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PREPARATION OF CaF_2:Dy CHIPS AS THERMOLUMINESCENT DOSIMETERS FOR ENVIRONMENTAL MEASUREMENTS USING A HARSHAW 2080 TL PICOPROCESSOR

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ABSTRACT

This work presents the necessary steps to prepare the dosimeters for application in environmental measurements. The material used was calcium fluoride dopped with dysprosium (CaF_2: Dy), commercially supplied by Harshaw as TLD-200. It were carried out four initial irradiations (Co-60 source gamma – 0.8mGy from Dosimetric Material Laboratory LMD/GMP) in a new batch of TLD-200 dosimeters to achieve a stable response and four more irradiations to separate them, according to their response range. A batch of pre-selected 197 dosimeters from 263 was used to finally obtained two batches of 99 (9.2±0.7nC) and 93 (10.5±0.6nC) dosimeters. A calibration curve was prepared relating the thermoluminescent response of the phosphor and the radiation exposure (0 to 600µGy), obtained by irradiation with Co-60 source of 4Π geometry and measurements using a Harshaw Picoprocessor 2080 TL equipment. Repeatability of the measurements was determined for 100µGy (CV= 5.09%) and 300µGy (CV=4.18%) and the minimum detectable dose was evaluated as 2µGy. The fading was determined by irradiating the dosimeters with 0.8mGy and reading then after 1, 5, 7, 8, 15, 29, 50, 70 and 90 days.

1. INTRODUCTION

Most of the ionizing radiation when absorbed by matter goes into heat, while a small fraction breaks chemical bonds. In some materials, part of the absorbed energy is stored in metastable states and can be recovered later as visible photons when these materials are heated.[1] Such phenomenon is called thermoluminescence of solids. This phenomenon can be explained by the model of bands in ionic solids. In this model, the electrons are in the valence band, the conduction band is empty and there is a gap of energy that separates them. When a crystal is exposed to ionizing radiation, energy is transferred to an electron located at the valence band that goes to the excited state (conduction band), leaving a hole in the valence band. The electron and the hole move until they are trapped in a metastable state (forbidden band), see Figure 1.[1-4] These metastable states are presumed to be associate with defects in the crystal, as impurity sites. When the crystal is heated, the electron receives enough energy to rise to the conduction band and moves until its recombination with a trapped hole and a thermoluminescent photon is emitted.[3]

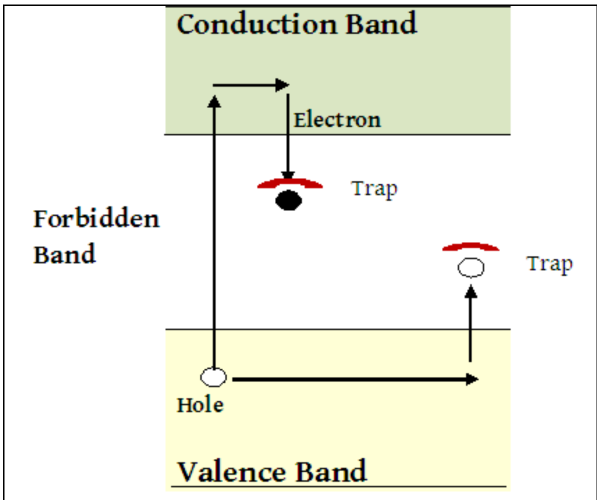

Figure 1: Exposition to radiation[1]

