LUMINESCENCE PROPERTIES OF GAMMA IRRADIATED AMPHIBOLITES AND β-ANHYDRITE RELEVANT TO DOSIMETRY

[1]Obed R.I*, [1]Olatunji A, [2]Fasasi M.K, [3]Dejene F.B & [4]Adebiyi A.A

[1]Department of Physics, University of Ibadan, Ibadan, Nigeria.
[2]Centre for Energy Research and Development, Obafemi Awolowo University, Ile-Ife, Nigeria.
[3]Department of Physics, University of the Free State, Private Bag X13, Phuthaditjhaba 9866, South Africa.
[4]Abdus Salam International Centre for Theoretical Physics (ICTP), Diploma Progamme, Trieste, Italy.
*E-mail: rachelobed@yahoo.com

ABSTRACT

The thermoluminescence (TL) characteristics of amphibolites and β-anhydrite have been investigated after gamma-ray irradiation. Both materials presented glow curves as a consequence of their trap system. The amphibolites have two peaks at low doses which convoluted to one peak at high doses. The β-anhydrite also has two main peaks that are prominent at high doses. Linearity index analysis showed that the amphibolite is supralinear while the β-anhydrite is either sublinear or supralinear depending on the dosimetric peak used. The amphibolites faded by 35% per month, while the β-anhydrite faded by 67% in a month. The two materials studied presented different TL properties.

Keywords: *Thermoluminescence, dose, amphibolites, β-anhydrite, annealing*

1. INTRODUCTION

Studies and applications of thermoluminescence (TL) dosimetric materials have been actively developed in the past three decades and traditionally, TL materials have been used for the detection of x-ray, gamma and beta radiation. Some of the areas where TL dosimetry has been successfully applied include: radiation monitoring of personnel working with ionizing radiation, environmental radiation monitoring, radiation therapy and quality assurance of therapy machines [1], [11] and [10]. Studies dealing with retrospective dosimetry and the search for materials with a high TL yield are also ongoing [5]. Therefore studies into the TL properties of some naturally occurring materials such as amphibolites and β-anhydrite are of relevance. Amphibolites can be found in many geological formations. The TL properties of natural samples have been found to depend upon their genesis, chemical composition, impurity content and geological history, requiring a TL study for each sample [7]. The TL analysis is widely used as an effective tool in the determination of trapping parameters in order to find a material's suitability for various applications [12].

In the present study, the TL properties of amphibolites and β-anhydrite are reported. Amphibolites are metamorphic rocks that consist mostly of amphiboles and plagioclase feldspar and little or no quartz. Amphiboles are mineral components of the metamorphic rocks while plagioclase feldspars are sodium/calcium aluminium silicates such as albite, ologoclase, andesine, labradorite, bytownite and anorthite. Amphibolites are used as aggregate for road stone and also for building construction. They can also be used for geochronology when samples are collected in the dark and not exposed to light. The dihydrate ($CaSO_4.2H_2O$) occurs naturally as gypsum and can be cast into various shapes which includes sheets (for drywall), sticks (for blackboard chalk) and molds (to immobilize broken bones, or for metal casting). On heating above 250 ^{0}C, the completely anhydrous form called β-anhydrite or "natural" hydrite is formed. Natural anhydrite does not react with water, even over geological timescales. Both amphibolites and gypsum are found and widespread all over Nigeria.

The effect of pressure on the luminescence properties of gypsum ($CaSO_4$) has been reported by Lakshmanan [6] but to the best of our knowledge there has been no published work on the TL properties of amphibolites relevant to radiation dosimetry.

2. MATERIALS AND METHODS

The materials used in this study were amphibolites (rock sample) and β-anhydrite (blackboard chalk). The amphibolites were mined from Ilesa while the blackboard chalk was purchased at Ile-Ife; both cities are located in Southwestern Nigeria. Both environmental materials were ground to fine powder. The amphibolites samples are dark-coloured characterized by a clear anisotropic crystalline structure and fabric while the blackboard chalk is a white powder.

2.1. Elemental analyses

Since the luminescence spectra were expected to be dependent on the major elemental analyses (K, Na, Zn and Ca) as well as certain trace element contents (*e.g.* Mn and Pb), elemental analyses were performed for the rock (amphibolites) and β-anhydrite samples. The elemental analyses of the samples were carried out using the Atomic Absorption Spectrophotometer; Alpha 4 ChemTech Analytical of the Centre for Energy Research and Development (CERD) Obafemi Awolowo University, Ile-Ife, Nigeria to find out the elemental compositions in each sample.

