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Optimizations of teflon embedded $CaSO_4$: Dy based TLD for environmental monitoring applications

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Abstract This study discusses the optimization of $CaSO_4$: Dy phosphor for its use in environmental monitoring program. In India environmental gamma radiation monitoring, using TLDs is being carried out since last four decades on regular basis. Investigations are made to determine optimum thickness of teflon disc and doping concentration. Teflon disc 0.8 mm thick was found optimum with the 0.2 mol% Dy concentration for highest sensitivity for environmental gamma radiation monitoring.

Keywords Environmental TLD · Gamma radiation · Dosimetry · Teflon disc

Introduction

Energy development plays an important role in the economic growth of nation. Nuclear power is environment friendly, technologically proven and economically competitive with the advantage of energy security. Besides this an important factor which must be taken into consideration is protection of environment around the operating nuclear power station. Department of atomic energy, since inception of Indian Nuclear Program, pays utmost attention to the safety and protection of radiation workers, public and environment. The greatest concern about the ionizing radiation stems from its potential to cause malignant disease in people exposed to it and inherited defects in later generations. However, radioactivity and radiations are universal phenomenon. Natural primordial radioactive elements such as Uranium, Thorium and their daughter products, potassium-40 occur not only in earth crust and environment around man but in man itself. These radionuclides significantly contribute to natural background radiation exposure to everyone throughout the world. Additionally other natural source of radiations and radionuclides is showers of cosmic radiation from outer space. The level of background radiation varies from place to place depending on concentration of primordial radioactivity in soil and altitude. The doses due to external gamma radiation arising from cosmic rays and primordial radio-nuclides and inhalation dose due to radon, thoron and their progenies are the two major components contributing nearly about 85 % of the total dose. The gamma rays are the most significant part of environmental dose due to its large range and deep penetrating power. Protection of the environment against radiation is given top priority in the development of Indian nuclear power program. Environmental gamma radiation monitoring in and around the environs of nuclear installations under Department of Atomic Energy (DAE), forms an important function of Health, Safety and Environment Group, BARC. This is essential not only for studying the long term trends of the radiation levels around the installations if any, but also for assuring the population at large about the negligible extent of the environmental impact due to the operation of nuclear facilities. The programme assures the public at large that the nuclear industry does not add to the radiation levels in the environs of the establishment. A variety of instruments like G.M. counters, high pressure ion chambers, portable and airborne gamma spectrometers and scintillation survey meters have been used for these measurements [1]. These instruments are all capable of recording only the transient fields at the time of measurement. The simple,

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inexpensive and precise measurement of small, integrated, external radiation doses is important, for studies of normal and increased radiation fields around nuclear installations. Thermoluminescent dosimeters (TLDs) are the best instruments to measure the dose and are widely used for radiation detection in the fields of environmental, medical, industrial and personal applications [2]. The advantages of passive detectors for environmental monitoring are that they are small, cheap, and reusable, do not require in situ electronic power supply and can be used in a large dose range. They provide measurements of the integrated dose during the time interval. The advantage of accumulated dose over the instantaneous dose is that it can be safely normalize to annual dose levels. Thermo luminescent dosimeters have found increasing use for personnel and environmental monitoring in many countries [3, 4]. Many inorganic compounds are known to exhibit thermo luminescence properties. Dosimeters have been fabricated in a number of shapes to suit specific applications. However, a TL phosphor for environmental monitoring should combine the dosimetric properties like high sensitivity as dose levels to be measured are low, resistance against disturbing environmental factors such as light, humidity etc., simple annealing procedure, TL spectrum of wavelengths in the wavelength region 300–500 mm in which the photomultiplier tube responds well, energy independent, linear dose response, isotropic in nature for directional independence, low fading rate, etc. These considerations limit the choice to only a handful of inorganic compounds, the important characteristics of which are described in literature [5–7]. Environmental gamma radiation monitoring, using TLDs around all the nuclear installations in India is being carried out using powder based CaF_2 dosimeters [8]. In order to cater to large scale requirement this system was labor intensive. Phosphor embedded teflon discs provides one of the key as it offers many advantages over loose powder handling. A study has compared the performance of $CaSO_4$: Dy, CaF_2: Dy and LiF: Mg, Cu, P for environmental gamma ray dosimetry and concluded that $CaSO_4$: Dy teflon TLD discs perform the best [9]. In their environments, CaF_2: Dy was found to exhibit relatively higher fading and LiF: Mg, Cu, P was found to give poor reproducibility. $CaSO_4$: Dy teflon TLD discs, has been found to be 35 times more sensitive than TLD 100 [10]. $CaSO_4$: Dy is widely used for Thermoluminescence dosimetry in the various fields of Radiation monitoring due to its better sensitivity, simple heat treatment and it can be easily prepared in laboratory at low cost [11–13]. All round efforts were made in order to use $CaSO_4$: Dy phosphor in environmental monitoring applications. The TL sensitivity of $CaSO_4$: Dy phosphor highly depends on grain size and the dopant concentration of Dy which act as activator and wavelength shifter. The TL sensitivities of particles smaller than 63 μm were only about the half of those at the higher particle sizes, while the activator concentrations remained almost constant [14]. To facilitate easy handling and to avoid weighing the phosphor for each measurement, dosimeters in the form of teflon discs were studied for dosimetric use. In order to meet the international standard requirements the American national Standards Institution (ANSI) [15], a suitable dosimeter was designed, provided with metallic filter aimed to compensate and assure a quasi constant response on the whole energy interval, 30 keV–3 MeV. A very sensitive, fully automatic TL reader to integrate the output of the photo multiplier for 30 s was chosen which covers the dosimetric peak [16]. The paper describes the main dosimeters technical performances and the method of characterization. The reported characteristics define the suitability of the system to be used in the survey of the environmental radioactivity in open areas.

