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1 **Exploring sources of variation in thermoluminescence emissions and anomalous fading in alkali**
2 **feldspars**

3

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12

13 **Highlights**

14 - TL emission spectra dependent on mineral phases, framework disorder and interfaces

15 - Single-phase feldspars show little anomalous fading

16 - Framework disorder increases intensity of blue TL emission and IRSL fading rate

17

18 **Abstract**

19 Alkali feldspar is routinely used in retrospective dosimetry using luminescence methods. However
20 there is a signal loss over time, termed ‘anomalous fading’, which results in age underestimation if
21 uncorrected. Although significant improvements have been made in recent years, luminescence
22 dating of feldspars remains challenging. This paper investigates the relationships between chemistry,
23 structural state and the scale of exsolution with thermoluminescence (TL) emission spectra and
24 infrared stimulated luminescence (IRSL) fading rates.

25 We measure TL emission spectra, where possible linking the recombination site to physical features
26 of the feldspar crystals. We show that fading rates are lowest in ordered end-member Na- and K-
27 feldspars but significantly greater in disordered end-members, showing that Al-Si order influences
28 fading. As well as having very low fading rates, ordered end-member samples have distinctive TL
29 emission spectra, with the yellow-green emission dominant, while all other samples have a dominant
30 blue emission. Perthite, i.e. exsolved members of the (Na,K)-feldspar solid solution, show greater
31 fading than disordered end-members and fading is greatest in semi-coherent macropertthite. We
32 propose that the state of Al-Si-order, and the occurrence of defects and dislocations at the perthite
33 lamellar interfaces influence anomalous fading rates in feldspar.

34 **Keywords**

35 Alkali feldspar, thermoluminescence, emission spectra, fading rate, framework disorder

36 **1 Introduction**

37 Luminescence dating of feldspars is routinely conducted using the infrared stimulated luminescence
38 (IRSL) signal and by measuring the emission in the blue (~410 nm) (e.g. Buylaert et al., 2012; Reimann
39 et al., 2012; Smedley et al., 2016; Riedesel et al., 2018). However, feldspar IRSL dating has a major
40 limitation, because it suffers from a signal loss at ambient temperatures, termed anomalous fading
41 (Wintle, 1973; Visocekas, 1985), which results in age underestimation if uncorrected. A range of
42 correction methods has been developed (e.g. Huntley and Lamothe, 2001; Kars et al., 2008), but these
43 often result in large uncertainties on the final age. To circumvent the issues associated with having to
44 quantify and try to correct for fading, research over the past decade has focussed on the isolation of
45 an IRSL signal from feldspar that minimises anomalous fading. Lower fading rates have been obtained
46 by increasing the stimulation temperature (Thomsen et al., 2008) and reduced further by using the
47 signal arising from a second IR stimulation at elevated temperature (termed post-infrared infrared
48 stimulated luminescence signal, post-IR IRSL, e.g. Thomsen et al., 2008; Thiel et al., 2011; Buylaert et

49 al., 2012), or by using a series of IR stimulations at increasing temperatures (termed multiple-elevated-
50 temperature protocols, MET, e.g. Li and Li, 2011).

51 The cause of anomalous fading has been debated for more than 40 years (e.g. Wintle, 1977; Templer,
52 1986; Sanderson, 1988; Visocekas et al., 2014) and a loss of charge from electron traps over time, due
53 to quantum mechanical tunnelling, is the most widely accepted explanation (e.g. Visocekas, 1985).

54 The rate of anomalous fading has been linked to the density of available recombination centres
55 (Huntley, 2006) and significant variations in the rate of fading have been observed in single-crystal
56 feldspars (e.g. Spooner, 1992, 1994; Huntley and Lian, 2006) and in feldspar (single) grains extracted
57 from sediments and bedrock (e.g. Neudorf et al., 2012; Trauerstein et al., 2012; Valla et al., 2016).

58 Visocekas and Zink (1995) measured tunnelling afterglow in microcline and sanidine feldspars and
59 inferred that increased anomalous fading of volcanic feldspars arises due to large concentrations of
60 defects in the crystal caused by Al^{3+} and Si^{4+} disorder on the framework, and additionally due to the
61 inclusion of Fe^{3+} on tetrahedral sites. Based on this observation, Visocekas and Zink (1995) suggested
62 that ordered feldspars, such as microcline, could be dated, because their ordered structure would
63 prevent fading. Indeed, Spooner (1994) observed a stable IRSL signal with the emission recorded
64 between ~ 330 nm and ~ 630 nm (using 2 mm Schott BG39 filter) for one of his microcline feldspars and
65 also for an Amelia albite specimen. In contrast, Huntley and Lian (2006) measured IRSL fading rates of
66 the ~ 360 to ~ 620 nm emission (using BG39 and Kopp 4-97 filters) of up to 12 % in feldspars that they
67 describe as microclines.

68 However, in most studies the samples are only characterised by their chemical composition, and
69 although a description, which includes the structural state of the feldspar is often included
70 (terminologies such as microcline or sanidine), no data are shown that proves this association. Thus it
71 is difficult to compare fading rates measured with any other mineralogical property of the samples
72 other than the chemical composition. Anomalous fading has often been observed and associated with
73 the volcanic origin of the samples (e.g. Wintle, 1973; Visocekas and Zink, 1995, 1999; Guérin and

74 Visocekas, 2015). However, few fading feldspars used in luminescence dating are of volcanic origin
75 (e.g. Valla et al., 2016; King et al., 2016; Jenkins et al., 2018).

76 Due to exsolution in alkali feldspars during slow cooling in plutonic rocks, such as granite, feldspars
77 are often perthitic (e.g. Parsons et al., 2015). A relationship between interfaces in perthites and the
78 intensity of the UV emission (~290 nm) has been found (Garcia-Guinea et al., 1996). Baril (2004)
79 observed phase-dependent variations of violet and yellow luminescence emissions in perthites, and
80 Correcher et al. (2000) linked the blue luminescence in feldspars with alkali self-diffusion along
81 interfaces offset by hole centres. Spooner (1992) observed variable fading in his feldspars, which
82 contain Na and K. However, no clear relationship between exsolution features and anomalous fading
83 rates has yet been established. Additionally, earlier studies often analysed the IRSL signal of feldspars
84 with a wide emission range (e.g. Spooner, 1992, 1994; Visocekas and Zink, 1999; Huntley and Lian,
85 2006), likely including emissions arising from different defect types (cf. Krbetschek et al. (1997) for a
86 review on emission centres and associated crystal defects in feldspars). Developments within the last
87 decade have changed measurement protocols (e.g. Thomsen et al., 2008; Li and Li, 2011) and a wide
88 range of available optical filters enables the isolation of a narrow emission window, which allows a
89 specific emission to be selected. Thus, there is the need for a re-investigation of potential causes of
90 anomalous fading of the blue IRSL signal in chemically and structurally different feldspars.

91 This paper investigates the links between the blue luminescence emission, the anomalous fading rate
92 of the blue IRSL signal and multiple mineralogical characteristics in alkali feldspars. It explores
93 temperature- and wavelength-resolved variations in the blue luminescence intensity and variations in
94 anomalous fading rates of 12 single-crystal alkali feldspars, and three feldspar grain mixtures extracted
95 from bedrock and sediment. This suite of samples enables the investigation of potential relationships
96 between variations seen in the intensity and anomalous fading rate of the blue luminescence emission
97 and (i) the chemistry of the samples, (ii) the number of phases present in a single crystal specimen (i.e.
98 whether they are single phase or perthitic) and (iii) the degree of framework disorder of the samples.

100 **2 Brief overview of the chemistry and structure of feldspars**

101 The crystal lattice of feldspar is made up of (Al,Si)O₄ tetrahedra, each connected to four others by
102 shared or “bridging” oxygen ions at their vertices, forming a continuous, interconnected framework.
103 There are four tetrahedral positions within the framework (two of each of the so-called T₁ and T₂-
104 sites), which are occupied by Si⁴⁺ or Al³⁺. The framework has a net negative charge, which is offset by
105 large cations (predominantly Na⁺, K⁺, Ca²⁺) occupying cavities in the framework termed M-sites (Deer
106 et al., 2013). The compositional variability of most feldspars is accommodated by three end members,
107 each with a different M-site cation: KAlSi₃O₈ (K-feldspar), NaAlSi₃O₈ (Na-feldspar) and CaAl₂Si₂O₈ (Ca-
108 feldspar). The solid solution between K- and Na-feldspars produces the alkali feldspar series; the solid
109 solution between Na- and Ca-feldspar is termed plagioclase. There is very limited solid solution
110 between K- and Ca-feldspar (<10 %). At high temperature (> 1000 °C), complete solid solution across
111 the plagioclase and alkali feldspar series occurs (see Parsons 2010).

112 Here we consider the alkali feldspar solid solution series between K- and Na-feldspar. The structural
113 state of alkali feldspar is an important indicator of the temperature of crystallisation and subsequent
114 thermal history (Deer et al., 2013) and hence alkali feldspar is divided broadly into low- and high-
115 temperature members. The structural state of low- and high-temperature alkali feldspars is dictated
116 by the degree of ordering of Al and Si within the framework and, in the case of albite (Na-feldspar),
117 coupled with displacive distortions to the framework. Ordered feldspar is equilibrated at low (e.g.
118 <300°C) temperatures. Generally, increased ordering of Al and Si within the framework takes place
119 during slow cooling, e.g. in plutonic rocks. During rapid cooling a disordered distribution of Al and Si
120 at high temperatures is quenched (e.g. sanidine, the monoclinic K-feldspar in volcanic rocks) (Deer et
121 al., 2013).

122 The progressive ordering in alkali feldspars can be understood in terms of the distribution of Al³⁺ on
123 the framework. In the high-temperature K-feldspar (monoclinic high sanidine), Al³⁺ has an equal (25 %)

124 chance of occupying any of the four tetrahedral sites ($T_1(0)$, $T_1(m)$, $T_2(0)$ and $T_2(m)$) within the more
125 open alkali feldspar framework. During slow cooling, as the framework contracts, Al^{3+} diffuses to
126 occupy preferentially one of the two T_1 -sites (giving the monoclinic low sanidine). With continued
127 slow-cooling, Al^{3+} preferentially diffuses into one of the T_1 -sites ($T_1(0)$, see Parsons 2010), causing a
128 change in symmetry from monoclinic (sanidine, with the greatest disorder) to triclinic, the ordered K-
129 feldspar maximum microcline (Deer et al., 2013). Associated with this drop in symmetry, microcline
130 adopts repeated twinning in two different orientations visible in a polarising microscope, termed
131 'tartan' or 'tweed' twinning (Deer et al., 2013).

132 Although alkali feldspar is a continuous solid solution at high temperature, the small ionic radius of
133 Na^+ (0.118 nm) triggers a displacive collapse of the framework around Na^+ as the structure attempts
134 to maintain bonding to the smaller Na^+ ion. The feldspar framework cannot collapse around the larger
135 K^+ ion (0.151 nm), hence the alkali feldspar solid solution becomes unstable on cooling and unmixing
136 ('exsolution') of the K- and Na-rich phases in alkali feldspar occurs. Such alkali feldspars are single
137 crystals comprising intergrowths of separate K- and Na-rich feldspars, termed 'perthite'; perthites are
138 further divided into crypto-, micro- and macroperthite, dependent on the scale of the intergrowths
139 (from sub-optical, nanometre scale cryptoperthite, microperthite with lamellae visible under a
140 petrographic microscope, and macroperthite whose lamellae are visible to the naked eye). The cooling
141 rate and fluid interaction after crystallisation of the magma influence the size of the perthite lamellae,
142 with cryptoperthite forming earlier in cooling (Brown and Parsons, 1984a) and coarsening over time.
143 Fluid interaction is involved in the coarsening process of micro- and macro-perthite (e.g. Parsons,
144 1978). Near-pure Na and K feldspars may occur in a variety of environments including deep, high
145 pressure (>0.5 GPa) or high-water content crystallisation of granites/pegmatites (including samples
146 used here e.g. from Minas Gerais, Brazil, CLBR, Table 1, Cassedanne and Roditi, 1996) or wet (low-
147 intermediate grade) metamorphic settings (e.g. albite Al-I, Table 1, Govindaraju, 1995). In these cases,
148 the alkali-feldspar phase relationships during various combinations of slow-cooling, water content and
149 magma/protolith composition can produce almost pure end-member ordered alkali feldspars (see

150 Deer et al., 2013, Parsons, 1978; Parsons et al., 2005). The homogeneity of samples from some of
151 these “exotic” environments (e.g. pegmatites) mean that such pure end-member samples are often
152 used as reference materials or samples for experimentation.

