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Quartz radiofluorescence: a modelling approach

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ABSTRACT

Modelling (natural) quartz luminescence (TL/OSL) phenomena appears to be quite common nowadays. The corresponding simulations are capable of giving valuable insights into the charge transport system. By contrast, simulating radiofluorescence (RF) of quartz has rather been neglected in the past. Here we present and discuss (1) the RF signals of natural quartz measured in the UV band and (2) simulations of these experiments executed using a three-energy-level model to explain the experimentally obtained results.

Two natural quartz samples were investigated at room temperature (RT) following different preheat procedures: (a) consecutively increasing preheat temperatures from 50 °C to 700 °C and (b) repeating a 500 °C preheat with subsequent UV-RF measurement at RT for eleven times. Based on the measurement and modelling results, we finally confirm theoretically the dependency of the UV-RF signal of quartz on the burial dose.

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1. Introduction

Numerical simulations pave the way for a better understanding of luminescence phenomena, such as thermally stimulated and optically stimulated luminescence (TL, OSL) of various dosimeters (e.g., [1–8]). By contrast, simulating radioluminescence / radiofluorescence (henceforth radiofluorescence: RF) of natural quartz appears to have been neglected in the past.

RF is the luminescence emitted during exposure to ionizing radiation and for quartz believed to result from direct recombination of electrons with holes captured in recombination centres [9, cf. for a review]. While quartz RF spectra are reported in the literature (e.g., [7,10–13]) simulation studies for a specific emission wavelength are missing so far. One recent study on simulating RF was published by Pagonis et al. [14], but it is limited to Al₂O₃:C.

While the comprehensive quartz model developed by Bailey [15] is capable of successfully simulating common TL and OSL luminescence phenomena (such as dose response, dose quenching, phototransfer, thermal activation) for the UV band, it fails for simulating experimentally obtained quartz RF signals.

The results obtained by Bailey [15, Section 3.4.4.] suggest that the shape of the simulated RF is correlated to the population of the

so called reservoir centres. In more recent publications a link between the pre-dose effect [16] and the RF behaviour is mentioned [13,17]. The successful simulation of pre-dose effects on TL signals was published by, e.g., Adamiec [18], Pagonis and Carty [19], Itoh et al. [20] but not the effect of different preheat treatments on the RF signal.

This study is separated into two parts. The first part presents experimental results obtained by measuring quartz RF in the UV band (UV-RF) for different preheat temperatures as well as repeated cycles of heating and subsequent UV-RF measurement for a preheat temperature of 500 °C for two natural quartz samples. In the second part the empirical results are complemented by numerical simulations, i.e., three parameters from the original model [15] are adapted and modified to reproduce the signal dynamics seen in the experiments. To allow an understanding of the charge transport during heating and UV-RF, a simplified one-trap-two-centres model was developed.

Our numerical simulations demonstrate the potential of quartz UV-RF as a method of retrospective dosimetry, which so far has been almost neglected. While the study by Marazuev et al. [21] appears to successfully demonstrate its general applicability, an elaborated explanation to understand the physical background of the obtained results is still missing.

To the best of our knowledge, the RF signal dynamics in the UV and the burial dose estimation for natural quartz samples using RF

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signals have not been simulated and presented in the literature before.

2. Material and methods

2.1. Quartz samples

Two natural quartz samples were chosen for the measurements: (1) the quartz fraction of sample BT586 was extracted from a colluvial sample originating from the Trebgast valley in the north-west of Bayreuth (Germany) [22]. For this sample a paleodose of ~ 24 Gy was obtained. (2) a second quartz sample (BT1195) was extracted from the quartz ridge 'Pfahl' (Bavarian Forest, Germany), which is one of the largest hydrothermal quartz veins in Germany. This sample was extracted under daylight conditions and subsequently gently crushed with a steel mortar with frequent sieving in between. Subsequent chemical treatments followed routine preparation procedures for luminescence dating samples (e.g. [23]). These are: HCl (30 %), H₂O₂ (30 %), density separation using sodium-polytungstate, HF (40 % for 60 min). In contrast to BT586 the sample BT1195 was bleached in a home made solar simulator (2 h with an Osram Duluxstar lamp). For both samples the used grain size fraction is 90–200 μm . Two different pretreatments (natural and bleached) were used to investigate differences in the RF behaviour concerning these pretreatments.

