IRSAR – A single-aliquot regenerative-dose dating protocol applied to the infrared radiofluorescence (IR-RF) of coarse-grain K feldspar

G. Erfurt* and M.R. Krbetschek

Sächsische Akademie der Wissenschaften zu Leipzig, Quaternary Geochronology Section at Institute of Applied Physics, TU Bergakademie Freiberg, Bernhard-von-Cotta-Str. 4, 09596 Freiberg, Germany

*corresponding author: Gunter Erfurt, Phone +49 3731 392163, Fax +49 3731 39 4004, Email address: gerfurt@physik.tu-freiberg.de

Abstract

Since the infrared radiofluorescence (IR-RF) at 1.43 eV emitted by potassic feldspars has been proven to be suitable for sediment dating, all instrumentation requirements for this new dating technique have been met. This paper describes the single-aliquot regenerative-dose protocol IRSAR (infrared radiofluorescence single-aliquot regenerative-dose) applied to coarse-grain K feldspar. Based on state-of-the-art knowledge of the physical background and the methodology, this new promising technique can be used for the dating of clastic sediment samples in the age range up to 250 ka.

1. Foreword

First papers on the radiofluoresence of potassic feldspar and the potential of an application in luminescence dating used the synonym "infrared radioluminescence (IR-RL)" (e.g. Trautmann 1998). From a physical point of view and, furthermore, to avoid misunderstanding we suggest the abbreviation "IR-RF", what stands for "infrared radiofluorescence". This luminescence phenomenon arises by interaction of the samples with ionising radiation and is, therefore, a fluorescence process.

2. Introduction

Since Trautmann et al. (1998) reported on the pioneering work of investigations of the spectral and dosimetric radiofluorescence (RF) properties of feldspars and on the potential of the dating application of the infrared radiofluorescence (IR-RF) only occuring in potassium feldspars such as microcline and orthoclase, considerable results have been obtained. The IR-RF can be understood as the radiative transition of electrons from the conduction band via the exited to the ground state of the 1.44 eV IR-OSL trap, emitting characteristic luminescence light at 865 nm. This conclusion explains the strict single exponential IR-RF dose curve decay function for all K feldspars. Trautmann et al. (1999) and Trautmann (2000) presented dating results which mostly agreed very well with other dating results. Krbetschek et al. (2000) summarised methodological aspects of the IR-RF technique and Krbetschek and Trautmann (2000) investigated the dosimetric RF behaviour of other materials such as quartz and halite. Trautmann et al. (2000) presented a spectral study of single feldspar grains and pointed out the potential of IR-RF single grain dating. The challenge of a very precise beta source calibration for the IR-RF technique has been solved due to the indroduction of a method using Al_2O_3:C crystals as probe dosimeters (Erfurt et al. 2000; Erfurt et al. 2001). Together with an automated multi-spectral radioluminescence (RL) reading system for geochronoloy and dosimetry, Erfurt et al. (2002) precisely desribed the calibration protocol using the RF of

Al_2O_3:C dosimeters. They also reported on dating results which correspond very well to independent ages. Furthermore, the physical background and the methodology of the IR-RF dating technique was investigated by Erfurt and Krbetschek (2002) and, among other things, they found out, that the IR-RF dose curves can be more exactly described if fitted to a stretched single exponential decay function. Besides, they suggested the use of a combination of additive dosing and regeneration (slide technique) for the estimation of the palaeodose. Schilles and Habermann (2000) and Schilles (2002) reported on an experimental device for the measure of IR-RF of K feldspar and on physical features of this emission.

In this paper we present the state-of-the-art knowledge on the methodology of the IR-RF dating technique, resulting in a standard single-aliquot regenerative-dose protocol, called IRSAR.

3. Experimental notes and sample preparation

All measurements were carried out using an automated multi-spectral radioluminesence (RL) instrument as described by Erfurt et al. (2002). The geological settings and the detailed sample preparation descriptions of the samples GOS4, HURL1, GRÖ8 and OOK1 can be found elsewhere (Preusser 1999; Erfurt et al. 2002; Trautmann et al. 1999). Sample SHO was grain size fractionated (100..150 µm) and underwent additional flotation to extract the whole feldspar fraction before separation of K feldspars and plagioclases by density using standart procedures (Aitken 1985). This sample originates...