153

154

155 **3 Material and methods**

156 **3.1 Samples**

157 The samples used in this study can be split into two groups, single crystal feldspar specimens and
158 examples of feldspars routinely used for sediment dating and luminescence thermochronometry. The
159 chemical composition of the samples and the mineral phases present were determined by X-ray
160 fluorescence (XRF) and X-ray diffraction (XRD), respectively. Additionally, the samples were examined
161 optically for their colour and the coarseness of any perthite lamella that may be present. We combined
162 our visual inspection, following the definition of perthite microstructures by Deer et al. (2013, p. 253),
163 and the results of the semi-quantitative phase analyses using XRD to separate the perthitic samples
164 into two groups: Both groups have in common that they were determined to be two-phase using XRD.
165 One group of perthites has K- and Na-feldspar lamellae on the optical scale, so that we were able to
166 identify them by eye or using a hand lens. We refer to these feldspar samples as macroperthite. The
167 second group consists of perthite where XRD results defined them as two-phase, but where we were
168 unable to see the lamellae with the naked eye or even through an optical microscope. We refer to
169 these feldspars as cryptoperthite.

170 The chemical composition of the samples was determined by XRF. For XRF measurements 2 g of
171 cellulose binder was placed into an Al sample boat. Powdered sample (0.1 g) was then mixed with
172 cellulose binder (0.1 g) and this mixture was spread on top of the cellulose binder in the Al sample
173 boat. Each boat was then pressed at 20 t to pellets for XRF measurements using a PANalytical MagiX

174 PRO XRF spectrometer equipped with a Rh anode X-ray source. The spectra were collected over ten
175 different energy ranges and then analysed to obtain semi-quantitative results. For the analyses, it was
176 assumed that all elements, except for Cl, were present as oxides. The chemical composition of all
177 samples is summarised in Table S1. Individual sample compositions were calculated to give the
178 molecular formula of the feldspar based on eight oxygen, following stoichiometry. The results are
179 shown in Figure 1A and given in Table 1.

180 Semi-quantitative phase analysis results of the samples were obtained using XRD. The measurements
181 were carried out using a PANalytical X'Pert MPD powder diffractometer operating with Cu K α X-rays
182 and a PIXCEL-1D area detector. For these measurements 0.1 g of powdered sample was mounted on
183 a low-background silicon XRD sample holder using acetone. Data were collected over the range of 5-
184 100 degrees 2 θ , each scan took just over 1 hour.

185 Because of the limited availability of sample material for samples R1-11A and CLBR, and since very
186 detailed characterisation is already published, detailed XRF and XRD measurements were not
187 performed. The chemistry and mineralogy of R1-11A was investigated in detail by Harrison et al.
188 (1990) and the results from their study are used here and are presented in Table 1. The chemical
189 composition determined by Harrison et al. (1990) is based on electron probe microanalyses. Rendell
190 and Clarke (1997) and Garcia-Guinea et al. (1999) performed luminescence experiments and
191 mineralogical investigations of CLBR and determined its structural state as low albite. Additionally,
192 Riedesel et al. (2019) assessed its chemical composition using an XRF detection head attached to a
193 Risø TL/OSL reader (Kook et al., 2012; and see the supplementary material of Stevens et al., 2018). For
194 this measurement a shard of the sample (~1 mm diameter) was placed on a Mo cup (without silicone
195 spray) and the chemical composition was measured three times. The result of this measurement
196 procedure is given, alongside the chemistry of the other samples, in Table 1.

197 Details regarding the origin of the samples, their chemical composition and mineralogical properties
198 as determined using the analyses described above are given in Table 1 for the single crystal specimens,

199 and in Table 2 for bedrock and sediment extracts. Single-phase feldspars included in the sample suite
200 are single-phase microcline (FSM-13), and single-phase albite (CLBR and Al-I). Note that Al-I includes
201 5 % quartz, based on semi-quantitative XRD measurements. Photoluminescence and excitation
202 spectra of CLBR were presented by Riedesel et al. (2019, referred to as Cleavelandite).

203 FSM-5 and FSM-11 are macroperthite, with the perthite lamellae visible with the naked eye. Based on
204 semi-quantitative phase analyses using XRD the phases present in these two samples are albite and
205 microcline. Cryptoperthite are R1-11, FSM-3, FSM-7, FSM-8 FSM-14 and FSM-15. The perthite phases
206 are microcline and albite in the case of R1-11A, FSM-3, FSM-7, FSM-8, FSM-14 and FSM-15, and
207 orthoclase and albite for FSM-6, as determined using XRD. Photoluminescence, excitation spectra and
208 cathodoluminescence of R1-11A were presented by Riedesel et al. (2019) and Finch and Klein (1999).

209 WHB-7, the density separated feldspar extract from unconsolidated sediment, consists of microcline
210 and albite, but XRD also showed the presence of muscovite and quartz. HAM-5, another feldspar
211 extract from sediment, consists of orthoclase, albite, quartz and illite, based on semi-quantitative XRD
212 results. The thermochronometry sample MBT-I-2430 consists of feldspar only, in this case microcline
213 and albite. Excitation spectra of HAM-5 and MBT-I-2430 were presented by Riedesel et al. (2019).

214 Whilst all these samples allow us to investigate the influence of differences in the chemical
215 composition and the mineral phases, laboratory heating experiments on FSM-6 and FSM-13 were
216 conducted to enable the investigation of potential effects of framework disorder on the luminescence
217 emissions and fading rates.

218 To explore the effect of Si and Al disorder on luminescence properties, 0.5 g of powdered sample
219 material of samples FSM-6 and FSM-13 (referred to as FSM-6LH and FSM-13LH after heating) were
220 placed in acid cleaned Pt crucibles and heated to 105 °C overnight to remove any potential liquid
221 present in the sample material. The Pt crucibles with the sample material were then placed in a
222 preheated furnace at 1050 °C and covered with fitted Pt lids. The samples remained in the furnace at
223 1050 °C for 5 days (FSM-6LH) and 10 days (FSM-13LH) to allow disordering of Si and Al on the

224 framework. To keep the disordered structure, the samples were rapidly cooled by placing the Pt
225 crucibles in cooled sample holders. Comparison of X-ray diffraction patterns of the unheated and
226 heated material of samples FSM-13/FSM-13LH and FSM-6/FSM-6LH reveals differences linked to
227 states of Al-Si order (Fig. 1B, C). The (131) diffraction peak indicates the triclinicity of the material (a
228 narrow single peak if the sample is triclinic, two peaks if the sample is monoclinic, e.g. Garcia-Guinea
229 et al., 1999). Firstly, Figure 1B and C indicate different degrees of disorder of the two starting materials
230 of FSM-13 and FSM-6 in their X-ray diffraction pattern. Whilst the XRD pattern of FSM-13 shows two
231 fully defined diffraction peaks (131 and $\bar{1}\bar{3}\bar{1}$) between 29° and 32° 2θ , FSM-6 shows only a single peak
232 (131) at 29.8° 2θ with a minor peak ($\bar{1}\bar{3}\bar{1}$) around 30° 2θ . This supports results from semi-quantitative
233 phase analyses, based on which FSM-13 was defined as microcline and FSM-6 as two-phase consisting
234 of albite and orthoclase (Table 1). Samples described as orthoclase are microcline with incomplete
235 order of Al^{3+} on T_1 -sites (Deer et al., 2013, p. 256-257) Secondly, as a result of the heating experiment,
236 the two individual peaks in the diffractogram of FSM-13 (Fig. 1B) move closer together (see red line)
237 and the minor diffraction peak in the diffraction pattern of FSM-6 (Fig. 1C) disappears and only a single
238 diffraction peak around 30° 2θ is visible after heating (FSM-6LH, Fig. 1C, red line). This shows that the
239 heating experiment is successful in disordering both samples, but that the heated material of FSM-
240 6LH is relatively more disordered compared to FSM-13LH.

241 In addition to changes of tetrahedral site occupancy by Al^{3+} , prolonged heating is expected to influence
242 the distribution of K^+ and Na^+ ions within the perthitic sample FSM-6. Whilst semi-quantitative phase
243 analyses of the unheated sample FSM-6 characterised it as two-phase, with albite and orthoclase as
244 the phases present, the heated sample FSM-6LH is defined as sanidine by XRD, where Na^+ and K^+ ions
245 are distributed randomly throughout the crystal and not separated in perthite lamellae of K- and Na-
246 feldspar. This becomes apparent when comparing the position of the ($\bar{2}01$) diffraction peak (Deer et
247 al., 2013, p. 260). In the X-ray diffraction pattern of perthitic sample FSM-6 are two ($\bar{2}01$) peaks, one
248 for each phase (at 20.98° 2θ for the microcline phase, at 22.01° 2θ for the albite phase). In contrast,
249 there is only a single ($\bar{2}01$) diffraction peak in FSM-6LH at 21.25° 2θ (Fig. 1D).

250 We also attempted Rietveld refinement, to refine the crystal structure and to quantify the tetrahedral
251 site occupancy by Al³⁺ and Si⁴⁺ ions. However, due to a strong preferred orientation of the sample
252 material a quantification of the tetrahedral site occupancy was not possible. When using XRD in this
253 manner, we recognise that XRD is a measure of long-range tetrahedral site order (i.e. the development
254 of large, e.g. 100s nm, ordered domains) and relatively insensitive to local short-range order which is
255 better explored by local probes such as Nuclear Magnetic Resonance (NMR, Xiao et al. 1995).

256

257 **3.2 TL emission spectra**

258 Thermoluminescence (TL) emission spectra were measured using an Andor spectrometer (Prasad,
259 2017) attached to a Risø DA20 TL/OSL reader equipped with a ⁹⁰Sr/⁹⁰Y beta source delivering ~0.1 Gy/s
260 at the sample position and a DASH (Lapp et al., 2015). The single-photon EMCCD-based spectrograph,
261 used to record the spectra, is attached to the Risø reader via a fibre optic bundle cable. The
262 thermoelectrically cooled EMCCD camera (Andor iXon Ultra 888) has a 1024 x 1024 sensor format and
263 a pixel size of 13 µm. The spectral resolution of the system is 13 nm for all spectra recorded. The TL
264 signal was recorded without any optical filters, to allow the emission spectrum to be recorded in a
265 wide range of wavelengths (~280 to ~720 nm). The grating of the spectrograph was set to 150
266 lines/mm and 500 nm blaze. The resulting spectra consist of 1024 bins, ranging from 280.68 nm to
267 718.94 nm, with a bin width of 0.43 nm. The electron-multiplying gain (EM) varied for individual
268 measurements, dependent on the sample brightness, except for the unheated and heated material of
269 FSM-13/FSM-13LH and FSM-6/FSM-6LH, where the EM gain was kept the same, to allow absolute
270 signal intensity comparisons.

271 For TL spectra measurements, samples were placed on stainless steel cups and were beta irradiated
272 using different doses (ranging from 60 to 1,000 Gy), to yield optimal signal output for the samples
273 analysed. To be able to compare absolute intensities for the unheated and heated material of FSM-13
274 and FSM-13LH as well as FSM-6 and FSM-6LH the same sample mass (10 mg) and irradiation dose

275 were used (400 Gy for FSM-13 and FSM-13LH; 200 Gy for FSM-6 and FSM-6LH). A 1000 s pause was
276 inserted after irradiation to allow potential phosphorescence to decay prior to the TL measurement
277 up to 380 °C (heating rate 1 °C/s). A second TL measurement was performed, immediately after the
278 first and used for background subtraction. Repeat measurements were performed and the impact of
279 changing the size of the given dose was tested, to check for reproducibility. All spectra were corrected
280 for the response of the instrument.

281 **3.3 IRSL fading measurements**

282 Infrared stimulated luminescence (IRSL) fading measurements were performed using Risø DA20
283 TL/OSL readers equipped with ⁹⁰Sr/⁹⁰Y beta sources delivering ~0.03 Gy/s and ~0.1 Gy/s at the sample
284 position. Sample material was placed on stainless steel cups. Typically, three aliquots per sample were
285 measured.

286 Fading was measured for a signal in response to ~90 Gy using a post-IR₅₀IRSL₂₂₅ protocol (Table 3) with
287 a preheat at 250 °C for 60 s. The test dose signal was measured after a beta dose of ~30 Gy. The IRSL₅₀
288 and post-IR IRSL₂₂₅ signals were recorded through a combination of a Schott BG39 and a Corning 7-59
289 filter (transmission greater than 10 % from 340 nm to 470 nm) using a UV-sensitive photo-multiplier
290 tube (PMT). To ensure stable heating for the IRSL signal measurements, the sample was held at the
291 measurement temperature for 30 s, prior to switching on the IR LEDs. Due to the high signal intensities
292 of FSM-13LH, a ND 1.0 filter was added to the blue filter combination. Delays between different fading
293 measurements ranged from prompt measurements with delays of ~500 to ~800 s between the end of
294 irradiation and luminescence measurement, depending on the instrument, to at least 1,000,000 s. For
295 both signals investigated, the first 10 s were integrated as a signal, and background corrected by
296 subtracting the signal integrated over the last 20 s of each decay curve. Fading rates, expressed as g-
297 values, were calculated using the Analyst software (Duller, 2015) and all g-values presented are
298 normalised to two days (Huntley and Lamothe, 2001).

