# Some Observations on Tunnelling of Trapped Electrons in

## Feldspars and their Implications for Optical dating

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## Abstract

Anomalous fading in feldspars is now understood to be caused by the tunnelling of electrons from one defect site to another. Here we present some experimental observations concerning the phenomenon. The fading rates of a variety of feldspar crystals and K-feldspar separates from sediments are reported. It is found that (1) the fading rates of 77 K-feldspar extracts from sediments range from 1 to 10 %/decade, with an average value of about 5 %/decade, (2) the fading rates of K-feldspars extracted from sediments derived largely from volcanic bedrock are not higher than those from non-volcanic bedrock as is widely thought, (3) the fading rates of 31 individual feldspars range from 1 to 35 %/decade, (4) in plagioclase feldspars the fading rate increases with increasing Ca and/or Fe content, (5) the fading rate increases with laboratory radiation dose at large doses, (6) for samples for which the time elapsed since burial is long enough for their luminescence to be in saturation, the fading rate is correlated with the ratio of the field saturation intensity to the laboratory saturation intensity; extrapolation to zero fading rate shows that trap emptying as a result of thermal eviction is not significant, and that the mean thermal lifetime in temperate environments of electrons in traps relevant to dating is \$4 Ma., and (7), different aliquots of a sample can have fading rates that differ by a factor as large as two even if the aliquots contain several thousands of grains, thus it is necessary to ensure when correcting ages for anomalous fading that the fading rate used is that applicable to the aliquots on which the equivalent dose is measured.

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## 1. Introduction

When a feldspar is given a radiation dose electrons are freed from their normal atomic positions and some of them become lodged at impurities or structural defects in the crystal referred to as traps. Traps can be crudely divided into two classes, shallow and deep. Deep traps are those for which thermal excitation at ~20 °C is insufficient to excite a significant number of electrons out of them, and the thermal lifetime of the trapped electrons is at least a few Ma. Shallow traps are those with shorter thermal lifetimes; these can be emptied of their electrons in the laboratory by heating to ca. 150 °C. Those deep traps from which an electron can be excited by 1.45 eV (infrared) radiation will be referred to as the principal traps, since it is these that are made use of in optical dating using feldspars. In practice, however, it is found that a significant fraction of electrons trapped in the principal traps escape from them at ~20  $^{\circ}$ C; this phenomenon has long been referred to as anomalous fading. In recent years sufficient understanding has been gained that the phenomenon is now recognized as being due to the quantum-mechanical tunnelling of electrons from the principal traps to nearby defects. This realization is due mainly to the work of R. Visocekas. Reviews can be found in Visocekas (2002) and Aitken (1985, Appendix F: 1998, Appendix D), and limited application of the ideas to correcting optical ages for the effect is described in Huntley and Lamothe (2001). Some fading data for individual feldspars were presented by Spooner (1994), from which it appeared that most feldspars showed fading; for some the fading was linear on a log(time) scale, but the data were not adequate for making a generalization to this effect. Spooner also reported that "the only unambiguously nonfading signals originate from highly ordered end-members of the alkali series".

The objective of the present work was to obtain a wide variety of experimental data in order to learn more about the tunnelling process with the objective of discovering what the relevant defects are and finding a theory which explains all the observations. It is found that Ca and/or Fe plays an important role in tunnelling, though the role is probably indirect. It is also found that the observation that the field saturation intensity is less than the laboratory saturation intensity is due to tunnelling and from this a lower limit of 4 M a can be placed on the thermal lifetime of electrons trapped in the principal traps in temperate environments.

### 2 Samples

Feldspars of specific mineralogy were obtained from Ward's Natural Science<sup>1</sup>, the University of Adelaide collection, and various colleagues. Thin slices were prepared from them using a fine low-speed diamond saw. Some granitic rocks and one sanidine were collected in the field; in order to obtain material from them not exposed to light, cores were obtained using a diamond core drill and slices cut from them using the fine low-speed diamond saw. Natural sediment samples were collected by the authors or provided by colleagues from a wide variety of geographic locations. K-feldspar grains were separated from bulk sediment using standard methods (HCl, sieving, 10% HF etch, magnetic separation and density separation at 2.58 g.cm<sup>-3</sup>). Preparations of all samples except for the specific feldspars were performed under dim orange light.

<sup>&</sup>lt;sup>1</sup> Ward's Natural Science Ltd., 397 Vansickle Rd., St. Catherines, Ontario, L2S 3T5, Canada.

### 3. Experimental

Radiation doses were given using a <sup>60</sup>Co Gammacell<sup>2</sup>, at a dose rate of ~ 10 Gy/hr. Excitation was with ~1.45 eV (840 nm) photons from light-emitting diodes. The 3.1 eV (400 nm, violet) luminescence was measured using an EMI-Thorn 9635 photomultiplier tube behind Kopp 4-97 and Schott BG-39 filters. It should be noted that for samples for which the 2.2 eV (570 nm) emission is dominant (e.g. Baril and Huntley, 2003b, Fig.4), as is usual for plagioclase feldspars, the high-energy tail of this band may be included; this is presumed not to be significant as it seems likely that the emission is a secondary photoluminescence.

