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# COMPARATIVE STUDY OF DIFFERENT LiF TLDs FOR NEUTRON DOSIMETRY

Submitted to *Radiation Protection Dosimetry*

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## Abstract

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Intensive investigations of new materials for thermoluminescence dosimetry (especially of LiF:Mg,Cu,P) are under way. Therefore a comparison of dosimetric characteristics for different TLDs on the basis of LiF is of great interest. Home TLDs: $^{6}$LiF:Mg,Cu,P, $^{7}$LiF:Mg,Cu,P (MSU – "Practica", Moscow); $^{6}$LiF:Mg,Ti, $^{7}$LiF:Mg,Ti (Institute of Geochemistry, Irkutsk) and TLD-700, TLD-600 ("Harshaw") are compared. The comparison has been performed on the basis of neutron dose equivalent measurements in the IHEP neutron reference fields. Prospects of using TLD for area, environmental and individual monitoring are analized with account of new ICRP recommendations.



## Аннотация

Алексеев А.Г., Карпов Н.А. Сравнение характеристик ТЛД, используемых для нейтронной дозиметрии.: Препринт ИФВЭ 94–130. – Протвино, 1994. – 8 с., 3 рис., 5 табл., библиогр.: 7.

В настоящее время проводятся интенсивные исследования новых материалов для термолюминесцентной дозиметрии (особенно LiF: Mg, Cu, P). Поэтому представляет интерес сравнение дозиметрических характеристик разных ТЛД на базе LiF. В работе представлено сравнение дозиметрических характеристик ТЛД: $^{6}$LiF:Mg,Cu,P, $^{7}$LiF:Mg,Cu,P (МГУ – "Практика"); $^{6}$LiF:Mg,Ti, $^{7}$LiF:Mg,Ti (Институт геохимии, Иркутск); ТЛД-600, ТЛД-700 (HARSHAW). Сравнение выполнено на базе измерения с помощью ТЛД эквивалентной дозы нейтронов в нейтронных опорных полях. Представлен анализ перспективности использования этих ТЛД в зонном, экологическом и индивидуальном контроле в ИФВЭ с учетом новых рекомендаций МКРЗ в области радиационной безопасности.


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# 1. INTRODUCTION

The passive neutron dosimetry based on the thermoluminescent detectors (TLD) is a promising trend in the area, enviromental and personnel radiation monitoring of accelerators. However, a successful TLDs application for the neutron dosimetry is limited by their low neutron response. The necessity of increased TLD response is associated with the new national standard in radiation protection, based on the recommendations of the International Commission on Radiological Protection (ICRP, Recommendation ICRP-60) [1].

The required low detection limit in an individual monitoring should be decreased by a factor of 5, when the ICRP-60 Recommendations are applied instead of the former National Radiation Norms and Standards (NRB-76) [2].

In recent years some new TL materials (such as LiF:Mg,Cu,P) having higher sensitivity as compared to the well-known LiF:Mg,Ti have been developed. The characteristics of the LiF:Mg,Cu,P and LiF:Mg,Ti TL-detectors from the point of view of neutron dosimetry are compared. The absolute sensitivity and the ratio of neutron and gamma sensitivities have been estimated from the experimental data for different TL-detectors.

# 2. DETECTORS

The following TL-detectors are compared:

- DTG-4-6 ($^6$LiF:Mg,Ti)
  DTG-4-7 ( $^7$LiF:Mg,Ti)
  (Institute of Geochemistry, Irkutsk, Russia);
- TLD-6011 ($^6$LiF:Mg,Cu,P)
  TLD-7011 ( $^7$LiF:Mg,Cu,P)
  (Moscow State University — "Practica",Moscow);
- TLD-600 ($^6$LiF:Mg,Ti)
  TLD-700 ($^7$LiF:Mg,Ti)
  ("HARSHAW", USA ).

Some parameters of the TLDs, the concentration of $^6$Li and $^7$Li isotopes in detectors, mass and sizes of the detectors are shown in Table 1.