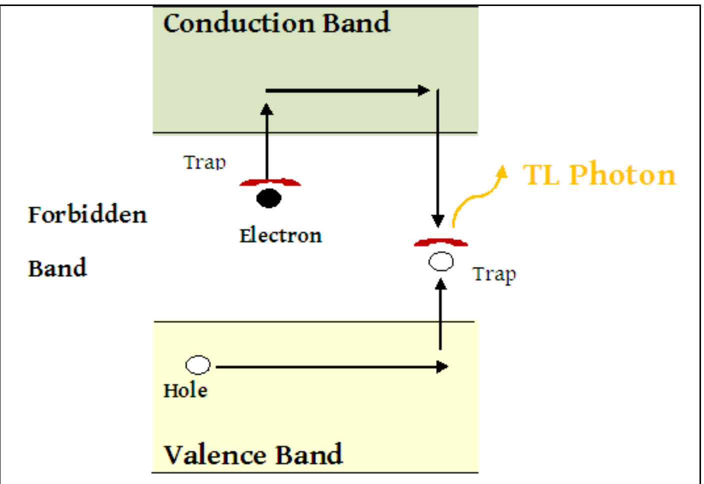

Figure 2: Heating[1]

The intensity of the emitted photons (thermoluminescence intensity) is proportional to the radiation absorbed by the material (called phosphor)[2] In this way, it is possible to apply this property to measure the radiation dose to which the phosphor has been exposed.[3]

The environmental dosimetry is the measurement of radioactivity from natural sources, as cosmic rays, and produced by man. It can be used in the surroundings of nuclear and radioactive facilities, in order to monitor the release of radioactive gases and effluents of these locations. Its purpose is to measure the radiation dose received by the public, who live or work near the installation.[2,4]

The crystal used for environmental dosimetry needs to have high sensitivity, because the measured doses are lower, compared with the personal dosimetry, and high heat, optical and moisture resistance, because the measurements periods, which the dosimeter remains exposed in the open areas, are much longer.[2] The material used in this work was the CaF_2: Dy, commercially supplied as Harshaw TLD-200, which has the required sensitivity but high optical fading and should thus be handled in the dark.

The purpose of this work was to prepare the dosimeters to be used in environmental measurements and for this were carried out the following steps: 1) irradiation of a new batch of dosimeters TLD-200 (CaF_2: Dy) to achieve a stable response and separate them according to the response range; 2) obtaining a calibration curve relating the response of the thermoluminescent crystals with the exposure dose (irradiation in the Co-60 source) using an equipment Harshaw Picoprocessor 2080 TL; 3) acquisition of the crystal fading factor as function of time and 4) determination of the measurements repeatability and the minimum detectable dose.

2. MATERIALS AND METHODS

The thermoluminescent material used for dosimetric purposes has to receive a previous treatment called annealing. This preparation has the function of stabilizing the structure of the crystal traps and eliminates any residual energy of previous irradiation. It is performed by heat treatment that, in general, consists of a heating at high temperature, then again at low

temperature to increase the sensitivity and to reduce signal loss due to thermal or optical fading[5].
Annealing is divided into:
• **Initialisation treatment**: it is used for new TL dosimeters or for those ones which have not been used for a long time. The aim of this procedure is to stabilize the trap levels, in order to maintain constant the intrinsic background and the sensitivity during subsequent utilization. The parameters (time and temperature) are the same as Standard Annealing or Pre-irradiation annealing[5].
• **Standard Annealing (pre-irradiation annealing)**: it is used to eliminate any residual irradiation effect which may remains in the dosimeters after the readings. It is carried out before using the crystals and has the aim to achieve the initial trap levels.[5]
• P**re-readout Annealing**: it is used to eliminate low temperature peaks from glow curve, since they are propitious to fade causing errors in the measurements.[5]

Following the annealing used in this work[5-8]:

Annealing	**Treatment carried out in the dark**
Initialisation	1- Furnace: 400°C - 1h withdrawing directly to oven. (A) 2-Oven: 100°C - 2h 3- Cooling on a stone Both furnace and oven are in the stable temperature before entrance of the crystals. 4-Cooling in a desiccator
Pre-irradiation	Same as Initialisation
Pre-readout	Oven: 100°C - 10 minutes