Rates of fading due to tunnelling were measured as described for method 'b' of Huntley and Lamothe (2001). The feldspar slices or grains were attached to aluminium planchets using a thermoplastic polymer (Crystalbond<sup>3</sup>) dissolved in acetone. To prepare an aliquot, two drops of the liquid were placed in a 13 mm diameter planchet and a slice of crystal or 12 mg of grains added; this was then dried at 50 /C for 1 hr. Five such aliquots were prepared for each sample. These were then given a red/IR bleach if they had not been bleached recently in nature or the laboratory. Each aliquot was then given a gamma dose of ~175 Gy, and heated at 120 /C for 16 hours to empty shallow traps filled during the irradiation. The luminescence from each aliquot was then measured using short shines<sup>4</sup> at successive times of ~2, 4, 8, 16 etc days, to about a year, after irradiation. In each run, the 5 aliquots of each of 7 samples were measured along with 8 aliquots of grains of K-feldspars separated from a Tertiary sandstone (CBSS). This sample was used to act as a reference as it had not been given any laboratory dose and therefore was expected to show no decay. Numerical values of fading rates are given for a starting time 2 days after the midpoint of the irradiation period. Some of the data of Fig. 6 required irradiations lasting several days, in which cases the data were extrapolated back to 2 days in order to evaluate g.

Figure 1 shows four examples of measurements that exhibit the range of fading rates observed.

## Figure 1 about here

The use of five aliquots allows the determination of the variability of the fading rate within a given sample. For most samples the variability was within the uncertainties of the measurements; however, we have observed variabilities of at least a factor of two for three samples, plagioclase feldspar #19, granodiorite BSG1, and feldspars separated from a sediment sample from the Queen Charlotte Islands, British Columbia; such variability was inferred earlier from data obtained from two sediment samples (Huntley, 1997) and is not unexpected because Lamothe and Auclair (1999) found that individual grains of a sample can have a very wide range of fading rates. It is not clear whether or not this variability has a significant effect on the data that follow; in some cases aliquots for the different experiments were selected in such a way as to avoid systematic effects caused by this.

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<sup>&</sup>lt;sup>4</sup>A short shine refers to an exposure of the sample aliquot to 1.4 eV excitation of intensity and duration that causes the luminescence to decay by only a small amount, usually less than 1%.

## 4. Fading Rates of sediments

The fading rates observed for K-feldspar grains separated from sediments are summarized in Fig. 2a. The main histogram shows the distribution of fading rates for all 77 samples measured. The range is from 1 to 10 %/decade, and the average value is about 5 %/decade. Since 30 of the samples are from the three prairie provinces of Canada, and their similar mineralogy could have influenced the distribution, the fading rates for these are shown by the cross-hatched area in the figure; they form a slightly narrower distribution, but the main distribution is little influenced by this.

### Figure 2 about here

It is often stated that volcanic feldspars have high fading rates compared to those of feldspars formed by non-volcanic processes (e.g. Aitken 1985, p.275). The original basis for the statement is that Wintle (1973) found that apparent thermoluminescence ages obtained from feldspars from a variety of lavas were significantly lower than the accepted ages of them. This observation has since been confirmed by Visocekas (2002). In order to test the implications of this further we included 7 sediment samples specially chosen from areas where the sediments are derived from volcanic material. Three are from British Columbia, and are sediments mainly derived from 1-2 Ma Fraser Plateau flow basalts (sample BLRL2 of Lian and Huntley, 1999), the Pennsylvanian and Permian Cache Creek group volcanics (sample CCL1 of Lian and Huntley, 1999), and the Upper Cretaceous Kingsvale Group volcanics (Coutlee sediments near Merritt, Fulton et al., 1992). The other four are from North Island, New Zealand, and are two sediment samples from the volcanic Great Barrier Island (late Miocene, samples GBDS4 and HBTS1), a sediment sample from One Tree Point, Whangarei Harbour (also derived from late Miocene volcanics), and a ~24 ka ignimbrite from Poihipi Road near Lake Taupo (sample PRI1). The fading rates for these samples are shown by the dark shading in Fig.1a. It is readily seen that these samples have fading rates similar to those of the other, non-volanic, samples, and it is concluded that K-feldspars extracted from sediments derived mainly from volcanic rock do not necessarily have unusually high fading rates.

### 5. Fading rates of Plagioclases and Alkali feldspars

We have measured fading rates for a wide range of feldspars for which we have major element analyses; for most we also have analyses for the lanthanides and 21 minor elements. The results are summarized in Table 1 and Fig. 2b. For plagioclase feldspars there is a positive correlation between fading rate and calcium content, as shown in Fig. 3a. This result was hinted at by Akber and Prescott (1985) who, with respect to thermoluminescence measurements, stated "the higher-Ca plagioclases usually show anomalous fading". It is clear, however, from the data presented here that there are samples that do not follow that general trend, and that there must be at least one other variable determining the fading rate. Fig. 3b shows that there is also a tendency for the fading rate to increase with increasing Fe content, though the correlation is not as good as it is for Ca. For all of these samples the analyses and fading rates were measured on a different parts of the same crystal, and the variability mentioned earlier may have introduced some of the scatter appearing in the figure.