Table 1. The characteristics of TL-detectors.

| Characteristics | Detectors | | |
|---|---|---|---|
| | DTG-4-6<br>DTG-4-7 | TLD-6011<br>TLD-7011 | TLD-600<br>TLD-700 |
| | monocrystal chip | high-strength chip | chip |
| Size,diameter | 4.34 | 4.5 | 3.2·3.2 |
| Thickness, mm | 0.99 | 0.98 | 0.91 |
| Mass, mg | 43 | 36 | 22 |
| $^6$Li concentration in $^6$LiF | 91.1 | 95.62 | 95.62 |
| in $^7$LiF, % | 0.1 | 0.07 | 0.07 |
| $^7$Li concentration in $^6$LiF | 8.9 | 4.38 | 4.38 |
| in $^7$LiF, % | 99.9 | 99.3 | 99.3 |
| Heating rate, $^0$C/s | 7.14 | 14.3 | 7.14 |
| $t_{max}$,$^0$C | 240 | 265 | 250 |
| Post-heating time,s | 50 | 60 | 50 |

The annealing procedure for all detectors was carriedout in accordance with the recommendations of the TLD manufactures. The HARSHAW 2080 Reader was used for the readout. The heating involves the preheating up to 100$^0$C, the linear heating up to $t_{max}$,then postheating at the constant temperature $t_{max}$. The heating rate, $t_{max}$ and the postheating time for their detectors are presented in Table 1. The thermoluminescence glow curves of TL-detectors are presented on Fig. 1. The dose measurements are performed according to:

- Dosimetric main peak intensity.
- The glow-curve integration. The integration has been performed above 100$^0$C distiminator level for the glow curve.

## 3. MEASUREMENT METHOD

The comparison of TLD characteristics has been performed on the basis of neutron dose equivalent measurements with the IHEP passive radiation monitors (PMS). The PMS used for area and enviromental radiation monitoring at the IHEP accelerator and consists of the LiF pairs located in the 25.4 cm in diameter spherical polyethylene moderator (see Fig. 2a).

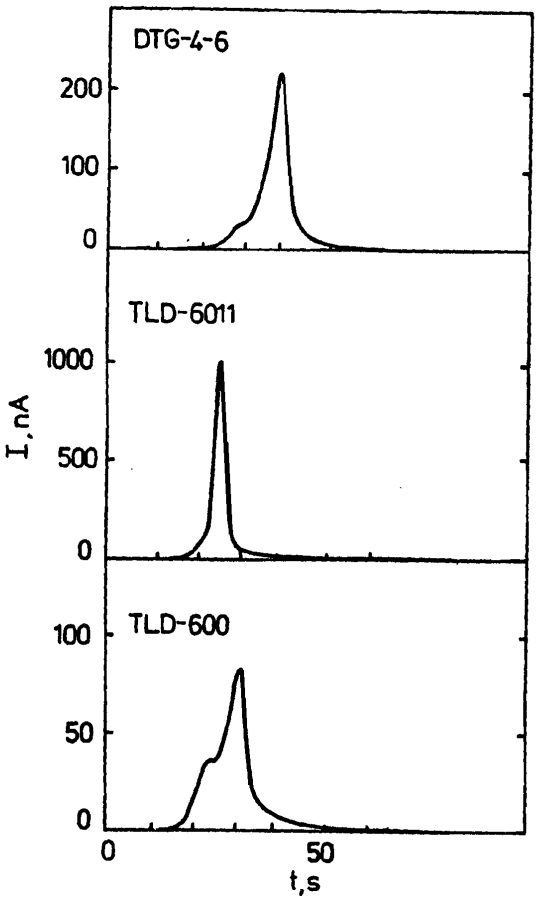


Fig. 1. Glow curve for DTG-4-6, TLD-6011, TLD-600 after $^{252}$Cf neutron irradiation (neutron dose equivalent $H_n$=2.8 mSv).

