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VALIDATION OF THE STAMPING METHOD FOR $CaSO_4$:RE + TEFLON® PELLETS PRODUCTION

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ABSTRACT

The IPEN method for the $CaSO_4$:RE + Teflon® production, developed and patented at the Dosimetric Materials Laboratory – LMD/IPEN in the earlier 1980's, [1] is highly time-demanding, so that the use of the stamping method, already widely industrially applied, would enhance the $CaSO_4$:RE + Teflon® pellets production. Thus, validating the stamping method, by comparing the dosimetric properties of a batch of pellets produced by each method, became a must. The stamped batch presents the same mean non-irradiated signals either after sintering or annealing while IPEN batch mean non-irradiated signals vary in 23%. The mean TL signal after irradiation was about 50 nC, but the standard deviation varies from 20% to 33% for IPEN batch and keeps in 10% for the stamped batch. 24 h after the irradiation, the TL signal decreased to about 35 nC, with no differences in IPEN batch standard deviation and a decrease to 5% in stamped batch standard deviation, for the five performed essays. Calibration curves present a linear behavior over the entire studied dose range and the same coefficients for both methods, however, the uncertainties in the coefficients determined to the calibration curve obtained with stamped pellets are significantly smaller, leading to a more precise dose determination. This results show that the stamping method produces more homogeneous batches, with pellets that maintain the dosimetric characteristics of the detectors produced by IPEN method, in such a way that the stamping method can substitute with advantages the IPEN method in the $CaSO_4$:RE + Teflon® dosimetric pellets production.

1. INTRODUCTION

Thermoluminescent materials have been applied in the radiation detection since the beginning of the nuclear tests, and its success, especially in the individual and environmental monitoring, [2] led to an intense development of the thermoluminescent dosimetry during the last three decades. The Instituto de Pesquisas Energéticas e Nucleares (IPEN-CNEN/SP) has actively contributed to it mainly through the Laboratório de Materiais Dosimétricos of the Gerência de Metrologia das Radiações, (LMD-GMR) with the development of thermoluminescent detectors based on the calcium sulphate activated with dysprosium ($CaSO_4$:Dy) using the free-flow Teflon® as the binding material destined to the X and gamma radiations dosimetry [3, 4] and to the beta dosimetry. [4, 5, 6]

All these varieties of detectors have been produced through the IPEN patented method, which consists in single weighing the mass of the chosen mixture that will constitute a single pellet after pressuring and then sintering them, and demands the efforts of a highly skilled worker for about a week in order to produce a batch of 200 pellets.

On the other hand, the stamping method consists in industrially producing a film with the desired thickness of the chosen mixture before stamping it with a cutter of appropriate size and shape to produce the pellets that will be later sintered, in a process able to produce up to 200.000 pellets in a day, depending on the available machinery.

Aiming to make the $CaSO_4$:RE + Teflon® pellets production a less time-demanding process, the dosimetric properties of a batch of 200 $CaSO_4$:Dy + Teflon® pellets produced by the stamping method were compared to the ones of a batch of 200 pellets of the well-known $CaSO_4$:Dy + Teflon® detectors produced by the IPEN method, with the same $CaSO_4$:Dy and free-flow Teflon® samples being used in the production of each batch, so that all pellets consist of the same 1:2 $CaSO_4$:Dy and free-flow Teflon® mixture weighing 50 mg shaped in a 6 mm of diameter and 0.8 mm of thickness disk.

2. $CaSO_4$:Dy + TEFLON® PELLETS PROPERTIES

Some dosimetric properties such as the mean non-irradiated signal after sintering and annealing, the mean TL signal after irradiation, the mean TL signal 24 h after the irradiation, the TL signal repeatability, the TL signal reproducibility and the intrinsic efficiency of the detectors are known to be related to their physical presentations and to significantly alter the calibration curve of the dosimeters. For this reason, the mentioned dosimetric properties of the $CaSO_4$:Dy + Teflon® batch of 200 pellets produced by the stamping method were compared to the properties of the batch of 200 pellets produced by the IPEN method.