## Figure 3 about here

In the case of alkali feldspars we have found a range of fading rates, but have not established any correlation between them and the major or minor element contents, or structural state of the feldspar. Visocekas and Zink (1995) reported from their observations that non-fading feldspars are microclines, and relate that to the structural order of microclines. The data of Table 1 show that microcline fading rates vary from 1 to 12 %/decade, thus one cannot conclude that microclines are non-fading, and thus that structural order does not necessarily lead to non-fading behaviour.

Eleven of the feldspars that we tested emitted too few photons for us to measure their fading rates. These were 2 sanidines, 2 anorthoclases, 1 albite, 3 labradorites, 1 bytownite and 2 anorthites. The low intensity is not unexpected (cf. Spooner, 1992), except for the albite, and this is the reason for the absence of fading rates for samples at higher Ca contents in Fig. 3. It should be possible, however, to measure fading rates for some such samples using the method of Auclair et al. (2003).

### 6. Field Saturation vs Laboratory Saturation

Before discussing the experimental data it is necessary to introduce the concepts of field saturation and laboratory saturation. Consider feldspar grains that have received a radiation dose of several thousand grays or more in the environment since burial. In practice this means one for which the principal traps were last emptied at least a few million years ago. Now consider the dose response of the feldspar, both as is (N+dose), and after bleaching it first (N+bleach+dose). Examples of these two dose responses are shown in Fig. 4, in which the former has been shifted parallel to the dose axis so that it matches the latter. It is observed that saturation occurs for both data sets at high doses, and that the intensity here is the same for both; this is referred to as laboratory saturation, and the intensity here will be denoted  $I_{LS}$ . It is also observed that the intensities of the undosed aliquots are significantly less than this value, even though the radiation dose the sample received in the environment should have been sufficient for saturation to have been attained. Such a sample is said to be in field saturation, and the intensity measured for it will be denoted by  $I_{FS}$ . A sample in field saturation would appear to be in a state of dynamic equilibrium in which electrons are leaving traps at the same rate as the traps are being populated as a result of the radiation. For sediments,  $I_{FS}$  is typically found to be 50-70 % of  $I_{LS}$ .

### Figure 4 about here (field saturation vs lab saturation)

 $I_{FS}/I_{LS}$  ratios were measured as follows. Several aliquots were prepared, and the luminescence measured using a short shine; this intensity was used later for normalization. Half of the aliquots were given a gamma dose of ~ 5000 Gy, at ~ 10 Gy/h for about 3 weeks. All were then heated for 16 hr at 120 °C, and the luminescence measured after a delay of ~3 months. The  $I_{FS}/I_{LS}$  ratio was calculated as the ratio of the normalized intensities obtained from the unirradiated and irradiated aliquots, after a small correction for the decay that resulted from the normalization measurement. In the case of the granitic samples there was no mineral separation, but from the excitation used and the emission band measured it is expected that it is the luminescence from alkali feldspars that was being measured.

Values of these quantities are shown in Table 2 and plotted in Fig. 5.

Table 2 about here

Figure 5 about here

Except for the four sediment samples, the experiment was designed to avoid any inherent sample variability having a systematic effect on the results. To achieve this 15 aliquots were prepared and a normalization measurement made using a short shine. The aliquots were then sorted by normalization value, the 5 middle ones used for the anomalous fading measurements, and the aliquots for the  $I_{FS}$  and  $I_{LS}$  measurements each selected symmetrically from those with the low and high normalization values.

The first thing to note is that, except for the two samples from permafrost, there is a good correlation between  $I_{FS}/I_{LS}$  and fading rate. The smaller the fading rate, the larger the ratio. We infer from this that the dynamic equilibrium that leads to  $I_{FS}$  being less than  $I_{LS}$  is primarily due to tunnelling.

Any reasonable extrapolation of the data of Fig. 5 to zero fading rate yields a ratio > 0.9. One can calculate what this ratio should be if the traps were being emptied by thermal excitation (see Appendix), and from this deduce that the thermal lifetime of the trapped electrons at environmental temperatures in temperate regions is > 4 Ma. Using the data from kinetics experiments on a single sample, Li and Tso (1997) deduced trap parameters from which the calculated thermal lifetime is 600 Ma at 10  $^{\text{e}}$ C and 60 Ma at 20  $^{\text{e}}$ C. Our result is consistent with these. We note, however, that Li and Tso's calculated lifetimes involve an extrapolation of over 12 orders of magnitude in time and thus are less reliable than ours.