Note: (A)- Furetta[5] suggests a rapid cool down after the furnace, on a cold metal block, to increase sensitivity in a reproducible and unchanged procedure. However in this work it was easier to keep reproducibility removing the crystals from furnace directly to the oven.
The readings were carried out one day after the irradiation, using a Harshaw 2080 TL Picoprocessor, in the following conditions:

Parameters	**Set-up**
Tension	620V
Pre-heating	100°C
Heating time	30 s
Acquisition time	40 s
Final Temperature	290°C
Heating rate	8°C/s

3. RESULTS

3.1. Separation of the crystals

The crystals which received the annealing process, were put between two 3mm thickness acrylic plates, to ensure the electronic equilibrium and sealed with a black plastic. The irradiation (0.8mGy) was carried out with a Co-60 source of 4Π geometry (LDT-IPEN). This procedure was repeated 4 times to assure better reproducibility and then more four times, but now with readouts one day before irradiation to separate the crystals in batches with similar response. The results are show in Table 1 and the Histogram in the Figure 3.

Table 1: Separation of crystals in batches – 1th Irradiation

Readings range (nC)	Number of crystals	%
7.75 – 8.25	1	0.4
8.25 – 8.75	1	0.4
8.75 – 9.25	10	3.8
9.25 – 9.75	22	8.4
9.75 – 10.25	41	15.6
10.25 – 10.75	35	13.3
10.75 – 11.25	53	20.2
11.25 – 11.75	46	17.5
11.75 – 12.25	26	9.9
12.25 – 12.75	15	5.7
12.75 – 13.25	8	3.0
13.25 – 13.75	3	1.1
13.75 – 14.25	1	0.4
19.25 – 19.75	1	0.4
Total	263	100

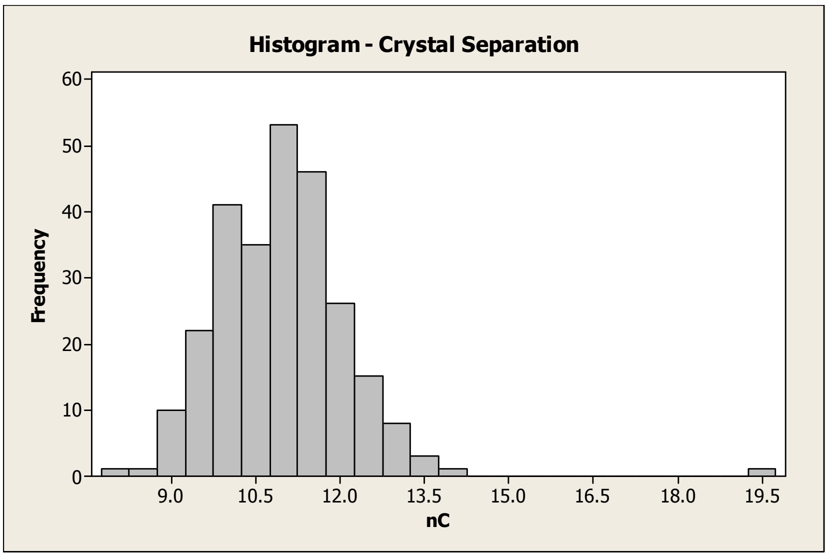

Figure 3: Histogram (1th Irradiation) for crystal separation

It was carried out three more irradiations and the final separation was:

Batch 1: 99 crystals – range 8.5 to 9.9 nC (9.2±0.7nC)

Batch 2: 93 crystals – range 9.9 to 11.1 nC (10.5±0.6nC)

3.2. Calibration Curve

Twenty six dosimeters were irradiated (two per dose) with three crystals each in a Co-60 source of 4Π geometry (LDT-IPEN). The results are shown in Table 2 and Figure 4.