A metrologically traceable ^{60}Co gamma radiation source of 4π geometry was used for every irradiation, held at 23 cm of the source in air and electronic equilibrium conditions, an automatic thermoluminescence reader model 5500 from Thermo/Harshaw performed every TL signal measurement and the graphical analysis software OriginPro 8 was applied to determine the desired parameters.

2.1. Mean Non-irradiated TL Signal After Sintering

The batch of 200 pellets was kept inside a lead shielding during its 1 h transportation from the production laboratory to the Laboratory of Thermoluminescent Dosimery (LDT) where it was read, being kept shielded for infrared (IR), visible (V) and ultraviolet (UV) lights while the readout, which lasts 4 h, was performed.

While non-irradiated TL signals after sintering of IPEN pellets, shown on the left in Fig. 1, vary from 0.0780 (10) nC to 0.4290 (10) nC, the non-irradiated TL signals after sintering of stamped pellets, on the right side in Fig 1, vary from 0.0389 (10) nC to 0.0494 (10) nC. The mean non-irradiated TL signal after sintering, TL_0, of 0.19 (4) nC and 0.0443 (23) nC of IPEN and stamped batches, respectively, and the region defined by its dispersion, $TL_0 \pm \sigma_{TL0}$, are also presented in Fig 1.

These results suggest both a more homogeneous distribution of the TL material through the batch and a lower contribution of triboluminescence signal generated by the occupation of high energy depth traps during the batch production in the stamping method.

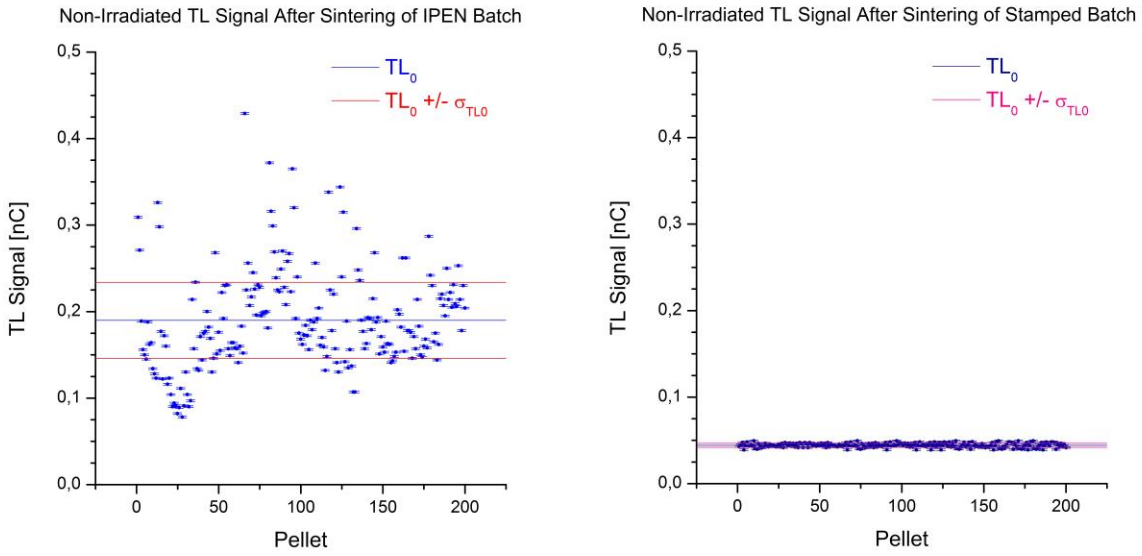

Figure 1. Non-irradiated TL signals after sintering of IPEN, on the left, and stamped, on the right, batches of $CaSO_4$:Dy + Teflon® pellets.