2.2. Annealing and irradiation of samples

Before use, the samples were annealed in the oven at 380 ^{0}C for 1 hr. After this annealing, the TLDs were cooled to room temperature before being subjected to irradiation. For γ-ray irradiation of the samples, a calibrated ^{60}Co gamma chamber with a dose of 4.5 Gy min^{-1} was used. In order to investigate the dose response, several portions of the annealed sample, each portion weighing 15 mg, were irradiated to different doses ranging from 10 Gy to 3 kGy. Doses between 10 Gy and 300 Gy were classified in this work as low doses while doses from 0.5 kGy and 3 kGy as high doses. The TL glow curves were measured using a heating profile similar to the usual one used for routine dosimetry, consisting of a pre-heat to 50 ^{0}C and a linear heating rate of 10 ^{0}C/s up to 400 ^{0}C and kept at the maximum temperature of 400 ^{0}C using a TLD Reader (Model 3500, Harshaw) at the Centre for Energy Research and Development (CERD). The TL reader is interfaced to a personal computer for data acquisition in the laboratory.

3. RESULTS AND DISCUSSION

The results and discussion of the elemental analyses, TL glow peaks, dose response, batch homogeneity and fading are presented for the two naturally occurring materials.

3.1. Result of the Elemental Analysis

The results of the elemental analyses of amphibolites and β-anhydrite by Atomic Absorption Spectrophotmetry (AAS) are presented in table 1. From the table, it is observed that for the amphibolites, K has the highest concentration followed by Na, Zn, Cu and Mg, Ca and Fe in that order as the major elements present in the samples. Apart from the major elements, amphibolites contain some other minor elements such as Co, Pb, Cr and Cd as presented in table 1. The absence of Mn content in this sample is consistent with the absence of calcite, in which Mn readily substitutes.

For the β-anhydrite sample, K also has highest concentration followed by Na, Mg, Zn, Cu and Ca as the major elements present in it. The minor elements shown are: Co, Mn, Pb, Cr, Fe and Cd. It is already established in the literature that impurity atoms affect thermoluminescence; that some impurities may or may not activate TL.

Elements	Amphibolites, ppm	Blackboard chalk, ppm
Zn	2.154 ± 0.03	2.557 ± 0.01
Cu	1.99 ± 0.02	2.45 ± 0.01
Co	0.05 ± 0.00	0.12 + 0.00
Mn	Nil	0.01 ± 0.00
Pb	0.03 ± 0.00	0.05 ± 0.01
Cr	0.10 ± 0.00	0.14 ± 0.00
Ca	0.92 ± 0.00	1.54 ±0.04
Mg	1.50 ± 0.06	2.87 ± 0.07
Na	5.69 ± 0.00	9.32 ± 0.00
K	17.82 ± 0.58	16.05 ± 1.94
Cd	0.02 ± 0.00	0.02 ± 0.00
Fe	0.23 ± 0.01	0.29 ± 0.03

Table 1. Results of elemental analysis of Amphibolite and blackboard chalk by Atomic Absorption Spectrophotometry (AAS).

3.2. TL glow curves

The TL glow curves of amphibolites for samples exposed to gamma radiation of between 10 and 300 Gy are shown in figure 1. From the figure it is observed that amphibolites has a distinct peak around 120 ^{0}C and the peak temperature shifts to higher temperatures as the dose increases. Also a broad emission with maximum peak at around 177 ^{0}C was detected as the dose increased. It is also observed that TL intensity increases as the dose increases. The TL glow curves of amphibolites are shown in figure 2 for samples exposed to gamma radiation in higher dose ranges. From figure 2, it is observed that the peak structure of TL glow curves of amphibolites is almost independent of gamma dose even up to exposures of 3 kGy, except for the height of the main dosimetric peak (≈ 135 ^{0}C) and this peak temperature remains constant. The dosimetry peak for this material is lower than those of other common TL dosimeters, such as LiF:Mg,Ti, LiF:Mg,Cu,P, CaSO$_4$:Dy, CaSO$_4$:Tm and so forth. These have the temperature of the main dosimetric peaks to be between 200 and 250 ^{0}C [3]. It is also observed that the TL intensity at the peak temperature increases as the dose increases.

From the results obtained, it can be deduced that the amphibolites presents a convolution of many peaks at low doses ($<$ 0.5 kGy) and that the material could be used for dosimetric applications at high doses ($\geq$ 3 kGy) at approximately 135 ^{0}C. Figure 3 shows typical TL glow curves of β-anhydrite for samples exposed to gamma radiation in different dose ranges (0.5 – 3 kGy). The TL curves presented two peaks around 105 ^{0}C and 275 ^{0}C. Each peak may have different dosimetric characteristics and may be differently dependent on a variety of factors such as the annealing procedure and type of radiation. From results obtained, the two peaks are not very conspicuous at low doses (10 – 300 Gy). The glow curves were found to be dose dependent. It was also observed that the TL intensity at the peak temperature increased as the dose increased.

