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Variations in Luminescence Sensitivity of Various Quartz Samples from Southwestern Nigeria

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ABSTRACT

This research was undertaken to investigate the sensitivity of Thermoluminescence (TL), Optically Stimulated Luminescence (OSL) and Infra-Red Stimulation Luminescence (IRSL) of twelve (12) quartz samples from different locations in the Southwestern part of Nigeria. This was done with a motive to survey the provenance suitability of each one of them for luminescence dating. All the luminescence measurements on the 12 quartz samples were carried out using a Risø TL/OSL read (model TL/OSL-DA-15). Each of the samples displayed different range of luminescence responses to the same test dose. The sensitivity of the TL and OSL did not follow identical pattern for all the samples but that of LM-OSL and CW-OSL demonstrated nearly identical pattern. Effect of feldspar inclusion seemed passive in TL but highly active in OSL most especially fast component. The two samples that exhibited highest OSL signal failed to pass feldspar inclusion test. The significance of feldspar inclusion in SAR protocols was stressed. Further studies on the samples at varying test doses may improve the understanding of the behaviour of these samples.

Keywords: Quartz, Thermoluminescence (TL), Continuous wave optically stimulated luminescence (CW-OSL), linearly modulated optically stimulated luminescence (LM-OSL), Infra-Red Stimulated Luminescence (IRSL), Sensitivity, Feldspar inclusion, Nigeria

1. Introduction

Luminescence techniques that are employed in retrospective dosimetry involve stimulation of light from some insulators (called phosphors) that had been exposed to ionizing radiation earlier [1]. When the stimulation is achieved by application of heat at a linear heating rate, (Ks^{-1}), resulting in the temperature varying as $T = T_0 + \beta t$, where T_0 is temperature at time t = 0 (K), the technique is termed thermoluminescence (TL). The technique is known as optically stimulated luminescence (OSL) when it is accomplished by optical stimulation [2]. Under this method, the irradiated phosphor is exposed to light (UV, visible or infrared) under a constant temperature and the OSL emission is recorded as function of stimulation time. The integral of the OSL emitted during the stimulation period is a measure of the dose of irradiation absorbed by the sample since it was last exposed to light. Unlike in the case of TL in which heat is applied only at a constant heating rate, there are three popular modes of stimulation in OSL. The first one is continuous-wave OSL (CW-OSL) in which the stimulation light intensity is kept constant and the OSL signal is monitored continuously throughout the stimulation period [3]. In linear-modulation OSL (LM-OSL), the stimulation intensity is ramped (increased) linearly while the OSL is measured [4]. In the case of the last and the least popular among them, pulsed-OSL (POSL), the stimulation source is pulsed and the OSL is monitored only between pulses. This method has been developed into time-resolved OSL (TR-OSL) which provides information about luminescent centres [5-8].

Generally, the quantity of acquired dose due to the earlier exposure to radiation by the phosphors is estimated by measuring the amount of light emitted during the stimulation [7]. This is based on the assumptions that the acquired dose is proportional to the quantity of the emitted light. Therefore, the measured

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acquired/equivalent dose is then utilized in retrospective dosimetry either in evaluation of dates in archaeological and geological dating or reconstruction of accident dose in accidental dosimetry. The most widely used phosphor in retrospective dosimetry is quartz [7]. An important reason for this is its abundance in nature. This mineral is one of the more abundant rock-forming continental crust minerals [9]. It is readily found in archaeological artifacts and geological materials like rocks and sediments [7]. Furthermore, quartz samples are always present in building bricks or blocks in which accident dose assessment could be based in the case of accident dosimetry.

Luminescence sensitivity of a given sample is defined as the amount of luminescence emitted per unit sample mass in response to a fixed laboratory dose [10]. Ideally, this quantity is expected to be constant for quartz but it is not in practice. Quartz of diverse kinds and origins possess different luminescence sensitivity. Better still, change in sensitivity stands as the most widely encountered problem in luminescence dating applications [11]. This attribute among other features of quartz that vary from 'samples to samples' is ascribed to different crystallization environments during formation of quartz samples [7,12].