Experimental

Procedure for material preparation and its measurement

$CaSO_4$ phosphors doped with Dy were prepared using the re-crystallization technique as reported by Yamashita et al. [13, 17]. Dy concentrations added were 0.05, 0.1, 0.2, 0.3, 0.5, 1.0 mol%. The samples were heated in a furnace at 650 °C for 2 hours. Finally the samples were again cleaned and sieved through 75–210 μm mesh size before use for TL measurement. For irradiation a ^{60}CO-γ- source with a dose rate 4 Gy/min was used. The gamma dose delivered was 1.33 Gy. For gamma irradiation, all the samples were kept inside a Bakelite capsule having 4 mm wall thickness. TL measurements were made with 4 mg of powder, 24 h after irradiation with a linear heating rate of 5.2 °C/sec. The standard deviation of TL measurements was within ±5 % for gamma irradiated samples.

Procedure to study performance of dosimeter on various teflon thicknesses

The TLD discs were made from a homogeneous mixture of $CaSO_4$: Dy powder and teflon resin taken in a weight ratio of 1:3. Discs of this mixture weighing 280 mg each were first cold pressed and then given a 400 °C h^{-1} heat treatment so that they become strong as well as flexible. $CaSO_4$: Dy phosphor embedded teflon discs of 0.1, 0.2, 0.5, 0.8, 1.2, 1.8 and 2.3 mm thickness and 13.3 mm dia were made using respective teflon tape. These discs were fixed in machine identifiable aluminum card. The set of twenty cards of each thickness were annealed at 250 °C for 3 hours and irradiated to various exposures by ^{137}Cs gamma radiation source. They were read off using automated TLD badge reader after 2 days, by integrating the output of the photo multiplier for 30 s, which covered the dosimetric peak at 210 °C only.

Characterization studies

The phosphor containing 0.2 % Dy was therefore used in the new dosimeters. The phosphor was mixed with teflon powder in the ratio of 1:3 and a discs of 13.3 mm diameter were made so that they can be read on PC based automatic TLD badge reader Two such discs were mounted on a machine identifiable TLD card enclosed in a plastic TLD cassette. This TLD was then subjected to further characterization studies for its application in environmental monitoring. It may be noted here that all the studies were carried out using freshly annealed TLD cards at 250 °C for 3 hours and read off with a delay of 2 days (time sufficient for low temperature TL signal to decay) after respective exposure.

