based program.

placement on the time axis which gave minimum deviation between the two intensities (best fit) was accepted as the D_e . The errors were calculated from the coefficient of variance. In this interpolation, the analytical form of equation to be fitted to IRRL decay curve was not required. We adopted this approach for the present study.

Protocols by Erfurt and Krbetschek (2003) and by Buylaert *et al.* (2012) recognized the presence of radio-phosphorescence along with IRRL signal as interference. A pause for 1 hr in protocols was therefore introduced to reduce the radio-phosphorescence associated with main IRRL signal. As for bleaching, Erfurt and Krbetschek (2003) proposed 30 min sun bleach and Buylaert *et al.* (2012) prescribed 1500 s bleaching using 395 nm light. Additionally both groups observed sensitivity changes due to repetitive measurements but did not examine this aspect further. With a view to further optimize these protocols, the objectives of the present study were:

- 1) assessment of interference of radio-phosphorescence during IRRL measurement,
- 2) estimation of optimum bleaching time for IRRL,
- 3) estimation of the magnitude of change in sensitivity during repeated measurements and estimation of appropriate correction factor,
- 4) examination of the reliability of new modified protocol through dose recovery tests.

2. SAMPLE PREPARATION AND EXPERIMENTAL DETAILS

For the present study, a museum microcline mineral sample (Morthekai *et al.*, 2012) and four K-feldspar samples from sediments with different provenances were examined. Most experiments were first conducted on museum samples and then applied to sediment samples.

Sample preparation

The museum microcline mineral was gently crushed in the presence of alcohol, dried and sieved to collect grains of 90-210 μ m size for IRRL measurements. Extraction of feldspars from sediments followed the standard treatment with 1 N HCl (to remove carbonates), 30% H₂O₂ (to remove adhering organic fraction), sieving for 90-210 μ m grains, extraction using Frantz magnetic separator (Porat, 2006), and finally a density separation using sodium polytungstate (2.58 g/cc). Both, mineral and sediment feldspar grains were mounted on stainless steel discs over 3 mm diameter using *Silkospray*.

Measurement conditions

IRRL measurements were carried out using an automated Risø TL/OSL reader with a RL attachment (Buylaert *et al.*, 2012; Lapp *et al.*, 2012). For stimulation, a ⁹⁰Sr/⁹⁰Y β source delivering a dose rate of 0.055 Gy/s was used, and the experiments were performed at room temperature. The system was fitted with a Hamamatsu

H7421-50 photomultiplier tube with a spectral response in the region 380-890 nm. An interference filter, Chroma D 900/100 with a band pass transmission at 850-945 nm was used and the net transmission of filter and the photomultiplier tube was in the region 855 ± 27 nm. Bleaching was done using UV LEDs of 1 W optical power, emitting at 395 nm with 700 mW/cm^2 irradiance on the sample over the disc.

Experiments

Radio-phosphorescence and bleaching

The radio-phosphorescence and optimization of bleaching time were studied using the sequence given in **Table 1a**. Initially the aliquots were bleached to remove any stored dose. Buylaert *et al.* (2012) proposed that a 1.5 ks of UV exposure was sufficient for complete bleach of IRRL signal. We applied an even longer bleaching time of 2 ks for initial bleach. After bleaching, the aliquots were given 800 Gy β dose. Then these aliquots were immediately bleached for different durations, ranging from 0 to 2 ks to estimate its effect on IRRL signal. Radio-phosphorescence was then recorded for 3 ks keeping β source in OFF position followed by IRRL for 5 ks keeping β source ON and then again radio-phosphorescence for 3 ks keeping β source OFF. Thus radio-phosphorescence signal before and after IRRL read-out was measured. The above protocol was performed using two approaches – multiple measurements on single aliquot and single measurement on multiple aliquots. Thus the complete protocol was executed for single aliquot and among the multiple aliquots, each aliquot represented one bleaching time after irradiation i.e. 7 aliquots for 7 different bleaching times.

