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ORIGINAL PAPER

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MTS-6 DETECTORS CALIBRATION BY USING ^{239}PU-BE NEUTRON SOURCE

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Abstract

Background: Thermoluminescent detectors, type MTS-6, containing isotope ^{6}Li (lithium) are sensitive in the range of thermal neutron energy; the ^{239}Pu-Be (plutonium-and-beryllium) source emits neutrons in the energy range from 1 to 11 MeV. These seemingly contradictory elements may be combined by using the paraffin moderator, a determined density of thermal neutrons in the paraffin block and a conversion coefficient neutron flux to kerma, not forgetting the simultaneous registration of the photon radiation inseparable from the companion neutron radiation. The main aim of this work is to present the idea of calibration of thermoluminescent detectors that consist of a ^{6}Li isotope, by using ^{239}Pu-Be neutron radiation source. **Material and Methods:** In this work, MTS-6 and MTS-7 thermoluminescent detectors and a plutonium-and-beryllium (^{239}Pu-Be) neutron source were used. Paraffin wax fills the block, acting as a moderator. The calibration idea was based on the determination of dose equivalent rate based on the average kerma rate calculated taking into account the empirically determined function describing the density of thermal neutron flux in the paraffin block and a conversion coefficient neutron flux to kerma. **Results:** The calculated value of the thermal neutron flux density was 1817.5 neutrons/cm^2/s and the average value of kerma rate determined on this basis amounted to 244 μGy/h, and the dose equivalent rate 610 μSv/h. The calculated value allowed for the assessment of the length of time of exposure of the detectors directly in the paraffin block. **Conclusions:** The calibration coefficient for the used batch of detectors is $(6.80\pm0.42)\times10^{-7}$ Sv/impulse. Med Pr 2017;68(6):705–710

Key words: thermoluminescence, calibration, thermoluminescent detectors, neutron, neutron source, MTS

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INTRODUCTION

The role of thermoluminescent detectors (TLD) based on lithium fluoride (LiF) in ionizing radiation field dosimetry is recognized and widely used by many dosimetry services in Poland and abroad [1–4]. Aside from natural lithium (consisting of 92.41% ^{7}Li and 7.59% ^{6}Li) the production of thermoluminescent detectors also uses lithium fluoride enriched and depleted in the isotope ^{6}Li. The use of 2 Li isotopes (^{6}Li and ^{7}Li) extends the use of detectors for individual dosimetry in mixed ionizing radiation fields.

The spectrum of neutrons energy is usually widely, starting from the thermal-energy neutrons through intermediate-energy neutrons to the high-energy neutrons. Due to: the fact that values of cross section strongly vary in energy for various reactions, the differences in these reactions in the tissue and the detector, the 2-step process of transferring the energy of neutrons to the medium, and the weighting factor (w_R) varying over a wide range of radiation – w_R means that the dose measurements of neutron radiation are difficult and the result is not as accurate as in the case of photon-radiation dosimetry.

One of the several methods that use the thermoluminescence phenomenon for individual neutron dosimetry method is the albedo dosimeter method, that is – the back-scattering of neutrons from the body. One of the simpler dosimeters in this regard includes 2 pairs of ^{6}Li – ^{7}Li detectors, one of which is covered in a cadmium layer from the body side, the other from the side of incident radiation. Detectors with the isotope ^{6}Li have a high value of cross section for reaction with thermal neutrons via the ^{6}Li$(n,\alpha)^{3}$H reaction, while detectors with ^{7}Li are practically insensitive to neutrons in that range of energy. That is the so called pair method, in

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which the detectors used record the thermal neutrons scattered from the body as part of its electromagnetic radiation.