4. Physical background

This new dating technique is characterised by the following methodological features:

- Due to the steady irradiation and fluorescence reading of the samples at once, the method is a single-aliquot technique with only one regenerative dosing cycle comparing the commonly used SAR protocols applied to quartz and feldspar (Murray and Wintle 2000; Wallinga et al. 2000)

- Since the IR-RF is a steadily measured luminescence during interaction with ionising radiation, all transitions of charges in the material are characterised by a transient equilibrium which compares to processes taking place in the radioactive environment of the sediment (Erfurt and Krbetschek 2002)
- Comparing secondarely excited TL and OSL methods, the dosimetric information bases on primary electronic transition effects (Erfurt and Krbetschek 2002)
- The spectral RF behaviour of K feldspars shows an overlapping RF peak at about 710 nm what has to be considered when filters for the separation of the IR-RF peak are chosen (figure 1)
- The method does not need thermal pre-treatment of samples and shows no significant sensitivity change during artificial dosing (figure 2)
- The IR-RF dose curves follow a stretched single exponential dose curve function (Erfurt et al. 2002) with non-significant fit residues (see figure 3)
- The mean saturation dose is about 1200..1500 Gy, but the dose curves are nowadays only resolvable up to about 600..800 Gy due to the small dynamic range of the IR-RF process (Erfurt et al. 2002)
- After bleaching by light having solar emission characteristics, the samples exhibit a strong phosphorescence (figure 4) which has to be considered during the dating performance
- Extracted K feldspars which underwent flotation prior separated by density showed stronger IR-RF intensities, which may indicate enhanced enrichment of K feldspars (Erfurt et al. 2002)

4.1 Filter choice

Krbetschek et al. (2000) mentioned IR-RF detection problems which could occur due to the superposition of neighbouring RF emission bands. For a better understanding of the filter selection we have used a Gaussian distribution function in order to simulate different overlapping ratios for the RF Gauss peaks, separated by different filters (resulting "model" spectra in figure 1). To simulate the "worst case" we used the mean Gaussian fit data of saturated K feldspars (about 1500 Gy) with very strong emission at 710 nm.

Figure 1

A special designed Andover interference filter (200FC35-25/8650) with a FWHM of 18.7 nm and 27.9 nm at ten percent of the central wavelength transmission (after Erfurt et al. (2002)) and the characteristics of the filters Schott RG850 and Schott RG830 (suggested as detection window by Schilles (2002)) have been used as integration interval. This Gaussian distribution of the spectrum was integrated (fig. 1) at 50 % transmission for all filters (see fig. 1: $x_0..x_3$ as integration intervals for the simulation of different filter transmissions). The results of this simulation were corrected for the changing detector sensitivities in this wavelength interval. The computed percentage of the simulated signal overlapping are for the Andover 865 nm interference filter (200FC35-25/8650) 5.2% for Schott RG850 6.4% and for Schott RG830 12.1%. Typical values for signal overlapping (e.g. sample Bur10, described in terms of signal overlapping by Krbetschek et al., 2000) were estimated by this spectra simulation to be not more than 0.9% for the 200FC35-25/8650, 1.0% for RG850 and 1.8% for RG830. In contradiction to Schilles (2002) we do not recommend the use of filters which transmit below 840 nm. Optimal results can be reached using an interference filter centred around the IR-RF emission at 865 nm. Moreover, the IR-RF signals are the strongest RF emissions of microcline and orthoclase. Therefore, the typical transmission percentage of interference filters does not lead to any statistical inaccuracies.

The emission at 920 nm in potassic feldspar (Erfurt and Krbetschek 2002) does not lead to serious problems because of the very low sensitivity of the used Hamamatsu PMT R943-02 in this wavelength region (Erfurt et al. 2002).

4.2 Changes in the dose curve response

If absorbed doses in K feldspar samples are regenerated twice, the resulting dose curves showed no significant difference. Figure 2 gives a typical example of a sediment sample (SHO). A change in the dose curve response is commonly addressed to sensitivity changes.

Figure 2

As one can see in figure 2, those effects may take place to a small amount but do not change the dating result adversely. If a palaeodose would be estimated using these two regenerated dose curves applying the IRSAR protocol (see section 5), a deviation of 3 % does occur (D_{black}=508.9 and D_{grey}=525.9).