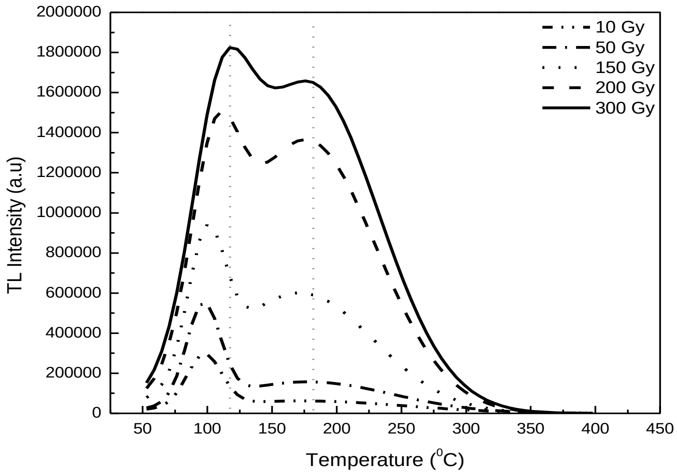

Figure 1. Glow curves of Amphibolites irradiated from 10Gy to 300Gy (low doses).

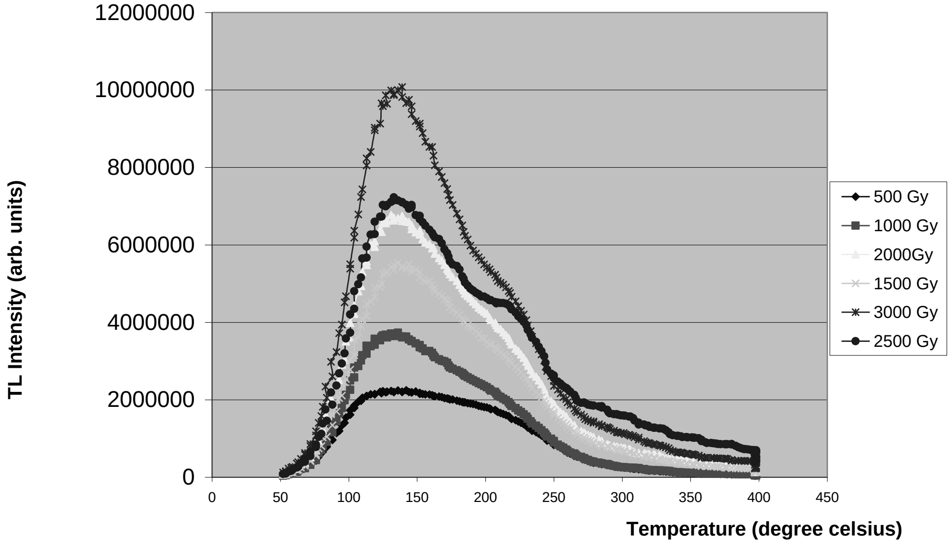

Figure 2. Glow curves of Amphibolites irradiated from 500Gy to 3000Gy (High doses).

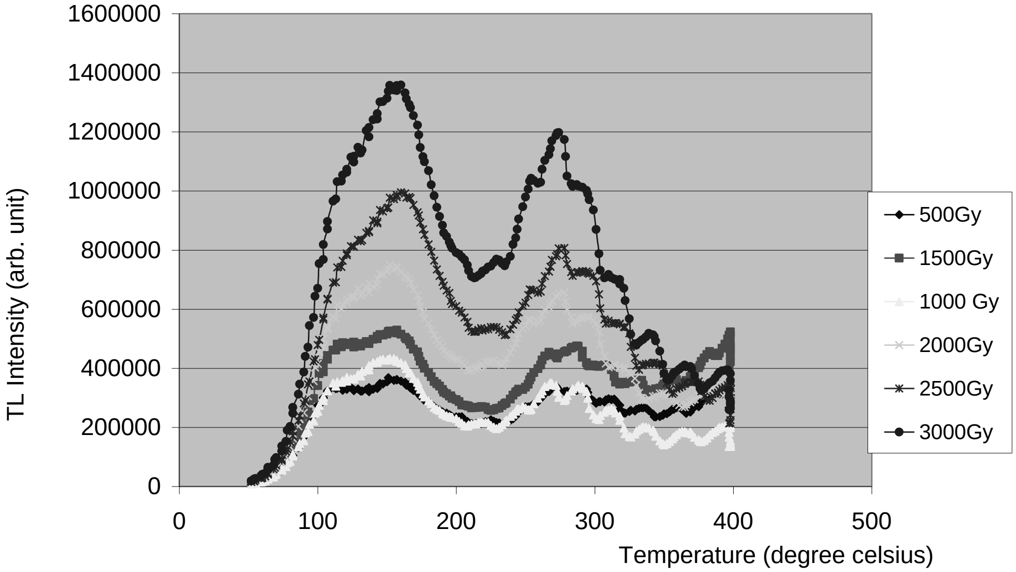

Figure 3. Glow curves of β-anhydrite irradiated between 500Gy and3000Gy (High doses).

