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■ 論 文 ■

Characteristics of $CaSO_4$:Dy TL Dosimeters by Determining LLDs, Fadings and Sensitivity Changes in Repeat Uses

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최소검출한계, 잠상퇴행 및 반복사용으로 인한 감도변화 결정에 의한 $CaSO_4$:Dy 열형광선량계 특성

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Abstract－Theoretical and experimental determination of the lower limits of detection (LLD) of C-300-A $CaSO_4$:Dy TL dosimeters which are currently used for the personnel monitoring in Korea Atomic Energy Research Institute(KAERI) are described with a critical level which is defined as the signal level above which a result has a probability of being due to a fluctuation of the background.

The personnel monitoring processors can derived easily the LLD of their system using this method with the background readings of their service interval and the irradiation readings of the known doses.

Experimental studies were also conducted for the fading rates of the dosimeters with the temperatures and humidities for 3 months. Finally sensitivity changes in repeat uses were measured for 40 times consecutive uses of the dosimeters. The applications of the experimental results of fading rates and sensitivity changes in real personnel monitoring services are discussed briefly.

Key words:lower limits of detection, fading rates, sensitivity changes in repeat uses

요약－현재 한국원자력연구소에서 개인방사선피폭관리용으로 사용되고 있는 C-300-A $CaSO_4$:Dy 열형광선량계의 최소검출한계(LLD)를 계산 및 실험에 의하여 구하였으며, 이때 오차의 확율이 자연방사선의 측정변화에 기인되는 선량준위로 정의되는 임계준위도 함께 구하였다. 개인방사선피폭서비스를 수행하는 기관에서는 이를 이용하여 개인선량계 패용 기간동안의 자연방사선 판독결과와 기지의 선량을 조사시킨 선량계의 판독결과로부터 쉽게 LLD를 구할수 있다.

또한 3개월간에 걸쳐 온도 및 습도의 변화에 따른 개인선량계의 잠상퇴행률을 실험적으로 구하였다. 마지막으로 40회 연속사용시 개인선량계의 반복사용에 의한 감도변화를 측정하였다. 본 실험결과 얻어진

잠상퇴행률과 연속사용시의 감도변화를 실제 개인방사선피폭 평가시 적용방법에 대하여 간략하게 논의하였다.

중심어:최소검출한계, 잠상퇴행율, 반복사용에 의한 감도변화

INTRODUCTION

The optimum choice of personnel dosimetry system could be considered the detection capability of the unexpected exposures involved the measurement of low dose level, the long-term stability of the sensitivity of the dosimeters under severe climatic conditions and the repeat use of the dosimeters without any corrections. But there should be no doubt the dosimetry system finally selected have to meet certain minimum performance criteria. The reliability requirements vary for different applications but a ±50% overall accuracy and precision of dose measurement has to be satisfied for personnel dosimetry[1]. The extent to which a couple of common characteristics of performance mentioned above under realistic conditions of personnel dosimetry may contribute to a given interpretation of dosimeter response, and through the exact assessment of these terms the evaluated results will be maintained more accurately and precisely.

For the lower limit of detection(LLD) it is emphasized in ANSI N13.11[1, 2] through the low limit of the test range and in the testing standard for the Department of Energy Laboratory Accreditation Program for personnel dosimetry[3]. If the LLD is at or above the low limit of the testing range, there is an unacceptable large risk of reporting a test dosimeter as a zero.

Many articles (e.g. Ref. [4, 5]) described the fading rates for the TLD phosphores and this rates also provided by the manufacturer, but the actual field conditions are different from constant laboratory conditions mostly in the extremes in temperature and humidity which are frequently encountered during the personnel dosimeter wearing period even if the average temperature and humidity are close to normal. Actually speaking no fading rates to be applied in the dose evaluation are provided by the manufacturer of the dosimeters.

We do not still know quantitatively how many times we can use the TL dosimeters repeatedly without any correction. Element correction factors (ECFs) provide an excellent tool to examine the dosimeters and the ECFs are measured for each dosimeter by processor or manufacturer before the dosimeters are used in the fields. But without any experiment for the sensitivity change by repeat uses of dosimeters, unnecessary efforts would be consumed to generate new ECFs from too short interval.