The quantity of interest in TL/OSL 'retrospective dosimetry' or dating method is the absorbed or equivalent dose, ED (in Gy). In dating, the age of the material is evaluated by measuring the ED that the materials received from radiation owing to the natural background, since they were last heated (as in TL) or exposed to sunlight (as in OSL). Thereby, the age is calculated by dividing the ED by the dose rate (in Gy/s) of the natural background. In accident dosimetry, the goal is to reconstruct ED as a consequence of a radiation accident.

Quartz based luminescence dating that has gained global attention is still unpopular in majority African countries; Nigeria inclusive. The aim of this study was to investigate the sensitivity of TL, OSL and IRSL of twelve (12) quartz samples from different locations in the Southwestern part of Nigeria. This was done since it is important to survey the suitability of each one of them for luminescence dating and serve as the scientific baseline for the luminescence sensitivity provenance study for the study area. This is important since low luminescence sensitivity is a malign feature that makes the precise luminescence ages to be dated difficult.

2. Materials and Methods

Twelve different quartz samples collected from south west Nigeria were given laboratory names S1, S2, S3, S4, S5, S7, S8, S9, S10, S11 and S12 respectively as shown in the Table 1 and Fig. 1. Fortunately, some of the quartz samples are recovered as bulk materials, without being of sedimentary (wind transported) origin.

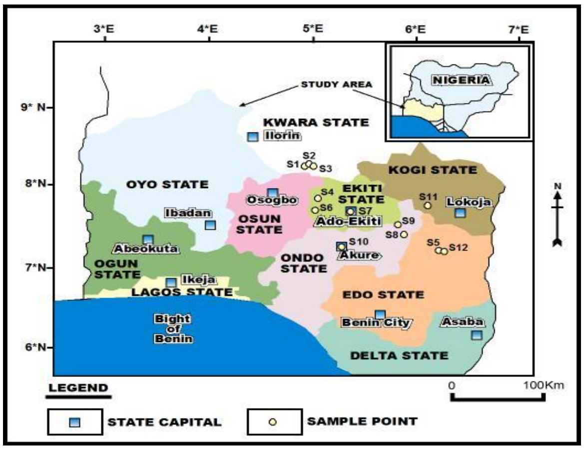

Fig. 1 *Map of Southwestern Nigeria, indicating the sampling locations*

The modus operandi behind sample selections in this study was to have quartz grains that possessed the same radiation, optical and thermal histories for each sample used. That was devised to rule out the possibility of grain to grain different sensitizations. Thus, instead of using sedimentary quartz samples, the original quartz samples were crystals of hydrothermal and metamorphic which occurred in veins associated with metamorphic rocks. S1, S2, S3, S4 (clear rock crystal type) and S6 (rose-pink type) are hydrothermal while S5, S7, S9, S8, S10, S11 S12 (milky quartz species), are of metamorphic associated with granites.
After collection, the samples were kept in the light proof containers while all handling and pre-treatment was performed in din red light conditions. These samples, which could be considered as poorly bleached quartz species, were then crushed and pulverized at the department of Geology, Obafemi Awolowo University, Ile-Ife, Nigeria.