3.3. Dose Response

The TL response of the samples was studied over the dose range of 10 Gy to 3 kGy of gamma irradiation. As shown in figure 4, the TL intensity of the glow peak increase almost linearly until 3 kGy of gamma dose for the main dosimetric peak of amphibolites (≈ 135 ^{0}C). Figures 5 and 6 show the plot of TL intensity against dose for peak I and peak II of β-anhydrite, respectively. The two peaks are actually convolution of many peaks. One of the most important properties of TL detector is that it should exhibits a linear relationship between TL intensity and absorbed dose according to McKeever [8]. Figures 4 through 6 reveal that the dose response of amphibolites is better in terms of linearity than that of β-anhydrite. This suggests that the amphibolites will be a better TL detector than the β-anhydrite. A further check on the linearity of the dose-response curve of amphibolites and β-anhydrite was performed using the linearity index f(D) proposed by Chen and McKeever [2]:

$$f(D) = \left[\frac{S(D) - S_O}{D}\right]\left[\frac{S(D_1) - S_O}{D_1}\right]^{-1} \qquad (1)$$

where in a graph of TL response against dose, S_0 is the intercept on the TL intensity axis as extrapolated from the linear region of the graph. S(D) is the sample TL response corresponding to dose D, and D_1 is a dose in the linear region,. S(D_1) is the sample TL response corresponding to dose D_1. By this definition *f(D)* > 1, *f (D)* = 1 and *f(D)* < 1, respectively imply supralinearity, linearity and sublinearity. Figures (7) – (9) show the plot of *f(D)* against dose. Using eq. (1) to fit the experimental data, it is concluded that the TL dose-response of amphibolites to gamma rays is supralinear at both low and high doses while the dose responses of β-anhydrite peaks 1 and 2 are sublinear and supralinear at low doses but mostly sublinear at high doses.

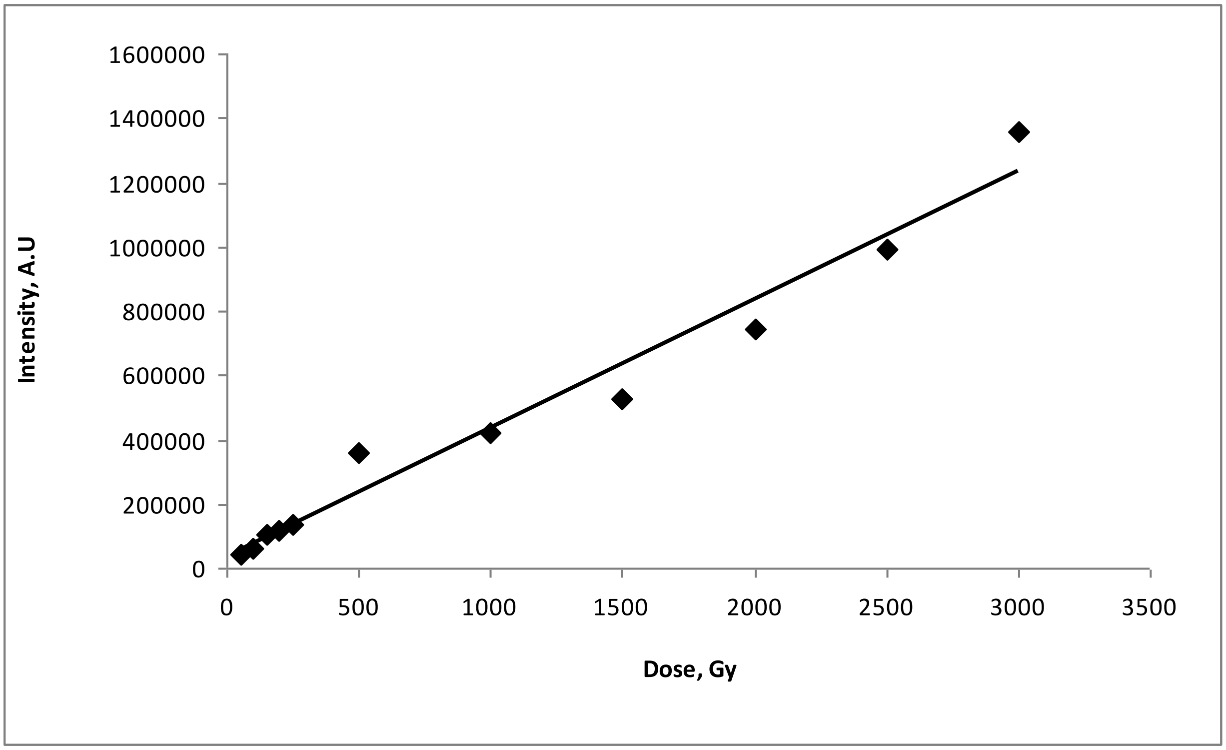

Figure 4. The Gamma-Dose Response for Amphibolites.