To characterize the above problems for Teledyne TLD system which is currently used as personnel monitoring system at KAERI this paper calculates the LLD by routinely collected data, measures the fading rates at several temperatures and humidities and investigates the change of the dosimeter response sensitivities with the repeat use.

THEORY OF LOWER LEVEL OF DETECTION(LLD)

The critical level(L_c) is defined as the signal level above which, at a predetermined confidence level, a signal may be considered to be outside the expected range of fluctuation of the background signal[1]. This signal provides a confidence level of 1-α, the value α is the significant level at which there is an acceptable risk of a false positive (the incorrect

The variance for the detection level ($\mu_H = L_D$) from eq.(10) is

$$\sigma_D^2 = \sigma_\mu^2 (L_D^2 + 2L_D \mu'_B) + \sigma_0^2 \tag{11}$$

where $\mu'_B \equiv \mu_B - \mu_N$. Inserting eq.(1) for L_c into eq.(11) to solve σ_D in eq.(2), then

$$(1 - k_B^2 \sigma_\mu^2) L_D^2 - 2(k_\alpha \sigma_0 + k_\beta^2 \sigma_\mu^2 \mu_{B'}) L_D + (k_\alpha^2 - k_\beta^2) \sigma_0^2 = 0 \tag{12}$$

For $k_\alpha = k_\beta = k$,

$$L_D = \frac{2(k\sigma_0 + k^2 \sigma_\mu^2 \mu_B)}{1 - k^2 \sigma_\mu^2} \tag{13}$$

For finite sample sizes, $\bar{H}_{B'} = \bar{H}_B - \bar{N}$ replaces $\mu_{B'}$, Student's t values replace the k values, and sample standard deviations (s) replace σ values. Using n background dosimeter readings to calculate S_0 and $\bar{H}_{B'}$, and m dosimeter readings of a known large dose (about 10 mSv) compared to background to calculate S_μ,

$$L_D = \frac{2(t_n S_0 + t_m^2 S_\mu^2 \bar{H}'_B)}{1 - t_m^2 S_\mu^2} \tag{14}$$

where t_n and t_m are Student's statistic for (n-1) and (m-1) degree of freedom, and S_μ is the relative sample standard deviation for (m-1) degree of freedom. A typical confidence level for t is 95%.

USNBS[9] includes necessary formula.

$$S_\mu^2 = \frac{1}{\bar{H}(m-1)} \sum_{i=1}^{m} (H_i - \bar{H})^2 \tag{15}$$

$$\bar{H} = \frac{1}{m} \sum_{i=1}^{m} H_i$$

$$S_0 = S_B(1 + 1/n)$$

$$S_B^2 = \frac{1}{(m-1)} \sum_{i=1}^{m} [(H_B)_i - \bar{H}_B]^2 \tag{16}$$

$$\bar{H}_B = \frac{1}{n} \sum_{i=1}^{m} (H_B)_i$$

$$\bar{H}'_B = \bar{H}_B - \bar{N}.$$

where the $(H_B)_i$ represents background readings and the H_i represents known dose readings. The reader noise level $\bar{N}$ may be measured or adjusted to zero before dosimeter readout.

EXPERIMENTS

Lower Limits of Detection(LLD)

The Teledyne C-300-A TL dosimeters consist of four TL chips with the first being 0.13 mm and the remainder being 0.4 mm thickness $CaSO_4$:Dy. The first chip is covered with a window thickness of approximately 7 mg/cm^2 which is used to report $H_p(0.07)$[10].

The LLD is calculated using reading data from 30 control dosimeters and 30 dosimeters irradiated to 3 different doses to compare the LLD differences with doses. Irradiation doses were 1, 5 and 10 mSv of Cs-137 gamma radiation, and 3 months time period for control dosimeters was chosen to match 3 months TLD processing cycle. The TLDs were then read by Teledyne A300 Reading System, the data was processed through the algorithm and $H_p(d)$ was recorded. Distribution for $H_p(d)$ are summarized in Fig. 1.