Dose recovery tests

Dose recovery tests for 200 Gy were carried out for 5-6 discs of mineral and sediment samples and the used protocol is shown in **Table 1b**. The samples were initially bleached for 800 s to remove any stored dose and then were given a β dose of 200 Gy. Considering 200 Gy as the natural dose of sample, natural IRRL was recorded for 10 ks (200 s radio-phosphorescence + 9.6 ks IRRL + 200 s radio-phosphorescence). Aliquots were then bleached for 800 s and regenerated IRRL was recorded for 30 ks (200 s radio-phosphorescence + 29.6 ks IRRL + 200 s radio-phosphorescence). Here, regenerated IRRL experienced a time delay of 1 ks (800 s bleaching + 200 s radio-phosphorescence) before its measurement, whilst the natural did not. Thus to make identical read-out conditions, we incorporated an extra pause of 800 s between irradiation and the read-out of natural IRRL (**Table 1b**, step 3). This protocol for dose recovery included 800 s bleach at initial stage to remove any stored dose in the sample and then a known dose was given. Another dose recovery was performed where 1 h UV light bleaching

was applied to samples at initial stage to assure the results (**Table 1b**). Additionally, changes in IRRL sensitivity were observed, when the IRRL signal was repetitively recorded, i.e. a loop of a bleach and IRRL measurement. Sensitivity changes during multiple read-outs were computed using 5 cycles of bleach-IRRL measurement (**Table 1b**).

For the dose recovery analysis, natural IRRL in the time interval of 1-8 ks was compared with the regenerated IRRL. The reason for using 1-8 ks signal was that the initial 200 s corresponded to radio-phosphorescence and the following 800 s IRRL data showed small increase in intensity (Buylaert *et al.*, 2012) and hence was not included in the data analysis. A program in MATLAB R2010a was developed to analyze the dose recovery data with the sensitivity corrections. The program code yielded results comparable with Risø RL analyst. The approach to calculate the equivalent dose and associated error in our program is the same as Buylaert *et al.* (2012).

3. RESULTS AND DISCUSSION

Radio-phosphorescence

The radio-phosphorescence decay curve along with IRRL of mineral K-feldspar immediately after irradiation of 800 Gy dose and with 0 s bleaching is shown in **Fig. 2a**. The figure indicates that typically, the initial radio-

Table 1. Experimental sequences.

Step	Treatment	Comment
a)		
1	2 ks Bleach	UV light (395 nm)
2	800 Gy β dose	
3	Variable bleaching time (t_i)	$t_i = 0, 50, 100, 200, 400, 600, 800, 1 \text{ k}, 1.2 \text{ k}, 1.6 \text{ k}, 2.0 \text{ ks}$
4	3 ks radio-phosphorescence	β source OFF
5	5 ks IRRL	β source ON
6	3 ks radio-phosphorescence	β source OFF
b)		
1	Bleach	UV light (395 nm) for 800 s or 1 h
2	200 Gy β dose	
3	800 s pause	To make identical read-out conditions for natural and regenerated IRRL
4	10 ks Natural signal (200 s radio-phosphorescence + 9.6 ks IRRL + 200 s radio-phosphorescence)	
5	800 s bleach	} 6 cycles
6	30 ks Regenerated signal (200 s radio-phosphorescence + 29.6 ks IRRL + 200 s radio-phosphorescence)	

phosphorescence signal corresponding to 0 s bleaching is $\sim 35\%$ of the laboratory regenerated IRRL signal. This could interfere with the succeeding IRRL signal. Thus earlier protocols included a 1 h pause even after application of bleaching time (1.5 ks), and concluded that the bleaching time for IRRL signal was not sufficient to reduce radio-phosphorescence. As from the sequence of **Table 1a**, analysis of initial 3 ks data indicated that in every case, the radio-phosphorescence signal corresponding to a particular bleaching decreased with measurement time of 3 ks as seen in **Fig. 2a**. And as the bleaching time increased this signal itself decreased (**Fig. 2b**). A decrease in radio-phosphorescence with bleaching time was seen and this was ~ 100 photon counts/10 s after ~ 500 s of bleaching (**Fig. 2b**), which is $<1\%$ of IRRL intensity of $\sim 10^4$ - 10^5 photon counts/10 s. Thus the radio-phosphorescence measurement time as well as bleaching time plays a role in reducing the radio-phosphorescence signal (**Fig. 2c**). The figure shows integrated radio-phosphorescence signal measured after each bleaching time plotted against the bleaching time. This also includes a radio-phosphorescence decay curve after 0 s bleaching plotted against its measurement time of 3 ks. Both the decays suggest that the bleaching time after irradiation does not influence radio-phosphorescence but rather acts as a time delay. Thus as mentioned the time delay > 500 s between cessation of irradiation and initiation of laboratory IRRL measurement is sufficient to reduce it to background level compared to IRRL. With 500 s bleaching, the reduction of signal while recording it for 3 ks is $\sim 0.08\%$ compared to IRRL. These imply that a saving of measurement time is possible by neglecting 1 h pause in the protocol if the bleaching time is > 500 s.

