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Thermally assisted OSL application for equivalent dose estimation; comparison of multiple equivalent dose values as well as saturation levels determined by luminescence and ESR techniques for a sedimentary sample collected from a fault gouge

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ABSTRACT

Equivalent dose estimation (D_e) constitutes the most important part of either trap-charge dating techniques or dosimetry applications. In the present work, multiple, independent equivalent dose estimation approaches were adopted, using both luminescence and ESR techniques; two different minerals were studied, namely quartz as well as feldspathic polymineral samples. The work is divided into three independent parts, depending on the type of signal employed. Firstly, different D_e estimation approaches were carried out on both polymineral and contaminated quartz, using single aliquot regenerative dose protocols employing conventional OSL and IRSL signals, acquired at different temperatures. Secondly, ESR equivalent dose estimations using the additive dose procedure both at room temperature and at 90 K were discussed. Lastly, for the first time in the literature, a single aliquot regenerative protocol employing a thermally assisted OSL signal originating from Very Deep Traps was applied for natural minerals. Rejection criteria such as recycling and recovery ratios are also presented. The SAR protocol, whenever applied, provided with compatible D_e estimations with great accuracy, independent on either the type of mineral or the stimulation temperature. Low temperature ESR signals resulting from Al and Ti centers indicate very large D_e values due to bleaching in-ability, associated with large uncertainty values. Additionally, dose saturation of different approaches was investigated. For the signal arising from Very Deep Traps in quartz saturation is extended almost by one order of magnitude. It is interesting that most of D_e values yielded using different luminescence signals agree with each other and ESR Ge center has very large D_0 values. The results presented above highly support the argument that the stability and the initial ESR signal of the Ge center is highly sample-dependent, without any instability problems for the cases of quartz resulting from fault gouge.

1. Introduction

Optically stimulated luminescence (OSL) as well as Electron Spin Resonance (ESR) have been both well recognized as important techniques for retrospective dosimetry and archaeological/geological dating of terrestrial materials. At the same time, both techniques have been reported as very effective tools towards understanding the trap-charge mechanism in the crystal structures [1–4].

Among the several minerals that were investigated within the aspect of luminescence features and dating implications, quartz and feldspar constitute the major, most dominant as well as important part of these researches. During recent years, quartz has become the main mineral used in dating of sediments by both techniques. Nevertheless, recently major effort has been devoted in using both feldspars and samples of mixed mineralogy as well for equivalent dose (D_e) estimation while applying luminescence. In order to obtain the natural dose received by grains, provided that they were last exposed to light or heat, it is necessary to compare the luminescence signal derived from electrons in traps that are sufficiently thermally stable with that of artificially applied known doses delivered by a laboratory source. In this respect, recent progress in laboratory methodologies and paleoenvironmental research has resulted in numerous advanced techniques and protocols to date mostly quartz as well as polymineral

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grains, each one incorporating a number of strict tests towards reliable estimations.

There is a high need, especially in the geochronology community, for further improving the luminescence age limits for covering at least the last few million years, in which the evolution of the humans has been taken place. Towards this direction, a large effort of luminescence research has been devoted on extending the upper age limits of luminescence dating applications. A luminescence age estimate equals paleodose (or equivalent dose D_e, derived from luminescence measurements) divided by dose rate (D, from elemental and/or isotopic data). Accurate luminescence dating of old sediments as well as extension of the current age limits require two main conditions: (1) luminescence signals with high stability and (2) high saturation dose levels. From a theoretical point of view, stable signals arising from traps with very long lifetimes could provide one of the main pre-requirements towards the extension of the luminescence age limits. These traps are usually characterized as either deep or Very Deep Traps, depending on their de-localization temperature. One such signal is the peak in thermoluminescence (TL) measurements at 325 °C (heating rate 5 °C/s) that was monitored in many sedimentary quartz samples [5,6]. Measurement of the kinetics of the electron trap giving rise to this signal led to estimates that the 325 °C TL peak has a lifetime in excess of 20 Myr at 20 °C [5]. Nevertheless, most importantly for dating of sediments, the 325 °C TL peak is bleached very rapidly by exposure to daylight [7,8]. Consequently, conventional OSL signals were reported to have their origin at the 325 °C TL peak in quartz [9], even though it has subsequently become clear that the OSL signal from quartz commonly consists of charge from more than one defect [10]. The lifetime of conventional OSL signal has been reported to be exceeding 1Myr [11,12].