4.3 Dose curve fitting

The experimental data and a luminescence model suggested by Trautmann (2000) resulted in a single exponential dose curve function which described the IR-RF data. The IR-RF was measured using RF spectra together with a Gaussian distribution of the spectra. Only a maximum of about 30 dose points were used for the single exponential fit function. Using the automated RL instrument (Erfurt et al. 2002), between 200 and 800 dose points are measured depending on the sample age. Such a high data density results in a very precise dose curve record. It has been observed by Erfurt and Krbetschek (2002) that the IR-RF dose characteristic does not follow a strict single exponential function. It has been suggested, e.g.

by Pavesi and Ceschini (1993), that a stretched exponential function (equation 1) can describe a behaviour which is often encountered in disordered condensed-matter systems.

(1) $\Phi(D)=\Phi_0-\Delta\Phi(1-\exp(-\lambda D))^{\beta}$

(Φ_0..initial IR-RF flux, $\Delta\Phi$..dose dependent change of the IR-RF flux, λ..exponential parameter, D..dose, β..dispersive factor)

They suggested that the dispersion of charge carrier transition or release rates and trap energies in multiple trapping-detrapping mechanisms can be expressed in the dispersive exponent β in equation 1. Erfurt and Krbetschek adapted such a function for the IR-RF dose curve description and discussed its influences of the model by Trautmann (2000).

Figure 3

Applying equation 1 to the data of figure 2, the dose curve of sample SHO can be described yielding residues close to zero (figure 3) (more examples in Erfurt et al. (2002) and Erfurt and Krbetschek (2002)).

4.4 Saturation dose and upper dating limit

Trautmann et al. (1999) concluded from spectral measurements, that the IR-RF saturation dose lies about 800 Gy. Measuring the IR-RF with higher dose resolution (number of measured dose points) using the automated RL instrument (Erfurt et al. 2002), a saturation dose of about 1200..1500 Gy was estimated (see figure 2). This would imply an upper dating limit of about 700 ka. On the other hand, the dating limit depends mainly on the dynamics ($d\Phi/dD$) of the IR-RF signals at high given doses. Based on state-of-the-art knowledge, it can be assessed that the IR-RF dose curves are resolvable up to about 800 Gy (Erfurt et al. 2002) what results in an upper dating limit of about 250 ka. However, the method has high potential to increase the upper age limit, because of the high saturation dose. If one can exclude influences of electronic instabilities which influence the transient equilibrium of the RF process, the age range might by enhanced. Figure 4 shows the thermal stability of the IR-RF signal after pulsed pre-heating of sample SHO to 100°C, 175°C, 250°C and 350°C. The equivalent dose of sample SHO is ≈ 600 Gy. The reconstructed dose after preheat does only show slight chances and are 100°C ≈ 595 Gy, 175°C ≈ 580 Gy and 250°C ≈ 565 Gy. After preheat up to 350°C, the signal is altered and the reconstructed dose equals to ≈ 515 Gy. This test implies, that the IR-RF signal seems to be thermally stable up to about 200°C. Straightforwardly, we conclude that the dating application of this new method is limited by the instabilities of all other radiative and non-radiative transitions within the minerals.

4.5 Phosphorescence effects after bleaching

Trautmann et al. (1999) pointed out the bleaching behaviour of the IR-RF signal and Krbetschek et al. (2000) concluded consequences for the dating methodology. If the samples are bleached using the Osram HTI lamp (which is part of the RL instrument described by Erfurt et al. (2002)) K feldspar show a very strong phosphorescence at 865 nm. Figure 5 illustrates this effect, observed on a saturated sediment sample OOK1 (about 1500 Gy given dose). The decay of the phosphorescence follows an exponential function. The phosphorescence was measured directly after the bleaching light was shut from the samples. Because of the time delay between shutter closing and the first phosphorescence measurement of 18 seconds, we extrapolated the exponential decay function to the intitial phosphorescence as shown in figure 5.

Figure 5

The phosphorescence in figure 5 was measured only 450 seconds. On the extrapolation of the phosphorescence characteristics, one can see that its intensity is not completely decayed. Therefore it is important to wait about one hour after bleaching, before the regeneration of the palaeodose can be started because of a possible superposition of the phoshorescence after bleaching and the radiofluorescence. We have not yet investigated the spectral optical excitation behaviour of the IR-RF emission. This IR emission of K feldspar is, however, not limited to the radiofluorescence.

5. The dating protocol IRSAR (Infrared Radiofluorescence Single-aliquot Regeneration)

Summarising all mentioned methodological aspects, we describe in the following the IRSAR (infrared radiofluorescence single-aliquot regeneration) dating protocol.