Fading Effect

The slow, thermally stimulated release of trapped electrons or holes in irradiated TLD materials has been described in terms of trap depth (activation energy) and frequency factor based on the classical Randall-Wilkins equations. The reliability of the fading predictions depends mostly on the prediction with which the trap parameters of a given phosphor are known, and on the assumed TL mechanism.

rejection of the null hypothesis) or Type I error. Values less than Lc is not reported as an indication of the detection of a positive signal, and signals above Lc due solely to background fluctuation should be reported as positive values accompanied by a confidence level.

Detection level(L_D) is defined as the dose equivalent for which the confidence level is 1-β that the result will be detected and properly reported as a positive results. Not reporting a positive value for a true dose equivalent is an error of the Type II or the incorrect acceptance of the null hypothesis (false-negative).

The critical level is determined by using the standard deviation of the readings from unirradiated dosimeters, σ_0[1,6,7,8],

$$L_C = k_\alpha \sigma_0 \tag{1}$$

where k_α is the abscissa of the standard normal distribution corresponding to a single-sided probability for a predetermined confidence level.

The detection level is greater than the critical level by an amount of dependent on the standard deviation of the signal of the confidence level,

$$L_D = L_c + k_\beta \sigma_D \tag{2}$$

where k_β is the abscissa of the standard normal distribution corresponding to a single-sided probability of 1-β, and σ_D is the standard deviation of the readings at the dose equivalent level L_D.

The total signal, μ_T, is the sum of a signal due to the effect to be measured (μ_H), a signal attributed to the dosimeter but not part of the effect being measured (μ_B-μ_N) and a signal not associated with the dosimeter, namely reader noise (μ_N),

$$\mu_T = (\mu_H + \mu_B - \mu_N) + \mu_N \tag{3}$$

where μ_B is the signal from the control dosimeter. The standard deviation of the total signal, σ_T, can be split into two components, one dependent on the dosimeter signal level ($\mu_B - \mu_N$) and the other identified with the reader noise level (μ_N),

$$\sigma_T^2 = \sigma_\mu^2(\mu_T - \mu_N)^2 + \mu_N^2 \tag{4}$$

where σ_μ is the constant value of the relative standard deviation for large signals. For the background readings,

$$\sigma_B^2 = \sigma_\mu^2(\mu_B - \mu_N)^2 + \mu_N^2 \tag{5}$$

The net dosimeter reading, μ_H, is the difference between the total value and the mean background value,

$$\mu_H = \mu_T - \mu_B \tag{6}$$

For one dosimeter readings, these values are replaced by observed values,

$$H = T - \bar{H}_B \tag{7}$$

where the background $\bar{H}_B$ is determined by n dosimeters and the bar denotes mean value.

The variance of the net signal is

$$\sigma_H^2 = \sigma_T^2 + \sigma_B^2/n \tag{8}$$

From eq.(4) and (5) for general readings,

$$\sigma_H^2 = \sigma_\mu^2(\mu_T - \mu_N)^2 + \sigma_N^2 + \sigma_B^2/n \tag{9}$$

For $\mu_T = \mu_H + \mu_B$ from eq.(6), eq.(9) becomes

$$\sigma_H^2 = \sigma_\mu^2[(\mu_H^2 + 2\mu_H(\mu_B - \mu_N)]^2 + \sigma_B^2(1 + 1/n) \tag{10}$$