Table 2: Results for calibration curve preparation

Theoretical Dose (μGy)	Exposition time (hh:mm:ss)	Real Dose (μGy)	Average readings (nC)	Standard Deviation (nC)
0	00:00:00	0.00	0.128	0.017
10	00:00:26	9.43	0.310	0.082
50	00:02:06	45.70	0.814	0.031
80	00:03:21	72.90	1.028	0.011
100	00:04:12	91.40	1.370	0.127
150	00:06:18	137.09	1.914	0.048
200	00:08:23	182.43	2.508	0.113
250	00:10:30	228.49	3.136	0.062
300	00:13:55	299.94	3.568	0.149
350	00:14:41	319.52	3.760	0.011
400	00:16:48	365.58	4.728	0.124
450	00:18:53	410.92	5.304	0.001
500	00:23:12	499.83	6.240	0.209
600	00:27:52	600.4	7.241	0.262

The dosimeter with 0μGy dose wasn't irradiated and the curve was drawn by subtracting the zero value from the doses readings.

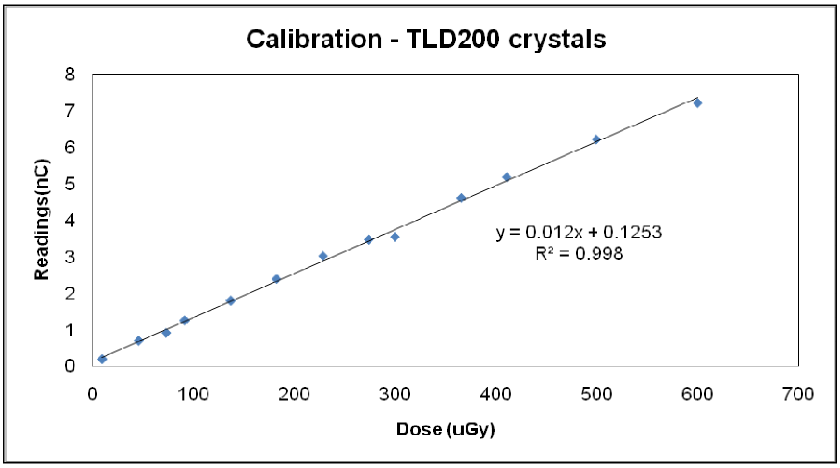

Figure 4: TLD 200 Calibration Curve

3.3. Repeatability

The repeatability was determined using a group of dosimeters from the same batch in which were applied two different doses (100 and 300µGy). The results are shown in Table 3.

Table 3: Repeatability determination

Readings	Dose 100µGy (nC)	Dose 300µGy (nC)
1	1.172	3.296
2	1.212	3.596
3	1.168	3.376
4	1.092	3.572
5	1.216	3.496
6	1.112	3.828
7	1.124	3.556
8	1.172	3.340
9	1.168	3.756
10	1.308	3.568
11	1.216	3.580
12	1.104	3.600
13	1.112	3.364
14	1.220	3.644
15	1.092	3.732
16	1.156	3.696
17	1.232	3.684
18	1.232	3.544
Average	**1.17**	**3.57**
Standard Deviation	**0.06**	**0.15**
Coefficient of Variation (CV)%	**5.09**	**4.18**

The results obtained for the coefficient of variation (CV) at doses of 100 and 300μGy may be considered acceptable since they are at the limit of CV ≤ 5%[5]

3.4. Minimum detectable dose

The zero dose (L_0) depends on the photomultiplier characteristics, the thermoluminescent material properties, annealing and previous utilization of the dosimeters. L_0 is obtained by the sum of equipment reading without the crystal (l_0) and non-irradiated crystal (l_1), as shown in Euation 1.[9]

$$L_0 = l_0 + l_1 \quad (1)$$

The minimum detectable dose (D_0) can be calculated as three times the standard deviation of zero dose (nC) which was changed to μGy using calibration curve[9] The results to l_1 and l_0 are shown in Table 4.