Bleaching time

The effect of bleaching using UV LED (395 nm) for IRRL signal was examined by analyzing 5 ks IRRL signal of the sequence shown in **Table 1a**. With the increase in bleaching time, IRRL intensity should increase for both the approaches; single and multiple aliquots. Results of sequence on single aliquot are plotted in **Fig. 3a**. Here no normalization was made, instead the maximum intensity was taken as unity to facilitate easy comparison between data. As seen from figure, IRRL intensity increased with increasing bleaching and attained a maximum value with 500 s of bleaching after which no substantial increase was observed. Any increase in normalized counts after the bleaching of 500 s could be due to a change in sensitivity.

To circumvent the sensitivity changes in single aliquot, we used multiple aliquots for the same sequence. Each aliquot was used for a particular bleaching time and the intensity was normalized by 2 ks bleached IRRL signal recorded after the measurement. **Fig. 3b** shows that normalized IRRL intensity increased up to 500 s of bleaching and then saturated at this value with no further increase for longer bleaching duration. From both, single

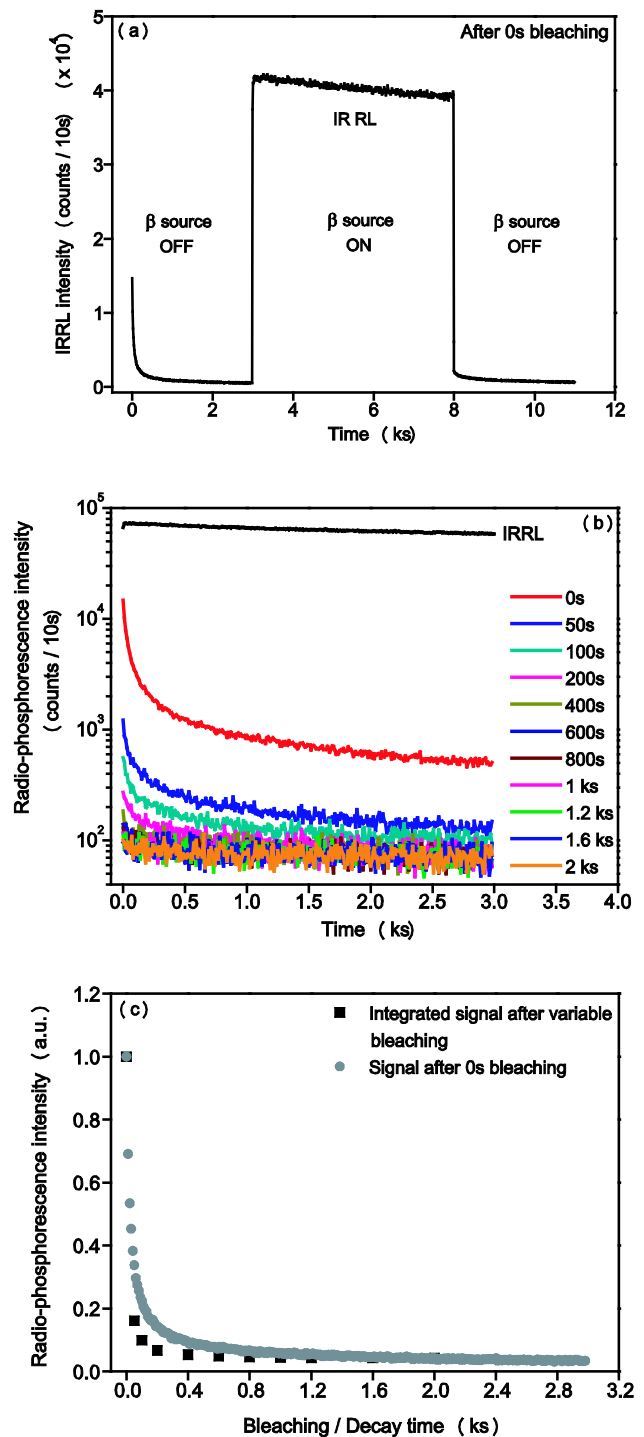


Fig. 2. a) IRRL signal (3 ks radio-phosphorescence + 5 ks IRRL + 3 ks radio-phosphorescence), immediately after irradiation. b) Radio-phosphorescence decay curves emitted due to 800 Gy dose measured for 3 ks after different bleaching time (0-2 ks). For comparison of intensities, IRRL curve is included. c) The two curves represent radio-phosphorescence decay. Grey circles represent signal against decay (measurement) time – 3 ks recorded after irradiation. The black squares represent the integrated (0-1 ks) signal corresponding to particular bleaching plotted against different bleaching time. The two curves are together to compare their nature of decay. Both the curves are normalized with respect to the initial intensity = 1.

