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Technical report

Low dose TL characteristics of Nigerian fluorite

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ABSTRACT

Nigerian fluorite has been characterized by β-irradiation for thermoluminescence in the low dose range (40 μGy–72 mGy). The glow curves exhibit 3 peaks recorded at 111 ± 11 °C, 196 ± 2 °C and 282 ± 4 °C at the heating rate of 5 °C s^{-1}. The two high temperature peaks exhibit a linear response over the range of study. The minimum detectable dose for each of the observed peaks has been determined and the lowest detection limit of fluorite was also determined. A complex fading pattern was observed for the phosphor and the possible source of the TL buildup has been discussed.

1. Introduction

Ever since the advent of thermoluminescence (TL) as a practical research tool in various fields of application by Farrington Daniels and colleagues in the 1950's, efforts are being made to find out natural as well as synthetic materials that would efficiently serve the purpose of radiation measurement. Among the earliest reports on the use of natural calcium fluoride (CaF_2:N), commonly known as fluorite, as dosimeter were those of Schayes and colleagues in the 1960's. It is well known that thermoluminescent characteristics of minerals are affected by a number of factors and among them are the origin, the composition, the radioactivity, the age, the temperature and the pressure (Blanchard, 1966).

In recent time, a number of articles have been published on the TL properties of fluorite of various origins (Polymeris et al., 2006; Tugay et al., 2009; Topaksu & Yazici., 2007).

In Nigeria, CaF_2 occurs naturally in the Middle-Benue valley (Balogun et al., 1997) including Jos Plateau area (Olabanji et al., 2005).

Nigerian fluorite was first characterized for glow peaks by Balogun et al. (1999). Thereafter, the effects of varying heating rates on the glow curve shape and the kinetic characterization of the two high temperature peaks of CaF_2:N of the same origin were studied by Ogundare et al. (2004) and (2005).

In the present study, the effect of extremely low dose of β-irradiation on the TL properties of naturally occurring calcium fluoride collected from Nigeria has been investigated. The glow curves and the dose response were examined in the dose range of 40 μGy–72 mGy. The minimum detectable dose (MDD) for each of the observed peaks as well as the lowest detection limit of CaF_2:N was determined. The fading pattern of the glow curves of irradiated fluorite was also examined over a period of 43 days.

2. Experimental procedure

Some amount of fluorite ore collected from the Middle-Benue valley was prepared following the method used by Balogun et al. (1999) and ground with the standard ring mill to fine powder. The obtained powder was annealed at 400 °C for 1 h in a standard TLD oven followed by slow cooling to room temperature.

The irradiation was carried out using a ^{90}Sr/^{90}Y β-irradiator (Thermo Fisher Scientific model 2210). The irradiator has a nominal activity of 33 MBq and a dose rate of 40 μGy min^{-1} rotation^{-1}.

The model 3500 Harshaw TLD reader was employed for the TL readout of irradiated sample. The powder dispenser of the reader was used to dispense 15 mg of the sample on the planchet of the reader. A constant fade time of 5 min was kept between the irradiation process and the TL readout. Throughout the study, the heating rate of 5 °C s^{-1} was used and samples were heated between 50 and 400 °C in a Nitrogen gas flow at a constant flow rate of 6 l/min.

For the fading study, some amount of the sample was irradiated to the dose of 43.2 mGy and then stored in a dark room at room

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temperature. The post-irradiation times of 1 h, 4, 11, 16, 29 and 43 days were considered for TL readout. While for the earlier post-irradiation times only one reading was taken, two readings, within an interval of 10 min, were taken on the 43rd day.

3. Results and discussion

3.1. Glow curve analysis

Fig. 1 shows the glow curves of fluorite irradiated with doses between 40 μGy and 72 mGy. Three (3) glow peaks (PI, PII and PIII) were observed and were replicated with increased absorbed dose. The high temperature peak (PIII) occurred between 275 and 288 °C with an average value of 282 ± 4 °C. The second peak (PII) occurred between 175 and 200 °C with an average value of 196 ± 2 °C. These deviations in peak temperatures are within the limit of experimental error of the TLD reader used. However, the low temperature peak (PI) was observed to be continuously shifting towards higher temperature with increased absorbed dose. This peak (PI) occurred between 97 and 125 °C with an average value of 111 ± 11 °C. It was also observed that for the doses of 40 μGy and 0.4 mGy, the glow curves were recorded with spikes and that could be attributed to the dark current effect of the reader because of the low dose involved. The high temperature peak was observed to be broader in shape than the other two peaks for the doses considered.

3.2. Dose response

The dose response curves of each of the observed peaks of CaF_2:N are shown in Fig. 2. The two high temperature peaks (PII and PIII) exhibited a linear response over the dose range of study. It was observed that PI had the same response as PII up to 2.4 mGy (1 h of irradiation). As the dose was increased beyond 2.4 mGy, the response of PI was no longer found to be linear due to partial fading in the peak's intensity which occurred during longer irradiation times.