Energy response

Due to the high effective atomic number of $CaSO_4$: Dy ($Z_{eff} = 15.3$) it shows significant over response for lower photon energies in the range of 25–200 keV in environmental gamma radiation energy spectrum. Without proper precaution, the TL response of the $CaSO_4$ phosphor based TLDs has been found to be higher for low energy X or gamma rays (<150 keV), by as much as ±65 % as compared to that for 660 keV. The American National Standards Institution (ANSI 2001) criteria recommends that the uncertainty due to photon energy dependence should not fall outside the limits of ±40 % over the entire range of energy for which dosimeter is intended to measure. Freshly annealed TLDs were divided in three sets, one without filter (bare card), and other with plastic cassette, and third with plastic cassette and 1 mm copper filter. They were given exposures of 20 mR using various energies of 24.85 keV (X-rays), 60 keV (^{241}Am), 65.9 keV (X-rays), 83.1 keV (X-rays), 117.6 keV (X-rays), 125 keV (X-rays), 223.9 keV (X-rays), 662 keV (^{137}Cs), and 1252 keV (^{60}Co). The exposures were performed, using the following facilities: a conventional X-ray machine (75–250 keV), ^{137}Cs source, ^{241}Am source, and ^{60}Co source in Radiation Safety System Division (RSSD), BARC. The effective photon energies for different kilo-voltages and filtration were obtained by means of HVT measurements. The exposure rate in air from the X-ray machine was determined using primary standard free air ionization chamber maintained at RSSD at BARC.

Detection limit

Sixty freshly annealed TLDs were taken and the background readings were recorded using the automatic TLD badge reader. Average background and the standard deviation were determined.

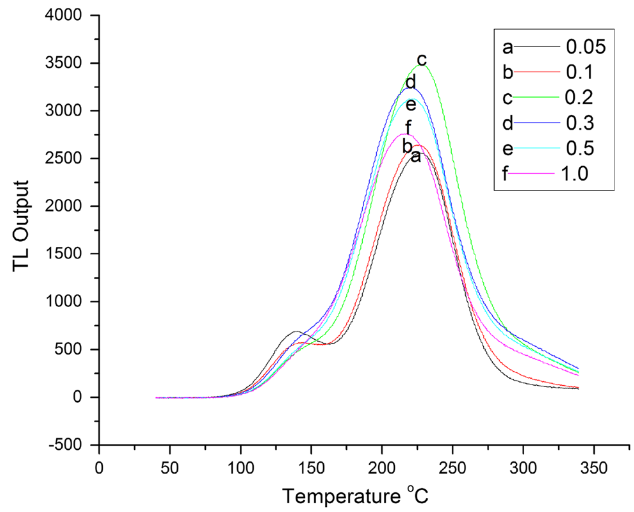

Fig. 1 Glow Curves of $CaSO_4$ doped with different Dysprosium concentrations (mole atom %)

Isotropy of response

In environment, as the radiation emanates from all the directions (4 π geometry), it is necessary that the response of a TLD is as isotropic as possible. As per the ANSI criteria, the uncertainty due to directional dependence, if the dosimeter assembly is rotated in free air around any axis during exposures of photons, should not be more than ±30 % from its response under static conditions at the angle of incidence used in calibration. The isotropic response of the TLDs was studied by exposing them to the calibration source at various angles viz. 0, 30, 45, 60, 90, 180, 270° and their response was normalized with that of normal incidence. TLDs were given an exposure at different angles of orientation by changing the orientation of TLDs with respect to source. The angular dependence of the badge about its vertical axis was studied for the exposure of 20mR of ^{137}Cs source.

Results and discussion

Optimization in dopant concentration

The effect of dopant concentration on the sensitivity of $CaSO_4$ has been investigated. Figure 1 shows the glow curves of $CaSO_4$: Dy with different dopant concentration. Figure 2 denotes the normalized TL output of $CaSO_4$: Dy with different dopant concentration. Figures 1 and 2 reflect the effect of gradual addition of dopant material on the TL-intensity of the prepared sample. It is clear from these