Table 1. Quartz sampling locations and laboratory code names

Samples	*Sample Locations*
S1	Okerimi Oro
S2	Ijomu Oro
S3	Olorunsogo Oro
S4	Ijero Ekiti
S5	Ojirami Igara
S6	Aramoko Ekiti
S7	Ado Ekiti
S8	Ugbe Akoko
S9	Okeagbe Akoko
S10	Akure
S11	Kabba
S12	Igwe Marble Deposit

Thereabout the pulverized samples were packaged into labeled airtight plastic dispensing sachets for analysis. Aforementioned different types of luminescence measurements were performed on each of the twelve samples. Each of the aliquots (sub-samples) was of equal mass of about 5mg. The TL, OSL and IRSL measurements on the 12 quartz samples were carried out using a Risø TL/OSL reader (model TL/OSL–DA–15) equipped with a 0.075 Gy/s $^{90}Sr/^{90}Y$ β ray source [13] at Archaeometry Laboratory, Cultural and Educational Technology Institute (C.E.T.I.), R.C. "ATHENA", Tsimiski 58, 67100 – Xanthi, Greece. The reader was fitted with a 9635QA photomultiplier tube. The detection optics consisted of a combination of a Pilkington HA-3 heat absorbing and a Corning 7–59 (280 – 500nm with pick transmission around 380nm) for the case of TL measurements while a Hoya U-340 (320–440 nm) blue filter was used for all OSL and the IRSL measurements. A heating rate of 1 ◦Cs^{-1} was used in all TL readouts in order to avoid significant temperature lag, up to a maximum temperature of 500 ◦C. The test dose applied was 7.5 Gy in all cases.

3. Results and Discussion

The TL sensitivity of 110 °C TL peak for all the samples resulting from step 1 of the experimental procedures of Table 2 is presented in Fig. 2.

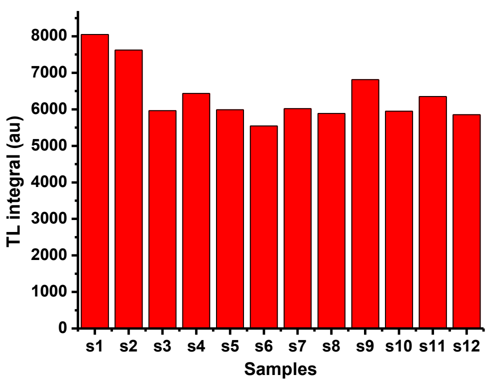

Fig.2 *Despite the lack of artificial irradiation, the 110 °C TL peak is still monitored. The response of this latter peak is presented for each quartz sample*

It could be observed that the 110°C TL peak signals for all the samples are very low. Still, despite the lack of any artificial irradiation, it is still monitored in extremely low intensity. This low response could be attributed to two reasons. The first reason is because these responses are as a result of the cumulative natural dose that each sample has received.

Table 2. TL measurement protocol

Step	Description
1	TL readout up to 500 °C was carried out on a sample
2	A test dose was given to the same sample
3	TL readout up to 500 °C was carried out on the same sample
4	Steps 1 to 3 were repeated for another sample

The stability of this peak is well known to be in the order of two hours in ambient temperature before it fades out [2]. Therefore, since there was no administration of laboratory dose prior to the TL measurement, the low response observed is justified. Secondly, the low response is also expected according to the pre-dose model since the samples have not been thermally sensitized prior to this step 1 (Table 2) [7].

The contributions of the two factors itemized above are conspicuous in the sensitization of Fig. 3 which is the output of the response of step 3 of the protocol in Table 2. The glow curves of sample S2 representing all the samples are presented in Fig. 4 for a better illustration. Curves (a) and (b) in this figure are the outputs of TL of Steps 1 and 3 respectively in Table 2.

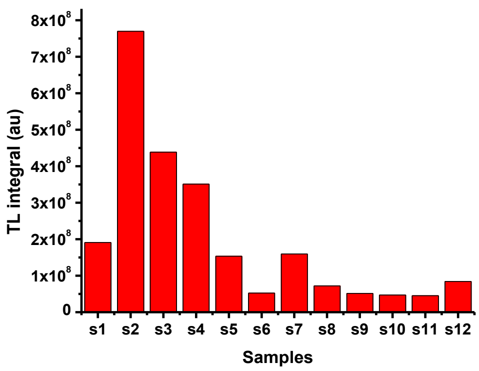

Fig.3 *Response of the 110 °C TL peak of all the samples to the test dose*

The sensitivity of 110°C TL peak (i.e response between 30 to 127°C) for Step 3 has increased in thousands folds as compared with that of step 1. The test dose of step 2 of the protocol is an indication that the TL was read immediately after the dose which rules out the possibility of fading. On the second factor, the TL readout of step 1 of Table 2 serves as a thermal treatment for sensitizing the samples. This singular treatment is enough to produce enormous signal as 110°C TL peak is known to be highly sensitized when heated beyond 200°C [14].