299 4 Results

300 4.1 TL emission spectra

301 TL emission spectra of all samples investigated are recorded in the wavelength range from 280 to
302 720 nm and will thus give insights into recombination centres which emit in this part of the
303 electromagnetic spectrum. The main emissions observed are centred at ~300 nm, ~400 nm, ~550 nm
304 and >700 nm (e.g. Krbetschek 1997). However, the intensity of these emissions and the temperature
305 at which the emissions occur varies between samples. The emission spectra from the samples can be
306 divided into four groups: those from single-phase feldspars, perthite with lamellae visible with the
307 naked eye (macroperthite), perthite with lamellae on the sub-optical scale (cryptoperthite) and
308 feldspar extracted from sediments and bedrock, which may contain grains of numerous, (possibly
309 different) feldspar group minerals.

310 The emission spectra recorded for the single-phase feldspars (FSM-13, Al-I and CLBR, Fig. 2), show
311 their brightest emission around 550 nm. Additionally, all three samples show an emission around
312 ~300 nm. The two albite specimens (CLBR and Al-I) also show an emission in the blue (~400 nm) and
313 sample Al-I a tail of a red emission around ~720 nm. The emissions at ~300 and ~550 nm of FSM-13
314 are most intense at ~100 °C (Fig. 2). The temperatures at which the emissions of Al-I and CLBR show
315 their maximum intensity differ slightly for each emission (Fig. 2). For Al-I, the peak of the red emission
316 occurs at the lowest temperature (~100 °C). This is followed by the relatively weak blue emission
317 around ~150 °C and the UV (~300 nm) and yellow-green emission (~550 nm) peak around ~170 °C (Fig.
318 2). Whilst FSM-13 and Al-I show one TL peak at any given emitting wavelength, CLBR displays a number
319 of TL peaks. The strongest emission for CLBR, the yellow-green, shows a low temperature peak around
320 ~95 °C, a second peak at ~170 °C and a shoulder around ~250 °C. Similar behaviour is also visible for
321 the weaker blue emission around ~400 nm. The UV emission (~300 nm) also emits around ~95 °C and
322 around ~170 °C (Fig. 2).

323 Perthite in this study are divided into macroperthite and cryptoperthite. Figure 3 shows the emission
324 spectra for all cryptoperthite used in this study. The emission spectra of these samples commonly
325 share a broad emission ranging from ~350 to ~600 nm. Mostly this emission occurs around ~100 °C
326 (Fig. 3), but R1-11A (Fig. 3B) and FSM-7 (Fig. 3F) also show an emission tail towards higher
327 temperatures, resulting in a minor peak around ~350 °C for the emission centred around ~400 nm
328 (Fig. 3). Additionally, all cryptoperthite show the short wavelength tail of a red emission (~700 nm).
329 This emission is strongest for sample FSM-15 (Fig. 3C), a pinkish cryptoperthite, which contains 0.2
330 wt% Fe (Table S1).

331 The emission spectra of the macroperthite samples FSM-5 and FSM-11 (Fig. 4A and B) are similar to
332 those of the cryptoperthite (Fig. 3), as the emissions of these two samples also occur at low
333 temperature (~100 °C). However, the macroperthite show a stronger red emission, compared to the
334 broad blue to yellow emission, which was observed as being dominant in the majority of the
335 cryptoperthite with the exception of FSM-15 (compare Fig. 3 and 4). All emissions for these two
336 samples exhibit a peak at the same temperature (Fig. 4). Sample FSM-11 contains 0.5 wt% Fe, which
337 is consistent with this dominant red emission, though the XRF data for FSM-5 registers no Fe above
338 the detection limit.

339 Further investigation focuses on the effect of prolonged heating to 1050 °C in a furnace and
340 subsequent rapid cooling on TL emission spectra of single-phase feldspars using samples FSM-6LH and
341 FSM-13LH. Figure 6 shows the TL emission spectra for FSM-13 and FSM-13LH, the latter was artificially
342 disordered through heating and rapid cooling as described in section 2.1. The TL spectrum of FSM-13
343 shows a dominant emission around ~550 nm and a dimmer one around ~300 nm (Fig. 5). A weak blue
344 emission (~400 nm) is also present. The maximum intensity of the TL spectrum of FSM-13 of just over
345 3000 cts/°C/0.4 nm is recorded around ~560 nm and at ~100 °C (Figs. 5A and C). The emission
346 spectrum of FSM-13LH differs from the spectrum of FSM-13. FSM-13LH shows its main emission in the
347 blue region of the spectrum, with a peak emission of over 40,000 cts/°C/0.4 nm around ~150 °C (Fig

348 5B). Additionally, the signal now spreads towards higher temperatures, with the blue emission
349 showing a secondary peak around 350 °C (Fig. 5D). A similar shift towards an emission at higher
350 temperature is observed when comparing the TL emission spectra of FSM-6 and FSM-6LH (Fig. 6).
351 Whilst FSM-6 already emitted in the blue region prior to disordering, the spectrum of disordered FSM-
352 6LH shows intensities in the blue emission twice as large as recorded for its unheated counterpart and
353 a narrowing of the emission peak is also observed (Fig. 6A and B). On the basis of the findings for
354 samples FSM-13LH and FSM-6LH, we observe that disordering results in i) an increase in the intensity
355 of the blue luminescence and ii) a shift of the TL emission peak temperature towards higher
356 temperatures.

357 To relate features observed from TL emission spectroscopy of single crystals to material generally used
358 in luminescence dating or thermochronometry, TL spectra were recorded from feldspar extracts from
359 sediments (WHB-7, Fig. 7A and HAM-5, Fig. 7B) and bedrock (MBT-I-2430, Fig. 7C). The emission
360 spectra of these sand-sized feldspar grain mixtures show similar characteristics to the perthitic
361 samples: TL maxima mainly around ~100 °C, a broad emission ranging from ~350 to ~600 nm, and the
362 tail of the red emission (Fig. 7).

363 **4.2 Fading rates**

364 Fading measurements were performed on all samples using a post-IR₅₀|IRSL₂₂₅ protocol (Table 3). This
365 protocol is chosen as it enables the measurement of two IRSL signals, one at low temperature (50 °C),
366 and a second IRSL signal measured at elevated temperature (225 °C). The data from all IRSL₅₀ fading
367 measurements, shown as L_x/T_x ratios after different delays, are presented in Figures S1 to S4 in the
368 supplementary material. In agreement with results shown by Thomsen et al. (2008), we observe
369 consistently lower fading rates for the post-IR IRSL₂₂₅ signal compared to the fading rates of the IRSL₅₀
370 signal of our samples (Table 4). We also observe a proportional relationship between the fading rates
371 obtained for the two IRSL signals (Fig. 8A). Şahiner et al. (2017) have shown that the first IRSL
372 measurement step in a post-IR IRSL or post-IR-MET procedure results in the recombination of

373 electrons with the most proximal recombination centres, implying that fading of this first IRSL
374 measurement can give information on the proximal recombination centres. All g-values (%/decade)
375 presented in the following discussion are those obtained for the IRSL₅₀ signal. The average g-value (\pm
376 1 standard deviation) for each sample is shown as a function of their K-feldspar content in Figure 8B.

377 Single-phase albite (CLBR and Al-I) show the lowest fading amongst this suite of samples for the IRSL₅₀
378 signal, resulting in g-values of 0.36 ± 0.60 and 0.58 ± 0.30 %/decade respectively. The other single-
379 phase sample (microcline FSM-13) was unique in giving very variable g-values, and hence nine sub-
380 samples were measured. Five aliquots yielded g-values of less than 1%/decade ($n = 5$, average value
381 is -0.23 ± 0.76 %/decade), one aliquot gave 1.14 ± 0.72 %/decade, and the three others showed fading
382 rates between 2.00 and 3.12 %/decade. Considering the data together by calculating the average and
383 standard deviation results in a very low g-value but with a high uncertainty (0.93 ± 1.52 %/decade, n
384 = 9). The reason for the variability is not known, but we hypothesise that it is due to small-scale
385 variations of structural state within the sample. Solid solutions of microcline are rarely found in nature
386 and, although this sample was determined as single phase microcline, we cannot exclude perthitic
387 patches or local Al disorder within the sample, which cannot be detected by XRD, but which may
388 influence the fading rates.

389 The second group of samples are perthites. The macroperthites (FSM-5 and FSM-11) exhibit the
390 highest fading rates (8.99 ± 1.82 %/decade, FSM-5, $n = 3$; and 12.42 ± 1.71 %/decade, FSM-11, $n = 3$).
391 Cryptoperthite show fading rates ranging from 1.80 ± 0.32 %/decade (FSM-8, $n = 3$) to 6.72 ± 2.51
392 %/decade (R1-11A, $n = 3$), but all the values are below those recorded for macroperthite.

393 The effect of framework Al disorder on the fading rate was explored using the artificially disordered
394 samples FSM-6LH and FSM-13LH. Heated sample FSM-6LH showed an increase in fading (4.99 ± 0.20
395 %/decade, $n = 3$), compared to the unheated material (FSM-6, 2.75 ± 1.47 %/decade, $n = 3$); similarly,
396 an increase in fading rate is observed for disordered FSM-13LH (3.24 ± 0.16 %/decade ($n = 3$),
397 compared to 0.93 ± 1.52 %/decade ($n = 9$) for FSM-13).

398 Fading measurements were also performed on feldspars extracted from sediment and bedrock
399 samples, to allow a comparison to material routinely used in luminescence dating studies. Here the
400 fading rates measured are within the range of those determined for cryptoperthite. IRSL₅₀ fading rates
401 for sediment samples WHB-7 and HAM-5 are 3.07 ± 0.74 and 1.86 ± 0.46 %/decade, respectively, while
402 the fading rate for the bedrock sample MBT-I-2430 is 3.28 ± 0.14 %/decade.

403 **5 Discussion**

404 **5.1 Thermoluminescence emissions in chemically and structurally different alkali feldspars**

405 The TL emission spectra reveal significant differences in emission centres for single-phase feldspars in
406 comparison to the perthites, particularly regarding the presence and intensity of the blue emission.
407 Single-phase microcline (FSM-13) and albite (CLBR and Al-I) show little blue emission, compared to a
408 relatively intense yellow-green emission, and additionally, an emission in the UV is present in the TL
409 spectra of these samples (Fig. 2). In contrast, in perthitic feldspars (Fig. 3 and 4) the blue emission is
410 dominant, only surpassed in intensity by the red emission in macroperthite (FSM-5 and FSM-11, Fig.
411 4) and cryptoperthite FSM-15 (0.2 wt% Fe, Fig. 3C). Disordering of samples FSM-13 (single-phase
412 microcline) and FSM-6 (perthite: albite and orthoclase) results in an increase in the blue emission
413 intensity recorded for FSM-13LH and FSM-6LH (Fig. 5 and 6). These results are consistent with the blue
414 emission in alkali feldspars being associated with (dis)-ordering of the framework (Finch and Klein,
415 1999) and with the observed sensitisation of the blue emission due to alkali ion leakage caused by
416 prolonged heating (Garcia-Guinea et al., 1999).

417 The defect giving rise to the blue emission in feldspars has been under investigation since the early
418 1980's and it has been suggested that it arises either due to a hole centre located on an Al-bridging
419 oxygen ion ($\text{Al}^{3+}\text{-O}^{1-}\text{-Al}^{3+}$, e.g. Speit and Lehmann, 1982a; Finch and Klein, 1999) or (for plagioclase) due
420 to Eu^{2+} substituting for Ca^{2+} on M-sites (Götze et al., 1999). A solution involving the silicate framework
421 is the most likely and in alkali feldspar, where Ca^{2+} concentrations are minor (e.g. Table 1), a hole

422 centre on an Al-bridging O is the most likely explanation and is consistent with the results of the
423 present study. The Al-O-Al bridge will only occur where there is a degree of Al-Si framework disorder
424 and it is hypothesised that it is stabilised at low temperatures by the addition of an electron hole (Speit
425 and Lehmann, 1976; Speit and Lehmann, 1982b, Finch and Klein 1999). This is consistent with the
426 spectra recorded for single-phase albite and microcline (CLBR, Al-I and FSM-13), which show very little
427 blue TL emission (Fig. 2). Artificially induced disorder in the microcline sample FSM-13 enhances the
428 blue emission in FSM-13LH, again supporting a model in which Al disorder is implicated in the blue
429 emission intensity. However, the perthitic feldspars are two-phase mixtures of albite and microcline,
430 both of which are partially ordered (Table 1). All perthitic feldspars investigated here emit in the blue
431 (Fig. 3).