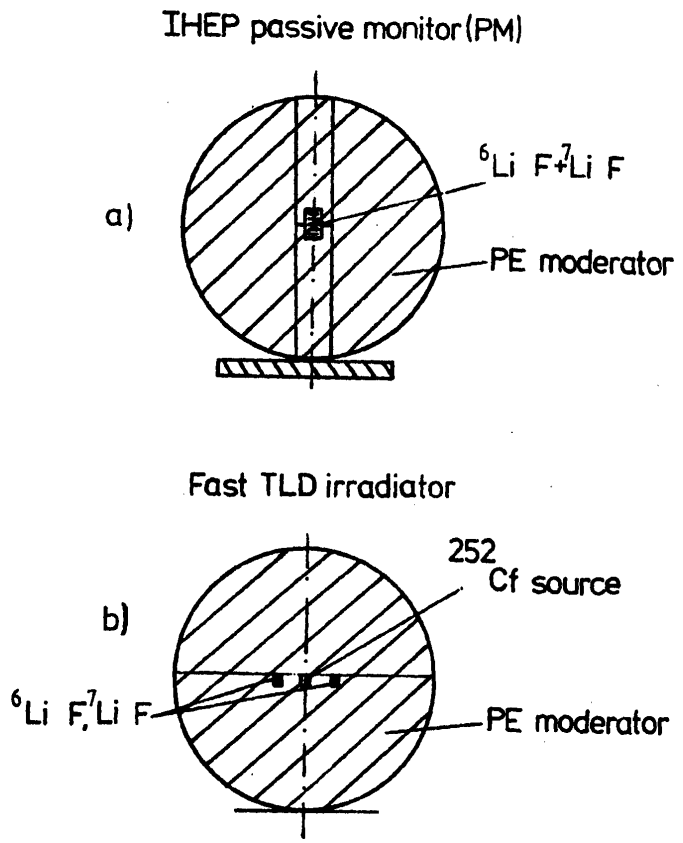


Fig. 2. a) IHEP passive radiation monitor. b) fast neutron irradiator for TLD.

The neutron dose equivalent $H_n$ is obtained according to :

$$H_n = \frac{D_n}{K_{n,Pu-Be}}, \tag{1}$$

$$D_n = D_{n,ph} - D_{ph}, \tag{2}$$

$$D_{n,ph}(^6LiF) = \frac{(I_6 - I_{f,6})}{K_{6,ph}}, \tag{3}$$

$$D_{ph}(^7LiF) = \frac{(I_7 - I_{f,7})}{K_{7,ph}}, \tag{4}$$

where $K_{n,Pu-Be}$ is the calibration factor (Gy·Sv$^{-1}$), obtained with Pu-Be neutron source; $D_n$ is the neutron dose for TLD; $D_{ph}$ is the photon dose for TLD; $I_6$, $I_7$ are TL-light peak intensity (nA) or the integral of glow curve (nC) for $^6$LiF and $^7$LiF;$I_{f,6}$ and $I_{f,7}$ are background signal; $K_{6,ph}$, $K_{7,ph}$ are the calibration factors (nA·Gy$^{-1}$ or nC·Gy$^{-1}$) obtained with $^{137}$Cs photon source for the $^6$LiF and $^7$LiF dosimeters. The photon dose $D_{ph}$ was obtained with the help of $^7$LiF.

## 4. RADIATION FIELDS

The PMS with TLD was irradiated in the following fields:

- The standard calibration field with $^{137}Cs$ proton source in a lead collimator. The standartization of this system was carried out by VNIIFTRI, the State Standard Committee of Russia (with 4% error). This system was used for the TLD calibration in units of photon tissue kerma.
- The thermal neutron field is obtained in the 25.4 cm diameter spherical polyethylene moderator. The $^{252}Cf$ neutron source was located at the sphere center ($^{252}Cf$ in $CH_2$). The TL-detectors were located in the circular position at the distance of 5 cm from source. This field is used for the fast TLD irradiation when selecting the $^{6}LiF$ detectors by sensitivity to the thermal neutrons.
- The reference field based on the $^{239}Pu$-Be neutron source. This field is used for PMS calibration in units of the neutron dose equivalent.
- The reference field based on the $^{252}Cf$ neutron source.
- The reference field based on the $^{252}Cf$ source in the 30 cm-diameter spherical iron moderator ($^{252}Cf$+Fe ).
- The reference field based on the $^{252}Cf$ source in the 30 cm diameter spherical polyethylene moderator ($^{252}Cf+CH_2$).