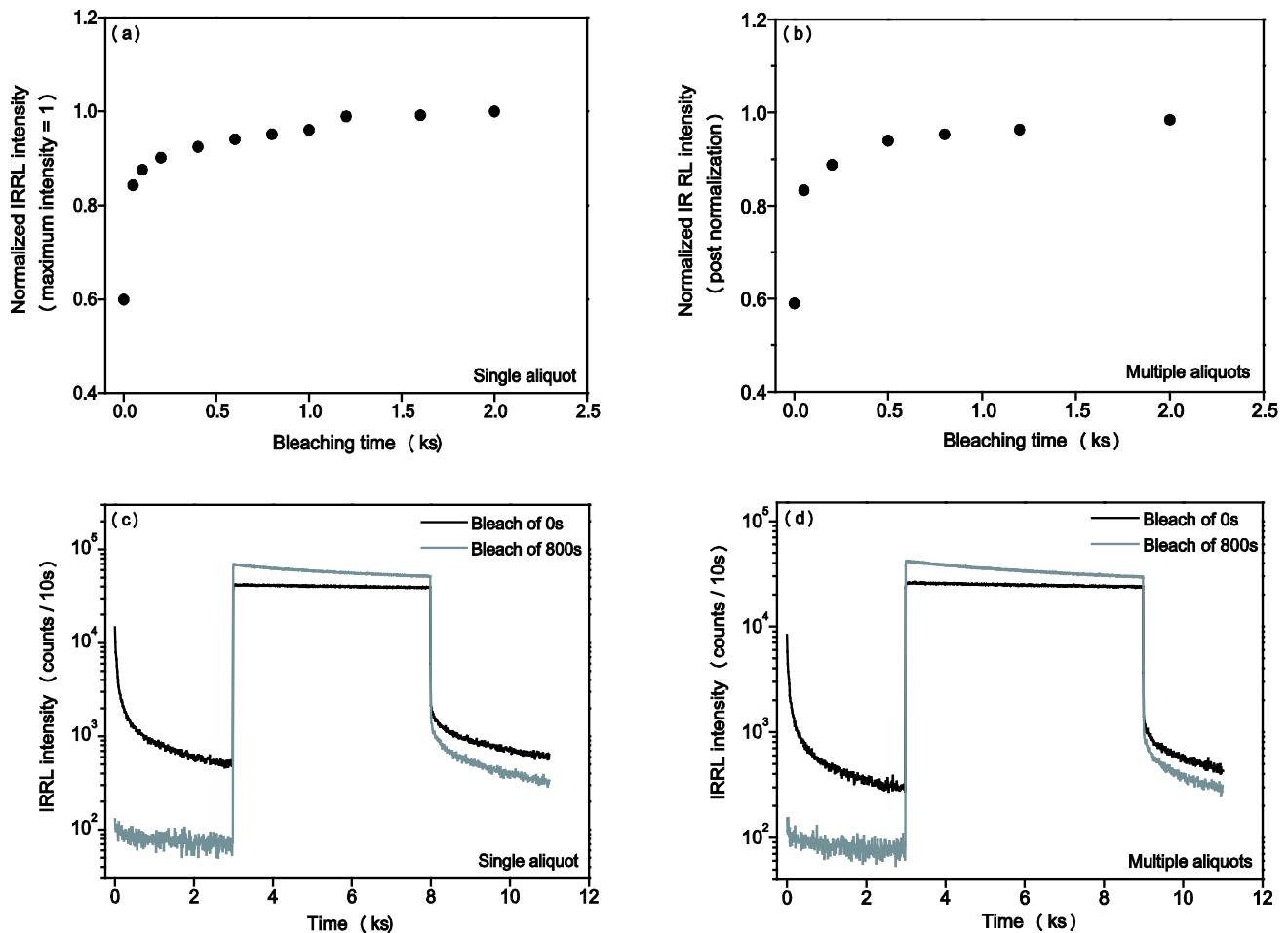


Fig. 3. Bleaching effect on IRRL intensity. An increase in intensity is observed with increase in bleaching time, when recorded for a) single aliquot and b) multiple aliquots. For single aliquot normalization is by taking maximum intensity = 1. In multiple aliquots, each aliquot is normalized using 2 ks bleached IRRL, recorded at the end of each measurement. This clearly indicates, after 800 s bleaching the IRRL signal reaches to maximum and the trend is parallel to x-axis. Two extremes (0 s and 800 s bleaching) IRRL curves (including radio-phosphorescence) are presented both for c) single aliquot and also for d) multiple aliquots to see the artifact of radio-phosphorescence and bleaching duration.

and multiple aliquots approaches the optimum time for bleaching IRRL signal completely was 800 s. A comparison between IRRL, immediately after irradiation and that after 800 s bleaching, both for single aliquot and multiple aliquots, is shown in Fig. 3c and d respectively wherein a reduction in radio-phosphorescence and an increase in IRRL signal is observed.