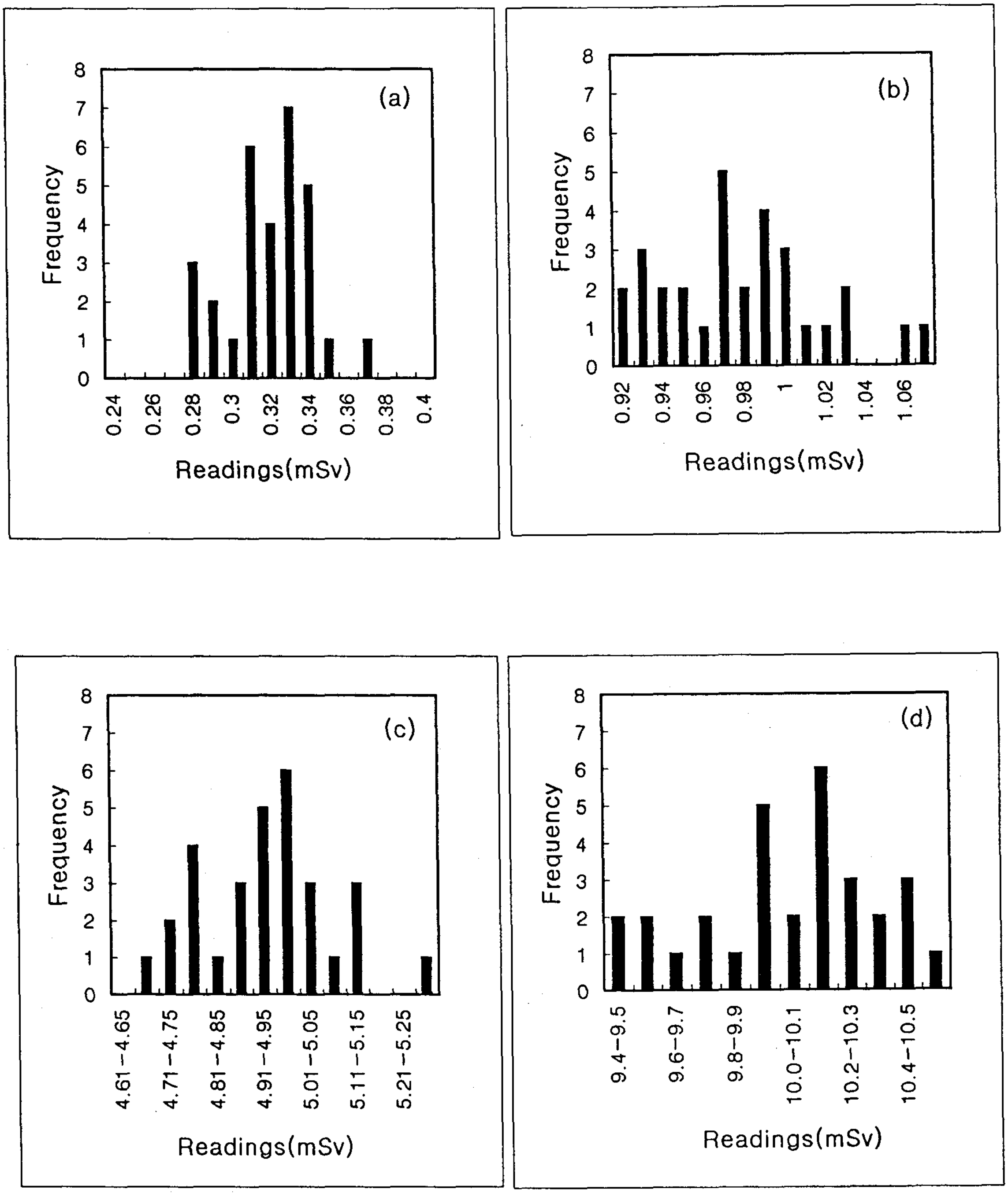

Fig. 1. Dosimeter readings for (a) unirradiatied dosimeters (b) Cs-137 1 mSv irradiation (c) Cs-137 5 mSv irradiation and (d) Cs-137 10 mSv irradiation

As a rule, the thermal stability of the radiation induced latent signal in a TL phosphor increase rapidly with increasing temperature, and it is true for the temperature dependence of the fading rates[4]. Although it is safe for some phosphors to assume a single linear extrapolation of fading rates which have been measured at elevated temperatures down to lower temperatures, such an extrapolation is not possible in many other cases. In no cases should calculated fading rates be assumed to be correct without verification. So the fading test was conducted to determine the fading rates of $CaSO_4$:Dy dosimeters at 4 different environmental conditions over a 3 months periods.