432 Garcia-Guinea et al. (1999) linked the UV emission to twin interfaces in the feldspar end-members.
433 Albite and microcline both have multiple twinning, the result of the inversion from monoclinic to
434 triclinic symmetry during cooling. Supporting this, Polymeris et al. (2013) observed more intense UV
435 TL emission in ordered microcline, compared to disordered sanidine. Microcline shows repeated
436 twinning, whilst sanidine does not (cf. Deer et al., 2013, p. 253). In perthite, the possible interfaces
437 include not only the twins within each component phase, but also the interfaces between the K- and
438 Na-feldspar end-members. According to heating experiments (Garcia-Guinea et al. 1999),
439 luminescence attributed to twins contributes to the UV and part of the broad blue emission in perthite
440 (cf. TL spectra in Fig. 3). This is supported by Correcher et al. (2000), who linked the blue luminescence
441 in feldspars with alkali self-diffusion along interfaces offset by hole centres.

442 Perthitic feldspar FSM-6 shows an emission in the blue region. Prolonged heating of the sample
443 material disordered both the Na- and K-feldspar end members, thereby reducing strain at the
444 interfaces and (if accompanied by Na and K diffusion) creating single-phase alkali feldspar with a
445 sanidine structure (FSM-6LH, Table 1, Fig. 1D). Associated with this heating, we observe that the blue
446 TL shifts towards higher temperatures, the emission band narrows and the signal intensity doubles

447 from 10,000 to 20,000 cts/°C/0.4 nm (Fig. 6). This increase supports the suggestion that disorder of Si
448 and Al is implicated in the intensity of the emission.

449 Whilst the blue emission is absent or weak in single-phase albite and microcline, the yellow-green
450 emission (~550 nm) is a key feature of their spectra (Fig. 2). Interestingly, this emission has been
451 associated with Mn²⁺ substituting for Ca²⁺ on M-sites in plagioclase (e.g. Geake et al., 1971; Telfer and
452 Walker, 1978), but it is also present in microcline sample FSM-13. This microcline sample has no
453 detectable CaO content (Table S1) and hence it is unlikely that the emission around ~550 nm of this
454 sample is related to Mn²⁺. Based on lifetime measurements of the green-yellow emission in a Na-
455 feldspar sample, Prasad et al. (2016) argued that Mn²⁺ substituting for Ca²⁺ on M-sites cannot explain
456 this emission; its recombination lifetime was too short for spin-forbidden Mn²⁺ transitions. Further
457 work is needed to identify the physical nature of this recombination centre but we concur with the
458 findings of Prasad et al. (2016) that it is inconsistent with Mn²⁺.

459 **5.2 Fading rates of chemically and structurally different alkali feldspars**

460 Fading rates measured for the IRSL₅₀ signal (Table 4, Fig. 8B) of chemically and structurally different
461 alkali feldspars range from 0.36 ± 0.6 %/decade (CLBR, n = 3) to 12.42 ± 1.71 %/decade (FSM-11, n =
462 3). Single-phase feldspars show the least fading, whilst macroperthites (FSM-5 and FSM-11)
463 demonstrate the highest fading rates. Cryptoperthite show a similar fading behaviour to samples
464 extracted from bedrock (MBT-I-2430) and sediment (WHB-7, HAM-5), and the fading rates observed
465 for these, range in between those measured for macroperthite and single-phase feldspars (Table 4,
466 Fig. 8B).

467 The low fading rates of the blue emission of the IRSL₅₀ signal for single-phase albite and microcline
468 samples coincide with the relatively low blue luminescence signal intensities of these samples, in
469 comparison to the green-yellow emission (compare Fig. 2, 3 and 4 for five samples as examples).
470 Isolated centres are typically modelled in a manner whereby luminescence intensity and defect
471 concentration are proportional (e.g. Speit and Lehmann, 1976, 1982a; Finch and Klein, 1999). Fading,

472 understood as tunnelling from an electron trap to a nearby recombination centre (e.g. Visocekas,
473 1985), is dependent on the density of recombination centres in the vicinity of the electron trap
474 (Huntley, 2006). We infer that where the density of recombination centres increases, the distance
475 between the electron trap as a donor and the recombination centre as the acceptor of the tunnelling
476 electron will be smaller (Jain et al., 2012). In contrast, fewer blue recombination centres will result not
477 only in lower signal intensities, but reduces the likelihood of a blue recombination centre being within
478 tunnelling distance of an electron trap and lower fading rates will be observed.

479 Heating of samples FSM-13LH and FSM-6LH increases the blue luminescence emission intensity (Figs.
480 5 and 6), which we interpret as an increase in Al-Si disorder, populating Al-O-Al bridges and thus
481 increasing the number of blue luminescence centres. Along with the increase in blue luminescence
482 intensity, the fading rate increases (Table 4, Fig. 8B) from 0.93 ± 1.52 %/decade (FSM-13) to $3.24 \pm$
483 0.16 %/decade (FSM-13LH) and 2.75 ± 1.47 %/decade (FSM-6) to 4.99 ± 0.20 %/decade (FSM-6LH).
484 Another interesting observation can be made when comparing L_x/T_x values of the unheated and
485 heated samples FSM-13/FSM-13LH and FSM-6/FSM-6LH during the fading measurements (Figs. S1 A,
486 B and S2 G, H). The unheated samples show very variable L_x/T_x values between the different aliquots
487 (for instance ranging from ~ 2.7 to 4.7 for FSM-13, Fig. S1 A) while the heated aliquots have very
488 consistent values (~ 2.7 for the prompt measurement of FSM-13LH, Fig. S1 B). It is also intriguing that
489 the L_x/T_x values of the prompt measurement of the heated samples are similar to the long delay L_x/T_x
490 values of the unheated counterparts. We cannot explain these two observations, but they may
491 indicate that prolonged heating influences the luminescence sensitivity of the sample material, in a
492 way which cannot be corrected by the test dose. A further observation based on the unheated and
493 heated sample pairs is that while we observe an increase in fading of the $IRSL_{50}$ signal for the heated
494 samples (FSM-13LH and FSM-6LH) compared with their unheated counterparts (FSM-13 and FSM-6),
495 for the post-IR $IRSL_{225}$ signal there is no significant change in fading (Table 4). We conjecture that this
496 could indicate that prolonged heating increases the density of recombination centres proximal to the
497 electron trap (and this causes the increase in fading of the $IRSL_{50}$ signal and the increase in TL signal

498 intensity), but that the heating does not change our ability to isolate distant recombination centres to
499 select a signal with lower fading rate. The observations described above require further investigation
500 to be able to explain fully their implications.

501 Differences in fading rates are also observed for single-phase feldspars and perthites; the single-phase
502 feldspars (CLBR, Al-I, FSM-13) show little signal loss, whereas perthites fade to varying, but
503 significantly greater degrees. A first order model for the behaviour of a perthite is that its properties
504 are a linear combination of the properties of the two end-members, since it is a physical intergrowth
505 of the two end-member phases. However, this is clearly not the case. We can explain the anomalous
506 behaviour of the intergrowths in two ways: a) the Na- and K-feldspar components of the perthites
507 have greater disorder than the pure end-members or b) that the fading is related to the interfaces
508 between the Na- and K-feldspar components. Hypothesis (a) is credible – we show above that
509 increased disorder promotes fading. Furthermore, the lattice mismatch between Na- and K-feldspars
510 increases as they become more ordered and lattice strain resists ordering (Deer et al., 2013, p. 263-
511 264). Hence end member Na- and K-feldspar samples will have greater Al-Si order than a perthite
512 grown under the same conditions. However, hypothesis (b) is also credible – interfaces are atypical
513 regions which stabilise or destabilise defects and whose states of order may be locally different to the
514 bulk feldspar. Garcia-Guinea et al. (1999) suggested that a component of the blue luminescence is
515 related to twins. Interfaces may stabilise different defect types; two different defects that are unlikely
516 to occur together in a random distribution in the bulk may occur in close proximity on an interface.
517 This suggests that a single-phase solid solution may have different properties from a perthite of the
518 same bulk composition.

519 To explore the two hypotheses a) and b) above, we contrast the behaviour of macropertthite and
520 cryptopertthite. Cryptopertthites have nm-scale intergrowths and there is a greater volume of the
521 feldspar bulk occupied by interface (Brown and Parsons, 1984b). Also, since the elastic interfacial
522 strain in cryptopertthite is so high, one expects the Al-Si disorder to be greater in cryptopertthite

523 compared to a macroperthite of the same composition. However, we observe that macroperthite
524 show greater fading rates than cryptoperthite (Fig 8, Table 4). If the state of order (hypothesis a) were
525 the sole driving factor, the opposite would have been observed. Our conclusion is therefore that the
526 interfaces between the Na- and K-feldspar phases are implicated in the fading of perthite. In
527 cryptoperthite, this may be elastic strain around the interfaces at which the two different feldspar
528 lattices are forced to remain coherent (Brown et al. 1997). Since the fading rates of cryptoperthite and
529 macroperthite are different, we explore the differences in the morphologies of the interfaces.
530 Boundaries between the K- and Na- feldspar phases in cryptoperthite are highly strained but fully
531 coherent (i.e. the framework passes continuously from one phase to another, Brown et al., 1997). In
532 macroperthite, interfaces are semi-coherent, i.e. the lattices are commonly joined across the
533 boundary but not always, creating a region of dislocations and dangling bonds (e.g. Lee et al. 1995,
534 Lee and Parsons, 1997; Parsons et al., 2015). Since fading is understood as the loss of charge from an
535 electron trap to a nearby recombination centre, we hypothesise that a higher density of defects
536 concentrated around semi-coherent interfaces in macroperthite are implicated in the higher fading
537 rates. It may be that defect families such as regions of local disorder, dangling bonds, element
538 substitutions etc. are all stabilised by their proximity to semi-coherent perthite interfaces, driving
539 defects to cluster, which in turn allows direct defect-defect interactions. Interfaces are thus proposed
540 as regions in which high defect densities result in shorter distances between donor and acceptor of a
541 tunnelling electron (Jain et al., 2012).

542 Thus, we propose that fading in feldspars is not only a phenomenon of feldspars with a high-
543 temperature (disordered) structural state, such as volcanic feldspars (e.g. Wintle, 1973; Visocekas and
544 Zink, 1995; Visocekas et al., 2014; Guérin and Visocekas, 2015), but also related to perthite interfaces.
545 Fading is controlled by the density of defects (Huntley, 2006) and defect clustering, which in turn is
546 inferred to be influenced by (i) the degree of Si and Al ordering on the framework, (ii) the interfaces
547 in perthites and (iii) elastic strain areas within the feldspar crystal. The only feldspars in this study,
548 which show fading rates below 1 % are ordered single-phase albite and microcline. This observation

549 of very low fading rates of ordered feldspars is in good agreement with suggestions by Visocekas and
550 Zink (1995) and fading measurements on some microcline and albite specimen by Spooner (1994).

551 Our fading results show that for alkali feldspars, the structural state and the nature of interfaces within
552 the crystal are more important than chemical composition when interpreting luminescence. The
553 observation of increasing fading with increasing Al disorder on the framework is consistent with the
554 properties of plagioclase in which fading rises almost linearly with increasing concentration of Ca^{2+}
555 (Huntley and Lian 2006; Huntley et al. 2007) since Ca^{2+} substitution on the M-site is offset by increases
556 in the Al content of the lattice and therefore Al-O-Al populations are dependent on Ca content.

557 Due to exsolution of feldspar during cooling in plutonic rocks (e.g. granite), most alkali feldspars in
558 rocks and unconsolidated sediments are perthites (e.g. Deer et al., 2013). Unfortunately, perthites
559 faded in our experiments. The feldspars extracted from unconsolidated sediments (WHB-7 and HAM-
560 5) and from crushed bedrock material used in luminescence thermochronometry (MBT-I-2430) in this
561 study exhibit fading rates of the IRSL_{50} signal within the range seen for cryptoperthites in this study
562 (Table 4, Fig. 8B). A similarity in fading rates and in TL emission spectra of sediment and
563 thermochronometry samples and perthite is consistent with similar mechanisms playing a role in
564 these sets of samples. The similarities hint that in feldspar grains extracted from sediment and rocks
565 for optical dating and thermochronometry, Al-Si order, interface morphology and elastic strain are
566 implicated both in the blue emission and in the fading rate.