The dosimetric characteristics of the IHEP neutron reference fields (the last 4 fields) have been given in [3]. The metrological certification of these fields has been performed by VNIIFTRI. The systematical errors of neutron dose equvalent measurement are lower than 10%. The ratios of neutron dose equivalent to photon kerma are given in Table 2.

Table 2. The ratio of neutron dose equivalent to photon kerma in IHEP reference neutron fields.

| Radiation | $K_{ph}/H_n$ ,Gy·Sv$^{-1}$ |
|---|---|
| Pu-Be | 0.037±0.006 |
| $^{252}Cf$ | 0.049± 0.008 |
| $^{252}Cf$+Fe | 0.007± 0.001 |
| $^{252}Cf+CH_2$ | 0.39± 0.05 |

## RESULTS

Table 3 presents the measurement results. The data for intensity of the maximum dosimetric mail peak and the data for glow curve integration method are presented. The data for DTG-4-7 and TLD-7011 are normalized to the TLD-700 values; the data for DTG-4-6 and TLD-6011 are normalized to the TLD-600 values. All the dosimeters were exposed simultaneously.

The ratio of neutron dose ($D_n$) to photon dose ($D_{ph}$) are given in Table 4. These data are in accordance with $K_{ph}$-to-$H_n$ ratios (Table 2) and are given in Fig. 3. The dose measurement method based on the peak intensity is commonly used for DTG-4 and

TLD-600,700. The neutron-to-photon response rations for this method are higher than for glow curve integration. Therefore the dose measurement using the peak intensity is more preferable to be used for neutron dosimetry.

Table 3. Measurement results.

| Irradiation field | Detectors | Dosimetric method: main peak intensity, relative units | Dosimetric method: glow-curve integration, relative units |
|---|---|---|---|
| $^{137}Cs$ | DTG-4-7 | 4.7± 0.2 | 3.4± 0.1 |
| | DTG-4-6 | 6.8±0.6 | 5.0± 0.5 |
| | TLD-7011 | 54±2 | 24±1 |
| | TLD-6011 | 23±1 | 12±1 |
| $^{252}Cf$ in $CH_2$ | DTG-4-7 | 5.0±0.2 | 3.0±0.1 |
| | DTG-4-6 | 2.4±0.2 | 1.8± 0.2 |
| | TLD-7011 | 52± 0.4 | 19± 1 |
| | TLD-6011 | 9.6± 0.6 | 4.8± 0.3 |
| Pu-Be | DTG-4-7 | 2.8±0.5 | 1.1± 0.2 |
| | DTG-4-6 | 2.4±0.1 | 1.8± 0.1 |
| | TLD-7011 | 32±6 | 5.7± 0.5 |
| | TLD-6011 | 9.0±0.5 | 4.0± 0.2 |
| $^{252}Cf$ | DTG-4-7 | 4.0±0.2 | 1.8± 0.2 |
| | DTG-4-6 | 2.4±0.2 | 1.9±0.2 |
| | TLD-7011 | 39± 3 | 8.2±1.2 |
| | TLD-6011 | 9±1 | 4.3±0.3 |
| $^{252}Cf$+ Fe | DTG-4-7 | 3.4±0.2 | 2.1±0.1 |
| | DTG-4-6 | 2.2±0.8 | 1.8±0.8 |
| | TLD-7011 | 41±4 | 12.1±0.5 |
| | TLD-6011 | 8.8±0.4 | 4.1±0.2 |
| $^{252}Cf$+ $CH_2$ | DTG-4-7 | 2.2±0.6 | 2.3±0.1 |
| | DTG-4-6 | 2.8±0.8 | 1.9 ±0.8 |
| | TLD-7011 | 34±2 | 12±1 |
| | TLD-6011 | 8±0.5 | 4.5±0.3 |

The photon sensitivity of DTG-4-6 is higher than for TLD-600 by 4-5 times. The ratio of TLD-6011 to TLD-600 sensitivity is higher by a factor of 40-50, when the data for dose measurements by peak intensity are used. For the glow-curve integration method the sensitivity ratios for all types of detectors are less then for the peak intensity method. The relative sensitivity for photon-neutron mixed radiation is equal to 2-3 (DTG-4-6 to TLD-600) and 8-9 (TLD-6011 to TLD-600). The effect when neutron-to-photon response ratio for LiF:Mg,Cu,P is less than for TLD-600 was obtained for the new Chinese TL-materials as well [6].

