INVESTIGATING THE DOSIMETRIC POTENTIALS OF NATURAL MARBLE

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ABSTRACT

This study examined the thermoluminescence (TL) dosimetric characteristics of natural marble with a view to ascertaining its potential as a TL based retrospective dosimeter. The marble samples from the Basement Complex of South Western Nigeria, were irradiated with a well calibrated ^{60}Co gammacell 220 irradiator. TL measurements were carried out using a ictoreen TL reader (Model 2800M).. The results indicate that the glow curve structure of this the material is independent of gamma dose except for the height of the main peaks. Its TL output following irradiation is reproducible with a reproducibility index of 0.32 ~ 0.3, the internationally recommended upper limit for most dosimeters. The TL– dose relationship is linear having the equation: $TL_{\text{int}(marble)} = 8.035E - 06D^2 + 0.06083D + 7.26677$ with the TL output being less sensitive than that of TLD-100 the 'standard' TL phosphor. Results also indicate that the glow curve is associated with high dose range and its linearity limit of 1.45 kGy is high compared to about 10 Gy of most common TL dosimeters. Furthermore, its fading feature is common to most TL detector materials. The minimum detectable dose for this phosphor is 0.03 kGy. It is therefore concluded that this material has the potential for high radiation dosimetry such as is found in nuclear accidents and radiation processing of food and medical appliances.

Keywords: Marble, Thermoluminescence characters, glow curve, dosimeter

INTRODUCTION

Consequent upon the immense direct benefits of radiation to man, there is an increasing spread of medical and industrial applications of ionizing radiation. Radiation we know is invisible, odourless and tasteless (Olomo, 2006) and as such, no human sensory organ is known to be sensitive to its presence. As a result of the consciousness of its documented deleterious effects, it is important its presence be both detectable and measurable since nuclear accidents/incidents never give prior notice.

Two purposes of detection and measurement are environmental dose control and personnel dose determination. These are vital in ensuring the radiological safety of the population and of individual radiation workers. Relevant and reliable dosimeters are required for these measurements. Amongst varying forms of dosimeters that could be employed, are those based on the phenomenon commonly referred to as thermoluminescence (TL). TL, the emission of light from an insulator or semiconductor when heated following previous energy absorption has found favour in diverse disciplines such as archaeology, geology, medicine, biology/biochemistry, industry etc. (Walker, 1992; Valkovic', 2000). Its application in

environmental monitoring followed soon after the introduction of its role in dosimetric purposes (Ranogajec-komor, 2002).
In the developing countries the cost of procurement of these dosimeters is high thus negatively affecting some aspects of research and training. This explains the antecedent quest by various investigators to source appropriate local TL materials in Nigeria for instance (Ogundare et al, 2004; 2006; Ige et al, 2006; Fasasi et al, 2007). Most importantly, Nigeria presently is geographically located at the centre of countries that have one or more nuclear reactors in the continent (Mokobia, 2009). These countries include Morocco, Algeria, Libya, Egypt, Ghana, Democratic Republic of Congo (DRC), Namibia and South Africa (IAEA, 1990; Tran, 2007). Meanwhile her first nuclear research reactor, the Nigeria Research Reactor 1 (NIRR-1) sited at the Centre for Energy Research and Training CERT, Ahmadu Bello University ABU Zaria is fully operational. Plans have also reached advanced stages for the country as well as others in her neighbourhood to acquire power reactors for the purpose of electricity generation.

Thus there is an expected upsurge of radiation services which may result in an increase in the exposure of the population to ionizing radiation since these facilities have tendencies of releasing radioactive effluents to their immediate environment and even beyond without respect to national and international boundaries. This situation demands that the Nigeria Nuclear Regulatory Agency (NNRA) ensures there is increased radiation surveillance in order to guarantee the fulfillment of one of the primary concerns of the national nuclear programme - the provision of adequate radiation protection.

The number of available dosimeters may be insufficient to cope with this anticipated upsurge. The implication will be that the nation may not be able to effectively monitor the radiation exposure of her populace as well as the radiation workers. Consequently, sourcing appropriate local materials that will introduce more available and well-understood dosimeter phosphors, which would be utilized by the country is certainly a viable and tangible option. This work then sought to investigate the TL dosimetric characteristics of marble of Nigerian origin with a view to determining its suitability for use as a retrospective TL dosimeter in the event of a nuclear accident. Specifically, the studied characteristics are Glow curve feature, TL – dose response, Batch homogeneity, fading characteristics, Dose threshold and Relative sensitivity.

MATERIALS AND METHOD

The marble samples were collected from Igarra in the South Western Basement Complex located between latitude 7-10^{o} N and longitude 2-5^{o} E. The as obtained samples were cleaned using acetone, crushed and then air dried to constant weight at room temperature. Pulverization was carried out using a thoroughly acetone cleaned pulverizer at the Department of Geology, Obafemi Awolowo University, Ile-Ife. The pulverized samples were then packed into a number of properly labeled polyethylene sachets and stored in a dessicator.

