J. Korean Assoc. Radiat. Protec.
Vol. 12, No. 1, 1987

Correlation Between Exposure Rate and Quasi-Effective Energy of Natural Radiation in Japan

—TLD Application—

Toshiyuki Nakajima

Division of Physics, National Institute of Radiological Sciences

=Abstract=

The quasi-exposure rate and the quasi-effective energy of the natural radiation in the field at 47 monitoring points around nuclear power plants have been studied with the pair filter thermoluminescence dosimeter system. The results of the six years observation showed that the relationship between the quasi-exposure rate X_q and quasi-effective energy E_q can be represented as a hyperbolic function: $X_q = A + C/(E_q - B)$, where the constants A and B correspond to the quasi-exposure rate of cosmic-rays and the minimum quasi-effective energy of natural radiation, respectivly. Furthermore, the constant A is in close agreement with the values obtained by using ionization chambers and scintillation detectors. The constant B is approximately 0.68 MeV, closely corresponding to the mean energy of the photons emitted from natural uranium.

1. INTRODUCTION

Thermoluminescence dosimeters(TLD) are widely used in the nuclear power industry, environmental science, dating of ancient pottery, medicine and in many other fields, and it has already been well established that these monitors and dosimeters serve usefully for estimating exposure.

It is to be noted that higly sensitive TLD phosphors such as CaF_2, $CaSO_4$ and Mg_2SiO_4 generally present a large atomic number, which causes their thermoluminescence (TL) response(TL intensity/exposure) to be strongly influenced by the energy of photon radiation. Consequently, unless the photon radiation of energy is known, accurate exposure evaluation can not be obtained with such TLDs. The expedient currently adopted to obviate this difficulty is to use a filter to lower energy-dependence of TL response. Such filtered TLD monitors are useful for detecting changes in the radiation exposure, but if the actual value of radiation energy could come to be evaluated by the same means, such a TLD system should find useful applications also in many other fields.

Two alternative methods are available for evaluating the effective energy of photon radiation by means of TLD. One is to use phosphors of different kind or of different effective atomic numbers(phosphor method)[1]. The other method is to use two or more TLD phosphors covered by filters of different material(filter method)[2].

The present work covers experiments to ascertain the possibilities presented by the filter method for evaluating with TLD the effective energy of photon radiation and the relationship between exposure rate and effective energy of natural radiation by using the pair filter TLD system.

2. PRINCIPLE

The energy of incident radiation to be measured is here understood to mean the effective energy of photon radiation evaluated with the half value layer method.

For filter-covered TLD monitors and dosimeters, the observed TL response

$$TL=f_j(E_j)\cdot R_j \exp\{-\mu(E_j)t\} \tag{1}$$

where R_j represents the corresponding exposure from radiations, $f_j(E_j)$ is the unit TL responses per exposure from the same source of radiation, which vary with the effective radiation energy, E_j, as well as with the kind of the TLD phosphor, $\mu(E_j)$ the atten uation coefficients of the filter material for photons of energy, E_j, and t is the thickness of the filter material.

When TLD phosphors covered by two filters of different materials or thicknesses are irradiated for the same duration and the same position, the ratio of TL response between phosphors in given by

$$TL_1/TL_2=\{f_1(E_j)/f_2(E_j)\} \exp\{\mu_2(E_j)t-\mu_1(E_j)t'\} \tag{2}$$

where the suffixes 1 and 2 respectively represent the values for lead and lucite filters. Equation (2) gives the TL response ratio TL_1/TL_2 as function of the effective energy of radiation and which is independent of exposure. This indicates the possibility of reliably determining the incident effective energy of photon radiation by choosing a suitable combination of materials and thicknesses for the filter pair.

3. Theory

Natural radiation, which is a mixture of terrestrial radiation and cosmic-rays consists of photons, charged and uncharged particles. Effective energy as well as exposure is introduced as a concept of photon radiation. The concept of effective energy and exposure is unsuitable to the mixed radiations like natural radiation. Therefore, the notation of quasi-effective energy and quasi-exposure are introduced for the mixed radiations except for neutrons.

In this work, the quasi-effective energy E_q of the mixed radiations such as natural radiation is defined as the effective energy corresponding to energies of photon radiation determined using the half-value thickness method.