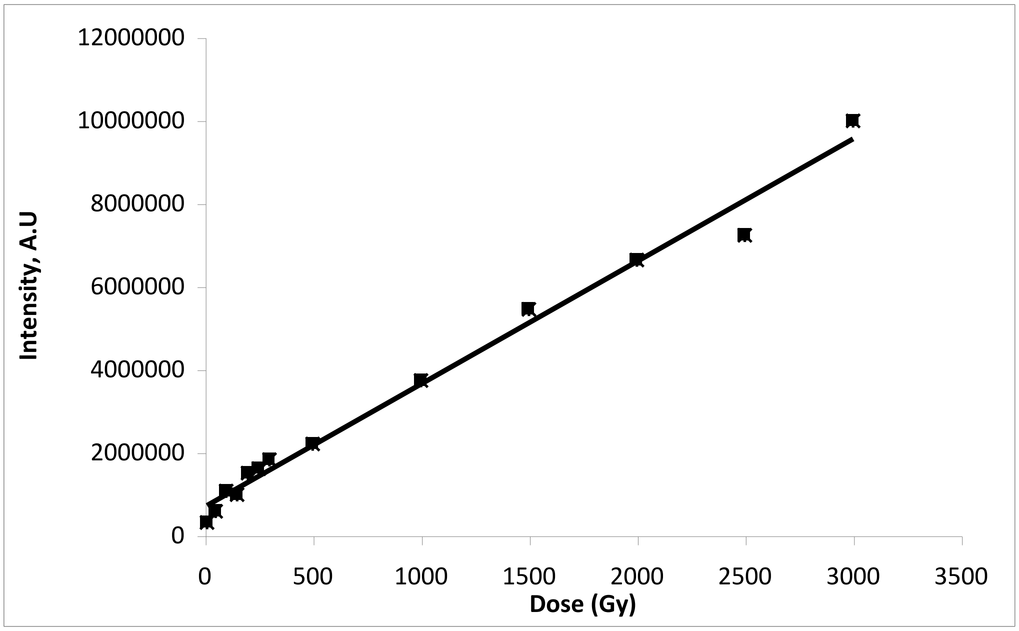

Figure 5. Dose response for β-anhydrite peak I.

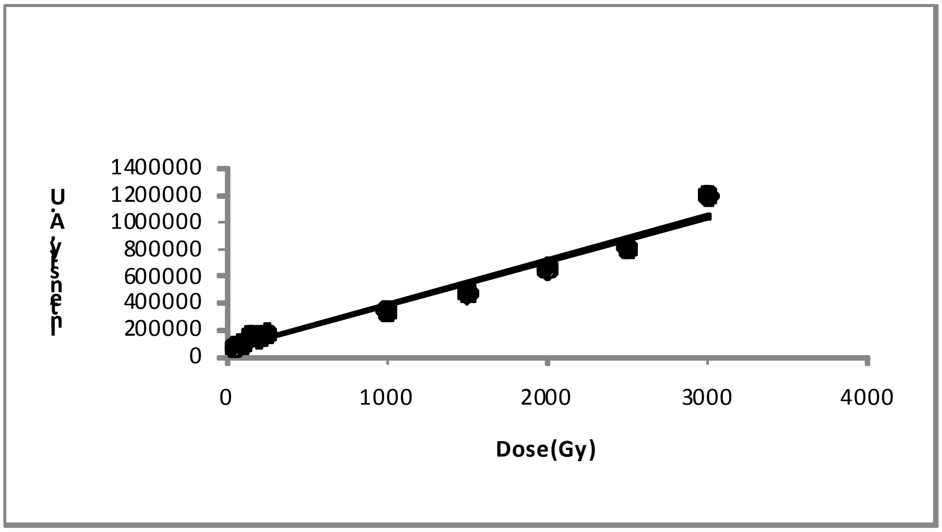

Figure 6. Dose response for β-anhydrite peak II.