Pre-irradiation annealing was carried out using the DA Pitman programmed thermoplate annealing oven at the Institute for radiation Protection and Research (IRPR), University of Ibadan, at a temperature of 400^{o}C for 2 hours. The annealed samples were then cooled at room temperature, repacked into the polyethylene sachets surrounded by double black polyethylene bags and replaced into the dessicator. Aliquots of the samples used for the various investigations undertaken in this work were subsequently taken from these.

To investigate the intended TL characteristics, a number of samples of the annealed phosphors each of appropriate mass in different batches were exposed to varying gamma radiation doses using the ^{60}Co gammacell 220 irradiator located at the Centre for Energy Research and Development CERD, Obafemi Awolowo University, Ile-Ife as described below. TL measurements (readouts) were performed at appropriate pre-heat temperatures for a maximum temperature of 400 ^{o}C using the Victoreen TL reader also in CERD. An appropriate linear heating rate was employed for each measurement.

GLOW CURVE ACQUISITION

Aliquots of five batches of equal masses (11mg) were separately exposed to varying gamma irradiation doses in the gamma cell. In each case, TL readout immediately preceded irradiation. The glow curves were then obtained by plotting the TL responses of these equal masses of the phosphor against temperature (^{o}C) (Figure 1).

TL RESPONSE WITH GAMMA IRRADIATION DOSE

The relationship between dose and the TL response was studied over the dose range 30.3 Gy to 3.8 kGy of gamma irradiation by irradiating aliquots of five batches of equal masses (11mg). The TL response was then plotted against gamma radiation dose. The graph obtained is shown in Figure 2.

BATCH HOMOGENEITY

Homogeneity of TL response is an important property of a material that could be used as dosimeter. Experiments have shown that the TL response of samples of same material that have undergone the same treatment may not necessarily be the same. Accordingly, The International Electrochemical Commission (IEC, 1991) recommends that for a material to qualify for use as a dosimeter, the homogeneity index for any one dosimeter in a batch shall not differ from that of any other in the same batch by more than 30%. The expression for this index, f is given by the equation [1] (!EC, 1991):

$$f \equiv \frac{TL_{max} - TL_{min}}{TL_{min}} \leq 0.3 \quad ....[1]$$

TL_{max}, TL_{min} represent the maximum and minimum net TL readouts respectively in a batch of dosimeters. This index was determined for the studied phosphor using 10 samples from the same batch subjected to the same treatments of annealing, irradiation and read out. The results are presented in Table 1.

FADING CHARACTERISTICS

For this investigation, 21 samples of the material in seven batches each containing three samples annealed under same conditions were irradiated for the same time but readout after different post- irradiation periods ranging from immediately after to greater than one month. The stability of the stored TL signal (fading) over a period of 40 days was investigated by plotting TL response against post irradiation storage period. This plot is given in Figure 3.

DOSE THRESHOLD

This is the lowest level of detection (minimum detectable dose) (El-Faramawy et al., 2000). It was determined using 20 unirradiated samples of the phosphor each of mass 20 mg. This determination was done employing the relationship: (Prokic, 2000)

$$D_{Th} = 2.26\sigma F \quad \text{.....}[2]$$

σ is the standard deviation of the background readout values of the 20 unirradiated samples in arbitrary units (AU) while F represents the conversion factor (59.579) in units of Gy $(AU)^{-1}$ obtained from the linear dose response curve. Each of the readouts was carried out at a pre-heat temperature of 50 ^{o}C, heating rate of 8 $^{o}Cs^{-1}$ and a cycle time of 45s.

RESULTS AND DISCUSSION

From Figure 1, it is obvious that the TL glow curve for marble exhibits four peaks. The structure of the curve is almost independent of gamma dose except for the height of the main peaks. This feature of the glow curve is expected for common dosimeters. Thus natural marble exhibits glow curve structure similar to common dosimeters such as LiF TLD-100 and LiF PTL-710. The high dose range gives the indication that the material could be suitable for both environmental and high radiation dosimetry.