2.2. Mean Non-irradiated TL Signal After Annealing

The pellets were irradiated with a ^{60}Co gamma radiation pre-dose of 10 mGy without undergoing a readout cycle, annealed at 300°C per 3 h using a resistance heated furnace model 3/550PD (Vulcan) and once again kept inside a lead shielding during 1 h and shielded for IR, V and UV lights during the 4 h readout performance, in order to reproduce the readout conditions of the non-irradiated TL signal after sintering essay. Non-irradiated TL signal after annealing was repeated five times, presenting no significant differences in mean non-irradiated TL signal after annealing, TL_{NIi}, and in its standard deviation, σ_{TLNIi}, as presented in Tab. 1. The average mean non-irradiated TL signal after annealing, TL_{NI}, and the mean non-irradiated TL signal after sintering, TL_0, are also presented in Tab 1 for comparison.

Table 1. Mean non-irradiated TL signal after annealing

	IPEN batch		Stamped batch	
Essay	TL_{NIi} [nC]	σ_{TLNIi} [nC]	TL_{NIi} [nC]	σ_{TLNIi} [nC]
1	0.044	0.010	0.0441	0.0023
2	0.045	0.010	0.0439	0.0023
3	0.042	0.010	0.0444	0.0023
4	0.046	0.010	0.0436	0.0023
5	0.043	0.010	0.0440	0.0023
TL_{NI}	0.0440	0.0012	0.04400	0.00020
TL_0	0.19	0.04	0.0443	0.0023

Although there are no differences in the non-irradiated signals of stamped batch, the mean non-irradiated TL signal after sintering of IPEN batch is 23% greater that its mean non-irradiated TL signal after annealing, reinforcing the suggestion that triboluminescence contribution is significant in the composition of the IPEN pellets TL_0 signal.

The suggestion of a more homogeneous distribution of the TL material through the batch in the stamping method is also reinforced, by the smaller dispersion of the stamped batch signals in comparison to the dispersion of the IPEN batch signals. The graphical comparison between TL_0 and TL_{NI1} for IPEN and stamped batches, respectively, presented in Fig. 2 help to clarify these points.

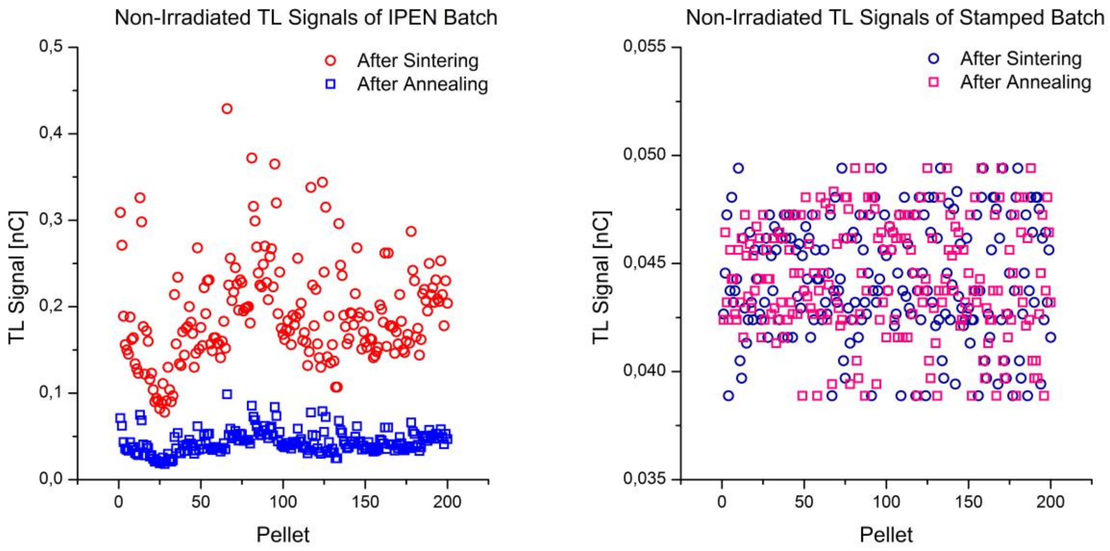

Figure 2. Non-irradiated TL signals after sintering and after annealing of IPEN and stamped batches.