The IRSAR protocol applied to the IR-RF on K feldspar:

1. Measuring of the IR-RF at natural dose
2. Bleaching of samples at solar conditions for 30 minutes without changing geometry
3. Waiting for at least one hour until complete decaying of the phosphorescence excited by bleaching light is reached
4. Regeneration of the IR-RF signal at natural dose with high number of dose points
5. Fitting data to a stretched single exponential function (equation 1) and controlling fit residues
6. Using fit parameters (Φ_0, $\Delta\Phi$, λ and β) and the IR-RF signal ($\Phi(D)$) at natural dose to calculate the palaeodose D

After an investigation by Erfurt et al. (2002), the application of the protocol is also shown in figure 6. First step is the measure of $\Phi_{natural}$ connected to palaeodose D_e. During bleaching for 30 minutes, the IR-RF increases up to the maxiumum IR-RF signal $\Phi_{bleached}$. After bleaching the regeneration starts until the IR-RF signal $\Phi_{natural}$ connected to palaeodose D_e is regenerated, measuring 200 dose points. The applied laboratory dose equals to the equivalent dose, absorbed during burial after the last light exposure of the sediment.

Figure 6

We like to propose to do a standart sample preparation together with additional flotation before feldspar separation by density to enrich K feldspars in the sample. Furthermore one should garantee to have a packed mono layer of sample grains because of dosimetric reasons. Therefore we suggest to use sample plates as described by Erfurt et al. (2002).

Figure 7

A comparison of ages determined by the IRSAR protocl and ages estimated using IR-OSL, U/Th and ^{14}C is presented in figure 7 for sample GOS4, GRÖ8 and HURL1 and underlines the good agreement of IRSAR data to other dating techniques.

7. Discussion and summary

We presented a single-aliquot regenerative-dose protocol applied to the infrared radiofluorescence (IRSAR) on coarse-grain K feldspar. It is a robust and precise method to determine the equivalent dose connected to the last light exposure of clastic Quaternary sediments. The physical and methodological charcteristics show that this new promising technique does not need special treatment of the samples due to e.g. sensitivity changes, comparing other luminescence methods (SAR on quartz and feldspar). Based on state-of-the-art knowledge on the physical background, we assume a dating range of up to 250 ka. Regarding the saturation dose levels as well as the thermal stability of the IR-RF signal, there is potential to increase the upper dating limit.

However, much more work is needed to investigate the IR-RF more precisely especially its behaviour at higher dose levels as well as on effects which may affect the stability of the transient equilibrium of the fluorescence occuring during interaction of the minerals with ionising radiation.

This paper should encourage the luminescence dating community to apply and to investigate this new IR-RF dating technique.

8. Acknowledgements

We gratefully thank Dr. F Preusser (Köln and Bern Universities) for providing material of samples GOS4 and HURL1. This research work is financed by the German Ministry of Education and Research (BMBF) under contract 03KR9FR2/7.

9. References

Aitken M. J. (1998) *An indroduction to optical dating.* Oxford University Press. Oxford, U.K.

Erfurt G.; Krbetschek M. R.; Trautmann T. and Stolz W. (2000). Radioluminescence (RL) behaviour of Al_2O_3:C - potential for dosimetric applications. *Radiation Measurements* **32**, 735-739

Erfurt G.; Krbetschek M. R.; Trautmann T. and Stolz W. (2001). Radioluminescence (RL) probe dosimetry using Al_2O_3:C for precise calibration of beta sources applied to luminescence dating. *Radiation Physics and Chemistry* **61**, 721-722

Erfurt G.; Krbetschek M. R.; Bortolot V. J. and Preusser F. (2002). A fully automated multi-spectral radioluminescence reading system for geochronology and dosimetry. *Nuclear Instrumentation and Methods in Physics Research B*, (submitted for publication)

Erfurt G. and Krbetschek M. R. (2002). Studies on the physics of the infrared radioluminescence of potassic feldspar and on the methodology of its application to sediment dating. *Radiation Measurements*, (submitted for publication)

Krbetschek M. R.; Trautmann T.; Dietrich A. and Stolz W. (2000). Radioluminescence dating of sediments: methodological aspects. *Radiation Measurements* **32**, 493-498

Krbetschek M. R. and Trautmann T. (2000). A spectral radioluminescence study for dating and dosimetry. *Radiation Measurements* **32**, 853-857