The conversion factor, F, which converts a given TL intensity into absorbed dose in mGy, was determined from the linear region of the dose response curve of the individual peaks. Similarly, F was also determined for CaF_2:N using the integrated TL response.

The standard deviation, σ, for the intensity of each peak was determined using a batch of 10 annealed non-irradiated fluorite samples. The standard deviation was also determined for the integrated TL intensities of the same samples.

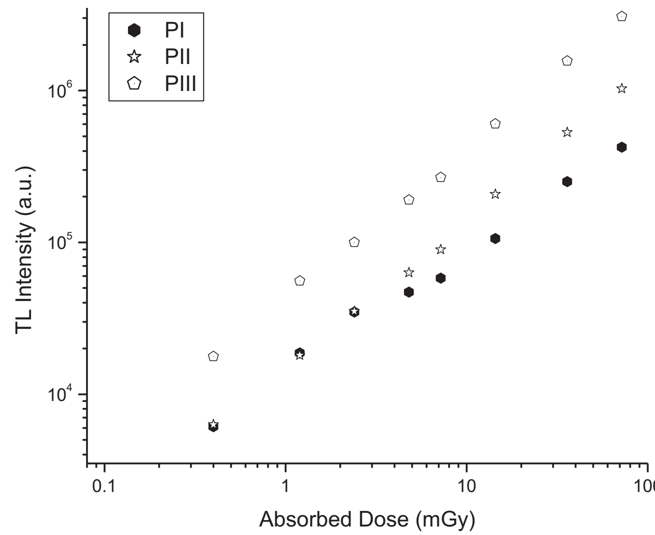

Fig. 2. Dose response curves of the observed peaks (PI, PII and PIII) of fluorite in the absorbed dose range of 40 μGy–72 mGy.

The obtained values of F and σ are listed below:

$$F(\mathrm{PI}) = (1.6 \pm 0.06) \times 10^{-4}\ \mathrm{mGy/a.u.} \quad \sigma(\mathrm{PI}) = 0.704\ \mathrm{a.u.}$$

$$F(\mathrm{PII}) = (6.96 \pm 0.06) \times 10^{-5}\ \mathrm{mGy/a.u.} \quad \sigma(\mathrm{PII}) = 0.983\ \mathrm{a.u.}$$

$$F(\mathrm{PIII}) = (2.33 \pm 0.015) \times 10^{-5}\ \mathrm{mGy/a.u.} \quad \sigma(\mathrm{PIII}) = 4.35\ \mathrm{a.u.}$$

$$F(\mathrm{CaF_2 : N}) = (6.51 \pm 0.16) \times 10^{-4}\ \mathrm{mGy/a.u.} \quad \sigma(\mathrm{CaF_2 : N}) = 0.104\ \mathrm{a.u.}$$

The minimum detectable dose is defined as (Prokic, 2001):

$$\mathrm{MDD} = 3\ \sigma\ F \tag{1}$$

Substituting for the values of F and σ in Equation (1), the following results were obtained:

$$\mathrm{MDD(PI)} = (0.34 \pm 0.013) \times 10^{-6}\ \mathrm{Gy},$$

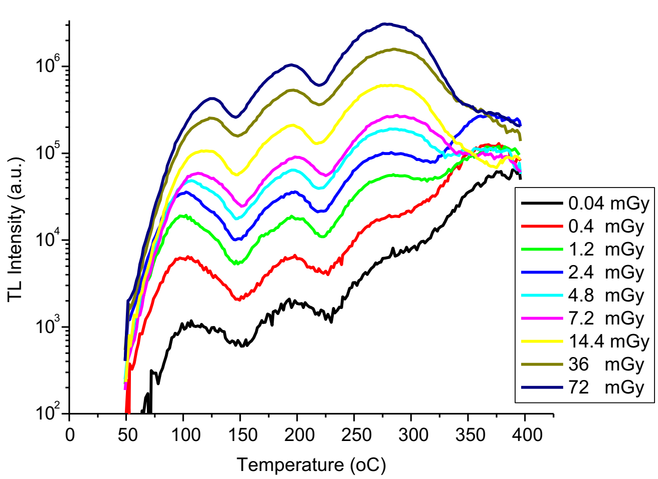

Fig. 1. Glow curves of fluorite in the absorbed dose range of 40 μGy–72 mGy acquired at the heating rate of 5 °C s^{-1}.

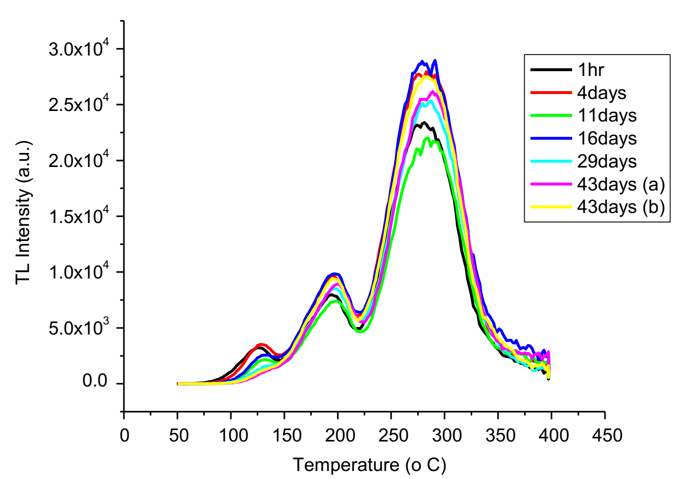

Fig. 3. Glow curves of fluorite irradiated to the dose of 43.2 mGy and read at the heating rate of 5 °C s^{-1} for different storage periods at room temperature.