All dosimeters in this study were processed at the same time on the same reader to minimize errors at each test interval and the test was conducted under very carefully controlled conditions. Conditions are included in Table 1. The storage conditions were maintained 24 hours a day throughout the duration of the study for temperature and humidity. Control dosimeters were maintained same as the exposed dosimeters used for the test for each test intervals and test conditions. Cs-137 source was used to perform all irradiations. The number of data points per test interval per condition was 20 and just same dosimeters per test interval were used repeatedly at each conditions to reduce the errors from the dosimeter variations.

Table 1. Storage Conditions of Dosimeters

Condition	Temperatures (℃)	Humidities (%)
TLD Storage Room	22	40~50
Refrigerator	4	40
Incubator(I)	32	35
Incubator(II)	32	90

Sensitivity Change in Repeat Use

The main advantage of the TL dosimeter is that it can be used many times without any correction with proper annealing procedures. Sensitivity change in repeat use is defined as the closeness of agreement in measurements of the output for the same value of input under the same operating conditions over a number of repeated trials. It is quite related to the element correction factor(ECF). As dosimeters are used in the field, their ralative response to radiation might change due to multiple readings and physical abuse. To compensate the dosimeter response decrease by repeat use, it is normal for processors of TL dosimeters to characterize each element they use by producing ECFs. The frequency for generating new ECFs for dosimeters depends entirely on how the dosimeters have been treated. Rather than an arbitrary schedule(i.e. many processors measure ECFs one or two year interval according to the their quality control program.), a rational schedule should be developed based on observed changes in ECFs by experiment of repeat use.

For this measurement 100 new C-300-A dosimeters were purchased from the Teledyne, and 60 dosimeters within 5% of unity in the dosimeters were selected for measurement of repeat use effect. The irradiation dose was 10 mSv on the PMMA phantom by Cs-137 gamma source and read after 24 hours for removing initial fading of the dosimeters and repeated the experiment 40 times.

RESULTS AND DISCUSSIONS

The dominant term in the expression for L_D in eq.(14) is $2tS_0$. This term represents the upper half of the background distribution plus the lower half of the L_D distribution. Since t at 95% confidence level is 1.7, L_D is approximately 3.4 times the standard deviation of the background(σ_0) and L_D was usually used to 3 times the σ_0 in KAERI according to the quality control program. Calulated input values for eq.(14), L_c and L_D from experimental data summarized in Table 2.

Table 2. Calculated Input Values, L_c and L_D

Input	Calculated Values with Doses		
	For 1 mSv	For 5 mSv	For 10 mSv
$\bar{H}_B$	0.320 mSv	0.320 mSv	0.320 mSv
S_B	0.022 mSv	0.022 mSv	0.022 mSv
$\bar{H}$	0.972 mSv	4.942 mSv	10.04 mSv
S_μ^2	0.002 mSv	0.004 mSv	0.012 mSv
t_m	1.7	1.7	1.7
Ouput			
L_c	0.037 mSv	0.037 mSv	0.037 mSv
L_D(LLD)	0.079 mSv	0.084 mSv	0.100 mSv

The critical level, L_c, is equivalent to 0.037 mSv. This indicates that when exposure is actually zero, the TLD may provide a reading in excess of 0.037 mSv in 5% of the time. The remaining 95% of the time the TLD will indicate an exposure of less than 0.037 mSv. Calulating the lower limits of detection, L_D, are equivalent to 0.079, 0.084, and 0.10 mSv for the irradiation doses of 1.0, 5.0 and 10.0 mSv respectively. The values of L_D are not quite different with the irradiation doses, because in these cases, as in most, all but the leading term $2tS_0$ are small in stable TLD system. One of the main issue to measure the L_D value that the exposure dose only above the level that can be identified as nonzero (above L_D) would be reported while retaining all other data for future evaluation. The good solution for this is to report minimal or zero in individual exposure record for the level of dose less than LLD while retaining the actual as-read value and the L_D value for the future epidemiological studies separately.

$CaSO_4$:Dy which is one of the most interesting of the modern phophors has only a little fading at room temperature (about 2~4% during one month, about 6% during 3 months) as shown in Fig. 2 and Table 3 comparing to the other phosphors[4].