The quasi-exposure X_q or quasi-exposure rate $\dot{X}_q$ of such radiations is the value of TL intensity from TLD in a lucite filter of the pair filter TLD system. The value is calibrated with gamma-rays from the ^{60}Co source. Terms of these quasi-effective energy and quasi-exposure rate were used for cosmic-rays and natural radiation in this work.

A soft component of the cosmi-ray dose is defined as a part of dose from cosmic-rays absorbed with a lead box of 5cm in thickness. On the other hand, a hard component is defined as the part from the cosmic-rays passed through the box.

Let us consider the relationship between the effective energy and the exposure rate of photon radiation from radionuclides at the position of dosimeter before discussion of such relationship on natural radiation.

When the mean effective energy and the total exposure rate of photon radiation consisting of some radionuclides are presented as E and X, these values are represented as follows:

$$\bar{E}=\sum\dot{X}_iE_i/\sum\dot{X}_i \qquad (3)$$

$$\dot{X}=\sum\dot{X}_i \qquad (4)$$

where $\dot{X}_i$ and E_i are the exposure rate and the effective energy of photon radiation from i-th radionuclide at the position of dosimeter or detector, respectively.

For simplification, the case of only two radionuclides with photon radiation is considered. It is assumed that the exposure rates due to these radionuclides with the effective energy of E_1 and E_2, are $\dot{X}_1$ and $\dot{X}_2$ at the position of dosimeter, respectively. From Eqs. (3) and (4), the relationship between the total exposure rate, $\dot{X}$, and mean effective energy, $\bar{E}$ of radiation due to such radionuclides will be given by:

$$\bar{E}=E_1+(\dot{X}_2(E_2-E_1)/(\dot{X}_1+\dot{X}_2)) \text{ or}$$

$$\dot{X}=\dot{X}_2(E_2-E_1)/(E-E_1). \qquad (5)$$

From Eq.(5), it is clear that the relationship between the total exposure rate and the mean effective energy of photon radiation from two radionuclides can be represented by a hyperbolic function. If it is assumed that X_2 is constant and X_1 approaches positive infinity or zero, Eq.(5) is presented as follows

$$\lim_{X_1/X_2\to\infty} X=X_1,\ \lim_{X_1/X_2\to\infty} \bar{E}=E_1 \qquad (6)$$

$$\lim_{X_1/X_2\to 0} X=X_2,\ \lim_{X_1/X_2\to 0} \bar{E}=E_2. \qquad (7)$$

Accordingly, if the exposure rate of one of two radionuclides is constant and if that of another radionuclide is varied, as given in Eq.(5), the relationship between the total exposure rate and the mean effective energy of photon radiation due to two radionuclides showed to represent a hyperbolic function and, under the conditions of Eqs.(6) and (7), Eq.(5) will be rearranged as follows;

$$\dot{X}=A+C/(\bar{E}-B), \qquad (8)$$

where A and B in Eq.(8) correspond to $\dot{X}_2$ and F_1 from Eqs(6) and(7).

The conception of this relationship can be applied to natural radiation consisting of both terrestrial gamma-rays and cosmic-rays. The exposure rate and effective energy of photon radiation from terrestrial radiation are $\dot{X}_t$ and E_t, respectively. It is assumed that the quasi-exposure rate, $\dot{X}_c$ of cosmic-rays with the quasi-effective energy of E_c is constnat and that a value of E_c-E_t is constant at all the monitoring points.

If $\dot{X}_t\gg\dot{X}_c$ is considered, for this limit as given by Eq.(6), $\bar{E}=E_t$ and $\dot{X}=\dot{X}_t$. Furthermore, if $\dot{X}_t=0$, the limit as given by Eq.(7) then get; $\bar{E}=E_c$ and $\dot{X}=\dot{X}_c$. Accordingly, the coefficients A and B in Eq.(8) become to be $\dot{X}_c$ and E_t.

This result reveals that if both values of E_c-E_t and $\dot{X}_c$ will be constant at all the monitoring points, or if a case of $E_c\gg E_t$ will be considered and $\dot{X}_c$ will be constant, the relationship between the values of $\dot{X}_q$ and E_q is presented as a hyperbolic function.