567 **6 Conclusions**

568 The present study analyses the effects of chemistry, the number of phases present within a single
569 crystal and the relative degree of framework disorder on TL emission spectra and fading rates of alkali
570 feldspars. The investigated sample suite includes Na- and K-feldspar end members, perthites with K-
571 and Na-feldspar lamellae on different scales, as well as material routinely used in luminescence dating
572 studies and luminescence thermochronometry. We observed variations in the intensity of the blue
573 luminescence emission and the rate of anomalous fading of the IRSL signal in our feldspars. We

574 associate the blue luminescence with the blue recombination centre being located on an Al-O-Al
575 bridge. The blue luminescence intensity increases as a function of the population of Al-O-Al bridges
576 which increases with Al-Si disorder. Ordered, single-phase feldspars, such as albite and microcline,
577 show very little fading, whereas the variations in fading rates in perthites are suggested to be linked
578 to a) the degree of Si and Al order on the framework and b) defect clustering associated with semi-
579 coherent perthite interfaces which modify the intensity of the blue emission and the extent to which
580 this emission fades.

581 Our study highlights the importance of detailed mineralogical characterisation of feldspar samples in
582 order to understand the complex drivers behind the variability seen in the blue luminescence intensity
583 and anomalous fading rates of the IRSL signal. This paper combines information on sample chemistry,
584 structural state, phases present, luminescence emissions and anomalous fading rate of alkali feldspar.
585 From these data we are able to differentiate between very low fading single-phase end members and
586 perthitic feldspars which all fade. Interestingly, the same differentiation can be made from TL emission
587 spectra, where single-phase feldspars show a more intense yellow-green emission, compared to the
588 blue emission, while perthites show the opposite, with the blue emission being dominant. Perhaps it
589 would be possible to identify those feldspar grains which are single phase (and hence likely to have a
590 low fading rate) by looking for those with a high value for the ratio of the yellow-green to blue
591 emission. Such measurements could be routinely feasible using automated filter changers in
592 combination with an EMCCD camera, and may avoid the need for costly and time consuming chemical
593 and structural analyses of individual grains.

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Table 1. Details regarding the origin, chemical composition, phases and size of perthite lamella for samples investigated. The chemical composition in % feldspars, was done using stoichiometric conversion of the semi-quantitative XRF data. Chemical composition and present phases of R1-11A was taken from Harrison et al. (1990). Present phases are based on semi-quantitative XRD analyses.

Sample ID	Origin	Chemical composition (FS %)			Phases						Extent of perthite development	Reference
		K-FS	Na-FS	Ca-FS	Microcline	Orthoclase	Sanidine	Albite	Anorthite	Other		
FSM-13	Brazil	98.5	1.5	0.0	100.0	-	-	-	-	-	none	-
FSM-13LH	Brazil	98.5	1.5	0.0	100.0	-	-	-	-	-	none	-
FSM-3	Granite pegmatite, Toe Head, South Harris, Scotland, UK (Cunningham, 1981)	82.5	17.2	0.3	78.0	-	-	22.0	-	-	cryptoperthites	-
R1-11A	Rapakivi Granite, South Greenland	81.0	19.0	1.0	Microcline and albite (Harrison et al., 1990)						cryptoperthite	Harrison et al., 1990; Finch and Klein, 1999; Riedesel et al., 2019
FSM-15	Buckingham, Quebec, Canada	80.4	19.6	0.0	82.0	-	-	18.0	-	-	cryptoperthite	-
FSM-14	Iveland, Southern Norway	78.3	21.2	0.5	54.0	-	-	46.0	-	-	cryptoperthite	-
FSM-8	Norway	77.1	22.6	0.3	68.0	-	-	32.0	-	-	cryptoperthite	-
FSM-7	Unknown	76.8	22.0	1.2	48.0	-	-	51.0	-	1.0	cryptoperthite	-
FSM-5	Unknown	74.8	25.20	0.0	57.0	-	-	43.0	-	-	Macroperthite	-
FSM-6	Granite pegmatite, Trezaise Quarry, Cornwall, UK (see Ussher et al., 1909)	74.4	25.3	0.3	-	38.0	-	62.0	-	-	cryptoperthite	-
FSM-6LH	Trezaise Quarry, Cornwall, UK	74.4	25.3	0.3	-	-	100.0	-	-	-	none	-
FSM-11	Perth, Canada	65.2	34.8	0.0	62.0	-	-	38.0	-	-	macroperthite	-
Al-I	Pinzele, Trente, Italy (Govindaraju, 1995)	1.0	97.0	2.0	-	-	-	-	-	-	none	-
CLBR	Pegmatite, Golonca District, Minas Gerais, Brazil (Cassadanne and Roditi, 1996)	0.5	99.3	0.2	-	-	-	100	-	-	none	Rendell and Clarke (1997); Garcia-Guinea et al., 1999; Riedesel et al., 2019

Table 2. Details on feldspar extracted from sediment and bedrock samples. The chemical composition in % feldspars, was done using stoichiometric conversion of the semi-quantitative XRF data. Present phases are based on semi-quantitative XRD analyses.

Sample ID	Origin	Chemical composition (FS %)			Microcline	Orthoclase	Phases			Other	Size of perthite lamella	Reference
		K-FS	Na-FS	Ca-FS			Sanidine	Albite	Anorthite			
MBT-I-2430	Calc-alkaline granite, Mont Blanc, Italy (Bussy et al., 1989; Bussy and Von Raumer, 1993)	86.1	13.4	0.6	85.0	-	-	15.0	-	-	-	Lambert, 2018; Riedesel et al., 2019
HAM-5	Lake Hamana, Japan	70.4	27.2	2.4	-	30.0	-	27.0	-	43.0	-	Riedesel et al. 2019
WHB-7	Channelled Scablands, Washington State, USA	63.9	31.5	4.6	37.0	-	-	30.0	-	33.0	-	-

Table 3. Post-IR₅₀IRSL₂₂₅ protocol used for the determination of fading rates.

Step	Details	Obtained
1	Beta dose ~90 Gy	
2	Preheat for 60 s at 250 °C	
3	IR stimulation at 50 °C for 200 s (IRSL ₅₀)	LX ₁ (IRSL ₅₀)
4	IR stimulation at 225 °C for 300 s (post-IR IRSL ₂₂₅)	LX ₂ (post-IR ₅₀ IRSL ₂₂₅)
5	Beta dose ~30 Gy	
6	Preheat for 60 s at 250 °C	
7	IR stimulation at 50 °C for 200 s (IRSL ₅₀)	TX ₁ (IRSL ₅₀)
8	IR stimulation at 225 °C for 300 s (post-IR IRSL ₂₂₅)	TX ₂ (post-IR ₅₀ IRSL ₂₂₅)

Table 4. Fading rates for the two luminescence signals recorded using a post-IR₅₀ IRSL₂₂₅ protocol. The samples were preheated to 250 °C for 60 s, prior to IRSL measurements.

Group	Sample ID	Fading rate (g2days, %/decade)	
		IRSL ₅₀	Post-IR IRSL ₂₂₅
Single phase			
	Al-I	0.58 ± 0.30	0.29 ± 0.24
	CLBR	0.36 ± 0.60	-0.08 ± 0.37
	FSM-13	0.93 ± 1.52	0.54 ± 1.10
Macroperthites			
	FSM-5	8.99 ± 1.82	2.74 ± 0.60
	FSM-11	12.42 ± 1.71	4.07 ± 1.39
Cryptoperthites			
	FSM-3	5.71 ± 0.12	2.19 ± 0.49
	R1-11A	6.72 ± 2.51	1.86 ± 1.39
	FSM-15	4.97 ± 1.98	0.79 ± 1.73
	FSM-14	3.51 ± 1.35	1.48 ± 1.63
	FSM-8	1.80 ± 0.32	1.23 ± 1.86
	FSM-7	4.13 ± 0.46	0.55 ± 0.38
	FSM-6	2.75 ± 1.47	1.47 ± 0.97
Artificially disordered			
	FSM-13LH	3.24 ± 0.16	-0.06 ± 0.13
	FSM-6LH	4.99 ± 0.20	1.68 ± 0.13
Sediments/rocks			
	HAM-5	1.86 ± 0.46	-1.01 ± 0.20
	WHB-7	3.07 ± 0.74	1.01 ± 0.26
	MBT-I-2430	3.28 ± 0.14	1.35 ± 0.30

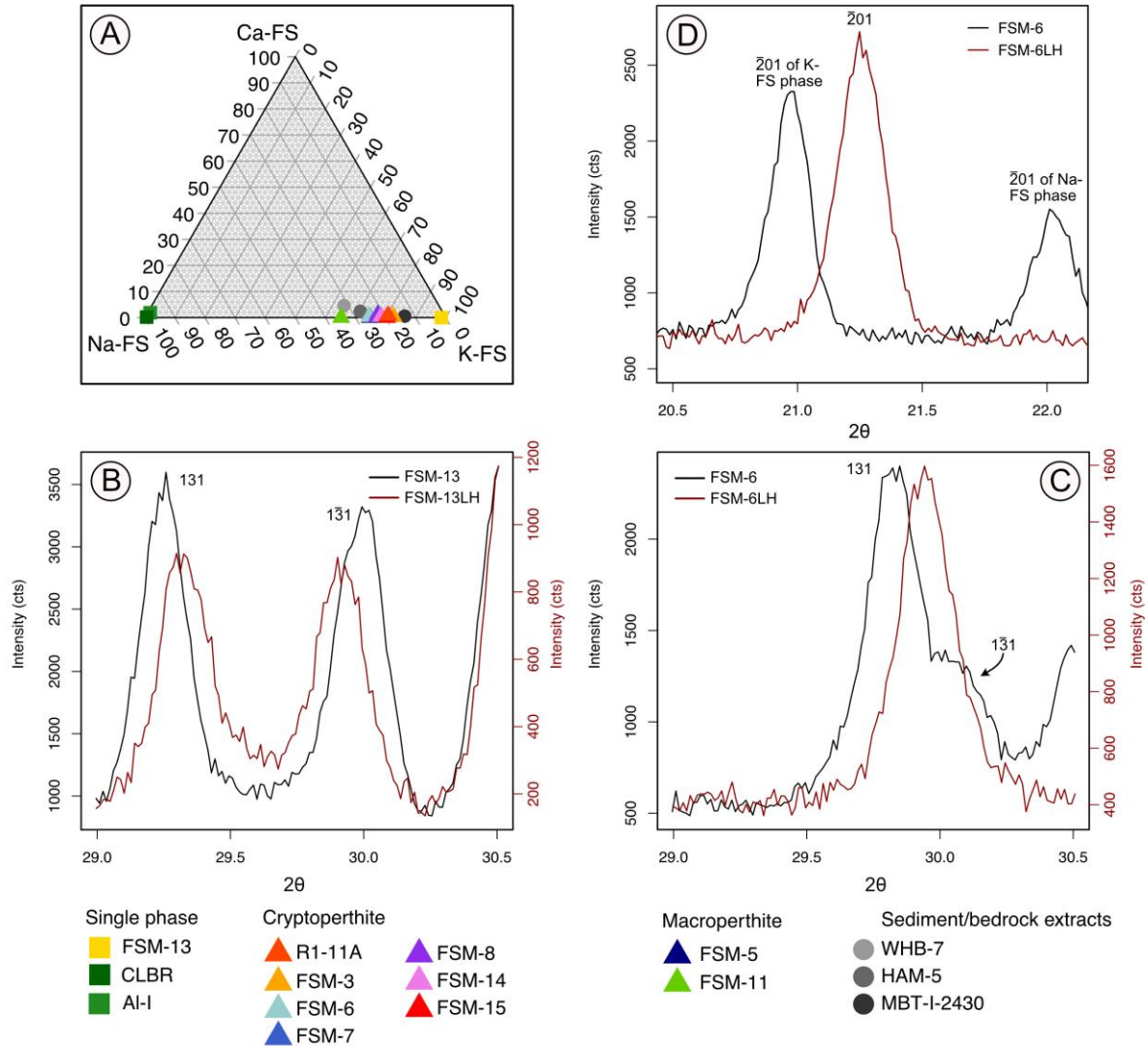


Fig. 1. A) Relative feldspar composition (%) of the samples. The relative compositions of K-, Na- and Ca-feldspar were calculated using stoichiometric conversion of XRF results. B) X-ray diffraction pattern of FSM-13 (black line) and FSM-13LH (red line) in the range of 29.0° 2θ to 30.5° 2θ . Visible are the (131) and $(1\bar{3}1)$ diffraction maxima. These two diffraction maxima move closer together due to the heating of the sample material. C) X-ray diffraction pattern of FSM-6 (black line) and FSM-6LH (red line) in the range of 29.0° 2θ to 30.5° 2θ . For FSM-6 (black line) a prominent (131) diffraction maxima is visible in the figure, and in addition a minor peak, which is the remainder of the $(1\bar{3}1)$ diffraction peak. After heating and rapidly cooling, only a single (131) diffraction peak remains in FSM-6LH (red line). This indicates that the original material of FSM-13 (black line in B) is more ordered than the starting material of FSM-6 (black line in C). For both samples, the X-ray diffraction pattern indicate the success of the heating experiment, as both samples are slightly more disordered than their starting material. However, it is expected that FSM-6LH is relatively more disordered than FSM-13LH. D) Comparison of the $\bar{2}01$ diffraction peak in perthite FSM-6 and in sanidine FSM-6LH. The heating experiment caused a homogenisation of the distribution of K^+ and Na^+ ions within the crystal. Whilst FSM-6 shows two $\bar{2}01$ diffraction peaks (one for each phase), FSM-6LH only shows a single diffraction peak, reflecting the homogeneous distribution of the cations. Radiation used in XRD analysis was $Cu\ K\alpha$.