2.2. Measurement conditions

RF measurements were carried out on a Freiberg Instruments *lexsyg research* reader [24] at the luminescence laboratory in Bayreuth. The reader is equipped with a $^{90}\text{Sr}/^{90}\text{Y}$ β -source (~ 3.6 Gy min^{-1}), calibrated for coarse grain quartz on stainless steel cups. The β -source is specifically designed for RF measurements [25]. Luminescence was detected through a Chroma BP 365/50 EX interference filter in front of a Hamamatsu H7360-02 photomultiplier tube allowing for a detection of the UV-RF signal between 315 nm and 415 nm. All measurements were performed in a nitrogen atmosphere. If not reported otherwise, preheating of the samples was performed with a constant heating rate of 5 K s^{-1} . The channel time for the RF measurements was set to 1 s. The experimental data presented in this study are the arithmetic mean of two aliquots for each measurement. Reproducibility of RF signals using different aliquots was better than 5%.

Further details on the UV-RF experiments are given in the text below.

2.3. Data analysis

Data analyses were carried out using the statistical programming environment **R** [26] and the **R** package 'Luminescence' [27,28]. For simulating the UV-RF signals the **R** package 'RLum-Model' [29,30] was used. The code for the simulations presented here can be found in the supplementary material. Simulation results were cross-checked with *Mathematica*TM and *MATLAB*TM.

3. Quartz UV-RF measurements

3.1. Preheat experiments

Martini et al. [13,31] reported that samples annealed at temperatures between 400 °C and 600 °C are showing an enhancement in the UV-RF intensity. To determine and better understand

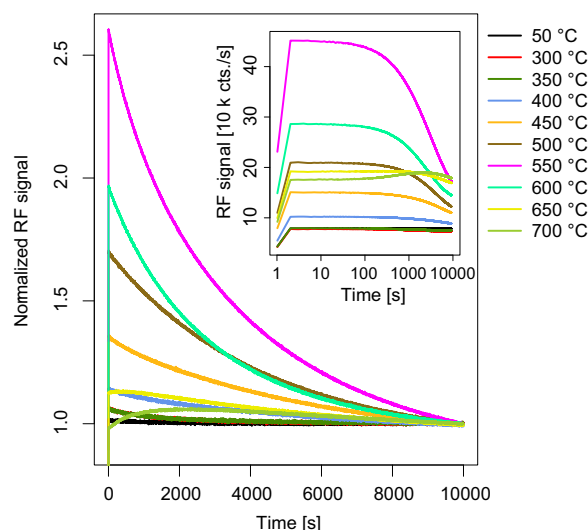


Fig. 1. UV-RF signal for sample BT586 for different preheat temperatures (hold for 120 s) prior to the RF measurements. For each temperature a new aliquot was used. The values are normalized to the last data point of each measurement and the total absorbed dose is 600 Gy during each RF measurement. For the sake of clarity the UV-RF curves for preheat temperatures from 100 °C to 250 °C were removed, because no change was observed. The inset shows the same data as the main figure but with absolute values and a logarithmic x-axis.

the correlation between preheat temperature and UV-RF signal intensity, UV-RF measurements were carried out for 10,000 s at room temperature (~ 20 °C) after preheating the samples to temperatures ranging from 50 °C to 700 °C using increments of 50 °C. The total absorbed dose after 10,000 s was ~ 600 Gy.

We expected a successive increase of the initial RF signal, triggered by the pre-dose effect, as described in Zimmerman [16] and Marazuev et al. [21]. A study by Krbetschek and Trautmann [32] showed that high temperature annealing of quartz up to 700 °C can lead to a UV-RF signal characterized by an exponential increase followed by a linear decrease. This behaviour was not observed in any of the studies by Martini et al. [31], although they used even higher temperatures (than reported by Krbetschek and Trautmann [32], up to 1100 °C). In these studies no exponential increase at the beginning of the measurement was observed, just a decrease of the UV-RF signal directly after starting the measurement.

Fig. 1 shows the UV-RF signals for sample BT586 after different preheat temperatures normalized to the final data point. For preheat temperatures from 50 °C to 350 °C no substantial differences within the signal shapes are visible and for the sake of clarity only the UV-RF curve for 50 °C is shown. The changes in these temperature interval are limited to a small decrease of the UV-RF signal in the first seconds followed by a stable signal until the end of the measurements.

In the range from 400 °C to 550 °C an increase by a factor of ~ 1.2 (400 °C) to ~ 2.6 (550 °C) of the initial UV-RF signal was observed. From 600 °C to 700 °C the signal dynamics decreased by a factor of 2 (600 °C) down to 1 (700 °C).

For the RF signal at 700 °C the maximum signal intensity is not observed at the very beginning of the measurements, but the signal builds up in the first channels (up to 3,000 s) and then decreases. A similar behaviour was described by Krbetschek and Trautmann [32] for a quartz sample, after annealing it for 3 hours at 700 °C followed by γ -irradiation.

The inset in Fig. 1 shows all measured data, but on a logarithmic x-axis and not normalized. The strong increase in the first channel is caused by the opening of the shutter of the β -source. This takes up to ~ 0.5 s and thus, the first channel comprises less

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