According to a result of calculation from data on ratio of uranium U, thorium Th and potassium K to gamma ray dose from terrestrial radiation at 56 monitoring points in Fukui[3], Kagoshim[4] and Ehime[5] Prefectures, a mean energy of gamma-rays was 1.099±0.031MeV and a variation of the coefficient on the mean energy was about 3 percent. Therefore, a value of E_c-E_t was assumed to be constant at all the monitoring points in this work, On the other hand, the value of $\dot{X}_c$ was assumed to be constant as will be mentioned in the Experiment. The coefficient A,B and C were obtained in the following experiment.

3. EXPERIMENT

The TLD phoshors used for the experiment was $CaSO_4$(Tm) (purchased from Matsushita Electric Industrial Co., Osaka), the compound having been chosen for the large atomic number and the consequent high sensitivity, indispensable for use in the filter method. The phosphor, received in powdered from, were encapsulated in small glass tubes measuring about 12 mm by 2 mm diameter, and nominally free of ^{40}K and other radioactive elements that can influence TL emission.

Lead and lucite were used as the filter pair, for the outstandingly high energy dependence shown by the TL_1/TL_2 ratio through the energy region of 100 keV~1.25 MeV. The 50 mm long cylindrical lead vessel serving as the filter was varied in wall thickness from 0.1 to 15 mm. The lucite vessel of the same 50 mm length as the lead filter, had the same shape and size for all runs, of 10 mm outside diameter, and with its axial pocket measuring 2.3 mm in diameter, 45 mm deep. In both kinds of vessel were contained three TLD elements of $CaSO_4$(Tm).

In order to determine the dependence of TL_1/TL_2 ratio on the radiation energy, the pair filter TLDs were irradiated by means of an ordinary X-ray generator as well as γ-rays from ^{137}Cs and ^{60}Co sources. The effective energy of X-rays was determined by half value layer method.

About five monitoring stations in the field at each prefectural Institute of Public Health in nine prefectures in Japan. The altitudes and the latitudes of all stations were in the range from a few to 30 meters, and 31 to 39 degrees, respectively. Two monitors per monitoring station were left in a wood or a polyvinyl chroride box in the field at 1.5 meters above-ground level. About three months later, both the monitor and background monitor were returned together to the author's Institute.

The TL response from irradiated TLDs was measured by integration method using Kyokko-TLD 1200 reader system.

4. RESULTS AND DISCUSSION

1) Energy Dependence of TL_1/TL_2 Ratio

The experimentally determined relation between the TL response ratio TL_1/TL_2 observed by pair filter TLD and the effective

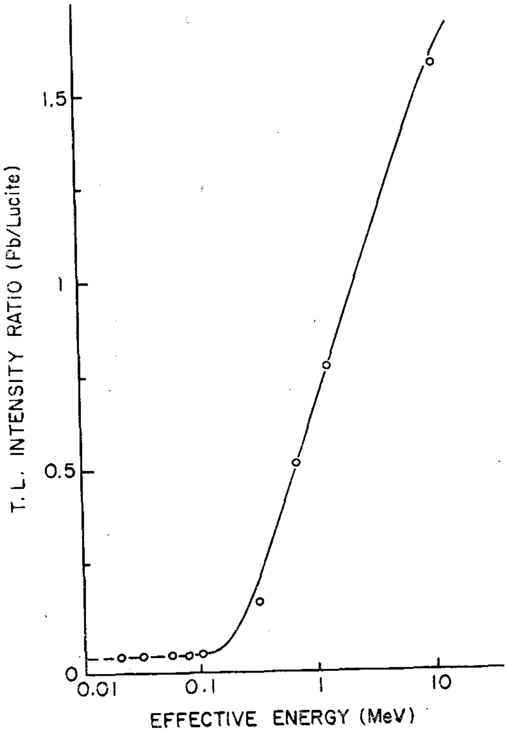

Fig. 1. Dependence of the ratio between TL intensities of the $CaSO_4$(Tm)TLDs in Pb and lucite filters upon a pair monitor on keV of photon beams. Wall thicknesses of Pb and lucite filters is 10g/cm^2 and 0.45g/cm^2, respectively.

photon radiation energy is presented in Fig. 1. The plots were obtained with $CaSO_4$(Tm) phosphor, both covered by filter pairs of 10 g/cm^2 thick lead and 0.45 g/cm^2 thick lucite. The plots reveal a sharp rise of the TL_1/TL_2 ratio in the range of effective energy above 200 keV, While below this range, TL_1/TL_2 is practically at zero level, which agrees with theoretical calculation.