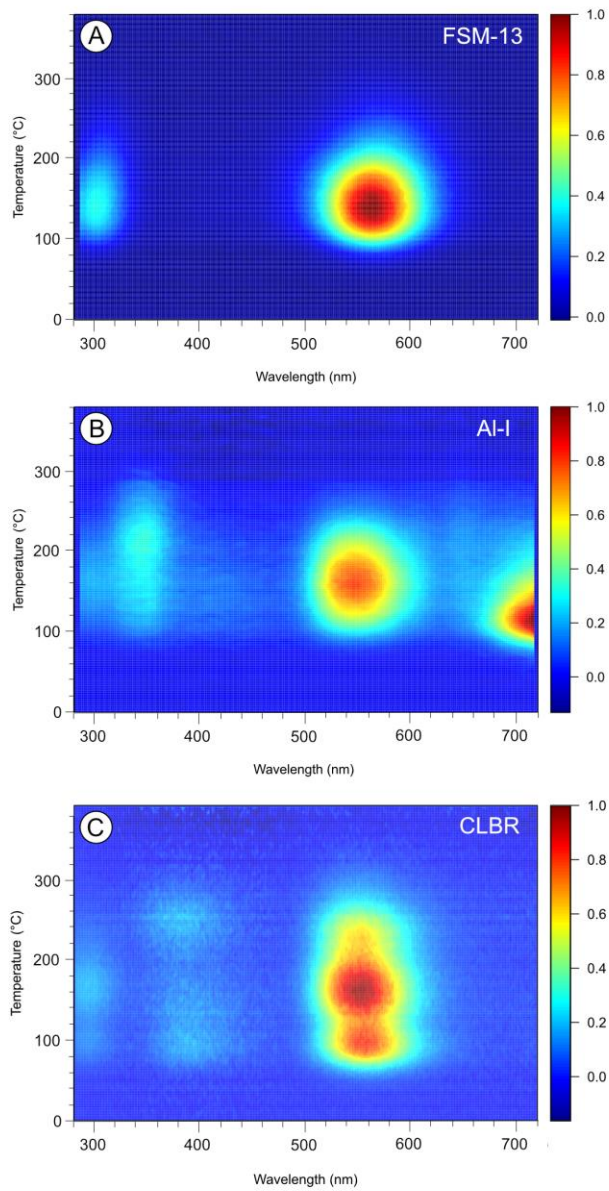


Fig. 2. TL emission spectra recorded up to 380 °C, from 280 to 720 nm. The samples shown in this figure are single-phase feldspar. FSM-13 is a microcline (A) and Al-I (B) and CLBR (C) are albite, with Al-I containing 5 % quartz. The TL intensities are normalised to the data point with the highest intensity.

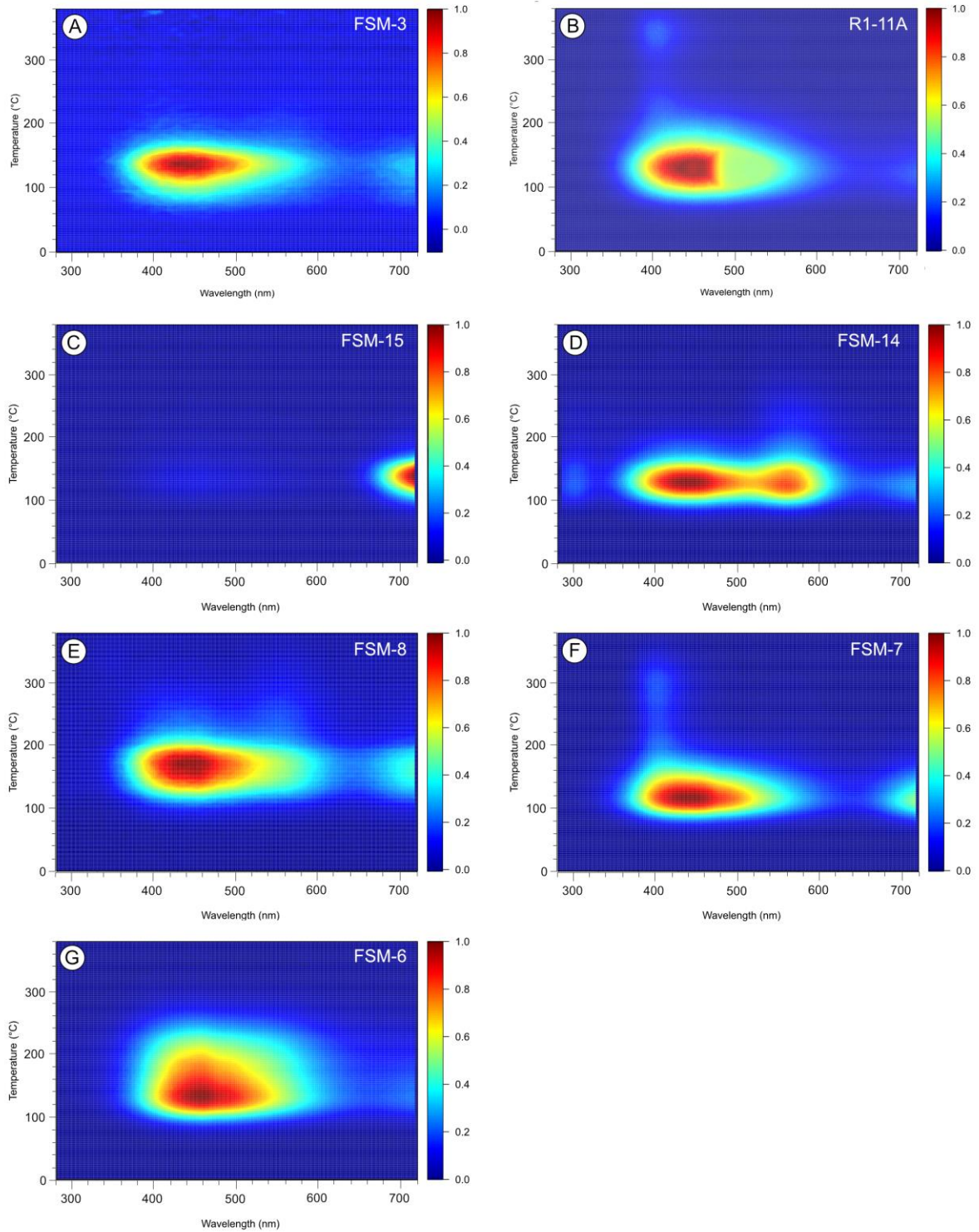


Fig. 3. TL emission spectra recorded up to 380 °C, from 280 to 720 nm. The samples shown in this figure are cryptoperthite: FSM-15 (C) is the only sample of the displayed samples, where Fe was detected during the XRF measurements. All samples consist of microcline and albite (A-F), except for FSM-6 (G), which consists of orthoclase and albite. The TL intensities are normalised to the data point with the highest intensity.

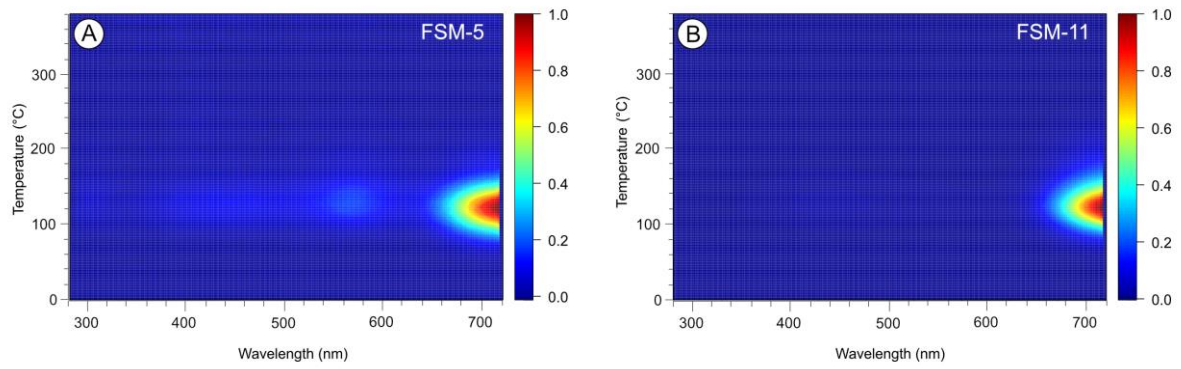


Fig. 4. TL emission spectra of perthites with coarse perthite lamella, visible with the naked eye: macroperthite. FSM-5 (A) is an amazonite sample, which shows twinning in the albite phase. XRF-measurements showed the presence of Fe in FSM-11 (B), which also is of orange colour in the microcline phase. The TL intensities are normalised to the data point with the highest intensity.

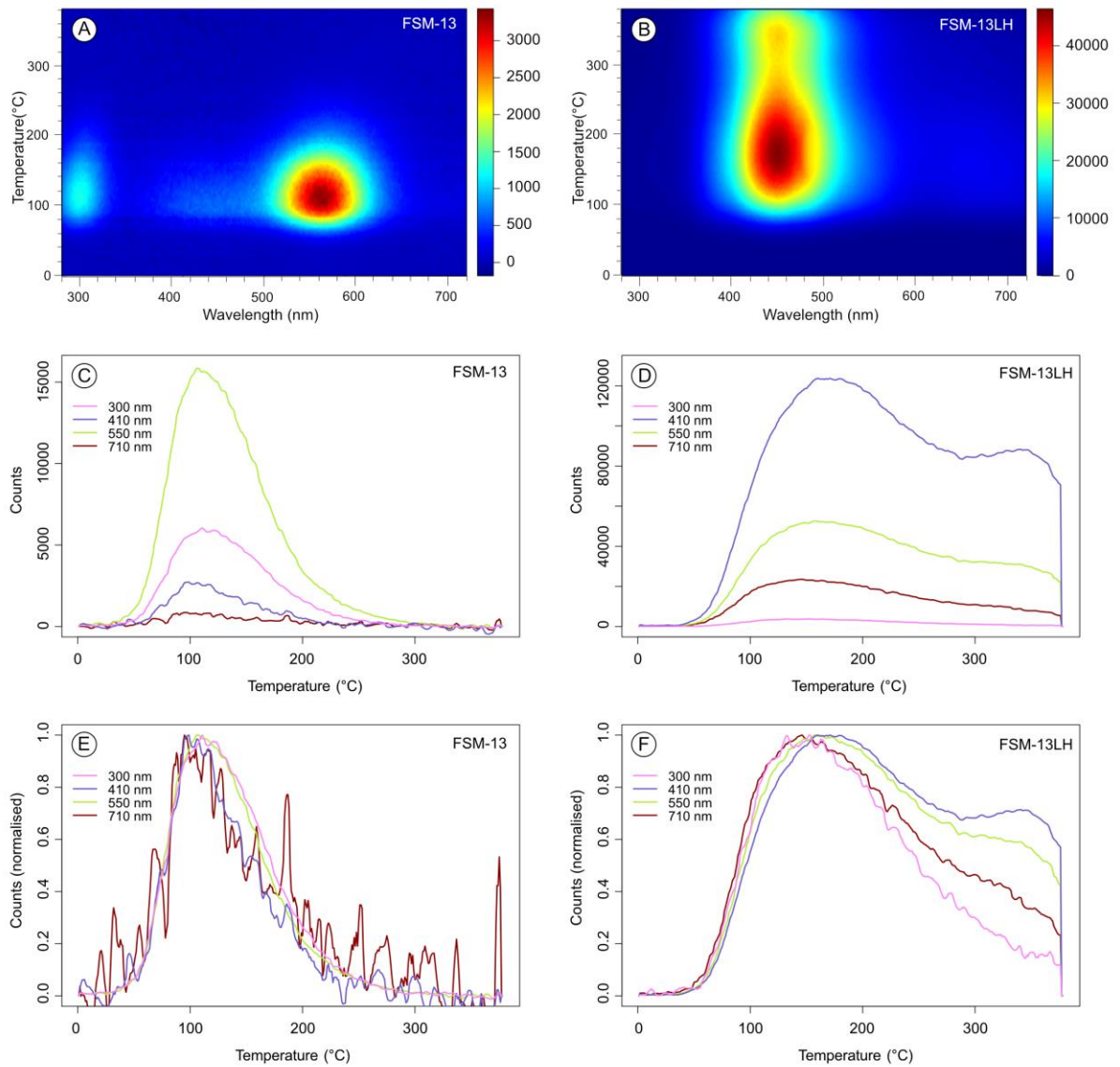


Fig. 5. Effect of heating sample FSM-13 (single phase microcline) at 1050 °C for 10 days. (A) TL spectra of the unheated material of FSM-13, (B) TL spectra of the heated fraction. (C) and (D) show the TL curve extracted from the TL spectra for different wavelengths for FSM-13 and FSM-13LH, respectively. (E) and (F) show the same data as (C) and (D), but normalised to the highest intensity for each TL curve. The measured intensities are absolute intensities in counts/0.4 nm/°C. For these measurements the same mass of sample material was used and the samples were measured during the same measurement sequence.