The two sediments collected from permafrost do not fit the trend of the other samples (Fig.5). It is possible that this is a result of the fading rate in the environment being lower than that measured in the laboratory because the fading rate decreases with decreasing temperature. Visocekas (2000, Fig. 8) found that fading of the thermoluminescence of a sanidine was greatly reduced by storage at 77 K; in contrast, our measurements on one of the samples, DY23, at -6, +20 and +52 °C show that any temperature dependence is insufficient to explain the observed effect, and we consider this matter unresolved. Although tunnelling itself is a temperature-independent phenomenon, a temperature dependence has been observed and can be accounted for by the non-linear response of the tunnelling rate to variations of barrier height and width due to lattice vibrations (Hurd, 1985).

# 7. Fading rate *vs* added laboratory dose for a sample in field saturation, and for a sample that has been bleached

Consider a sample in field saturation to which a laboratory dose is given; the luminescence intensity is increased by the dose (Fig. 4), and thus it is deduced that the laboratory dose fills traps that were empty during field saturation. If, as appears to be the case, the traps were empty because of tunnelling, one expects intuitively to find a higher fading rate for them because electrons in these traps are more likely to tunnel out than electrons in other traps.

Fig. 6 shows the measured fading rate as a function of laboratory dose for aliquots of a sample that is in field saturation, and for aliquots first given a laboratory red/IR bleach. It shows as expected that the fading rate for a sample in field saturation is much larger than that for the same sample after bleaching.

### Figure 6 about here.

Fig.6 also shows that for the bleached aliquots the fading rate increases with laboratory radiation dose. This may be due to an increase in the density of recombination centres. Visocekas (1988) found that the fading rate for alpha irradiation is higher than that for beta (or gamma) irradiation and related this to the saturation ionization density along an alpha track producing a high density of recombination centres within tunnelling distance of the trapped electrons. It is not

clear whether or not this is related to our observation, but whatever the cause it will complicate any detailed analysis.

## 8. Discussion

It is apparent from the wide range of feldspars measured that they all show fading due to tunnelling, and the conclusion of Huntley and Lamothe (2001) that fading is ubiquitous in feldspars is given further support. There are occasional claims in the literature that for the feldspars measured the violet emission does not display anomalous fading; so far, such claims have not been supported by adequate quantitative information and must be regarded with skepticism.

#### Fading for a sample in field saturation:

Consider taking a sample in field saturation, giving it a radiation dose, and measuring the fading of the component of the luminescence intensity that is due to this dose. To a first approximation the rate of fading is found to be independent of the sample. This seems surprising at first, but can be readily explained. Consider a rough division of the traps into two classes, those from which electrons do not tunnel significantly in the environment, i.e. those giving rise to  $I_{FS}$ , and the rest. If one assumes that there is no dependence of fading rate on dose or bleach, one can readily show that the rate of anomalous fading,  $g_{FS}$ , should be:

$$g_{FS} = g_o \frac{I_{LS}}{I_{LS} - I_{FS}}$$

Here  $g_o$  is the overall fading rate (i.e., the fading rate measured for aliquots that have been bleached sufficiently that the intensity is  $\langle I_{FS} \rangle$ , and  $g_{FS}$  is the fading rate for the component of I that is  $> I_{FS}$ .

The upper left part of the curve in Fig.5 is roughly linear and can be described approximately by  $I_{FS}/I_{LS} = 1 - 0.14 g_o$ . Substituting this into equation [1] yields  $g_{FS} = 7$ %/decade, independent of  $g_o$ . Hence it is predicted that the measured fading rates for all samples in field saturation should be the same!

Our values of  $g_{rs}$  for samples CBSS and DY23 are  $14 \pm 0.5$  and  $15 \pm 1$  %/decade respectively (Fig.6). The measured fading rates of individual grains of sample GP10 (Lamothe and Auclair, 1999, Fig 3A) are all very similar, 10-13 %/decade, despite the grains having widely different values of  $R_I$  (an indicator of fading), a result that was initially surprising. The fading rates for two samples shown by Lamothe et al. (2003, Figs. 2 & 3) are about 15 %/decade. The difference between these observed rates and the value of 7 %/decade predicted above presumably results from the crudeness of the model and the dependence of g on dose.

There is a very simple argument that the fading rate for a sample in field saturation should be roughly constant and equal to about 13 %/decade. The argument is based on the idea that it is the electrons with a particular range of tunnelling lifetimes that are measured during the fading measurements, and that this range is the same for all samples. Divide the electron traps into three groups. The first group is those for which the distance to the nearest centre to which a trapped electron can tunnel is such that the tunnelling lifetime is less than 2 days; such trapped electrons will all have tunnelled out by the time any measurements are made 2 days or more after irradiation, and thus play no role in the measurements. The second group is those traps for which the tunnelling lifetimes are long enough that the traps are all full when the sample is collected; these are the ones that give rise to  $I_{FS}$ , and also play no role in determining the fading rate. The traps can be considered to be all full when the probability per unit time of the radiation dose filling an empty trap exceeds the probability of emptying by tunnelling. The former is ~ 400 ka<sup>-1</sup> (see Appendix A). The third group is the rest of the traps, ie those with tunnelling lifetimes between 2 days and ~400 ka, and it is these that are measured during a fading measurement. The range is ~ 8 decades of time, and thus assuming a fading decay that is linear with log(time) one should expect a fading rate of 100 %/8 decades or 13 %/decade, which is about what is observed. Some variations from this figure are expected because of the crudeness of the model, because the above figure of 2 days is arbitrary (it is chosen because that is the convenient starting point,  $t_c$ , used for all our fading rates), because the figure of ~400 ka<sup>-1</sup> is sample dependent, and because the assumption of the form of the fading decay cannot be correct at all times.