The 800 s bleaching time was examined further with another experiment of dose recovery test for 200 Gy. Sample TR 9 was initially bleached with UV light for 1 h, sufficiently long enough to remove any stored dose and then dose recovery was performed where natural IRRL for 200 Gy was recorded; bleached for 800 s and then regenerated IRRL was recorded (Table 1b). The experiment resulted in dose recovery ratio of 1.13 ± 0.03 which confirmed 800 s as optimum bleaching time (Table 2).

Fig. 2 and the nature of radio-phosphorescence suggest that the required bleaching time (800 s) exceeds the time required for radio-phosphorescence to decay to

background level and therefore it is suggested that no extra pause as was suggested by Buylaert *et al.* (2012), is needed. Bleaching of 800 s is sufficient to bleach IRRL and to minimize the radio-phosphorescence to its residual level.

Thus the modified protocol based on the studies so far was: natural (additive) IRRL + bleach (UV; 395 nm; 700 mW/cm²) 800 s + regenerated IRRL

Dose recovery and sensitivity change

To study the reliability of new protocol, a dose recovery test of 200 Gy on mineral and sediment samples was done using 5-6 discs. The sequence included 200 s of radio-phosphorescence before and after IRRL measurement and hence effective pause between the cessation of irradiation and IRRL measurement was 1000 s (800 + 200) (Table 1b). Extra radio-phosphorescence read-out was included for observing the signal. The experiment