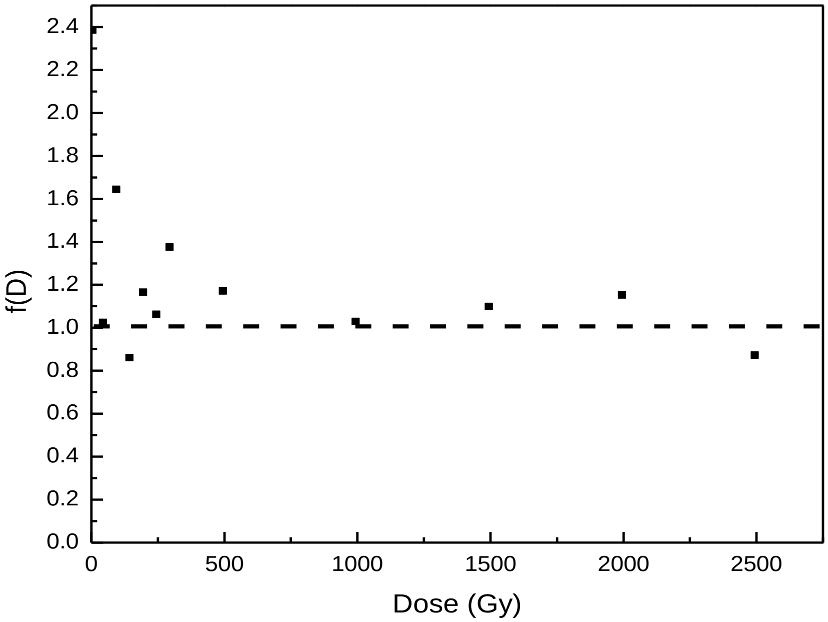

Figure 7. Plot of the Linearity Index function f(D) against dose of Amphibolite.

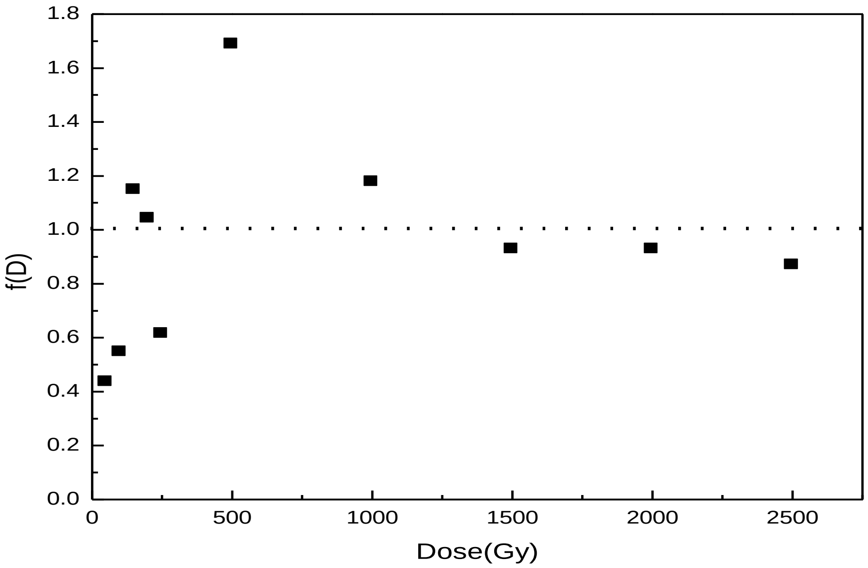

Figure 8. Plot of the Linearity Index Function f(D) Against Dose of β-anhydrite Peak 1.

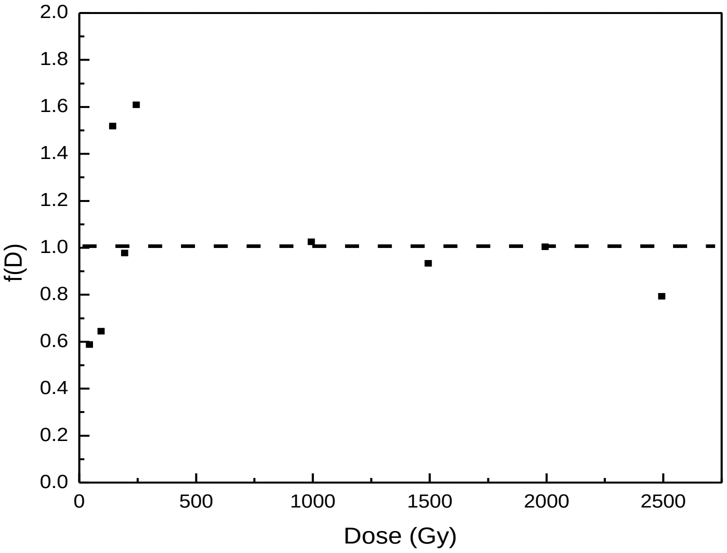

Figure 9. Plot of the Linearity Index Function f(D) Against Dose of β-anhydrite Peak 2.