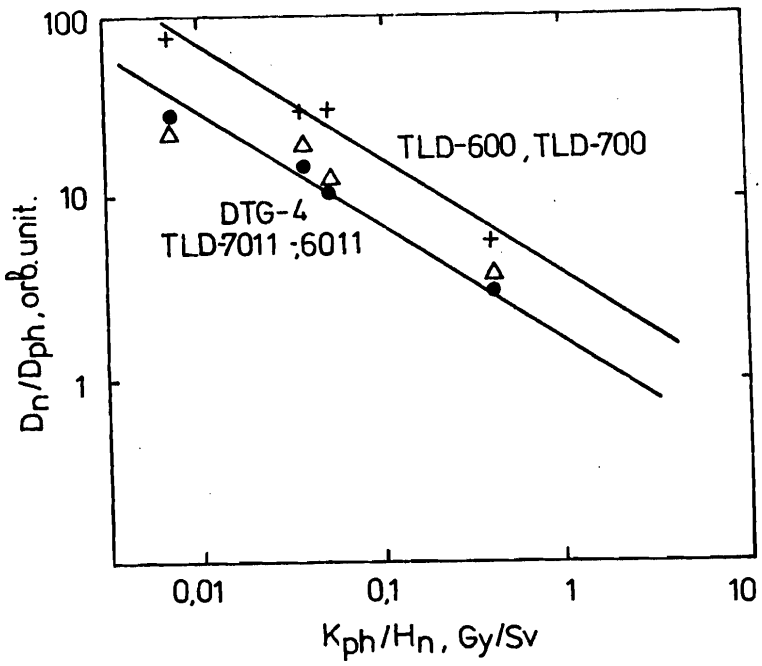


Fig. 3. The neutron - to - photon response ratio for TLD in PMS depending on the ratio of photon tissue kerma to neutron dose equivalent in the radiation fields.

Table 4. The ratio of thermal neutron dose to photon dose (in terms of the photon tissue kerma from $^{137}Cs$) for TLD in PMS.

| Irradiation fields | Detectors | $D_n/D_{ph}$, relative unit Dosimetric method: peak intensity | glow-curve integration |
|---|---|---|---|
| $^{252}Cf$ in $CH_2$ | DTG-4-7, DTG-4-6 | 17±2 | 18±3 |
| | TLD-7011, TLD-6011 | 23±3 | 21±2 |
| | TLD-700, TLD-600 | 53±5 | 44±4 |
| Pu-Be | DTG-4-7, DTG-4-6 | 17±2 | 13±2 |
| | TLD-7011, TLD-6011 | 18±2 | 16±2 |
| | TLD-700, TLD-600 | 31±2 | 13±2 |
| $^{252}Cf$ | DTG-4-7, DTG-4-6 | 11±1 | 9±1 |
| | TLD-7011, TLD-6011 | 16±1 | 13±1 |
| | TLD-700, TLD-600 | 33±1 | 17±1 |
| $^{252}Cf$+ Fe | DTG-4-7, DTG-4-6 | 28±2 | 28±3 |
| | TLD-7011, TLD-6011 | 31±3 | 24±2 |
| | TLD-700, TLD-600 | 81±6 | 63±5 |
| $^{252}Cf$+ $CH_2$ | DTG-4-7, DTG-4-6 | 3.4±0.4 | 2.1±0.2 |
| | TLD-7011, TLD-6011 | 3.6±0.3 | 2.9±0.3 |
| | TLD-700, TLD-600 | 7.9±0.7 | 5.7±0.5 |

The neutron-to-photon response ratio for DTG-4-6 (DTG-4-7) and TLD- 6011 (TLD-7011) are the same, but this ratio for TLD-600 (TLD-700) is higher by a factor of two as compared to other detectors. This effect can be explained by smaller thickness of TLD-600 and by lower effect of self-shielding for thermal neutrons. LiF:Mg,Cu,P have

lower efficiency to high LET radiation. And low neutron-to-photon responce ratio for LiF:Mg,Cu,P is an ionisation density effect [7] .