Nevertheless, the main drawback with both aforementioned signals arises from their growth characteristics. Growth of either the fast component or the 325 °C TL may be fitted for large attributed doses by a single saturating exponential equation of the form:

$$I = I_{max}(1 - \exp(D/D_0)),$$

where D is the dose used to obtain a signal I, D_0 characterises the growth towards saturation and I_{max} is the value of the signal for infinitely large D, i.e. at the saturation level. The value of the parameter D_0 of course is very important. For the case of OSL signal from quartz, this parameter, besides specific rare exceptions, do not exceed the value of 100 Gy [13], and according to Wintle and Murray [14] trustworthy ages could be yielded setting an upper threshold for the equivalent dose such as twice the value of D_0, since as this point is approached, the degree of curvature at such high doses means that the slope of the dose response curve is very low, incorporating thus large errors. A number of studies have been performed aiming towards other luminescence signals from quartz with larger D_0 values, such as the one by Singarayer et al. [15], showing the potential of one of the slow components with a D_0 value of several thousands of Gy, as well as the one by Jain et al. [16] using an isothermal TL signal (ITL) at 310 °C with a D_0 value of almost 500 Gy.

An alternative approach was reported by Lowick et al. [17]. These authors pointed out that, despite the general expectation that OSL growth should be described by a simple saturating exponential function, an additional high dose component is often reported in the dose response of the OSL signal of quartz. Although often reported as linear [18], it appears that this response is the early expression of a second saturating exponential [19,20]. While some studies using equivalent doses that fall in this high dose region have produced ages that correlate well with independent dating, others report that it results in unreliable age determinations. Nevertheless, the process that gives rise to this additional component still requires investigation [4,21].

In 2006, Wang et al. [22,23] described measurements of a thermally transferred optically stimulated luminescence (TT-OSL) signal from quartz which continued to grow at doses of many thousands of Gy, long after the fast component OSL signal had become saturated. Out of the different methods investigated, this TT-OSL signal exhibits growth to the highest dose and has the potential to extend the age range of luminescence dating by at least an order of magnitude [13,24,25]. Besides TT-OSL, a great deal of work has been focused on extending the age limits of luminescence dating, including post infrared-infrared stimulated luminescence (post IR IRSL) in feldspars [26,27] as well as stimulation of quartz by using violet light [28,29]. Nevertheless, still work is further required.

The conventional OSL techniques and applications take exclusively advantage of the electron trapping levels that can be thermally excited at temperatures well less than 500 °C. As Very Deep Traps (VDT) are considered the traps which give rise to TL glow peaks having their peak maximum temperature, T_{max} beyond 500 °C [30]. Due to a number of reasons including thermal quenching, interference of high IR background, large thermal depth in conjunction with low photo-ionization cross section and instrumental limitations imposed for the commercially available luminescent readers, the signal from such deep traps is very difficult to measure. An alternative experimental method was suggested, termed as thermally assisted OSL (TA-OSL), in order to not only measure the signal of the deep traps according to dose response beyond the 500 °C, but also use this signal for estimation of the equivalent dose in the SAR protocol implication of luminescence dating. This method, consisting of photo transfer OSL measurements in the continuous wave configuration performed at elevated temperatures using the blue LEDs housed at commercial Risø systems, comprises combined action of thermal and optical stimulation. The presence of VDT has been experimentally verified in many cases of luminescent materials via intense TA-OSL, such as $CaF_2:N$ [31], $Al_2O_3:C$ [30,32–35] sedimentary quartz [36,37] and apatites [38,39] as well as to some materials consisting the ground layers of wood and canvas paintings, such as yellow ochre, $BaSO_4$, gypsum and chalk [40]. Very recently, intense TA-OSL signal was monitored for the case of potassium feldspars and polymineral samples [41]. TA-OSL signals from quartz, $CaF_2:N$ and $Al_2O_3:C$ crystals exhibit a number of interesting properties which could be effectively used towards dosimetry purposes, especially for large accumulated doses [10,30,32,34]. Among these properties, the most notable are the straightforward relation, even though not linear, observed between either the initial or the integrated TA-OSL intensity and the dose, along with the simple TA-OSL curve shape. Therefore, the role of the VDT in dating very old samples could be very important as well as significant. Moreover, the TA-OSL is expected to be stable, since the origin of the signal is the traps which give rise to TL glow peaks at higher peak maximum temperature, beyond 500 °C. Recent works indicated that the TA-OSL signal in materials exhibiting anomalous fading is much more stable compared to both TL as well as OSL signals [38,39]. Despite the fact that recently Polymeris et al. [37] have provided with a number of arguments towards establishing a robust protocol using this TA-OSL, applications of D_e estimations using the TA-OSL signal have not yet been reported in the literature.

In the present study, different approaches for equivalent dose estimation have been independently applied both to quartz as well as polymineral grains received from the same sample towards a comparative study. A number of related protocols have been incorporated, including conventional luminescence measurements such as OSL and IRSL measured at various temperatures, as well as TA-OSL. All the aforementioned protocols and stimuli have been used because they have been extensively applied for the case of moderate and large age assessment; in each one of these approaches a