3.4. Batch homogeneity

TL response of same samples subjected to the same TL measurement conditions may not necessarily be the same. The International Electrochemical Commission (IEC) [4] has recommended that the evaluated value for any one dosimeter in a batch shall not differ from the evaluated value of any other in the same batch by more than 30%, that is,

$$f \equiv \frac{TL_{\max} - TL_{\min}}{TL_{\min}} \leq 0.3 \qquad (2)$$

Amphibolites and β-anhydrite has been investigated using 13 samples of each phosphor from the same batch and subjected to the same treatment of annealing, irradiation and read out. For the amphibolites, f has been calculated to be 0.2, a value which is lower than the upper limit recommended by IEC whereas for the β-anhydrite, 0.3 has been estimated, a value slightly higher than the upper limit recommended by IEC. This indicates the homogeneity of the samples used. To further show the homogeneity of the phosphors within a batch, the percentage standard deviation of the 13 TL responses from their mean was estimated and obtained to be 20% for amphibolites and 30% for the β-anhydrite. The deviation for amphibolites is lower than recommended while the deviation for β-anhydrite is the upper limit recommended, showing the homogeneity of the samples used.

3.5. Fading

Fading study was carried out by keeping the irradiated samples in the dark, (to prevent UV light) for different number of days at room temperature before read out in the TLD reader. The samples were kept over a storage period of four weeks at a gamma radiation dose of 1000 Gy. Figure 10 shows that amphibolite's TL responses decays with storage time. It is observed that after 4 weeks of storage, the TL response has reduced by 35%. The majority of the commercial TL dosimeters such as TLD-100 have small fading (not over 5% annually), except CaF_2 dosimeters whose monthly fading may be as high as 25% [9]. Also, it is observed that there is an increasing change in peak temperature with storage time. A shift in the glow peak maximum towards high temperature indicates that low-energy traps are damaged more than high-energy traps. It is known that deep traps are present in many TL dosimetric materials [5].

Figure 11 shows that the β-anhydrite also decays with storage time. Unlike the amphibolites, it has shallow trap because the TL yield after 4 weeks of storage is about 67%. The decay of β-anhydrite does not follow a specific trend like that of amphibolites. This means that β-anhydrite cannot store dosimetric information for a long time. The available results show that amphibolites could be used as TL material for high-dose dosimetry both for industrial and scientific applications.