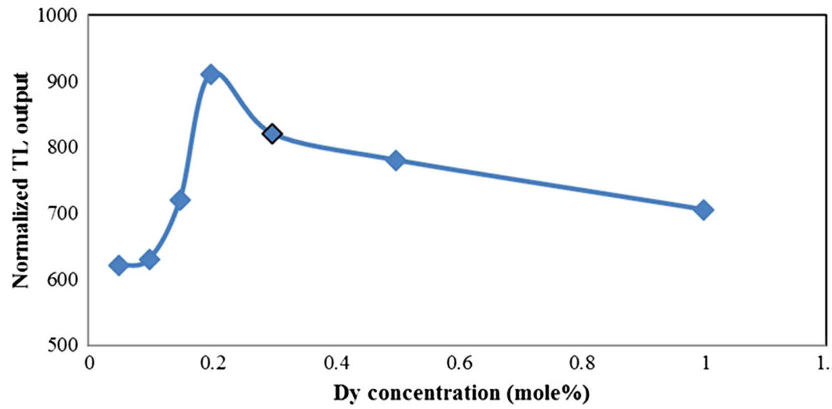

Fig. 2 Normalized TL output of $CaSO_4$ with different dopant (Dy) concentrations

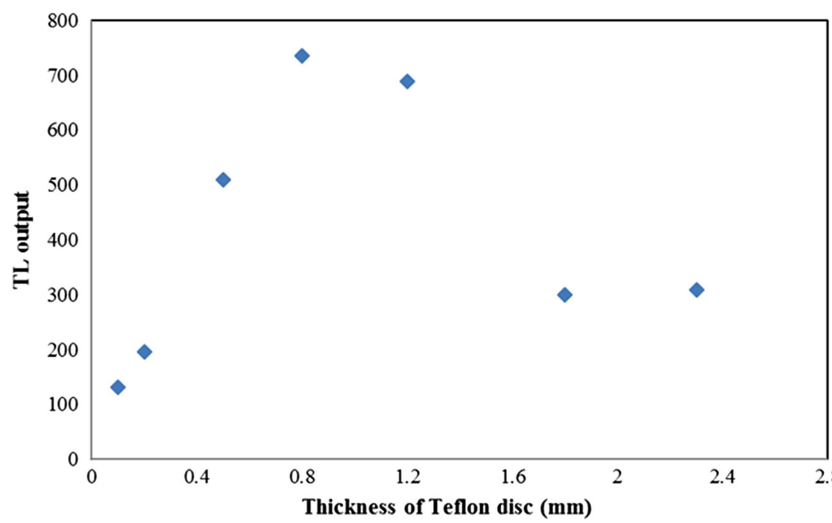

Fig. 3 Variation of TL response with respect to thickness of teflon discs

figures that 0.2 mol% of Dy is the optimum concentration that characterized by the maximum TL-intensity and after which a concentration quenching occurs. Hence $CaSO_4$: Dy with 0.2 mol% dopant concentration was found to be most suitable for environmental gamma radiation monitoring.

Optimization of thickness of teflon embedded discs

The variation of TL response with respect to thickness of teflon discs for 400 µGy gamma exposure is shown in Fig. 3. It is seen that response to gamma ray increases as thickness increases and after reaching a peak at 0.8 mm the increase in thickness results in response declined. The dose response curve drawn using the TLD output against the exposure imparted is as shown in Fig. 4. It is observed from the figure that 0.8 mm disc shows the linear dose response in the range up to 400 µGy, the range useful for routine environmental gamma radiation monitoring. It is clear from the Fig. 3 and Fig. 4 that teflon disc with 0.8 mm thick is the optimum thickness for highest sensitivity with linear dose response.

Optimization of filter element to achieve energy independence

Table 1 shows the photon energy response of the TLDs with and without filters, normalized to ^{137}Cs source. The normalized response thus plotted in Fig. 5. Due to high atomic number of $CaSO_4$: Dy TL phosphor (effective atomic no for $CaSO_4$: Dy is 15.3), bare cards (without any filter) show significant over response for lower photon energies (25–200 keV). Plastic filter alone does not seem to affect the over response. However flattening of energy dependence curve was achieved using 1 mm copper filter. The TLDs with copper filter of 1 mm thickness is seen to be sufficient to make the TLDs almost energy independent of the gamma/X-ray energy and thus it was quite suitable for the environmental gamma radiation monitoring.