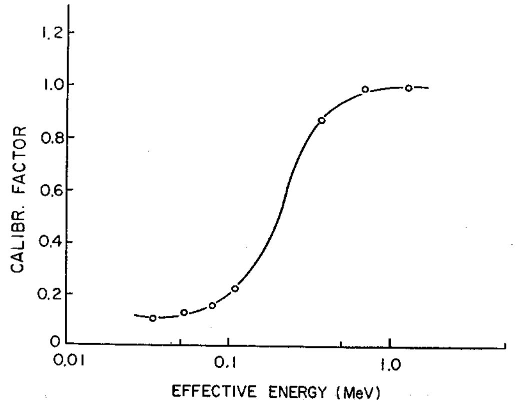

Fig. 2. Energy dependence of relative TL response obtained with lucite filter.

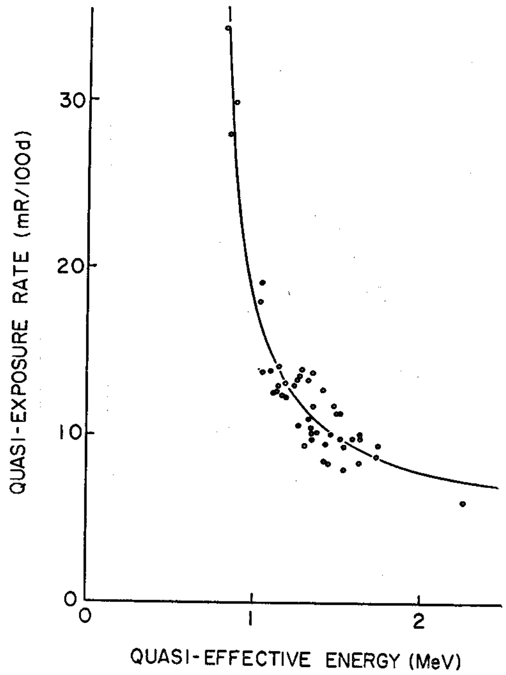

Fig. 3. Relationship between the quasi-exposure rate and the quasi-effective energy of natural radiation in the field by means of the pair-filter TLD system. The solid curve gives a hyperbolic function obtained using the least-square method. The present quasi-effective energy of natural radiation in the field excluded the hard component of cosmic-rays.

For extending the range of measurement below 200 keV, to permit monitoring the quasi-effective energy of natural radiation in the low energy region, the 10 g/cm^2 lead filter would require replacement. The pair filter method was therefore tried with this lead filter replaced by a commercially marketed UD 200 s TLD holder, purchased from Matsushita Electric Industrial Co.

In the range of low-energy radiation, the TLD phosphor with lucite filter will play an important role in estimating the exposure by pair filter method. For this reason, the energy-dependence of TL sensitivity was examined on the $CaSO_4$(Tm)-TLD placed in 0.45 g/cm^2 thick lucite vessel. The result is shown in Fig. 2, which indicates a TL response ratio somewhat lower than for a corresponding TLD without filter, in the energy range of 30~50 keV. This lowering of sensitivity by the presence of filter, however, is not to the extent of impairing the evaluation of exposure with the TLD, and multiplication of the energy calibration factor applied to the radiation and TL responses should permit estimation of exposure to required accuracy.

2) Application to Natural Radiation

The data on the exposure rate for 100days and the quasi-effective energy of natural radiation in the field have been plotted in Fig. 3.

The quasi-effective energy and the exposure rate of natural radiation, as shown in Fig. 3, are distributed in a wide range from about 0.8 MeV to 2.3 MeV and from about 6 to 35 mR/100days, respectively. The quasi-effective energy at a majority of the monitoring stations was in the range from 1.0 MeV to 1.5 MeV.