#### Trap filling vs radiation emptying:

It is widely held that laboratory saturation occurs as a result of all the traps being filled. An alternative explanation is that the radiation causes traps to be emptied as well as filled, and that laboratory saturation occurs when a dynamic equilibrium is reached. The mathematics of a simple trap filling model and the mathematics of a simple radiation emptying model both lead to a dose response which is a saturating exponential, thus it has not been possible to determine from the dose response which process is occurring. This can be readily seen by solving equation A1 of the Appendix and interpreting 1/J as the probability per unit time of an electron being evicted from a trap by either thermal excitation or by radiation excitation. If the radiation excitation process was responsible for saturation, all the traps would be constantly being emptied and refilled, and in field saturation there would be empty traps of both classes. One might therefore expect that  $g_{FS}$  should equal  $g_{\rho}$ . This is not so. The filled traps at field saturation are all nonfading, and during a  $g_{rs}$  measurement some of these are emptied by the laboratory radiation dose and some are filled, but the actual number does not change; thus the non-fading traps give rise to a constant component of the luminescence intensity,  $I_{ES}$ . The laboratory radiation dose also fills some of the fading traps. A simple mathematical analysis shows that equation [1] still holds. Thus one cannot determine the cause of saturation by this method.

## The centres to which the electrons tunnel:

There are several arguments that these centres are not impurities but structural defects. Firstly there is the observation that the fading rate increases with increasing Ca content in plagioclase feldspars (Fig.3a). However, the two labradorite samples with similar Ca contents but with very different fading rates is an indication that the Ca atom itself is probably not the centre to which an electron tunnels, but that the centre is a result of the replacement of Na by Ca or the replacement of Al by Si. It is possible that Fe, rather than Ca, plays an important role (Fig.3b). Dislocations or the boundaries formed by the lamellar intergrowths found in plagioclase feldspars are other possibilities.

Secondly, from the information available it seems that sanidines generally have higher fading rates than orthoclases and microclines. These feldspars only differ in the arrangement of the Al and Si atoms.

Thirdly, the two sanidine samples BRS and WCRS came from the same deposit within a few metres of each other and the chemical analyses show that their compositions are very similar; on the other hand their fading rates are very different, 17 and 10 %/decade respectively (Table 1). This is hard to reconcile with impurities being responsible. An exception occurs for an impurity that can be present in two different ionization states, such as  $Fe^{+2}$  and  $Fe^{+3}$ , with tunnelling being to one of them and the proportion of this ion being different in the two samples.

Finally, fading rates observed for alkali feldspars show a wide range, and there is no correlation of fading rate and any of the impurity elements analysed for.

## Age correction:

In thermoluminescence of feldspars, the field saturation intensity is also less than the laboratory saturation intensity, and the reason is likely to be the same tunnelling phenomenon discussed here. Mejdahl (1988) treated the difference as though it were due to thermal excitation and showed how to correct ages for it. We conclude that this is not correct.

Huntley and Lamothe (2001) showed how to correct ages for tunnelling for samples which are on the low-dose linear region of the dose response, but we do not yet know how to correct ages for older samples which are on the non-linear region of the dose response curve. The correction will be very different from that for thermal excitation. A detailed model of the tunnelling process is required, and it will have to include an explanation of the increase of fading rate with dose.

From the scatter observed in fading rates measured for individual aliquots, we deduce that it is necessary to ensure that the fading rate measured is that applicable to the aliquots used for the equivalent dose measurements.

## 9 Conclusions

Every feldspar and feldspar extract from sediments that we have analysed so far shows measurable tunnelling (anomalous fading). For plagioclase feldspars there is a strong tendency for the fading rate to increase with increasing Ca and/or Fe content. No generalization could be made for alkali feldspars.

For samples in field saturation it was found that the relative field saturation intensity is correlated with the fading rate, and from the data it is deduced that the mean lifetime of trapped electrons due to thermal excitation in temperate environments is > 4 Ma.

The measurements reported on here are a step towards a detailed understanding of fading due to electron tunnelling. At present the identities of the trap out of which electrons tunnel and the centre to which they tunnel are unknown, but arguments are made that it is likely that the centres to which the electrons tunnel are structural defects rather than impurities.

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# Appendix A. Calculation of the mean lifetime of a trapped electron due to thermal excitation.