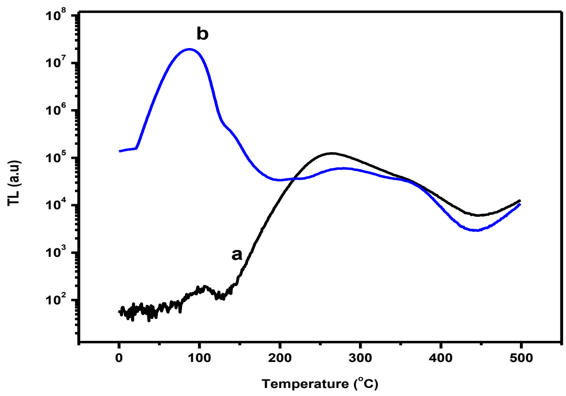

Fig. 4 *TL glow curves measured for the sample with code S2. Curve (a) corresponds to the sensitivity of Step 1 without previous thermal activation. Curve (b) corresponds to the glow-curve of Step 3 after the thermal activation*

An important observation in the sensitivity of this peak in question is the different levels of sensitization that each of the samples exhibited. This is despite the fact that the equal level of test dose was administered to all of them. Although, there is no much difference in the sensitivities of all the samples in Fig. 2 but each samples displayed different range of responses to the same test dose in the case of Fig. 3. Sample S2 followed by S3 and S4 are classified to be the highly sensitized while S1, S7 and S5 are in the intermediate group. The remaining samples are categorized to be poorly sensitized.

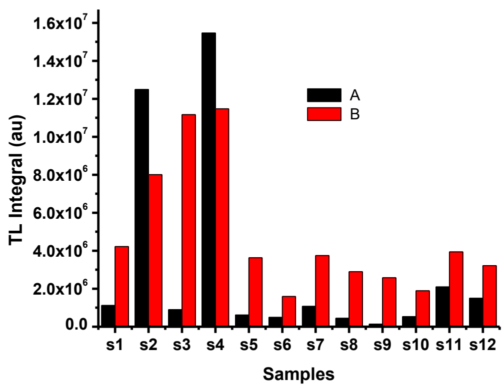

Fig. 5 *Response of the high temperature TL peak of all the samples to natural dose (A) and test dose (B)*

The sensitivity of the 110°C TL peak is very important in dating and luminescence protocols. This is because it is the key instrument used in monitoring sensitivity changes of higher TL peaks and OSL signal of quartz as used in Additive-Dose (AD) and Single-Aliquot Regenerative-dose (SAR) methods of dating [1, 15-16]. Furthermore, this technique has been effectively utilized in authenticity testing of artefacts and firing temperature measurements of pottery materials [17-20].