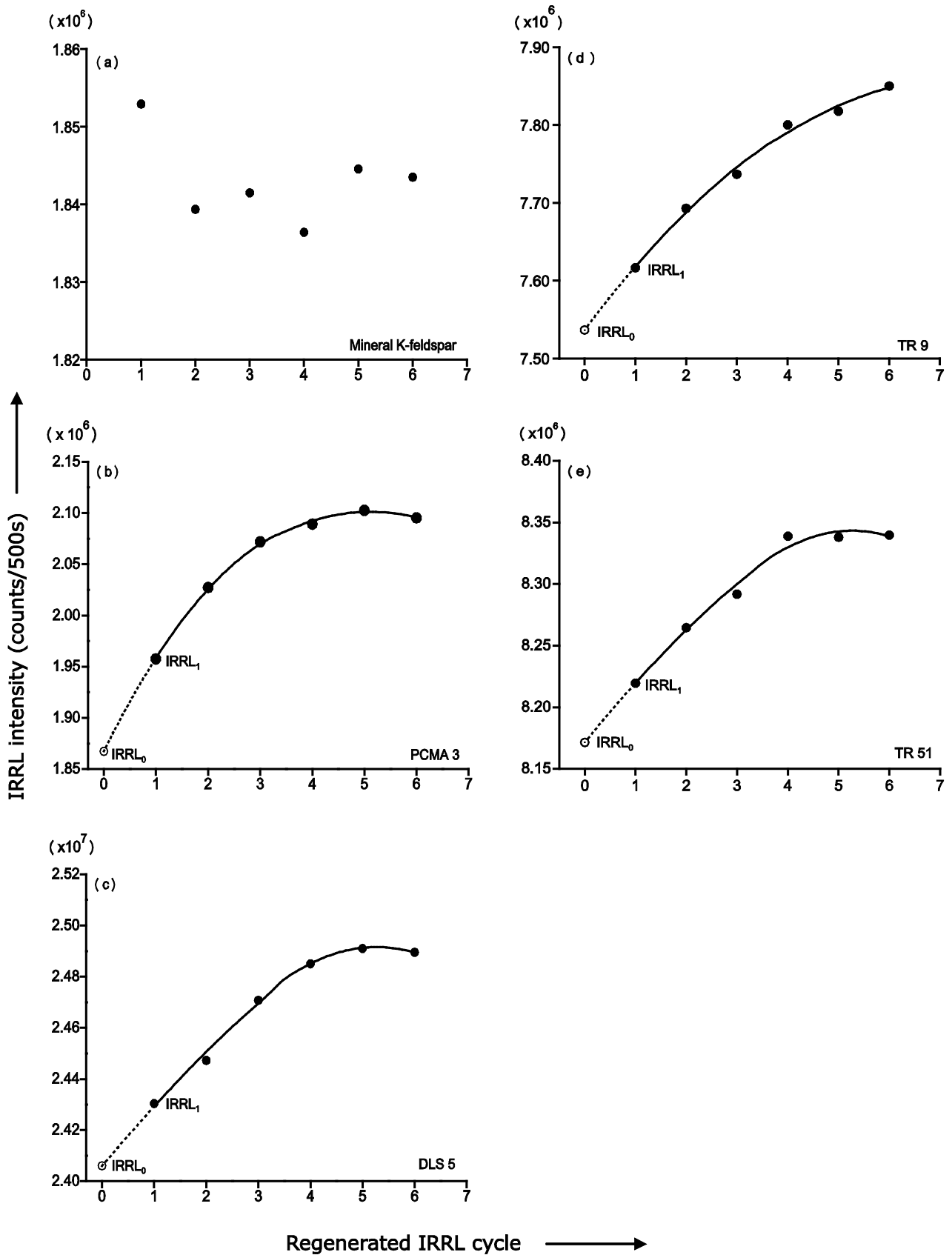


Fig. 4. Regenerated IRRL plotted against regenerative cycle for mineral which shows no trend, while sediment samples showing an increasing sensitivity change which is spline fitted to calculate sensitivity correction factor F_s .

Table 2. The result of dose recovery experiments with and without sensitivity correction. Here dose recovered is the average of 5-6 discs and the corresponding errors are standard error. The correction factor was measured for 2-3 discs and averaged F_S was assumed same for all discs.