2.3. Mean TL Signal Measured after a ^{60}Co Gamma Irradiation

In order to measure the mean TL signal of the batches right after their irradiation, the pellets were irradiated with 10 mGy and kept shielded for IR, V and UV lights during the 4 h readout performance. The mean TL signal obtained in each one of the five repetitions of this essay and its standard deviation, together the average TL signal measured after the irradiation are presented in Tab 2. The results obtained for the same readout cycle performed 24 h after the irradiation of the batches are presented in Tab 3.

Once again, the dispersion of the TL signal of IPEN batch is greater than the one of the TL signal of stamped batch, being in the range of 20% to 33% against 10% in every essay for the readouts performed right after the irradiation and in the same range against 5% in the readouts performed 24 h after the irradiations.

Table 2. Mean TL signal measured right after a ^{60}Co gamma irradiation

	IPEN batch		Stamped batch	
Essay	TL [nC]	σ_{TL} [%]	TL [nC]	σ_{TL} [%]
1	51	20	49.9	10
2	47	33	50.2	10
3	49	25	50.7	10
4	51	27	49.7	10
5	52	30	49.5	10
Average	50.0	3	50.0	0.7

Table 3. Mean TL signal measured 24 h after a ^{60}Co gamma irradiation

	IPEN batch		Stamped batch	
Essay	TL [nC]	σ_{TL} [%]	TL [nC]	σ_{TL} [%]
1	34	31	34.9	5
2	38	32	35.6	5
3	35	33	34.8	5
4	32	20	35.3	5
5	36	26	34.5	5
Average	35.0	5	35.0	1.0

2.5. TL Signal Repeatability and Reproducibility

The TL signals measured with the same pellet in the five essays present the same pattern than the mean TL signals determined for the whole batch, with the same relative dispersion; thus, the TL signal repeatability, considering a 95% confidence interval, is simply three times the relative dispersion associated to the average value of the mean TL signal calculated for every essay, also the dispersion associated to the average of the TL signal measured with every pellet. The repeatability of the TL signal measured Δt hours after a ^{60}Co gamma irradiation with pellets from IPEN and stamped batches are presented in Tab 4.

As every pellet can be considered a different body of proof, the reproducibility of the TL signals measured with the pellets of a batch is the 95% confidence interval of the distribution of the individual signals read with every pellet around the mean TL signal. In this case, the wider distribution was considered to determine the reproducibility presented in Tab 4.

Table 4. TL signal Repeatability and Reproducibility

	Repeatability [%]		Reproducibility [%]	
Δt [h]	IPEN batch	Stamped batch	IPEN batch	Stamped batch
0 – 4	9.0	2.1	99	30
24 – 28	15	3	99	15

This shows that the pellets produced, specially by IPEN method, cannot be considered identical for dosimetric purposes, so, selecting the identical pellets is needed, dividing the batch produced together in different new batches, so that the pellets can be applied as identical thermoluminescent detectors.

In the present study, this selection was performed in such a way that the reproducibility of the TL signal of the dosimetric batch is of 5% for the pellets produced by IPEN method and of 5% for the readout performed right after the irradiation for the pellets produced by the stamping method. In both cases, the intrinsic efficiency of the pellets to the ^{60}Co gamma radiation is of 0.0139 (15)%.

3. CALIBRATION CURVES TO ^{60}Co GAMMA RADIATION

As the physical presentation alters the calibration curve of the dosimeters, the calibration coefficient, the lower detection limit and the linear dose-response range of the dosimeters are also modified. The slightly different responses to the ^{60}Co gamma radiation presented by the $CaSO_4$:Dy + Teflon® pellets produced by IPEN and stamping methods for these calibration curve characteristics are due to the enhanced repeatability and reproducibility shown by the pellets produced by the stamping method.

The calibration curves presented in Fig 3 were constructed by the mean TL signal of five $CaSO_4$:Dy + Teflon® pellets irradiated with the same dose, using the same experimental setup applied to perform the previous dosimetric characterization and selection of the pellets, with doses ranging from 1 mGy to 2 Gy.

