

**Reprint**[Radiation Protection Dosimetry, **6**,333 (1984)]**SINGLE CRYSTAL LiF THERMOLUMINESCENCE DOSEMETERS**

T. NIWA, H. MORISHIMA, T. KOGA, H. KAWAI and Y. NISHIWAKI

**Abstract**—Commercial Thermoluminescence dosimeters (TLDs) currently used are finely powdered lithium fluoride (LiF), calcium sulphate or magnesium silicate etc. sealed in glass tubes or those melted with special resins. These are opaque or semi-transparent and therefore thermoluminescence light emitted inside does not come out efficiently due to the self-absorption.

To avoid the self-absorption LiF single crystals have been examined. A cylindrical graphite crucible including LiF mixed with trace amounts of magnesium, copper and phosphorus was heated in a vacuum stainless tube with a slowly moving heater coil along the axis. The LiF single crystal grew gradually from the bottom tip of the crucible.

Thermoluminescence emitted from a LiF single crystal after irradiation with  $\gamma$  rays increased as phosphorus in the LiF crystal increased. Thermoluminescence emitted from a single crystal with 0.25 mole percent phosphorus was one order higher than that emitted from powdered LiF with the same mole percent phosphorus and the same weight.

A single crystal could not grow with phosphorus of more than 0.25 mole percent. However, lithium phosphate appears to be a better activator material than the ammonium phosphate used in our experiments to overcome this the 0.25 mole percent barrier.

**INTRODUCTION**

The types of thermoluminescence dosimeters (TLDs) used at present are unsealed thermoluminescent (TL) powder<sup>1,2)</sup>, TL powder sealed in a glass tube or a TL mould mixed with heat proof resin<sup>3)</sup>. Since most of these are polycrystals they are opaque or semi-transparent. This leads to considerable self-absorption of the thermoluminescence light. To avoid this self-absorption we have made LiF single crystals which would be expected to have higher sensitivity due to their transparency.

LiF TLD's are widely used because they are tissue-equivalent. Although they are less sensitive than CaSO<sub>4</sub> and other TLDs, single crystals of LiF are considered to be promising.

**PRODUCTION OF A LiF SINGLE CRYSTAL**

The production of a LiF single crystal was performed using the method as shown in Figure 1. Materials for TLD were LiF powder made by Merck Co. (99.99% purity), MgCl<sub>2</sub>·6H<sub>2</sub>O, CuCl<sub>2</sub>·2H<sub>2</sub>O and NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> or Li<sub>3</sub>PO<sub>4</sub>. The mixing concentrations of activators Mg, Cu and P are 0.2, 0.05 and 0.1~0.25 mole percent respectively. The mixture was homogenised and put in a carbon crucible hung with a tungsten wire in a stainless tube evacuated up to 1.3 Pa (10<sup>-2</sup>Torr) with a rotary pump. A cylindrical heater, which surrounded the tube, moved upward at the speed of two millimetres per hour for thirty hours. LiF in the crucible (the melting point of LiF is 845°C) was heated to 900°C and cooled

slowly from the bottom tip of the crucible.

A single crystal thus produced was heated again up to 700°C in vacuum for annealing for 50 hours to eliminate strain in the crystal. The dimensions of the single crystal were 16 mm in diameter and 27 mm in length. Thermoluminescence dosimeters were made by cutting the crystal along the direction of crystal face sizes  $12 \times 3 \times 2 \text{ mm}^3$  and  $6 \times 6 \times 2 \text{ mm}^3$ .

### RESULTS AND DISCUSSIONS

The light released from TLD's which contained phosphorus of 0.10, 0.15, 0.20 and 0.25 mole percents after  $2.5 \times 10^{-4} \text{ C.kg}^{-1}$  (1 Roentgen) gamma ray exposure were compared (see Table 1). The amounts of the activators Mg, Cu and P in the single crystal was considered not to be uniform because of the zoning effect during crystallisation. So the TLDs were classified into four parts A, B, C,

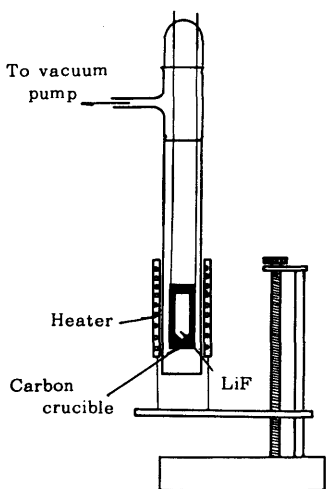


Figure 1 A single crystal production system.