The decrease of neutron-to-photon response ratio leads to the increase of the measurement errors of the neutron dose equivalent in fields under large photon contribution into a total dose ($\frac{K_{ph}}{H_n} > 1$). Usually the contribution of photons, muons and charged particles into the total dose equivalent is less than 10% behind the shield of high energy accelerators. The exception is the so-called "muons torch" at the IHEP experimental hall, when the neutron dose equivalent contribution is less than 5%. Therefore a decrease in ratio $\frac{D_n}{D_{ph}}$ for TLD-6011 does not restrict applications for the neutron dosimetry at accelerators.

The minimum measurable dose depending on the TLD type is the most important characteristic for dosimetric methods. The minimum measurable dose for photons was obtained as a "zero dose" of the TLD readings immediately after annealing. The neutron calibration factors $K_n$ are equal to 1.15 Sv·Gy$^{-1}$ for PMS obtained with the Pu-Be source. The $K_n$ is equal to 4.5 Sv·Gy$^{-1}$ for personal albedo-dosimeter [4]. The factors $K_n$ for PMS in other neutron spectra behind the shield of the U-70 IHEP accelerator range from 0.6 to 1.5 Sv·Gy$^{-1}$ and from 0.5 to 13 Sv·Gy$^{-1}$ for albedo-dosimeter [4]. If $\frac{D_{ph}}{D_n}$ ratio is less than 0.1 for PMS and this value is less then 1.0 for a albedo-dosimeters [4].

The minimum measurable neutron dose equivalent for different detector types and dosimetric method (HARSHAW 2080 reader) without nitrogen atmosphere are presented in Table 5.

Table 5. Minimum measurable neutron dose equivalent $H_f$ for different detector types ("zero dose" for HARSHAW 2080 reader without nitrogen atmosphere)

| Detector type | $H_f$,mSv | |
|---|---|---|
| | PMS | Albedo dosimeter |
| DTG-4-6,7 | 0.015-0.02 | 0.06 - 0.2 |
| TLD-6011,7011 | 0.004-0.006 | 0.016 - 0.05 |
| TLD-600,700 | 0.04 - 0.06 | 0.16 - 0.5 |

The application of new annual limits according to the ICRP-60 Recommendations [5] leads to a decrease of the required minimum measureable dose for dosimeters.

The lower limit of detection according to ICRP-35 Recommendations should be 0.1 of an annual limit. It should be borne in mind that comentionally administrative limits are less than annual limit. The annual level is equal to 2 mSv according to ICRP-60. Therefore in the individual monitoring a monthly measured dose at the lower limit of detection should be 0.17 mSv. It is TLD-6011, 7011 for the albedo dosimeter that provides the neutron equivalent dose measuring at the lower limit of detection. The PMS with TLD-6011, 7011 allows one to carry out the area monitoring with the minimum measurable dose 0.1 of the recommended dose limits for population (1 mSv a year).

At the same time the use of albedo dosimeters with TLD-6011, 7011 is in competition with MK-20 nuclear film which are still used in the IHEP individual monitoring at present. The minimum measurable neutron equivalent dose for MK-20 is equal to 0.05 mSv.

## CONCLUSION

The comparison of TLDs to be used for neutron dosimetry shows that photon sensitivity of a new TLD-6011 is higher than that of TLD-600 by a factor of 50. The neutron sensitivity of TLD-6011 is 9 times higher than that for TLD-600. The neutron sensitivity for TLD-6011 is higher than that for DTG-4-6 by a factor of 4-5. The neuton-to-photon response ratio for TLD-600 is about two times higher than that for TLD-6011 and DTG-4. At the same time the new TL-detectors TLD-6011, 7011 are intended to be used in area and individual monitoring even under the new ICRP recommendation in radiation protection.

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