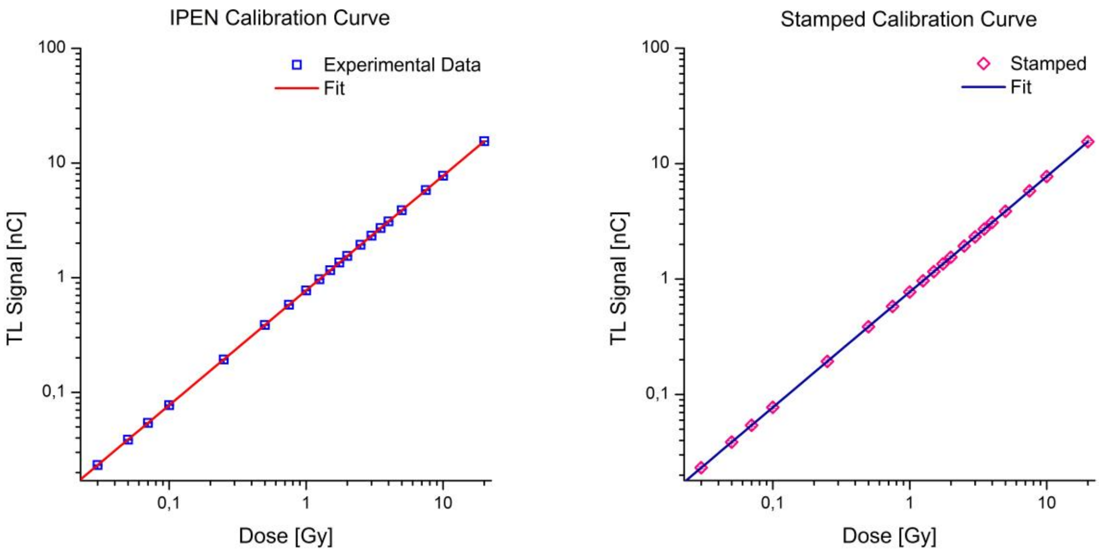

Figure 3. IPEN and stamped pellets calibration curves to the ^{60}Co gamma radiation.

The calibration factor or calibration coefficient, f_c, defined as the inverse of the angular coefficient calculated to the linear fit to the experimental points for the calibration curve, and the lower detection limits, defined by the calibration factor and the average non-irradiated mean TL signal measured after annealing presented in Tab 1 through Eq 1, for IPEN and stamped pellets are presented in Tab 5.

$$LDD = \left(TL_{NI} + 3.\sigma_{TLNI}\right).f_c \quad (1)$$

Table 5. Calibration factor and lower detection limit for IPEN and stamped pellets

	IPEN batch	Stamped batch
f_c [Gy.nC^{-1}]	1.29 (6)	1.29 (3)
LDD [Gy]	0.061 (4)	0.0576 (29)

4. CONCLUSIONS

The stamped batch presents the same mean non-irradiated signals either after sintering or annealing while IPEN batch mean non-irradiated signal after annealing is 23% smaller than its mean non-irradiated signal after sintering. The mean TL signal after irradiation was about 50 nC, but the dispersion varies from 20% to 33% for IPEN batch and keeps in 10% for the stamped batch. The TL signal decreased to about 35 nC with no differences in IPEN batch dispersion and a decrease to 5% in stamped batch dispersion 24 h after the irradiation in the five performed essays. These results suggest both a more homogeneous distribution of the TL material through the batch and a lower contribution of triboluminescence signal generated by the occupation of high energy depth traps during the batch production in the stamping method. Calibration curves present a linear behavior over the entire studied dose range and the same coefficients for both methods, however, the uncertainties in the coefficients determined to the calibration curve obtained with stamped pellets are significantly smaller, leading to a more precise dose determination and to a smaller lower detection limit. Stamping method produces more homogeneous batches, with pellets that maintain the dosimetric properties of the detectors produced by IPEN method, in such a way that the stamping method can substitute with advantages the IPEN method in $CaSO_4$:Dy + Teflon® dosimetric pellets production.

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