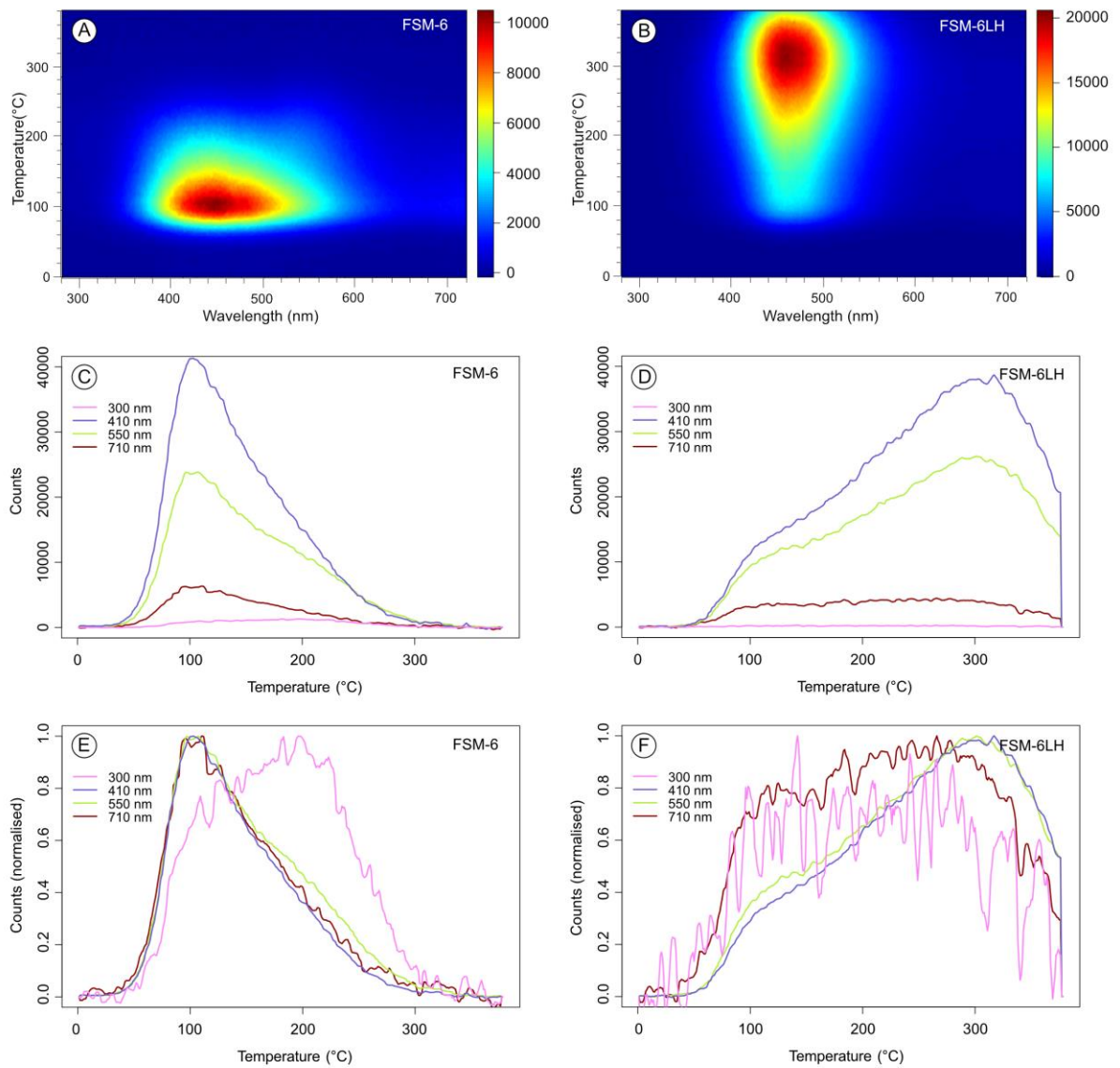


Fig. 6. Effect of heating sample FSM-6 (orthoclase and albite) at 1050 °C for 10 days. (A) TL spectra of the unheated material of FSM-6, (B) TL spectra of the heated fraction. (C) and (D) show the TL curve extracted from the TL spectra for different wavelengths for FSM-6 and FSM-6LH, respectively. (E) and (F) show the same data as (C) and (D), but normalised to the highest intensity for each TL curve. The measured intensities are absolute intensities (in $\text{cts}/0.4 \text{ nm}/^\circ\text{C}$). For these measurements the same mass of sample material was used and the samples were measured during the same measurement sequence.

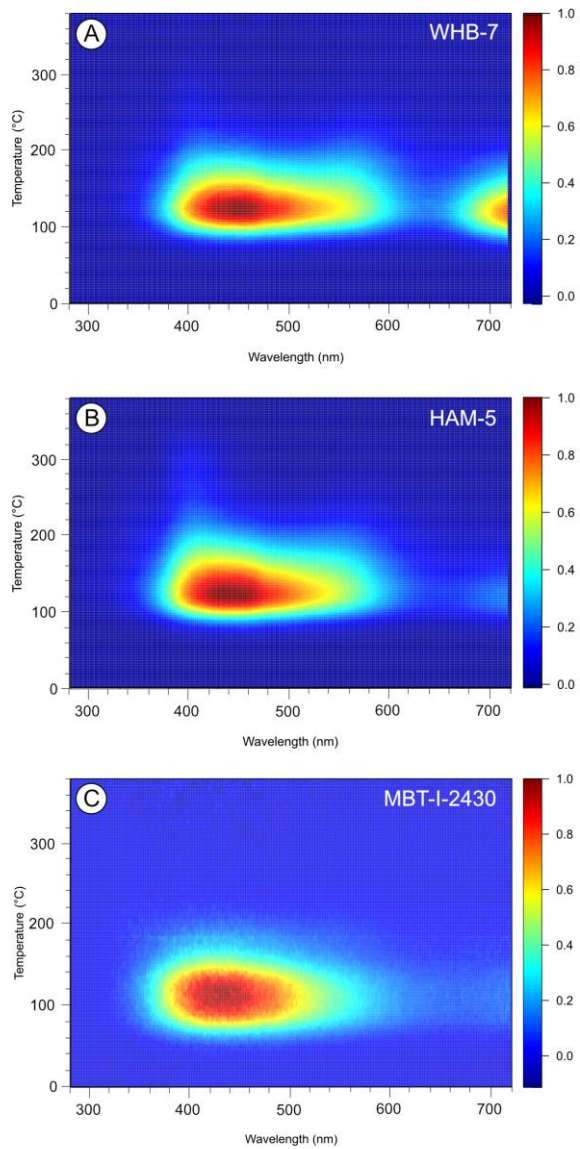


Fig. 7. TL emission spectra of alkali feldspars extracted from sediments (WHB-7, HAM-5) and bedrock (MBT-I-2430). The TL intensities are normalised to the data point with the highest intensity.

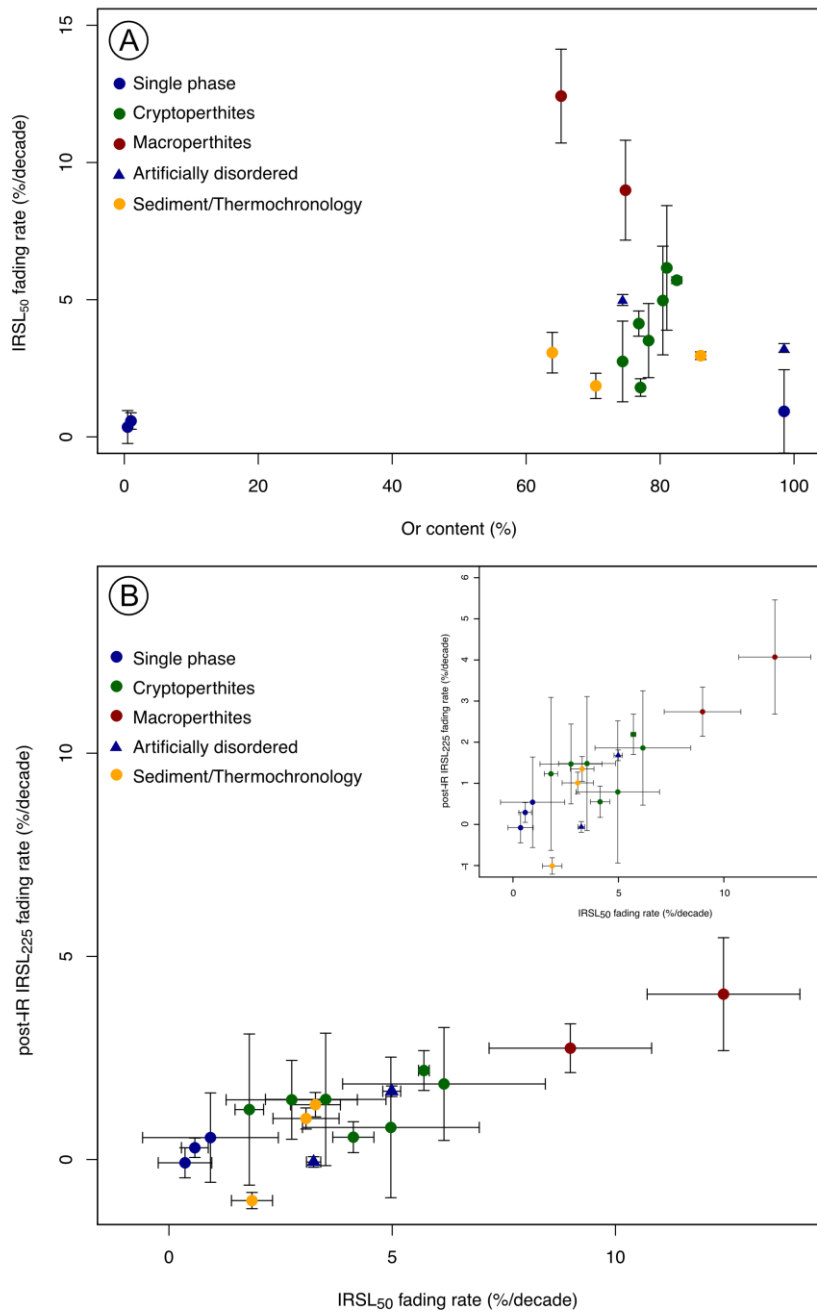


Fig. 8. A) Comparison of fading rates of the post-IR IRSL₂₂₅ signal and the IRSL₅₀ signal, obtained using the post-IRIRSL protocol. The inset is a close-up version of B. The data points in A and B are the average fading rate of the measured aliquots for each sample and the standard deviation of these measurements. B) Fading rates for the IRSL₅₀ signal measured using a post-IR₅₀IRSL₂₂₅ protocol, with a 250 °C preheat for 60 s.