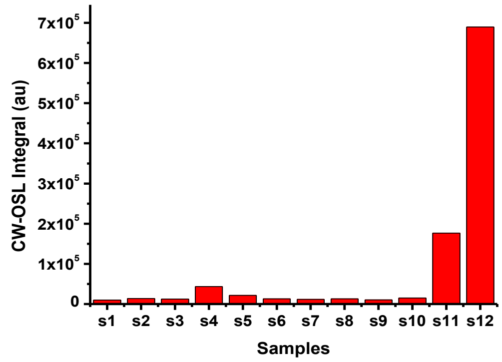

Fig. 6 *Fast component natural signal CW-OSL of all the samples*

The sensitivity of the high temperature TL peak (i.e response between 200 to 410°C) which is known to be stable and used for dating or dosimetry purposes is presented in Fig. 5. Unlike the case of 110°C TL peak, the sensitivities of step 1 are not of the same degree. Sample S4 takes the lead of sensitivity over S2 that was mostly sensitized for the case of 110°C TL peak. In fact, only these two samples, S4 and S2, recorded high TL signal in Step 1 and, at the same time, exhibited sensitivity of Step 1 signal than responses of Step 3. The sensitivity resulting from Step 3 (Fig. 5) followed nearly identical pattern with that of Step S1 with samples S2, S3 and S4 still leading. It must be stressed that the response from Step 3 that is observed to be more than that of Step 1 for all the samples except samples S2 and S4 is unexpected. This is on the basis of the stability of the high temperature TL peak and the low test dose (7.5 Gy) administered that is considered to be far lesser that the accumulated ED by each sample. However, this characteristic is an indication of the degree of unbleachable component of the high temperature peak of

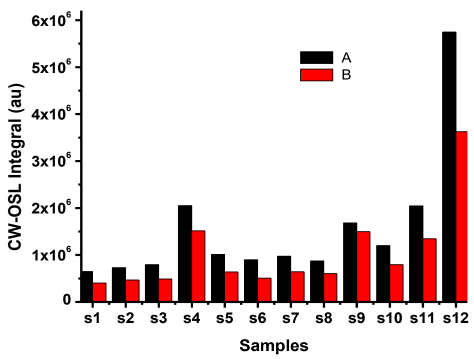

Fig. 7 *Response of the CW-OSL all the samples to residual dose + test dose (A) and test dose (B)*

these two samples comparably to the remaining samples [21].

The signals recorded in Step 1 of protocol presented in Table 3 for CW-OSL is depicted in Fig. 6. This is the typical response of the fast component of CW-OSL (signal recorded in the first 5 sec of measurement). As observed the responses for all the samples are very low except for the case of S11 and S12. This is expected since the samples are believed to have been exposed to light before sampling and the fast components are bleachable in few seconds of exposure to light. The signal recorded here for Samples S11 and S12 have been confirmed to not have emanated from quartz as it is going be soon revealed.

Table 3. CW-OSL Measurement protocol

Step	Description
1	CW-OSL at 125°C for 100s was carried out on a sample
2	A test dose was given to the same sample
3	CW-OSL at 125°C for 500s was carried out on the same sample
4	A test dose was given to the same sample
5	CW-OSL at 125°C for 500s was carried out on the same sample
6	A test dose was given to the same sample
7	LM-OSL at 125°C for 1000s was carried out on the same sample
8	Steps 1 to 7 were repeated for another sample

The signal resulting from Step 3 Table 3 for the case of the same CW-OSL is shown in Fig. 7. While Sample S12 outstandingly takes the lead in sensitivity over all the samples, followed by this are samples S4, S9 and S11 that compete among themselves. Much difference is not exhibited among the sensitivity of the remaining samples. It is important to note that responses of Step 3 are always higher than that of step 5 unlike the case of 110°C TL peak. Interestingly, the sensitivity of LM-OSL of Fig. 8 resulted from Step 7 of Table 4 follow the same pattern with that of CW-OSL of step 5.

Table 4. IRSL Measurement protocol

Step	Description
1	A test dose was given to a same sample
2	IRSL measurement was carried out on the same sample
3	Steps 1 and 2 were repeated for another sample

Inclusions of substantial quantity of feldspar in quartz material to be used for luminescence dating or study could be always problematic. This is because feldspar is relatively more sensitive than quartz [22]. Therefore, its considerable inclusion in quartz will definitely undermine the overall result of the experiment. Usually, this is checked by exposing aliquots to infrared (IR) at ambient temperatures, in order to check if there is an infrared stimulated luminescence (IRSL) signal.