Sample name	Sample type	Without sensitivity correction		With sensitivity correction		
		Recovered dose (Gy)	Dose recovery ratio	Correction factor (F_S)	Recovered dose (Gy)	Dose recovery ratio
Microcline	Museum	195±4	0.97±0.02	1.00	195±4	0.97±0.02
PCMA 3		331±5	1.66±0.03	1.04	190±3	0.95±0.01
DLS 5	Sediment	246±12	1.23±0.06	1.01	222±12	1.11±0.06
TR 9		256±5	1.28±0.02	1.01	232±5	1.16±0.02
TR 51		214±5	1.07±0.02	1.01	201±4	1.00±0.02
TR 9*		256±3	1.28±0.01	1.01	226±3	1.13±0.03

* With initial 1 h UV light bleaching.

resulted in over-estimation of recovered dose. Also when IRRL was recorded repetitively after same bleaching, a systematic increase in sensitivity between these regenerated IRRL was seen. Since the regenerated IRRL intensities increased with successive runs, we inferred that the natural IRRL was measured at a lower sensitivity compared to the first regenerative IRRL. Fig. 4 gives the trend of increase of IRRL sensitivity with cycle number and this could be used as a tool to trace back the sensitivity during natural IRRL read-out and incorporate a sensitivity correction factor, F_S in natural IRRL. By incorporating this, intensity of natural changes and the basic assumption, natural and regenerative readout should be made under identical sensitivity condition, is fulfilled.

Firstly without considering any sensitivity changes, average dose recovered with standard error from mineral K-feldspar was 195±4 Gy which gives average dose recovery ratio of 0.97±0.02. In the case of sediment feldspar, dose recovery gave an over-estimation such that the average recovered dose for a sample was 318±5 Gy (dose recovery ratio 1.59±0.02) (Table 2). This over-estimation implied the need for the sensitivity correction.

Using regenerated IRRL we aimed to estimate the sensitivity of sample at the time of measurement of natural signal. Thus graph of regenerative intensity against cycle of repetition was plotted. In the absence of a clear elucidation of the mathematical form of the trend followed by sensitivity (increasing intensities) of different samples, a numerical spline fitting was used to extrapolate the curve to natural or zeroth cycle (IRRL₀) using SigmaPlot 11.0 software. Equivalently, the ratio of first regenerated IRRL (IRRL₁) to the obtained IRRL₀ gives the required sensitivity correction factor $F_S = \text{IRRL}_1/\text{IRRL}_0$. This factor was then multiplied with measured natural and analyzed for dose recovery. In the present work, mineral feldspar showed no sensitivity change (Fig. 4a) resulting in $F_S=1$ and for sediments a trend was seen (Fig. 4b, c, d and e). For each sample F_S

was computed and the results of sensitivity corrected dose recovery are given in Table 2. From the table we can observe that sensitivity correction helped to identify sensitivity changes in the read-out of the natural IRRL signal. By incorporation of F_S , over-estimation reduced to acceptable values of recovered dose and hence we could appropriately trace the sensitivity changes between natural and regenerated IRRL. This could also be used to trace the sensitivity changes while measuring palaeodose in natural sediment samples.

4. CONCLUSIONS

The present study enables the following inferences based on our samples,

- 1) a bleaching of 800 s with 395 nm UV LED of power 700 mW/cm² is an optimum choice to bleach IRRL signal,
- 2) radio-phosphorescence signal reduces to minimum within 500 s and hence an extra pause of 1 hr after bleaching with 800 s, is not needed,
- 3) sample dependent sensitivity change was observed. Mineral sample did not show any change but was significant for sediment samples. This change can be traced using spline fitting for regenerative IRRL signals and using it sensitivity correction factor F_S can be calculated. Thus bleach-IRRL cycles led to improved results and this provides a means to ensure acceptable IRRL ages.
- 4) the modified protocol is: natural (additive) IRRL + bleach 800 s + regenerated IRRL + bleach-IRRL₅ cycles. The additional bleach-IRRL₅ cycles is a time consuming step. So F_S can be calculated for 3-5 discs, and the averaged F_S can be used for rest of the discs,
- 5) This new protocol including sensitivity correction factor (F_S) could be used for palaeodose estimation using IRRL for natural sediment samples.

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