As shown in Fig. 3, the relationship between the quasi-effective energy and exposure rate rate of natural radiation can be represented by a hyperbolic function.

From Eq. (8), it is clear that the relationship between the quasi-exposure rate and quasi-effective energy of natural radiation can be represented by a hyperbolic function under the two conditions, as above mentioned. Then, the coefficients of the hyperbolic function were calculated from the data on the quasi-effective energy and the quasi-exposure rate of natural radiation using the least-squares method. In the calculation, both the quasi-exposure rate from cosmic-rays and a farction of natural radioactive nuclides are assumed to be a constant at all the monitoring points. From Eq. (8), $\dot{X}_q$ is the quasi-exposure rate, (mR/100days) of natural radiation; then A (mR/100 days) is the quasi-exposure rate $\dot{X}_c$ due to only the cosmic-rays which is the function of solar flare activity, altitude, geographic latitude and other factors of the monitoring point; B (MeV) is the minimum quasi-effective energy of natural radiation, corresponded to the effective energy of photon from terrestrial radiation at the fields; C (mR MeV/1000 days) is a constant; and E_q(MeV) is the quasi-effective energy of natural radiation at any monitoring point in the field.

Eq. (8) was presented with the following constants: A=4.60 mR/100 days, B=0.679 MeV and C=4.27 mR MeV/100 days,

$$X_q = 4.60 + \frac{4.27}{E_q - 0.679} \qquad (9)$$

In Eq. (9), it is assumed that in case of E_q of 3 MeV or more, $\dot{X}_q$ is 4.60 mR/100 days because natural radiation almost consist of dose from cosmic-rays under such condition and second term seem to be zero.

In Fig. 3, the solid curve is a hyperbolic function with coefficients obtained using the least-squares method. All observed data falls within 24% of the value calculated using Eq. (9).

5. CONCLUSION

The relationship between the quasi-effective energy and quasi-exposure rate of natural radiation in the field with both same cosmic-ray dose and a constant of the energy difference, $E_c - E_t$, has been studied and the following items are concluded.

1) The relationship between the quasi-exposure rate and quasi-effective energy of natural radiation can be represented as a hyperbolic function.

2) The quasi-exposure rate of cosmic-rays can be obtained from Eq. (8) and, further, the value of 2.96 to 3.43 μR/h in the present work is in good agreement with values obtained using ionization chamber and scintillation detector methods.

3) The minimum quasi-effective energy of natural radiation is the effective energy of photon radiation from terrestrial radiation and is about 0.68 MeV

4) The relationship of a hyperbolic function between the values $\dot{X}_q$ and E_q of natural radiation can be applied under the above mentioned conditions in all cases.

ACKNOWLEDGEMENTS

The author wishes to thank the staff members of the Prefectural Institutes of Public Health in the following prefectures in Japan for their supporting of this work: Fukushima, Fukui, Shimane, Niigata, Shizuoka, kagoshima, Ehime, Saga and Miyagi. The author also wishes to express his thanks to Dr. T. Kumatori, Director General of his Institute, Dr. T. Terashima, Deputy Director General of his Institute, for their encouragement on the present work, and to Mr. M. Okazaki and Mr. K. Nemoto for assistance on the radiography of Pb filter and on observation from activities in the Pb filter using whole body counters and semiconductor detectors. The author would also like to thank Mr. T. Koshijima and Dr. K. Fujitaka for their helpful discussion concerning this work and to Mr. M. Nenoi for his assistance in the calculation of the least-squares method.

REFERENCES

1) S.G. Gorbics and F.H. Attix: *Int. J. Appl. Radiat. Isot.*, **19**, 81(1986).
2) T. Nakajima and M. Chiba: *J. Nucl. Sci. Techn.*, **23**, 258 (1986).
3) H. Tokuyama, et al., *Rept. Fukui Pref. Inst. Public Health* 31-46, (1983).
4) S. Shimozono, et al., "In-Situ measurment of the environmental gamma rays by portable Ge detector", *Ann. Rpt. Kagoshima Pref. Inst. Env. Sci.*, **2**, 236-243(1986).
5) T. Kaneko, Private communication, 1987.