Table 4: Readings of non-irradiated crystals (l_1) and (l_0) equipment without the crystal

Readings	**l_1 (nC)**	**l_0 (nC)**
1	0.140	0.064
2	0.116	0.028
3	0.032	0.012
4	0.032	0.016
5	0.032	0.008
6	0.032	0.004
7	0.040	0.008
8	0.040	0.000
Average	**0.058**	0.008
Standard Deviation	**0.044**	**0.021**

The zero dose standard deviation was calculated as shown in Equation 2[10], using de values s_{l0} = 0.021nC and s_{l1} = 0.044nC.

$$s_{L0} = (s_{l0}^2 + s_{l1}^2)^{1/2} \quad (2)$$

The result of 3*s_{L0} is 0.15nC, which leads to a minimum dose of 2μgGy by using the calibration curve [Reading (nC) = 0.012*dose(μGy) + 0.1253].

3.5. Residual dose

After the annealing procedure the readings of the crystals were carried out to verify if any residual dose remains. The results are shown in Table 5.

Table 5: Residual dose determination

Readings	(nC)
1	0.016
2	0.004
3	0.004
4	0.000
5	0.012
6	0.008
7	0.000
8	0.000
9	0.012
10	0.012
11	0.040
12	0.028
13	0.024
14	0.004
15	0.020
16	0.016
17	0.016
18	0.024
19	0.040
20	0.012
Average	0.015

Both the average value and individual values are below the minimum detectable dose, when the measure unit was changed to µGy using calibration curve.

3.6. Fading

The term fading can be described as the spontaneous release of charge carriers (holes and electrons) from their traps with consequent emission of light by the dosimeter. The main responsible factor for this phenomenon is the temperature increase that causes the emptying of shallow traps. For this reason the pre-readout annealing is applied in order to avoid readings errors.[9]

Another phenomenon that must be considered is the optical fading caused by light exposition (solar or artificial). In this case the electrons receive enough energy to escape from the traps.[9] In order to avoid this effect the dosimeters are wrapped in dark plastic.

While the dosimeter remains exposed in the open, two effects compete: the background radiation that creates traps and the fading caused by temperature.[5] In order to evaluate this fading 33 dosimeters (300μGy) were irradiated and the readings were carried out as following in Table 6 and the results are shown in Table 7. The calculation was performed considering the first day reading as 100%.

Table 6: Fading determination

Readings	Interval irradiation - reading (days)	Number of crystals	Number of non-irradiated crystals
1	1	3	3
2	5	3	3
3	6	3	3
4	7	3	3
5	8	3	3
6	15	3	3
7	29	3	3
8	50	3	3
9	70	3	3
10	90	6	6

Table 7: Results of fading determination (average of 3 readings)

Day	Irradiated crystal reading (nC) **(I)**	Non- Irradiated crystal reading (nC) **(NI)**	(I) – (NI) (nC)	Fading (%)
1	5.347	0.057	5.289	--
5	5.421	0.164	5.257	0.6
7	5.216	0.180	5.036	4.8
8	5.319	0.221	5.097	3.6
15	5.317	0.468	4.849	8.3
29	5.268	0.796	4.472	15.4
50	6.063	1.537	4.525	14.4
70	6.561	1.883	4.679	11.5
90	6.615	2.532	4.083	22.8

4. CONCLUSIONS

- The new batch of TLD-200 dosimeters achieved a stable response after eight irradiations and they were separated, according to their response range, in two batches: of 99 chips (9.2±0.7nC) and 93 chips (10.5±0.6nC).
- The calibration curve relating the thermoluminescent response of the phosphor and the radiation exposure (0 to 600µGy), obtained by irradiation with Co-60 source, reached a correlation of R^2=0.998, which is acceptable for the aim of this work.
- Repeatability of the measurements was determined for 100µGy (CV= 5.09%) and 300µGy (CV=4.18%). They are at the limit of CV ≤ 5% and thus be considered acceptable.
- The minimum detectable dose was evaluated as 2µGy.
- The residual dose after annealing was lower than minimum detectable dose and the fading after 90 days was 22.8 %

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