Consider N electron traps, n electrons in them, a constant probability of an empty trap being filled as a result of a constant radiation dose rate, D, and a constant probability per unit time, 1/J, of an electron being evicted by thermal excitation. Then

[A1] 
$$dn/dt = \dot{D}(N-n)/D_c - n/\tau$$

The solution for n vs t (time) is a saturating exponential with  $D_c$  being the characteristic dose in the exponential. At times sufficiently long that equilibrium is effectively reached, dn/dt = 0 and the fraction of traps occupied is:

[A2] 
$$n/N = 1/\{1 + D_c / D\tau\}$$

With typical values of  $D_c = 800$  Gy and  $D^2 = 2$  Gy/ka, for n/N = 0.9 one deduces J = 4 Ma. The probability per unit time of an empty trap being filled by the radiation is  $D^2 D_c = 400$  ka<sup>-1</sup>.

| Sample                  | Description  | g (%/decade)    | $I_{FS}/I_{LS}$   |  |  |  |  |
|-------------------------|--|-----------------|-------------------|--|--|--|--|
| granites                |  |                 |                   |  |  |  |  |
| BSG1                    | Baltic Sea granite, Åland, Finland   | $1.4\pm0.6$     | $0.694\pm0.015$   |  |  |  |  |
| CGS                     | Coryell granite, Westbridge, B.C.  | $5.83 \pm 0.44$ | $0.401 \pm .018$  |  |  |  |  |
| MCG                     | granodiorite, McKay Cr., North Vancouver, B.C.                                     | $1.0 \pm 0.4$   | $0.840\pm0.015$   |  |  |  |  |
| RMG                     | "red granite", Rannoch Moor, U.K.  | $4.95\pm0.35$   | $0.513 \pm 0.018$ |  |  |  |  |
| sanidine                |  |                 |                   |  |  |  |  |
| WCRS                    | sanidine crystal, Kettle River Valley, B.C.  | $10.1\pm0.3$    | $0.27\pm0.04$     |  |  |  |  |
| sediments               |  |                 |                   |  |  |  |  |
| CBSS                    | Eocene or Oligocene sandstone, China<br>Beach, Vancouver Island, B.C. <sup>a</sup> | $3.12\pm0.31$   | $0.542\pm0.012$   |  |  |  |  |
| GP10                    | Pliocene Yorktown Formation, Gomez Pit,<br>Virginia, U.S.A. <sup>b</sup>           | $4.37\pm0.36$   | $0.445\pm0.011$   |  |  |  |  |
| sediments in permafrost |  |                 |                   |  |  |  |  |
| DY23                    | red sand, Diring Yuriakh, Siberia, Russia ° $4.1 \pm 0.5$ $0.696 \pm 0.03$         |                 |                   |  |  |  |  |
| TML1                    | Pliocene Unit, Bluefish basin, Yukon <sup>b</sup>                                  | $3.66\pm0.094$  | $0.741 \pm 0.012$ |  |  |  |  |

TABLE 2

Table 2. Measured fading rates, g, and ratios of the luminescence intensities for field saturation and laboratory saturation. The value of g was evaluated at a starting point 2 days after the irradiation for a sample bleached and given a dose of 175 Gy.  $I_{LS}$  is the value corrected to 100 days after the end of irradiation. B.C. is British Columbia, Canada.

<sup>a</sup> Yorath and Nasmith 1995, p.129.

<sup>b</sup> Lamothe and Auclair 1999.

<sup>c</sup> Mochanov 1988.