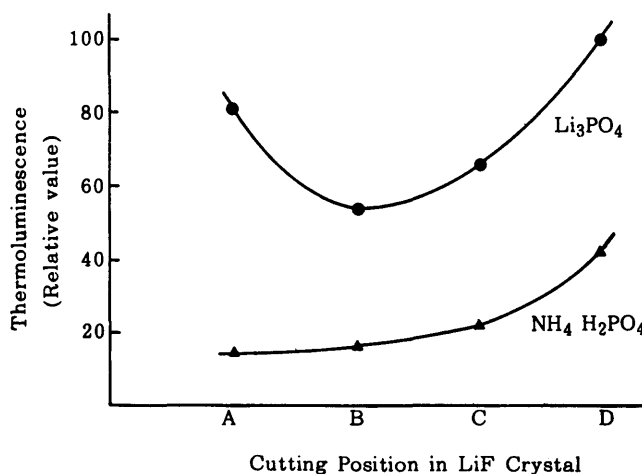


Figure 3 Comparison of thermoluminescence between  $\text{NH}_4 \text{H}_2 \text{PO}_4$  and  $\text{Li}_3 \text{PO}_4$ .

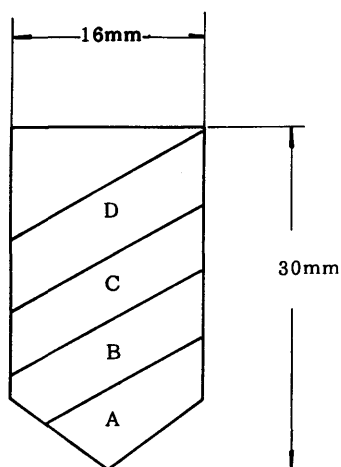


Figure 2 A, B, C, D, positions in LiF single crystal for TLD cutting.

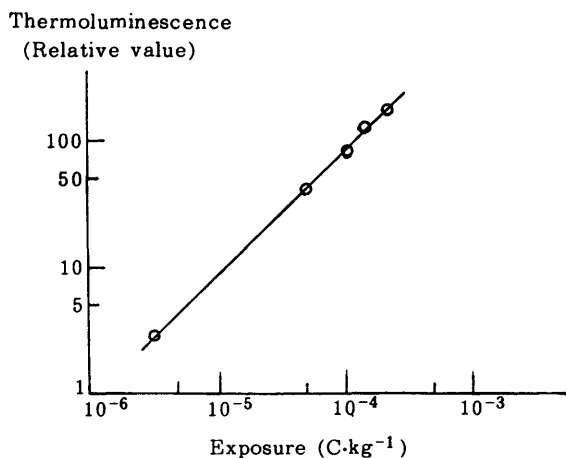


Figure 4 Relation between exposure and thermoluminescence

**Table 1** Thermoluminescence for different amounts of phosphorus activator.

Cutting Position	Mole percent of P			
	0.10	0.15	0.20	0.25
A	292	—	—	—
B	259	499	310	976
C	236	394	300	—
D	156	400	277	197

D along the axis as shown in Figure 2. Since the glow curve showed a peak at 200°C, the TLD was heated up to 300°C for readout in the reader. The amount of light (digits expressed in a reader) increased by increasing the phosphorus activator content as shown in Table 1. The differences of thermoluminescences at the same mole percent of activator (A, B, C, D) suggest a zoning effect.

Figure 3 shows the comparison between activators  $\text{NH}_4\text{H}_2\text{PO}_4$  and  $\text{Li}_3\text{PO}_4$  at 0.25 mole percent phosphorus. The sensitivity of  $\text{Li}_3\text{PO}_4$  is approximately twice as much as  $\text{NH}_4\text{H}_2\text{PO}_4$ . Another merit of  $\text{Li}_3\text{PO}_4$  is that it can be introduced into a LiF single crystal in amounts greater than 0.25 mole percent which is the maximum quantity than can be introduced as  $\text{NH}_4\text{H}_2\text{PO}_4$ .

Thermoluminescence emitted from a LiF single crystal TLD with 0.25 mole percent phosphorus ( $\text{NH}_4\text{H}_2\text{PO}_4$ ) was one order higher than that emitted from powdered LiF with the same mole percent phosphorus and normalised to the same weight.

The relation between the exposure and that of TL light emitted from LiF single crystal was confirmed to be linear as shown in Figure 4.

#### REFERENCES

- 1) Harvey, J. R. and Townsend, S. *The Response of Finely Powdered Thermoluminescent Lithium Fluoride to Beta Radiation*. Berkeley Nuclear Laboratories RD/B/N1372. (July 1969).
- 2) Oberhofer, M. and Scharmann, A. *Applied Thermoluminescence Dosimetry*. Adam Hilger Ltd. 97-99 (1981).
- 3) Hartin, W. J. *An Improved Thermoluminescence Dosimetry System*. Health Physics **13**, 567-573 (1967).