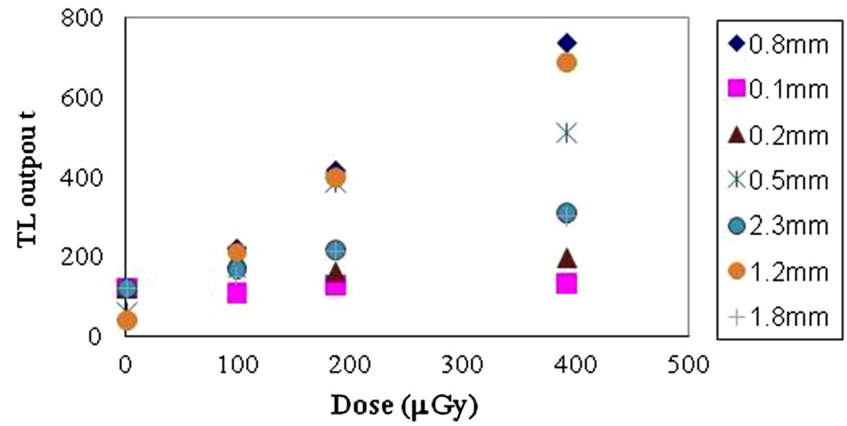

Fig. 4 Dose response curve for various thicknesses

Table 1 Results of studies on energy dependence of TLDs. The response is normalised with respect to that for 662 keV

Energy (KeV)	Normalized TL response			Energy (KeV)	Normalized TL response		
	Bare	Plastic	Plastic+Cu		Bare	Plastic	Plastic+Cu
24.85	7.97	8.35	0.17	125	1.43	1.41	1.27
60	5.81	5.95	1.12	223.9	1.33	1.32	1.06
65.9	3.79	3.65	0.93	662	1.00	1.00	1.00
83.1	2.46	2.48	1.35	1252	1.09	0.99	0.74
117.6	1.13	1.20	1.02				

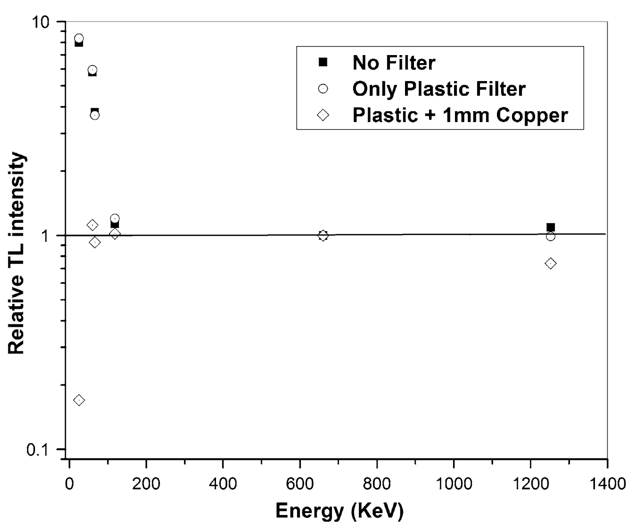

Fig. 5 Normalized response of three sets of TLDs against the various X-ray/gamma radiation energies

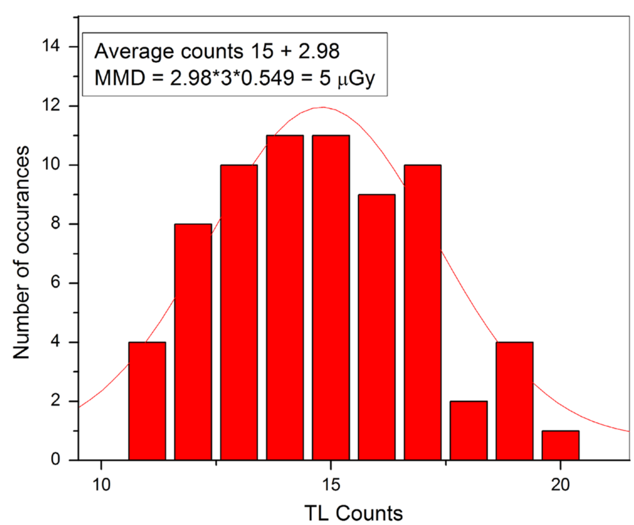

Fig. 6 Distribution of background TL signal of 77 freshly annealed TLDs. 90 % of the TLDs are seen to have the background between 15 ± 3. The standard deviation of 2.98 give the LDD of 5 μGy

Lowest detectable dose (LDD) achieved by optimization

Background reading of the sixty freshly annealed TLDs taken showed the average to be 15 ± 2.98 counts. Taking three times the standard deviation of readings corresponding to the zero dose, and multiplying it by the calibration factor. The LDD was found to be 5 μGy (Fig. 6).