- Fig. 1: Data showing the range of measured fading rates. MCG is a granodiorite, K13 is a microcline, A3 is an oligoclase and ML1 is gem-quality labradorite. The ordinate is the luminescence intensity, I, resulting from a short excitation by 1.4 eV photons (infrared), divided by the intensity,  $I_1$ , from the first measurement at ~ 2 days after laboratory irradiation. All samples were bleached, given a dose of ~175 Gy of <sup>60</sup>Co gamma rays and heated at 120 <sup>B</sup>C for 16 hours before the measurements.
- Fig. 2: a) Histogram of the fading rates measured for K-feldspars separated from sediments. The main histogram has 77 points; the samples are from a wide variety of provenances around the world and many of them are listed in Huntley and Lamothe (2001). The cross-hatched area shows fading rates for a subset of 30 samples from the prairie provinces of Canada (Alberta, Saskatchewan and Manitoba). The dark-shaded area shows fading rates for a subset of samples derived from volcanic source material. b) Histogram showing the range of fading rates measured for specific feldspars.
- Fig. 3: a) Fading rate vs Ca content for a variety of plagioclase feldspars. Ca contents are in % cation (albite is 0 % Ca and anorthite is 20 % Ca). The lower right point is for labradorite sample P10 which shows an exception to the correlation of fading rate with Ca content shown by the other samples.
  - b) The same as (a) except for Fe instead of Ca.
- Fig. 4: Dose response curves illustrating the meaning of field and laboratory saturation intensities,  $I_{FS}$  and  $I_{LS}$  respectively. The data are for silt grains from sample Davis Creek Silt, Saskatchewan, which is associated with a tephra bed with a fission track age of  $8.3 \pm$ 0.2 Ma (Vreeken and Westgate, 1992). The ordinate is the luminescence intensity caused by a short excitation by 1.4 eV photons (infrared). The crosses are data for aliquots that have had various laboratory doses. The solid symbols are for aliquots that were given a red/infrared bleach and then laboratory doses. For both sets, all the aliquots were then heated at 120 <sup>B</sup>C for 16 hours after irradiation to empty the shallow traps. The line is a saturating exponential fit to both data sets, with one data set shifted parallel to the dose axis, the shift being a parameter in the fit.
- Fig. 5:  $I_{FS}/I_{LS}$  (the ratio of the luminescence intensities at field saturation and laboratory saturation) vs g, the rate of anomalous fading evaluated at a starting point 2 days after the irradiation for a sample bleached and given a dose of 175 Gy.  $I_{LS}$  was evaluated 100 days after irradiation. The line is drawn to show that there is a plausible relation between  $I_{FS}$  $I_{LS}$  and g, if the permafrost samples are omitted. The line is drawn through (0,1) to show that it is plausible that  $I_{FS}/I_{LS} = 1$  if g = 0. Any reasonable extrapolation of the data to g =0 yields  $I_{FS} / I_{LS} > 0.9$ , from which a lower limit to the thermal lifetime of the trapped electrons can be deduced as described in the text.
- Fig. 6: Luminescence intensity as a function of laboratory dose for samples CBSS and DY23. Open squares are for aliquots that have first been given a red/infrared bleach. Solid circles are for aliquots that have not been given a bleach; these data have been shifted parallel to the abscissa by an amount that matches the two data sets as in Fig.4. N indicates aliquots given no laboratory dose; in this case the intensity is the field saturation intensity. Fading rates are shown beside the points. For the aliquots not bleached these fading rates are for the component of the intensity above the field saturation intensity (N).

Table 1. Fading rates and compositions for the feldspars. Concentrations are given as % cation. Ideally those for K, Na, Ca, Mg, Ba and Mn shown should total 20. Deviations indicate the presence of other minerals. A3 is known to contain quartz and the numbers have been adjusted to reflect this. A sample is from Canada if no country is stated. n.d.= not detected.

| sample   | g (%/decade) <sup>a</sup> | Κ    | Na   | Ca   | Fe   | Mg   | others > 0.01                       |  |
|--|---------------------------|------|------|------|------|------|-------------------------------------|--|
| alkali feldspars, in order of decreasing K content |                           |      |      |      |      |      |                                     |  |
| K13  | $5.0\pm0.3$               | 16.2 | 2.8  | 0.07 | 0.07 | 0.03 |                                     | microcline, Madawaska, Ontario. Ward s 46E 5124  |
| HFC  | $4.4\pm0.4$               | 14.5 | 4.5  | 0.10 | 0.08 | 0.01 |                                     | perthite, Ontario  |
| K10  | $4.4\pm0.2$               | 14.3 | 4.2  | 0.07 | 0.04 | n.d. |                                     | microcline, Keystone, S.Dakota, U.S.A. Ward s #1 of 45 E 2941  |
| K9   | $3.9\pm0.3$               | 14.1 | 4.3  | 0.02 | 0.06 | n.d. | 0.04 P                              | microcline, var. amazonite, Kola peninsula, Murmansk, U.S.S.R., Ward s #5 of 45 E 2941 & 46E 5164 $^{\rm d}$ |
| K6   | $0.9\pm0.3$               | 14.0 | 4.8  | 0.06 | 0.10 | 0.01 | 0.24 P                              | microcline, Ward s 45W 9252  |
| K8   | $2.8\pm0.2$               | 13.6 | 4.3  | 0.14 | 0.19 | 0.03 |                                     | perthite (microcline & albite) , Perth, Ontario, Ward s #4 of 45 E 2941 & 46E 0514 $^{\rm d}$                |
| K3   | 8.2 <sup>f</sup>          | 13.5 | 4.4  | 0.22 | 0.08 | n.d. | 0.07 Ba                             | orthoclase, Red Lodge, Montana, U.S.A., U.B.C. TC313A6 <sup>b,c,d</sup>                                      |
| K11  | $23.4\pm0.9$              | 12.1 | 3.7  | 3.3  | 0.77 | 0.33 | 0.10 Ti, 0.18 Ba<br>0.02 P, 0.03 Mn | orthoclase (Carlsbad Twin), Gothic Colorado, U.S.A., Ward s #3 of 45 E 2941 $^{\rm d}$                       |
| K7   | $12.2\pm0.6$              | 9.9  | 7.2  | 0.01 | 0.24 | 0.01 |                                     | microcline, Crystal Peak, Colorado, U.S.A. Ward s #2 of 45E 2941 $^{d}$                                      |
| A5   | $1.6\pm0.6$               | 9.0  | 10.4 | 0.40 | 0.26 | 0.05 | 0.03 Ba                             | albite (sic), Bancroft, Ontario, Ward s $\#7$ of 45 E 2941 <sup>d</sup>                                      |
| A4   | 3.0 ± 1.2                 | 4.4  | 10.8 | 3.5  | 3.3  | 1.65 | 0.74 Ti, 0.05 Ba<br>0.30 P, 0.10 Mn | anorthoclase, Larvik, Norway, Ward s #6 of 45 E 2941 <sup>d</sup>  |
| K12  | $10.0\pm1.2$              | -    | -    | -    | -    | -    |                                     | orthoclase, India, Ward s 49E 5919   |