Exploring sources of variation in thermoluminescence emissions and anomalous fading in alkali feldspars

Supplementary Material

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Figures

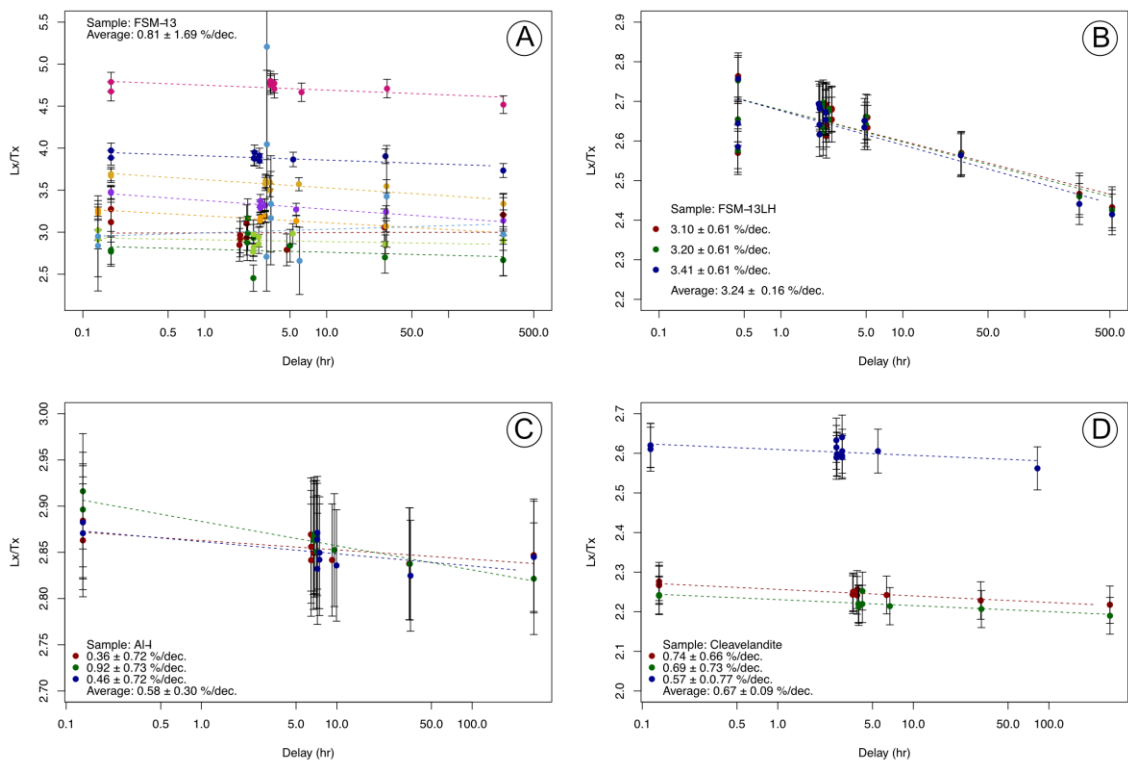


Fig. S1. Fading results as L_x/T_x versus delay time for FSM-13 (A), FSM-13LH (B), Al-I (C) and Cleavelandite (D).

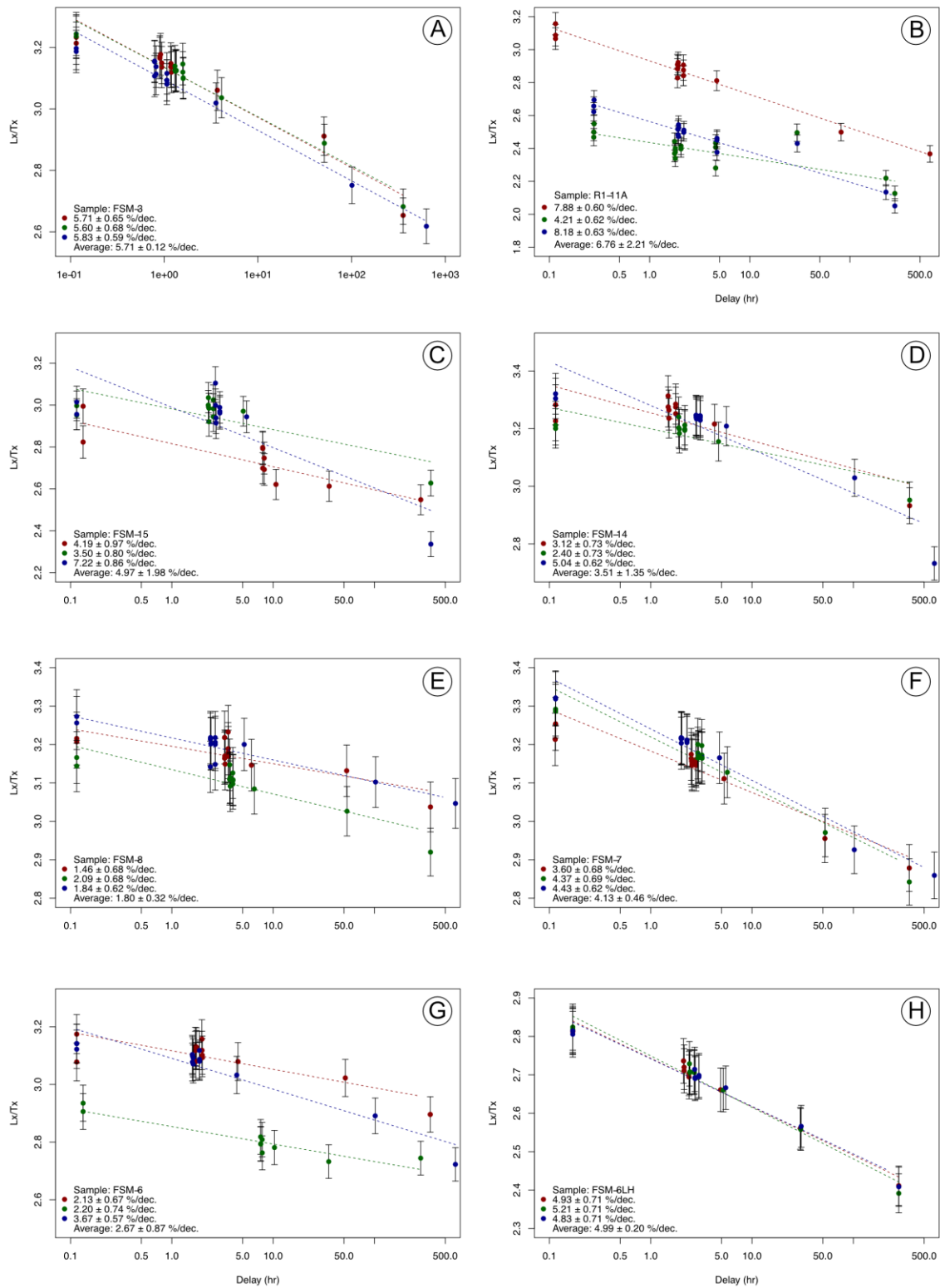


Fig. S2. Fading results as L_x/T_x versus delay time for FSM-3 (A), R1-11A (B), FSM-15 (C) FSM-14 (D), FSM-8 (E), FSM-7 (F), FSM-6 (G) and FSM-6LH (H).

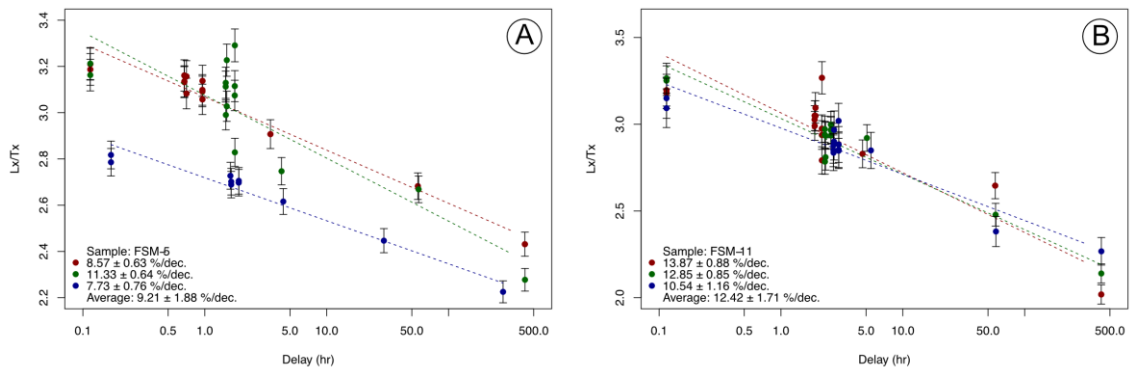


Fig. S3. Fading results as L_x/T_x versus delay time (hours) for FSM-5 (A) and FSM-11 (B).

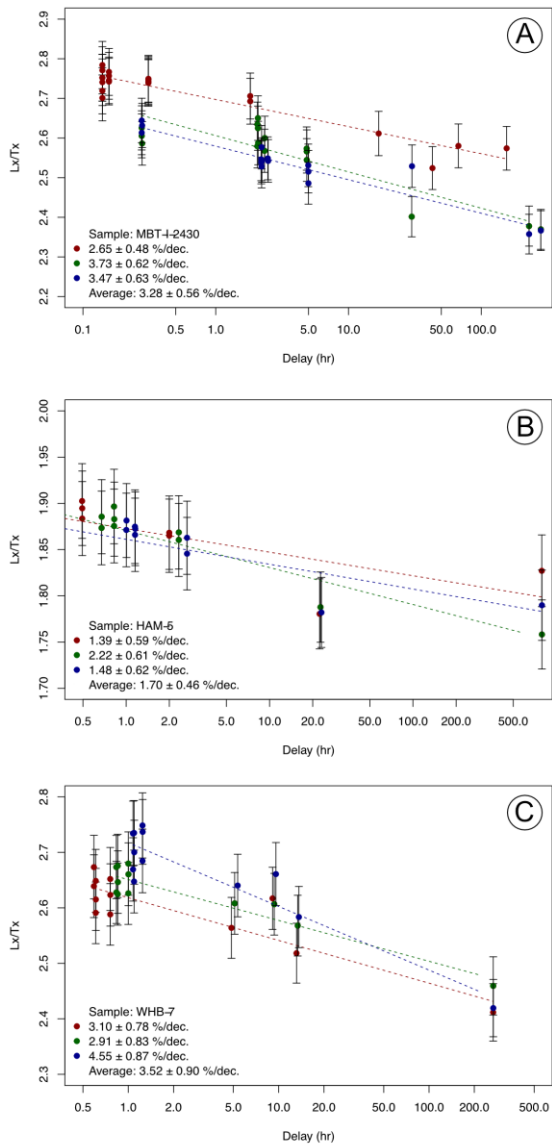


Fig. S4. Fading results as Lx/Tx versus delay time (in hours) for MBT-I-2430 (A), HAM-5 (B) and WHB-7 (C).

Tables

Table S1. Semi-quantitative XRF results (wt%) of samples used in this study, except R1-11A and Cleavelandite.

Oxide	HAM-5	241_WHB-7kf	MBT-I-2430	255_FSM-3	255_FSM-5	255_FSM-6	255_FSM-7	255_FSM-8	255_FSM-11	255_FSM-13	255_FSM-14	255_FSM-15
Al ₂ O ₃	33.80 ± 0.50	20.20 ± 0.60	22.10 ± 0.40	23.50 ± 1.00	21.30 ± 0.50	23.20 ± 1.00	21.20 ± 0.50	25.60 ± 2.00	20.00 ± 0.40	22.00 ± 0.30	21.80 ± 1.00	21.80 ± 1.00
SiO ₂	46.00 ± 0.40	65.90 ± 3.00	63.20 ± 2.00	61.40 ± 2.00	62.50 ± 2.00	62.70 ± 0.40	62.80 ± 2.00	55.60 ± 0.60	64.30 ± 2.00	62.00 ± 2.00	61.90 ± 2.00	63.10 ± 2.00
K ₂ O	14.40 ± 0.10	8.38 ± 0.10	13.00 ± 0.10	13.20 ± 0.10	13.10 ± 0.10	11.10 ± 0.10	12.70 ± 0.10	15.20 ± 0.10	11.20 ± 0.10	15.70 ± 0.10	12.00 ± 0.10	12.60 ± 0.10
Na ₂ O	3.66 ± 0.20	2.72 ± 0.30	1.33 ± 0.10	1.81 ± 0.10	2.90 ± 0.20	2.48 ± 0.10	2.39 ± 0.20	2.93 ± 0.20	3.93 ± 0.20	0.16 ± 0.03	2.14 ± 0.10	2.02 ± 0.10
CaO	0.59 ± 0.03	0.72 ± 0.04	0.10 ± 0.01	0.06 ± 0.01	0.00 ± 0.00	0.06 ± 0.01	0.24 ± 0.02	0.08 ± 0.01	0.00 ± 0.00	0.00 ± 0.00	0.09 ± 0.01	0.00 ± 0.00
Fe ₂ O ₃	0.40 ± 0.02	1.23 ± 0.03	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.46 ± 0.02	0.00 ± 0.0	0.00 ± 0.00	0.17 ± 0.01
TiO ₂	0.14 ± 0.01	0.02 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
MgO	0.47 ± 0.04	0.59 ± 0.07	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.0	0.00 ± 0.00	0.00 ± 0.00
Other	0.45 ± 0.02	0.29 ± 0.09	0.21 ± 0.01	0.07 ± 0.00	0.12 ± 0.01	0.40 ± 0.02	0.61 ± 0.02	0.62 ± 0.13	0.19 ± 0.01	0.18 ± 0.00	2.15 ± 0.12	0.26 ± 0.01