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Characterisation of clay samples from minerals–rich deposits for thermoluminescence applications

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Abstract Thermoluminescent (TL) characteristics of clay samples from Ijero-Ekiti and Isan-Ekiti, Ekiti State, Nigeria were studied for application in retrospective dosimetry. In order to increase the potentials of the clay material for relevant radiation measurement applications, characteristic glow curves of samples from the clay deposits have been obtained using a Victoreen (2800M) TL reader and the TL properties subsequently examined. The variable heating rate method was used to determine the glow peak shape and intensity by estimating the activation energy or trap depth, E and the frequency factor, S. Most samples demonstrated the dependence of their glow curve on the heating rate, with the peak temperature, T_m increasing with the heating rate. These glow peaks were noted to be a complex trapping system, consisting of several overlapping first order peaks and each with different TL behaviour, except for certain samples, which showed independence of peak temperature with dose. While recommending further heating protocol to isolate each peak for the calculation of other kinetic parameters, we present an approximate 0.29–0.84 eV for E and 3.7×10^1 to $4.0 \times 10^6\ s^{-1}$ for S. Though the samples with the TL dosimetric property of increased peak intensity with increasing excitation dose could be considered for TL applications, the reduction in TL intensity must be considered for high heating rates applications.

1. Introduction

Radiation emergencies such as nuclear war, warhead tests, nuclear strikes by terrorists and radiological accidents pose potential threat to the safety and survival of man and his environment. In these events, routine monitoring is often insufficient to provide estimates of doses received by the radiation workers, the general public and the environment. In order to effectively monitor doses from these sources, retrospective dosimetry using luminescence methods becomes relevant.

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These methods are based on heated materials such as the various ceramic materials. Ceramic materials are mostly clay and often contain quartz and/or feldspar that serve as the active phosphor for measurement. Recently, studies have been carried out to identify and characterise adequate radiation sensitive materials, which can be used as emergency dosimeters (Woda et al., 2002). These materials could be those carried close to the human body or universally available.

Efforts have been made to separate and employ naturally occurring local materials for the development of thermoluminescence dosimeters (TLD) and other applications. In this regard, many materials or minerals such as quartz, feldspar, fluorspar, dolerite, marble and muscovite have been variously studied to determine their suitability for such applications (Balogun et al., 1999; Ogundare et al., 2004, 2005, 2006a; Ige et al., 2006; Fasasi et al., 2007; Egbe et al., 2010; Mokobia, 2010, 2012). A variety of materials such as cane sugar, egg shell and chalk have also been examined for their TL potentials (Iyang et al., 2011). Ige et al. (2006) studied the TL response of gamma-irradiated muscovite samples from two granite pegmatite deposits in Ilesha and Ijero-Ekiti, Nigeria. While using a heating rate of 8 °C/s, the authors showed that the glow curves and the TL sensitivity were deposit dependent. This was due to the TL peaks observed around 238 and 330 °C in one and only 238 °C in the other. Furthermore, the glow peaks demonstrated a super-linear response at low doses while the response was between linear and sub-linear for the high doses. Relatively, the TL signal remained stable judging from the fading studies carried out over a period of one month. The results showed that muscovite has a lot of potential as a phosphor material in retrospective dosimetry and TL dating applications. Egbe et al. (2010) investigated the basic thermoluminescence properties of clay sourced from Calabar, Cross River State, Nigeria. The samples were irradiated with x-rays in the diagnostic energy range. The results showed that the TL output of the samples was very low, but demonstrated enhanced performance with the addition of common salt (NaCl). The clay samples' water absorbing property offered a means of overcoming the hygroscopic nature of common salt at those diagnostic radiological doses.

More than 80 deposits of clay raw material are widely distributed in Nigeria (Aramide et al., 2014) and the most prominent type is the clay kaolin (Okunola and Egbulem, 2015). Mineral ores hosted by these deposits include but not limited to tantalite, tin and other precious stones; and this has resulted in mostly skeletal artisanal mining for their economic benefits. Given the abundance and the vast industrial potentials of the available clay deposits, the Nigerian clay has been largely underutilised. Most other known characterisations, especially from the famous Ijero-Ekiti and Isan-Ekiti deposits, are either for other industrial applications such as ceramic and refractories (Oyinloye and Adebayo, 2005; OlaOlorun and Oyinloye, 2010; Ologe et al., 2014; Akinola et al., 2014) or background radiation level measurements (Olise et al., 2016). In order to widen the industrial potentials of these deposits, there is the need to further characterise the clay samples, especially from the deposits with minerals similar to those already studied.

Thus, this study is aimed at investigating the potential of the clay samples from the Ijero-Ekiti and Isan-Ekiti deposits for application in retrospective dosimetry. The features of clay specimens as good candidates for such dosimeters include increase in intensity peak with gamma excitation dose (McKeever, 1985). This study also investigates the dependence of the TL on the heating rates and the other glow curve factors.

2. Materials and methods

2.1. Description of the study areas

The study areas considered are located in Ijero-Ekiti and Isan-Ekiti, both in Ekiti State, South-western Nigeria. Ekiti State is entirely underlain by crystalline basement complex rocks of the gneiss- schist complex, the meta-sediments and meta-volcanic series and the Pan African granitoids–older granites (Malomo, 2011). These are composed of gneisses, schists, quartzite migmatite, charnockite, diorites, granites, granodiorites and pegmatites. The older granites are of the Precambrian-Cambrian age (Malomo, 2011).

Ijero-Ekiti plays host to a number of important minerals such as cassiterite and tin ore, columbite and foundry sand. Due to the available minerals in these deposits, Ijero area is characterised by frequent and different kinds of mining operations. The two deposits considered in Ijero are the Oke-asa and Oke-kusa deposits, both located on the hills. From the field observations, the deposits were characterised by well-dug mining pits. Oke-asa is known for its characteristic white colour clay. The minerals mined at Oke-asa include feldspar, marble and charnockite. The Oke-kusa clay deposit plays host to minerals such as kaolin, gemstones, mica, cassiterite and quartz.

Isan Ekiti in Oye local government area is considered a residual and partially reworked kaolin deposit varying from 10 to 20 m thick, with up to 1 m of overburden, overlying weathered gneiss and charnockite intruded by pegmatite (Malomo, 2011). Ceramic (ball) and kaolinite clays available in these deposits are used as raw material for abrasive, plastics, ceramic wares, pharmaceuticals, textiles, fertilizers, white tiles, insulator wares and pencils (Malomo, 2011).

2.2. Sample collection and preparation

Sample collection was conducted in February, 2015 during the dry season. In all, 13 samples, each weighing about 500 g was collected from the three different clay deposits using the standard geological techniques (Akinola et al., 2014) and the point sampling method. At all the sampling points, the exposed surface of the excavated clay was removed of overlaying organic matter using a hand trowel. The samples, which appeared in clustered form, were collected and sealed in separate polyethylene bags to prevent cross contamination. In Ijero, six (6) and two (2) samples were collected from Oke-kusa and Oke-asa deposits, respectively while five (5) samples were collected at Isan. The Oke-kusa samples presented white, grey and brownish-red colouration while those from Oke-asa presented a greyish, almost white colouration. The Isan samples were dark-grey, reddish-brown to off-white in colour. These suggest the presence of minerals such as Iron and Calcium.

The as-received samples were air-dried to achieve constant weights and subsequently pulverised using agate mortar and pestle. This was to facilitate the sieving process with a grain size of less than 200 µm. The sample preparation procedure was carried out at room temperature. In order to empty the trap sites of the stored energy due to natural radiation, the