# plagioclases, in order of increasing Ca content

| A2    | $1.3\pm0.2$    | 0.20 | 18.2 | 0.26 | 0.09 | 0.01 |                                     | albite, Amelia Courthouse, Virginia, U.S.A., Ward s 49E 5851 $^{\rm d}$                     |
|-------|----------------|------|------|------|------|------|-------------------------------------|---|
| A6    | $2.1\pm0.2$    | 0.25 | 17.9 | 0.26 | 0.07 | n.d. | 0.21 P                              | albite, var. Cleavelan dite, Keystone, S. Dakota, U.S.A., Ward s $^{\rm d}$ #9 of 45 E 2941 |
| P18   | $1.6\pm0.4$    | 0.45 | 18.6 | 1.2  | 0.01 | n.d. | 0.05 Ba                             | oligodase, Virginia, U.S.A. <sup>e</sup>  |
| #17   | $10.6\pm0.7$   | 1.8  | 14.1 | 3.2  | 0.61 | 0.2  | 0.03Ti                              | oli goc la se, Arendal, Nor way <sup>e</sup>  |
| A1    | $12.7 \pm .6$  | 1.4  | 13.1 | 4.4  | 0.20 | 0.16 |                                     | albite, Bancroft, Ontario, Ward s 46E 0234 <sup>d</sup>                                     |
| #18   | $12.8\pm0.5$   | 1.1  | 13.6 | 4.9  | 0.19 | 0.02 |                                     | oligoclase, Sweden <sup>e</sup>   |
| #19   | $7.1\pm0.8$    | 0.66 | 13.8 | 5.0  | 0.05 | 0.03 |                                     | oligoclase, Ren frew County, Ontario <sup>e</sup>   |
| #20   | $9.1\pm0.4$    | 0.5  | 13.3 | 5.2  | 0.15 | 0.01 |                                     | oligoclase, Mitchell County, N.Carolina, U.S.A. <sup>e</sup>                                |
| A3    | $14.2\pm0.4$   | 0.25 | 14.3 | 5.4  | 0.33 | 0.07 | 0.05 P, 0.03 Cr                     | oligoclase, Mitchell County, N.Carolina, U.S.A., Ward s #10 of 45 E 2941 $^{\rm d}$         |
| P10   | $3.1 \pm 0.8$  | 0.53 | 8.3  | 10.1 | 0.65 | 0.19 | 0.15 Ti, 0.03 Ba<br>0.03 P          | labradorite, Nain, Labrador, Canada, Ward s #11 of 45 E 2941 $^{\rm d}$                     |
| ML1   | $22.4\pm1.0$   | 0.61 | 8.4  | 10.3 | 0.85 | 0.81 | 0.35 Ti                             | labradorite, Madagascar   |
| P11   | $7.6 \pm 2.1$  | 1.3  | 7.8  | 10.7 | 1.6  | 1.00 | 0.27 Ti, 0.02 Ba<br>0.43 P, 0.03 Mn | and<br>esine, Essex County, N.Y., U.S.A., Ward s #12 of 45 E 2941<br>d                      |
| other |                |      |      |      |      |      |                                     |   |
| BRS   | $16.7\pm0.6$   | 14.2 | 2.2  | 7.7  | 0.29 | 0.09 | 0.04 Ti, 0.30 Ba<br>0.02 Mn         | sanidine, Kettle Valley, B.C.   |
| WCRS  | $10.1 \pm 0.6$ | 13.0 | 3.2  | 7.9  | 0.37 | 0.12 | 0.04 Ti, 0.30 Ba<br>0.03 Mn         |   |
| PLTF  | $34.5\pm4.2$   | 9.3  | 7.9  | 1.2  | 0.59 | 0.21 | 0.11 Ti, 0.14 Ba<br>0.05 P, 0.02 Mn | Puy de la Tache, France   |

<sup>a</sup>g is the fading rate evaluated at a starting point 2 days after the midpoint of the irradiation

<sup>b</sup> thermoluminescence spectra are in Huntley et al. (1988)

<sup>c</sup> an extensive study of this sample can be found in Short and Huntley (2000) and Short (2003).

<sup>d</sup> emission and excitation spectra are in Baril and Huntley (2003a, b) and Baril (2002)

<sup>e</sup> sample from University of Adelaide; thermoluminescence spectra are in Prescott and Fox (1993); concentrations from Akber

(1986). <sup>f</sup> the data for K3 are not linear; the value shown is an average.





Fig. 2







Fig. 6