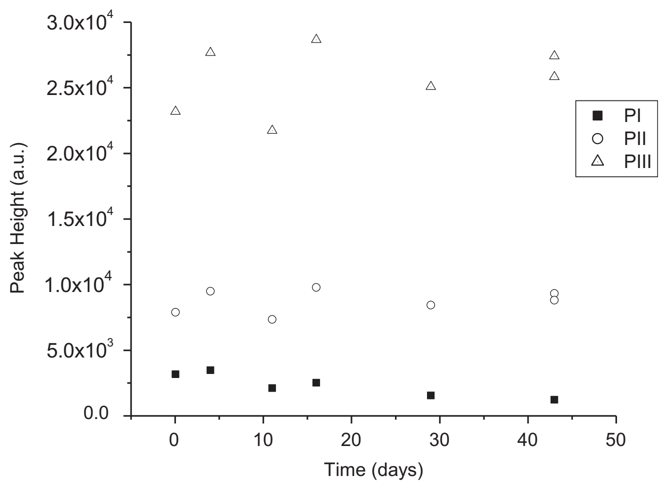

Fig. 4. Peak heights of PI, PII and PIII observed in Fig. 3 as a function of storage time (varying from 1 h to 43 days).

$$\mathrm{MDD}\left(\mathrm{PII}\right) = (0.21 \pm 0.002) \times 10^{-6}\ \mathrm{Gy},$$

$$\mathrm{MDD}\left(\mathrm{PIII}\right) = (0.3 \pm 0.002) \times 10^{-6}\ \mathrm{Gy} \quad \text{and}$$

$$\mathrm{MDD}(\mathrm{CaF_2 : N}) = (0.2 \pm 0.005) \times 10^{-6}\ \mathrm{Gy}.$$

The obtained values of MDDs are all in the order of 10^{-6} Gy and are comparable to the lowest detection limit of fluorite obtained by Polymeris et al. (2006) using optically stimulated luminescence (OSL) method.

3.3. *Fading*

Fig. 3 shows the glow curves of CaF_2:N (irradiated to the dose of 43.2 mGy) after different storage periods. The intensities of the peaks observed in Fig. 3 were plotted as a function of storage time in Fig. 4. It was observed that PI faded as much as 62% after 43 days of storage while PII and PIII did not follow any specific decay pattern. In order to confirm that the observed fluctuations in the peak heights with increased storage period are not solely due to experimental errors, the two readings taken on the 43rd day were examined. It was observed that the peak heights of PI in both cases were almost the same while those of PII and PIII showed significant differences. These observations could be attributed to self-irradiation of the natural phosphor as a result of the radiation emitted in the decay process of the radioactive elements present in it. From the elemental composition of Nigerian CaF_2:N (Balogun et al., 1997; Olabanji et al., 2005), ^{90}Y which is a pure β-emitter (Knoll, 2000) with a short half life of 64 h stands as the major contributor to the observed TL buildup. Balogun et al. (1999) have evaluated the specific gamma activity of natural calcium fluoride of the same origin to 13 Bq/Kg which suggests that the self-irradiation could not be from γ-rays. More so, the presumed γ-emission could only originate from thorium whose common isotopes are ^{230}Th and ^{232}Th. These two elements primarily emit α-particles followed by γ-emission. However, the half lives of 1.4×10^{10} years in the case of ^{232}Th and 7.7×10^4 years in the case of ^{230}Th make it improbable in our case due to short study period. It was also observed that no significant buildup was recorded after the TL readout of annealed non-irradiated fluorite upon storage over 1 week. Cherniak et al. (2001) showed that Y had the fastest diffusion rate of all five elements including Dy, Nd, Sr and Yb with a diffusion rate of $1.2 \times 10^2\ exp\,(-458\ kJ\ mol^{-1}/RT)\ m^2/s$ in treated fluorite. This could be a related phenomenon to the point hereby made.

4. Conclusion

CaF_2:N exhibits three glow peaks (PI, PII and PIII) upon irradiation with β-rays from 40 μGy to 72 mGy in the temperature range of 50–400° C. The high temperature peak was found to be broader in shape while the low temperature peak was observed to be continuously shifting towards higher temperature with increased dose. This is an indication of the complexity of these two peaks. On the other hand, the shape of PII and the relative stability of the temperature at which it occurred, with increased dose, suggest a first order kinetics. The linear response of this phosphor coupled with its lowest detection limit of the order of 10^{-6} Gy make it appropriate for background environmental monitoring.

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