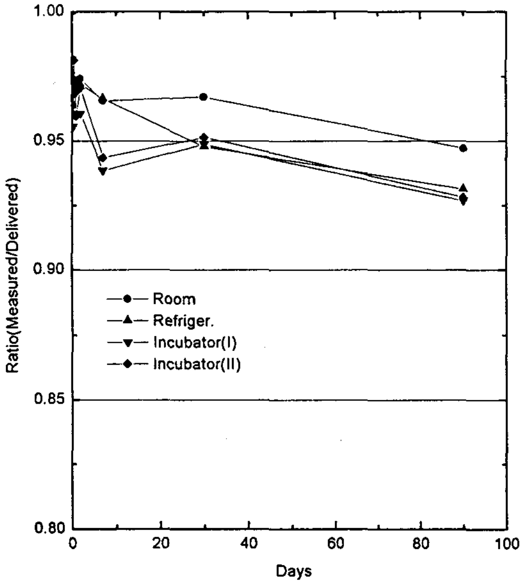

Fig. 2. Fading Rates of $CaSO_4$:Dy Dosimeters

Table 3. Fading Rates of the $CaSO_4$:Dy Dosimeters with the Storage Conditions

Duration	Room	Refrigerator	Incubator (I)	Incubator (II)
Reading just after Irradiation	1	1	1	1
6 h	0.9639*	0.9679	0.9556	0.9682
15 h	0.9726	0.9690	0.9718	0.9814
24 h	0.9691	0.9732	0.9607	0.9596
48 h	0.9741	0.9721	0.9606	0.9705
1 week	0.9656	0.9665	0.9385	0.8436
1 month	0.9672	0.9477	0.9486	0.9514
3 months	0.9471	0.9314	0.9267	0.9281

* The response 0.9639 indicates a 3.61% fade.

The fading rate increases slightly to about 5∼6 %/month and about 8%/3months at 32℃ but no significant fading effect observed in 4℃ and in high humidity. For personnel monitoring dosimeters are normally issued for a 1 to 3 month wear period. When dosimeters are being worn where the average temperature to which the dosimeter is normally exposed is less than 30℃, even if the dosimeter was to be continuously exposed to 30℃, the maximum fade over 3 months would only 8%. Moreover the fading rate of one day after irradiation is about 3∼4%, nearly more than 60% of the fading for 3 months of dosimeter wearing interval. It means that if the reader calibration would be performed 24 hours after reference irradiation with reference dosimeters it is not nearly necessary compensate fading effect for normal monitoring service. But the fading rate can give a good information for other cases, e.g. intercomparison study for which the irradiation date is exactly known.

Fig. 3 and Table 4 show the sensitivity changes of 4 elements of CaSO$_4$:Dy dosimeters in repeat use respectively.

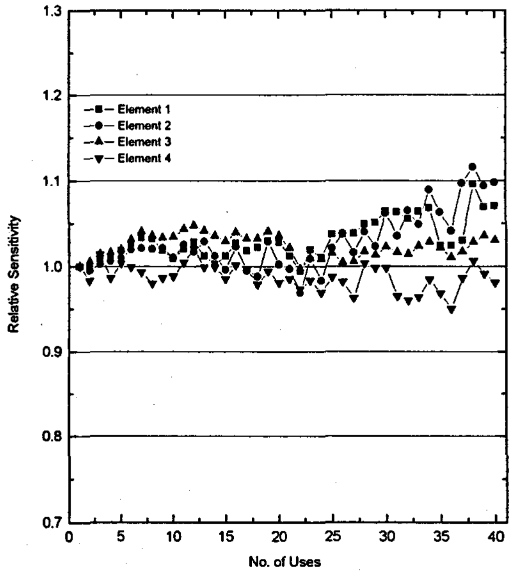

Fig. 3. Sensitivity Changes of CaSO$_4$:Dy Dosimeters in Repeat Use

Table 4. Sensitivity Changes of 4 Elements of CaSO$_4$:Dy Dosimeters in Repeat Uses