Murray A. and Wintle A. (2000). Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol. *Radiation Measurements* **32**, 57-73

Schilles T. und Habermann J. (2000). Radioluminescence dating: the IR emission of feldspar. *Radiation Measurements* **32**, 679-688

Schilles (2002). Die Infrarot-Radiolumineszenz von Feldspäten und ihre Anwendung in der Lumineszenzdatierung. *PhD thesis*. Ruprecht-Karls-Universität Heidelberg

Preusser F. (1999). Luminescence dating of fluvial sediments and overbank deposits from Gossau, Switzerland: fine grain dating. *Quaternary Geochronology* **18**, 217-222

Trautmann T.; Krbetschek M. R.; Dietrich A. and Stolz W. (1998) Investigations on radioluminescence of feldspar: Potential for a new dating technique.- *Radiation Measurements* **29**, 421

Trautmann T.; Krbetschek M. R.; Dietrich A. and Stolz W. (1999). Feldspar radioluminescence: a new dating method and its physical background. *Journal of Luminescence* **85**, 45-58

Trautmann T.; Krbetschek M. R.; Dietrich A. and Stolz W. (2000). A systematic study of radioluminescence properties of single feldspar grains. *Radiation Measurements* **32**, 685-690

Trautmann T. (2000). A study of radioluminescence kinetics of natural feldspar dosimeters: experiments and simulations. *Journal of Physics D: Applied Physics* **3**, 2304-2310

Wallinga, J., Murray A. and Wintle A. (2000). The single-aliquot regenerative-dose (SAR) protocol applied to coarse-grain feldspar. *Radiation Measurements* **32**, 529-533

Figure 1

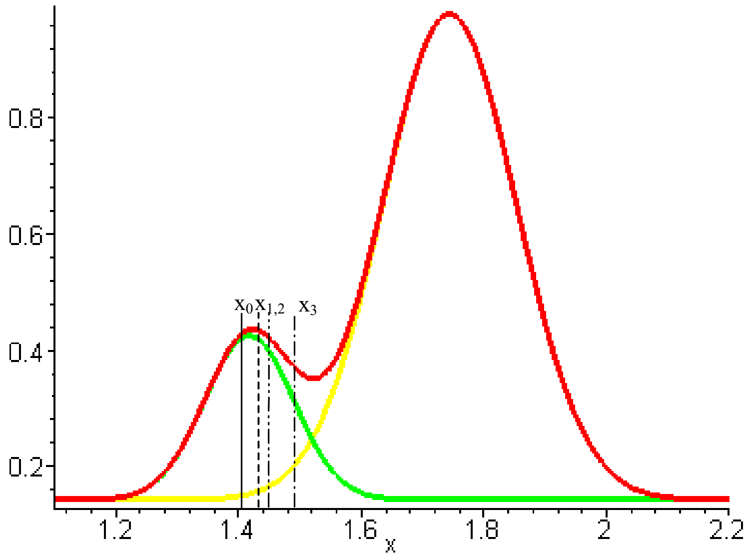
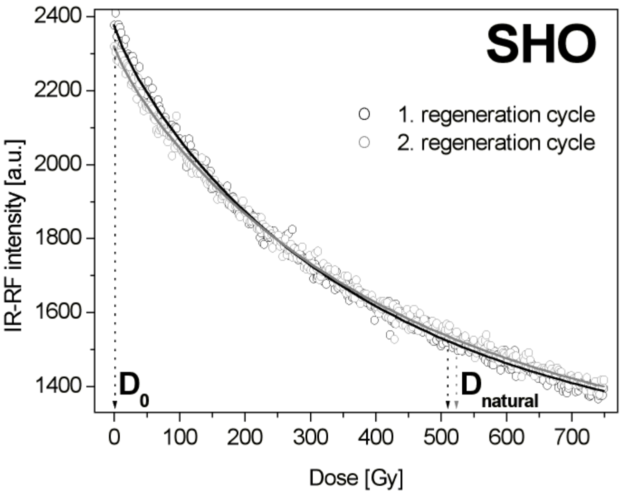
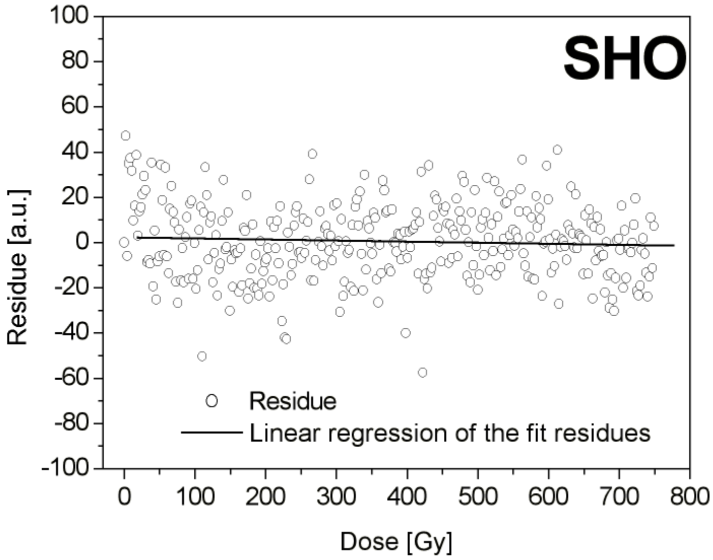