Accomplishment of isotropic response as per ANSI criteria

The angular dependence of the TLDs, normalized to perpendicular incidence, using ^{137}Cs source is shown in Fig. 7. It can be seen, from the figure, that the response at

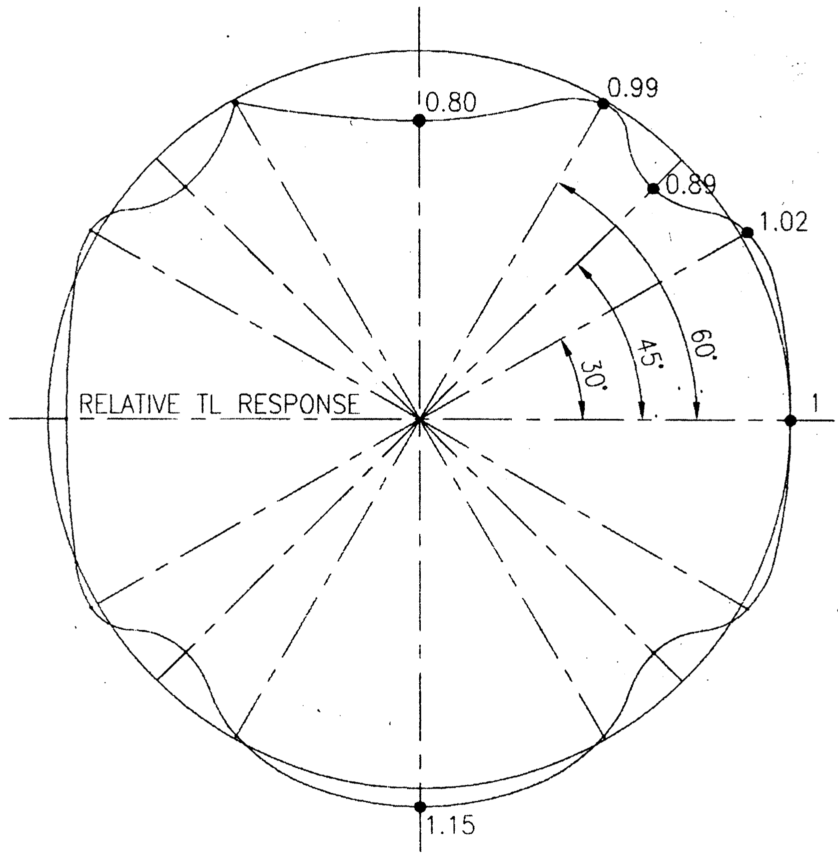

Fig. 7 Angular dependence of the TLD. TL response for all the TLDs was observed to be within 10 % of the normal incidence value excluding 90° angle

all the angles of incidence is very much within 30 % of the normal incidence and hence fulfills the ANSI criteria. It may be remembered here that the work was carried out using a calibration source that can be taken as a point source. Small variation in the distance between source and the individual disc can matter in the response in the laboratory experiment whereas the exposure in environment is from a infinite source and as such the variation in response with respect to the true value is expected to be much smaller. The TLDs thus can be said to be isotropic in response.

Accuracy and precision obtained after optimization

The calibration factor obtained as discussed earlier was used to evaluate the TLD readings recorded from the other set of TLDs exposed to another source. Accuracy and precision were then evaluated from the readings (Table 2). American National Standard Institute criteria for environmental TL dosimeters (ANSI N545) specify that the field results should be within 30 % of the delivered values. It is seen from the table that the accuracy of all the values is well within the limit specified in ANSI 545. The precision was found to be about 5 % of the delivered value.

Table 2 Determination of accuracy and precision of the TLD readings

Delivered Dose (μGy)	Observed dose (μGy)	Precision %	Accuracy %
123 ± 1.2	123 ± 4.3	4	0.5
184 ± 2.3	175 ± 8.3	5	4.7
246 ± 2.7	231 ± 4.9	5	5.7

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