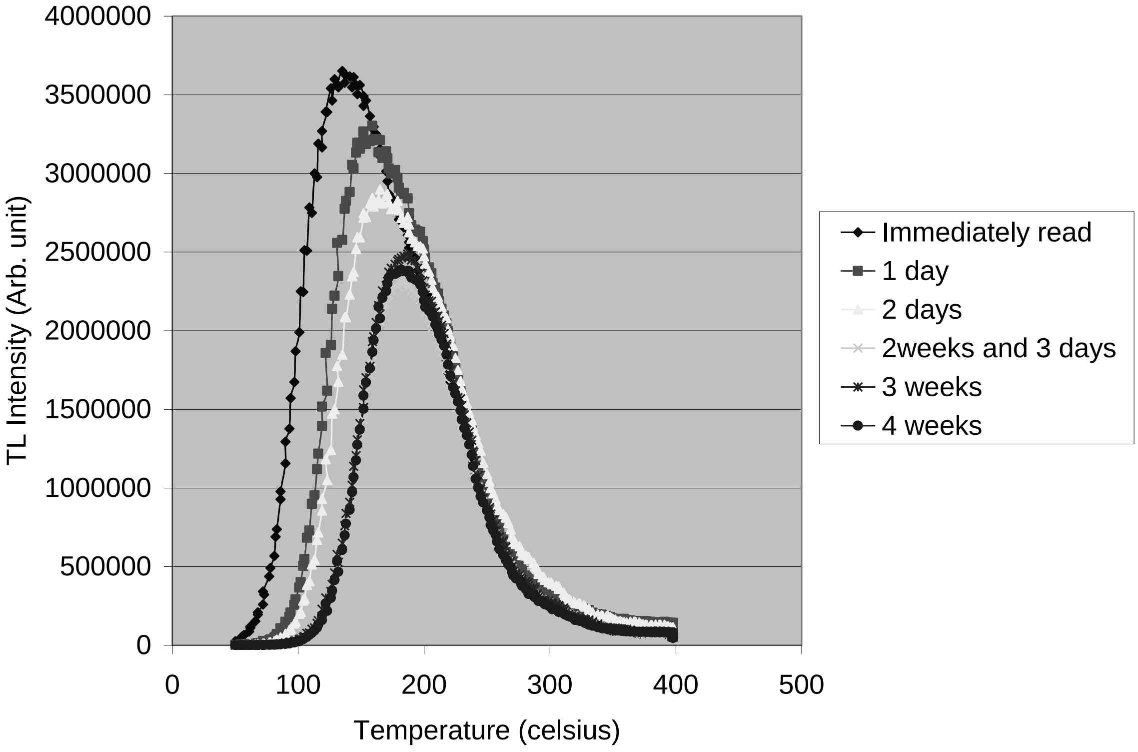

Figure 10. Fading characteristics of Amphibolites

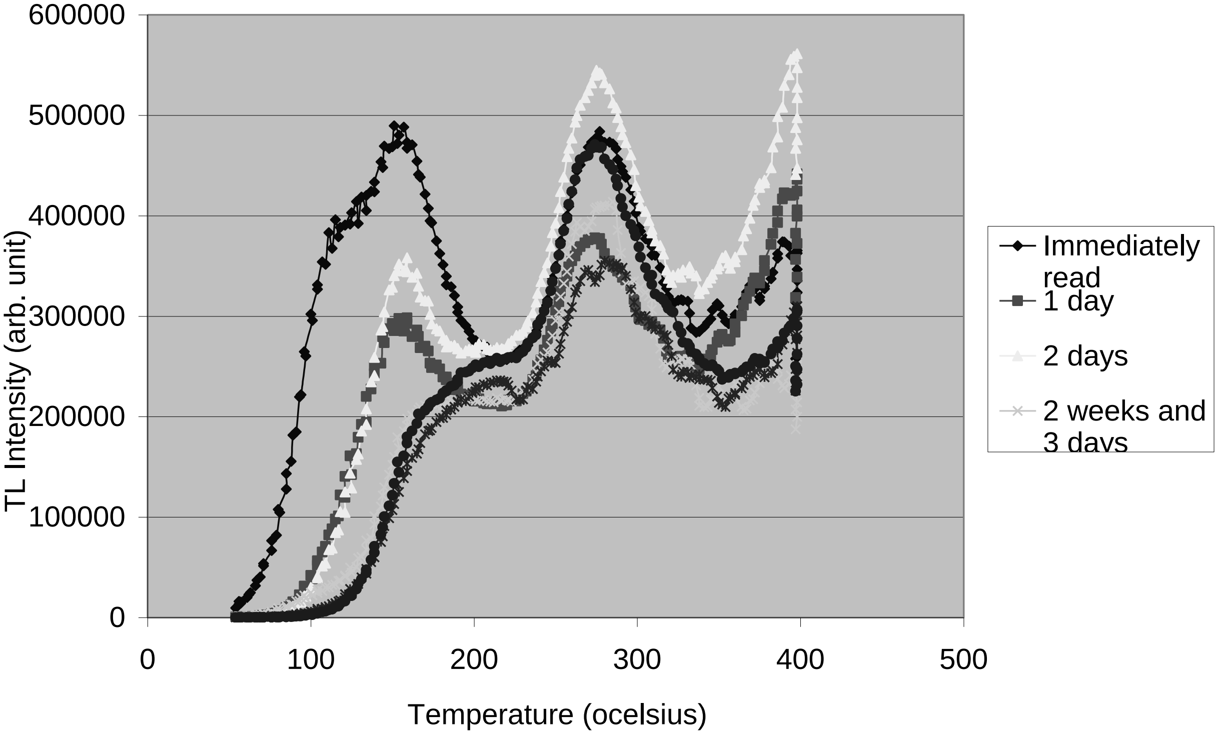

Figure 11. Fading characteristics of β-anhydrite.

4. CONCLUSIONS

A TL-dosimetric study has been carried out using some naturally occurring materials such as amphibolites and β-anhydrite. The TL properties of amphibolites and β-anhydrite have been investigated. Varying the gamma dose (0.5-3 kGy), thermoluninescence (TL) measurements were made and glow curve maximum is obtained at 135 ^{0}C for amphibolites, 105 ^{0}C and 275 ^{0}C for β-anhydrite peaks. The amphibolites could be used for dosimetric applications at high doses (≥ 3 kGy), at approximately 135 ^{0}C. The dose response of amphibolites is better in terms of linearity than that of β-anhydrite. The amphibolites faded by 35% per month, while the β-anhydrite faded by 67% in a month. The low temperature TL glow peaks and high fading of the materials act as disadvantages for their retrospective use for TL dosimetry purposes. However, with improved standardization of amphibolites, it could be used as TL material for high-dose dosimetry both for industrial and scientific applications.

5. ACKNOWLEDGEMENTS

Part of this work was carried out at the Abdus Salam International Centre for Theoretical Physics (ICTP), Italy. Two of the authors who are Associates and one who is a diploma student thank “ICTP, Trieste, Italy”. R.I. Obed, is grateful to ICTP for providing a research fellowship under the ICTP-TRIL programme.

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