Figure 2



SHO
1. regeneration cycle
2. regeneration cycle
IR-RF intensity [a.u.]
Dose [Gy]
D0
Dnatural
2400
2200
2000
1800
1600
1400
0
100
200
300
400
500
600
700

Figure 3



SHO
Residue [a.u.]
100
80
60
40
20
0
-20
-40
-60
-80
-100
0
100
200
300
400
500
600
700
800
Dose [Gy]
Residue
Linear regression of the fit residues

Figure 4

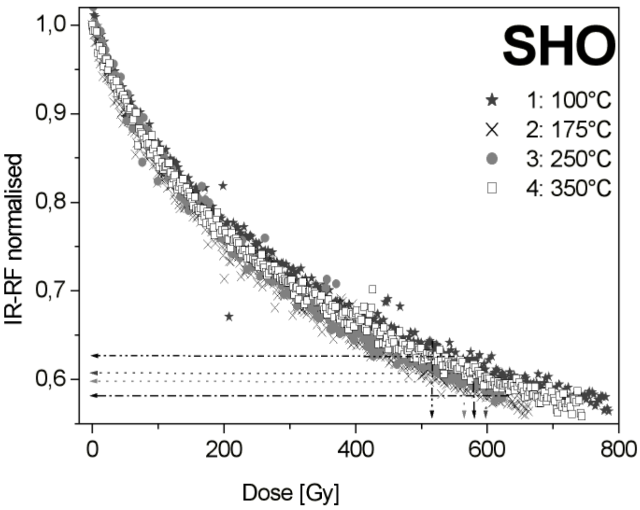
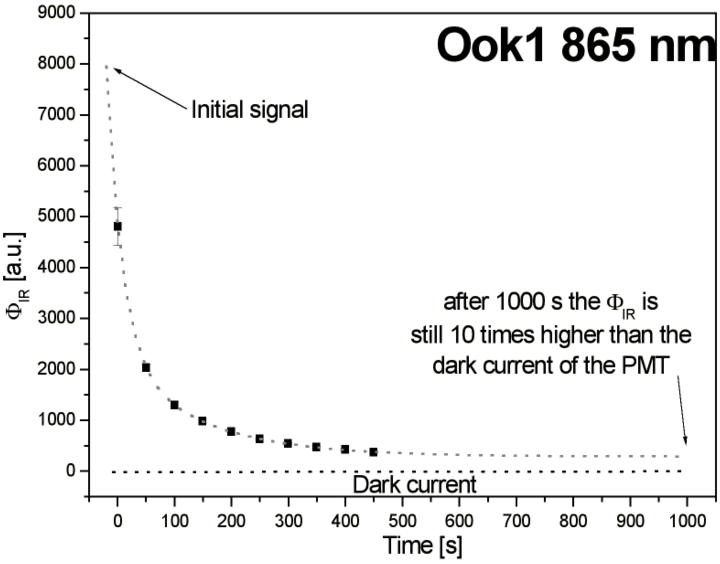
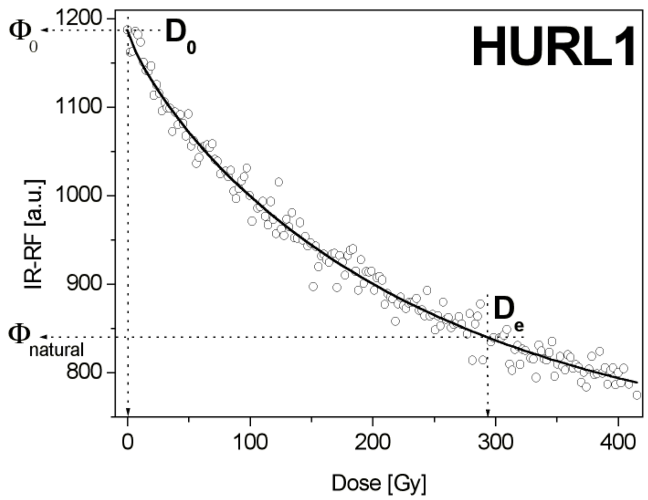