No. of Repeat Uses	Sensitivity Changes			
	Element 1	Element 2	Element 3	Element 4
1	1.000	1.000	1.000	1.000
2	0.998	0.995	1.005	0.983
3	1.007	1.002	1.014	1.014
4	1.010	1.006	1.016	0.986
5	1.018	1.010	1.019	1.003
6	1.027	1.019	1.031	0.999
7	1.033	1.021	1.040	0.993
8	1.032	1.020	1.035	0.980
9	1.018	1.021	1.033	0.986
10	1.009	1.010	1.034	0.988
11	1.020	1.025	1.043	1.004
12	1.028	1.017	1.047	1.020
13	1.012	1.029	1.041	0.999
14	1.002	1.012	1.035	0.999
15	1.012	0.996	1.029	0.985
16	1.027	1.023	1.039	1.001
17	1.018	0.995	1.032	0.997
18	1.022	0.988	1.032	0.979
19	1.030	1.029	1.040	0.994
20	1.028	1.002	1.035	0.981
21	1.012	0.997	1.021	0.985
22	0.994	0.969	0.997	0.973
23	1.019	1.009	1.009	0.983
24	1.010	0.983	1.008	0.969
25	1.037	1.021	1.015	0.987
26	1.037	1.038	1.004	0.982
27	1.038	1.016	1.005	0.963
28	1.049	1.040	1.017	1.003
29	1.051	1.023	1.013	0.998
30	1.064	1.062	1.022	0.998
31	1.063	1.035	1.016	0.965
32	1.055	1.065	1.014	0.960
33	1.064	1.049	1.023	0.964
34	1.068	1.089	1.028	0.984
35	1.024	1.063	1.021	0.968
36	1.024	1.041	1.010	0.950
37	1.030	1.097	1.017	0.986
38	1.096	1.116	1.028	1.006
39	1.069	1.094	1.035	0.991
40	1.070	1.098	1.030	0.981

These elements show the sensitivity change less than 5% within the reliability of 95% after the repeat uses of 40 times. So minimum 40 times repeat uses for C-300-A dosimeter which is uesd for a quarter annually (periodic) monitoring are assured. In the quality control program for TLD system of KAERI which was approved by Ministry of Science and Technology(MOST) according to the MOST Ordinance 96-7[11], the ECF should be measured every 2 year interval. But our experimental results show that the interval for measurement of ECF is too short because one dosimeter uses only 2 times a year (For one radiation worker, two dosimeters are used for monitoring service at every 3 months in turn.). Even though the most conservatively speaking, the ECF correction is enough to be made every 5 year interval because physical abuses for several uses, environmental conditions for working places and other factors by which would be affected the dosimeter sensitivity changes are not considered in this experiment. If a dosimeter is to be used more times than stated above, it should be carefully examined with enough consideration to its history in the past use.

CONCLUSIONS

Lower limit of detection (LLD) was determined with the goal of achieving a reasonably low dose measurement capability and it was indirectly emphasized by the specification in ANSI N13.11 through the low limit of the testing range. The calculated LLDs were in the range of 0.08 to 0.1 mSv with the irradiation doses from which the LLDs were not quite different. The calculated LLD can be used to control the zero dose equivalent readings reported, but as-read data should be retained separately for future evaluation. If the processors maintain their dosimetry system such that the LLD is less than half the low limits of the MOST proficiency test exposure ranges, they can avoid testing failures due to poor performance at low exposures.

The fading rates of $CaSO_4$:Dy phosphors are not changed severely with the temperatures and humidities and it could be negelected for personnel monitoring services if the reader calibration would be done at 1 day after reference irradiation as recommended in the quality control program of KAERI. But this information can be uesd for a better dose evaluation where the irradiation date is exactly known.

Severe sensitivity changes are not occurred in $CaSO_4$:Dy phosphores for 40 times repeat uses. So the measurement interval of ECF of every 2 year described in quality control program could be renewed to minimum every 5 year because one $CaSO_4$:Dy dosimeter is uesd for only 2 or 3 times a year.

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