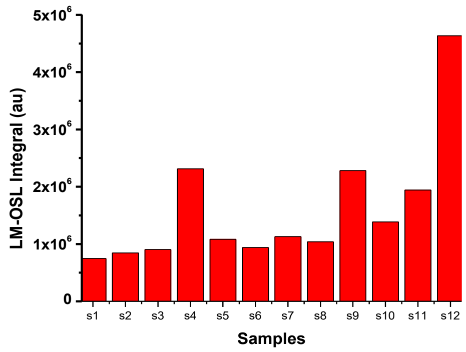

Fig. 8 *Response of the LM-OSL of all the samples to test dose (B)*

This is possible because feldspar is highly sensitive to IR stimulation to which quartz is insensitive. Thus, IRSL enables discrimination for the presence of feldspar. Alternatively, or in addition, a light microscope is normally used to estimate level of infections [23-24]. The feldspar infection test was carried out on all the quartz samples by following the IRSL method according to the protocol of Table 4. The result of this is shown in Fig. 9. It is evident from this that only samples S5, S11 and S12 are likely to be infected with feldspar while others proved not being contaminated. This may be the reason why Samples S12 and S11 still exhibited reasonable signal in Fig. 6 where very low signal is expected.

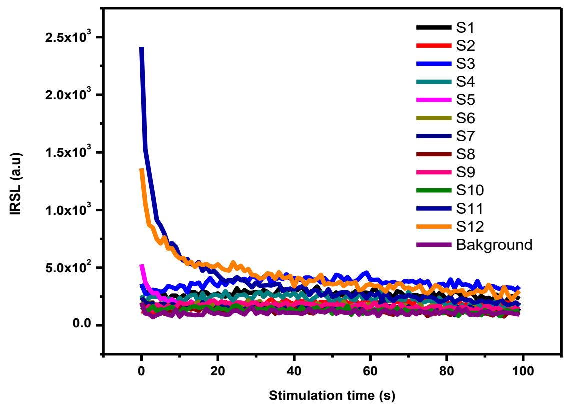

Fig. 9 *IRSL curves of all the samples*

Conversely, the sensitivity of these two samples (S11 and S12) in the case of TL, despite the fact that they are likely being infected with feldspar, are relatively low as compared with the remaining samples as confirmed in Figs. 3 and 5. Whereas, the contribution of the feldspar inclusion is conspicuous in the cases of CW-OSL and

LM-OSL of these two samples (S11 and S12) in Figs. 7 and 8, even more dominant in the case of their fast components. Feldspar inclusion may not be ordinarily expected in the three samples that proved infected in this study since they were not sedimentary origins. However, the probability of feldspar infusion associated different weathering processes of the samples cannot be totally ruled out since they are of metamorphic associated with granites origins. It is obvious from the present result that further investigation and treatments on these affected samples in necessary before their further applications. This observation reveals the extent to which feldspar infection could cause overestimation in luminescence dating. The is because SAR protocol of dating that is most popular among luminescence dating techniques [7], is majorly predicated on fast component that proved to be highly influenced by the feldspar infection. The importance of feldspar inclusion test must not be overlooked in SAR protocols.

4. Conclusions

1. The luminescence sensitivity properties of various quartz samples were investigated displaying different range of responses to the same test dose.
2. The sensitivity of the 110 ^oC TL peaks showed three of the samples (S2, S3, and S4) yield high sensitivities compared to the others after test dose was administered.
3. The sensitivity of LM-OSL and CW-OSL demonstrated nearly identical patterns.
4. The two samples (S11 and S12) that were mostly sensitized in the cases of OSL proved positive to feldspar inclusion test.
5. The significance of feldspar inclusion in SAR protocols was stressed.
6. Further studies on the samples at varying test doses may improve the general understanding of the behaviour of these samples.

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Conflict of Interest

The authors have no conflict of interest.

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