SHO
1: 100°C
2: 175°C
3: 250°C
4: 350°C
IR-RF normalised
1,0
0,9
0,8
0,7
0,6
0
200
400
600
800
Dose [Gy]

Figure 5



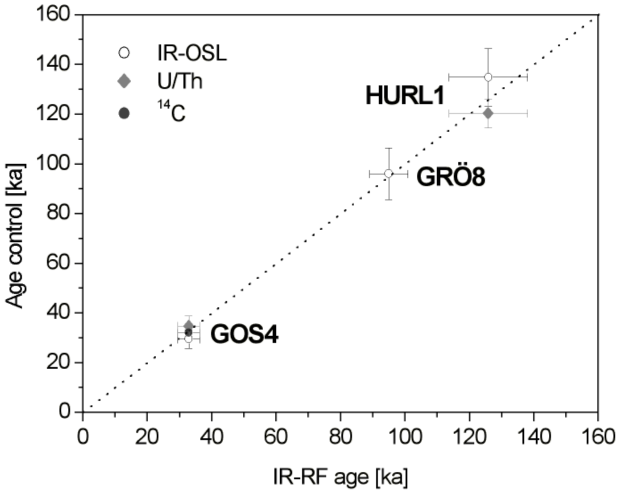
Ook1 865 nm
Initial signal
after 1000 s the Φ_{IR} is
still 10 times higher than the
dark current of the PMT
Dark current
Φ_{IR} [a.u.]
Time [s]
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
0
100
200
300
400
500
600
700
800
900
1000

Figure 6



HURL1
Φ_0
D_0
D_e
$\Phi_{natural}$
IR-RF [a.u.]
1200
1100
1000
900
800
0
100
200
300
400
Dose [Gy]

Figure 7



IR-OSL
U/Th
14C
HURL1
GRÖ8
GOS4
Age control [ka]
IR-RF age [ka]
0
20
40
60
80
100
120
140
160

Figure captions

Figure 1:

Simulated saturated spectrum generated by mean values of Gaussian distributions from an number of RF spectra of K feldspars in the energy range from 1.2..2.0 eV. The vertical lines indicate the integral intervals ($x_0..x_1$ = Andover 200FC35-25/8650, $x_0..x_2$ = Schott RG850, $x_0..x_3$ = Schott RG830)

Figure 2:

Dose regeneration cycles of sample SHO. One can see slight changes in the luminescence response resulting in only a very small change of the dose determination.

Figure 3:

Fit residues of the IR-RF dose curve fit using euqation 1. The residues were fitted using a linear regression function. One can see the precision of the parameter determination using this stretched single exponential fit function. Together with high number of measured dose points (a few hundreds) the IR-RF data show good agreement of experimental observation and theoretical considerations (Trautmann 2000, Erfurt and Krbetschek 2002).

Figure 4:

Comparison of IR-RF dose curves after preheat of 100°C (grey stars), 175°C (black crosses), 250°C (grey circles) and 350°C (open black squares). Comparing the determined equivalent dose for the unheated sample of 598 Gy, the recovered doses of the preheated samples at 100°C and 175°C does only change slightly. This indicates the high thermal stability of the IR-RF process.

Figure 5:

Typical phosphorescence kinetics at 865 nm measured directly on sample OOK1 after bleaching for 30 minutes. The decay follows an exponential function and complete depletion of the signal is measured only after a few thousands seconds. This may superpose the IR-RF signal, if regeneration is started immediately after bleaching.

Figure 6:

IR-RF dating example appying the IRSAR protocol to sample HURL1 (Erfurt et al. 2002). The dose curve was measured reading 200 dose points.

Figure 7:

Comparison of IR-RF ages with IR-OSL, ^{14}C and U/Th ages (data after Trautmann et al. (1999) and Erfurt et al. 2002) of samples GOS4, GRÖ8 and HURL1