The plot of marble integrated TL response against dose for the range 30.25 Gy up to 3.75 kGy as shown in Figure 2, predicts that this phosphor displays a linear TL- gamma dose relationship up to 1.45 kGy before supralinearity sets in. This relationship between TL intensity responses with gamma radiation dose was described using the second-degree polynomial:

$$TL_{\text{int}(marble)} = 8.035E - 06D^2 + 0.06083D + 7.26677 \quad \text{... ....}[3]$$

This observed linear relationship is a vital property of would be dosimeters. However, the 1.45kGy upper limit for linearity is high relative to the about 10 Gy limit for most common dosimeters. This is supportive of the earlier prediction that this phosphor portrays the potential for use for high dosimetric measurements. A further verification of this predicted linear TL - dose relationship for this rock carried out using the International Electrochemical Commission (IEC) stipulated condition that for linearity, the response of each detector among others, shall not vary from the conventional value by more than 10% (IEC, 1991) that is:

$$0.90 \le \frac{\overline{R}_i \pm l_i}{C_i} \le 1.10 \quad \text{.....}[4]$$

where l_i is the half-width of the confidence interval of $\overline{R}$ relative to a given set of measurements and is given by (IEC, 1991):

$$l_i = \frac{t_n s}{\sqrt{n}} \quad \text{[5]}$$

yielded an average value of 0.90. This value is greater than 0.81 but less than 1.10, the lower and upper recommended limits respectively. This further confirms that this material possesses a linear TL – gamma dose behaviour and therefore is a likely TL dosimetric material.

A homogeneity index f of 0.32 obtained in this work is approximately equal to the 0.3 upper limit recommended by IEC. It can therefore be adduced that samples of marble separately subjected to the same treatments of annealing and irradiation are capable of producing reproducible TL readouts and so qualifies for use as a TL dosimeter.
Figure 3 shows that this material exhibits exponential fading characteristics which is first fast, then gradual and almost constant after about one month post- irradiation storage. This feature is common to most TL detector materials. The minimum detectable dose (MDD) obtained for this phosphor is 0.03 kGy

CONCLUSION

From the results it is concluded that this natural rock has the potential for use as a TL dosimeter. It is however observed that its glow curve is associated with high dose range and that its linearity limit of 1.5 kGy is high compared to about 10 Gy of most common dosimeters. Thus it can be concluded that marble is a likely phosphor of choice for high dose measurements, such as is found in nuclear accidents and radiation processing of food and medical appliances.

Table 1: TL Responses of 10 Marble Phosphor Samples in a Batch

Sample ID	First Reading (AU)	Second Reading (AU)	Net Response (AU)
D01	1.06	0.0581	1.0019
D02	1.101	0.0897	1.0113
D03	1.232	0.0961	1.1359
D04	1.091	0.0869	1.0041
D05	1.004	0.0778	0.9262
D06	1.141	0.0731	1.0679
D07	0.996	0.0691	0.9269
D08	1.138	0.1256	1.0124
D09	1.036	0.0822	0.9538
D10	0.932	0.0727	0.8593

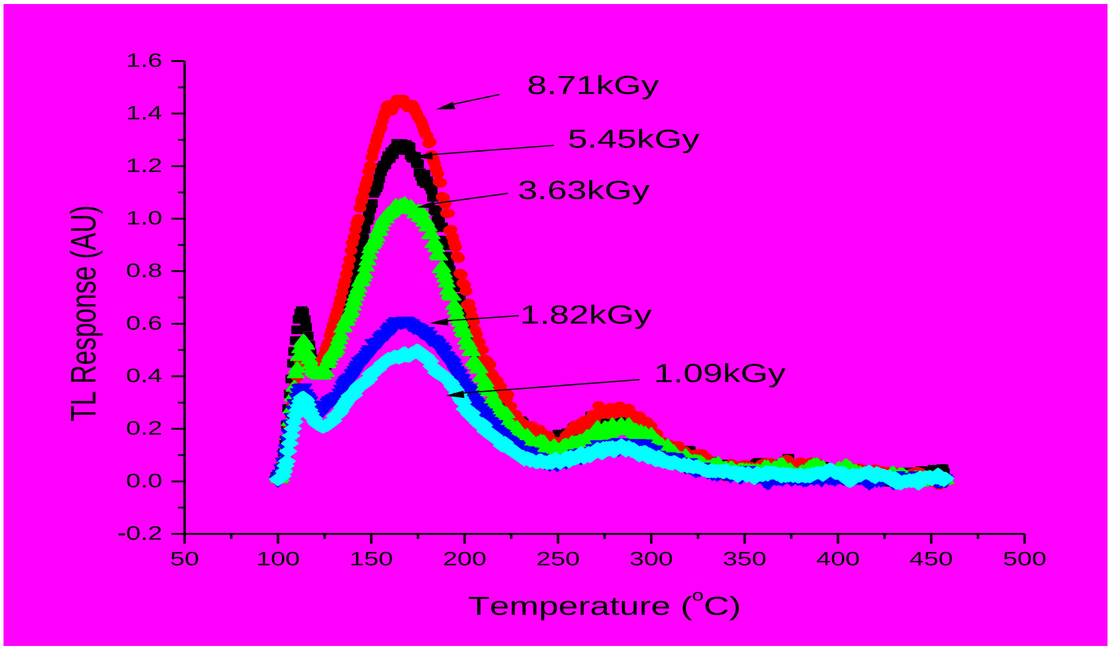

Fig. 1: Marble glow curves for different gamma radiation doses

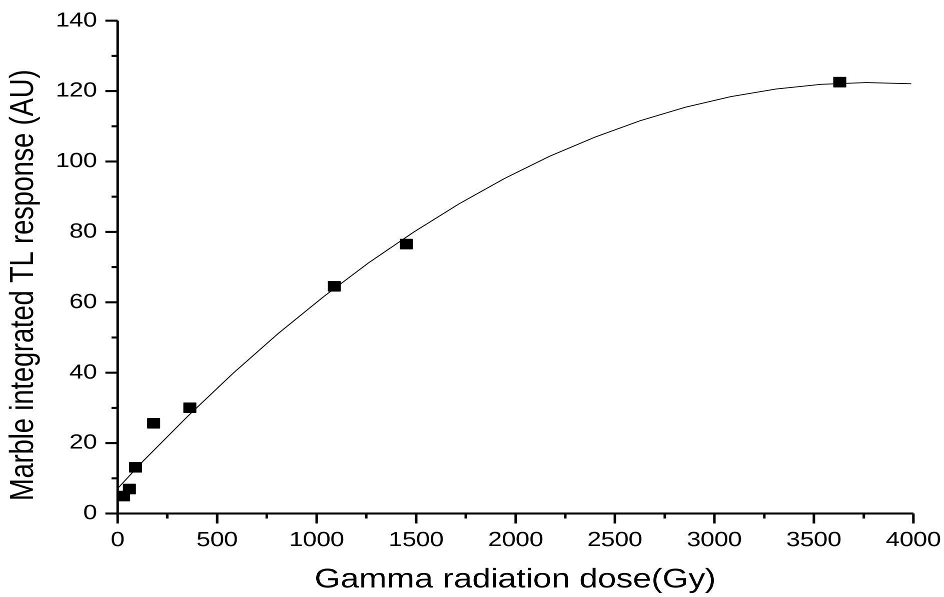

Fig. 2: Integrated TL-Gamma Dose Response for Marble

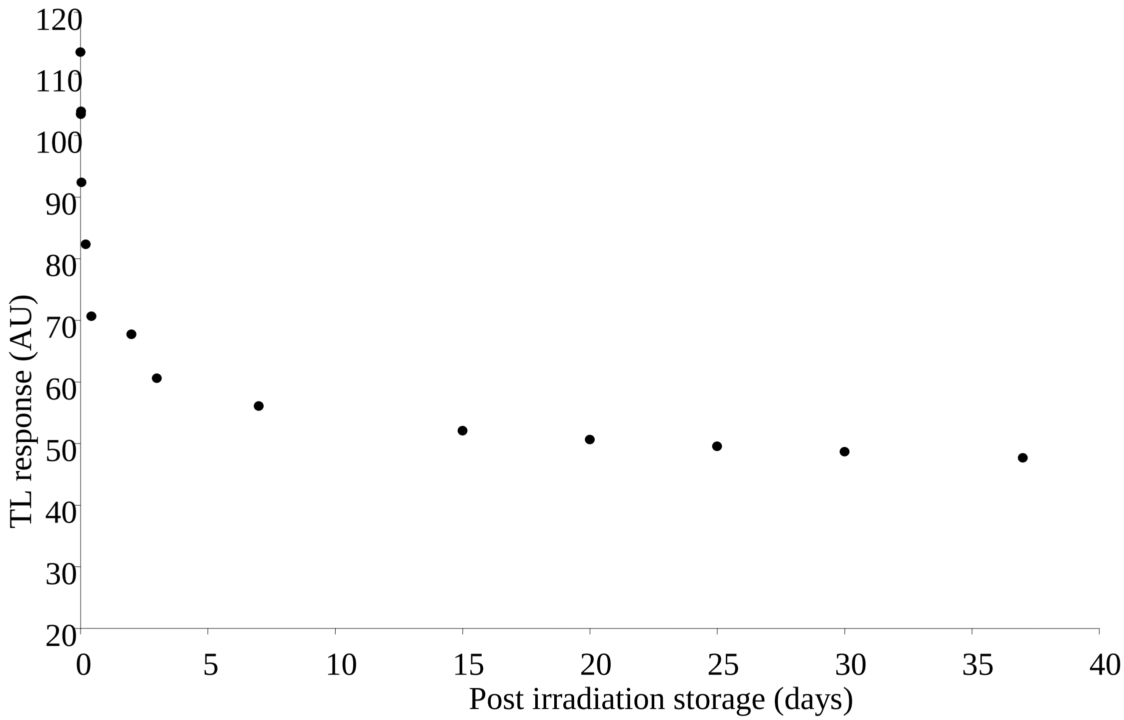

Fig. 3: Marble Fade with Post-irradiation Storage Time

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