Formally, the difference in the height of the main peak (or surface area) thermoluminescent curves for detectors with isotopes ^{6}Li and ^{7}Li, placed in the same measuring point, resulting from the differences in cross sections for ^{7}Li and ^{6}Li on the thermal neutron absorption, allows for the calculation of the contribution of thermal neutrons to the total TLD signal in that measurement point. Calibrating these differences of responses of the detectors with isotopes ^{7}Li and ^{6}Li in respective dosimetric units may, under certain conditions, determine the value of the neutron dose [5]. The calibration in this case is not straightforward. An additional difficulty is to obtain a result from the detector exposed to the neutron radiation in units of equivalent dose and also the respective energy of neutrons emitted by a neutron source. A pair of detectors ^{6}Li – ^{7}Li (MTS-6–MTS-7), without using the appropriate dosimetric cassette but with the high energy neutron source, may be used for calibrating one such detector. The paper presents a method of calibration of thermoluminescent detectors made on the basis of ^{6}LiF: Mg, Ti (MTS-6), using a plutonium-and-beryllium (^{239}Pu-Be) neutron source.

MATERIAL AND METHODS

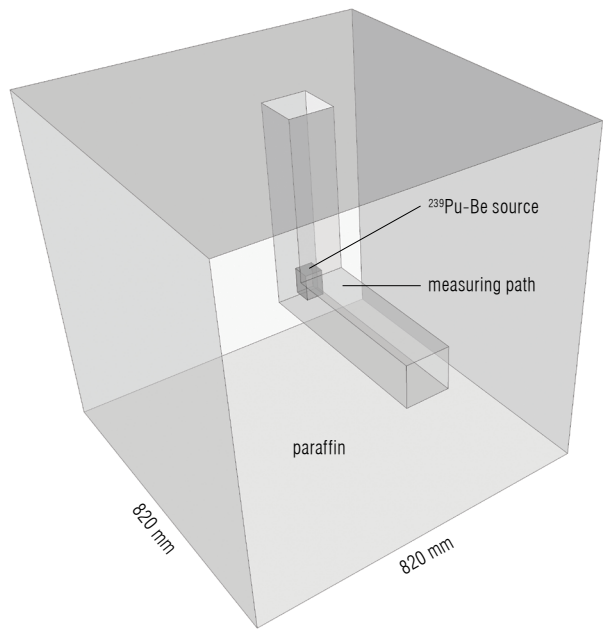
Thermoluminescent detectors – MTS

In this work, MTS-6 and MTS-7 thermoluminescent detectors were used, which are respectively ^{6}LiF: Mg (magnesium), Ti (titanium) and ^{7}LiF: Mg, Ti. Both detectors are manufactured by the Polish company RADCARD. One hundred type MTS-6 and MTS-7 detectors were used during the measurements. Before the measurements, each of the detectors had been assigned an individual number so as to distinguish the MTS-6 from MTS-7 detectors. The TLDs are in the form of small pastilles (4.5 mm diameter, 0.9 mm thick). These detectors require a two-step annealing process before exposure, at 400°C as well as for 2 h at 100°C. To read the exposed detectors, the Reader Analyser RA'04 was used. Isotopes ^{6}Li and ^{7}Li, used in these detectors, allows the recording not only in the field of neutron radiation corresponding to thermal neutrons, but also associated with electromagnetic radiation.

The cross section for the reaction of (n, α) for isotope ^{6}Li is 940 b. In contrast, cross-sections for the reaction of (n, γ) in ^{6}Li and ^{7}Li b are 0.039 and 0.045 b, respectively [6]. Thermoluminescent detectors MTS-6 are recording the neutron and photon radiation while MTS-7 only records photon radiation. The difference in the response of the detectors used at the same time provides the information about the number of counts coming from neutron radiation; after appropriate calibration the information about the dose that these detectors were exposed to is obtained as well.

Neutron source

The neutron sources used in the study was a plutonium-and-beryllium source (^{239}Pu-Be), which was a mixture of powdered plutonium (^{239}Pu) and beryllium (^{9}Be), enclosed in a capsule made of metal. Emission of neutrons is carried out via (α, n) reaction. The activity of the source is 10^{6} neutrons/s, emitted in the full solid angle [7]. Half-life of plutonium is 2.44×10^{4} years, which applies to the entire source [7]. The source is placed within a paraffin block in the form of a cube of side length about 820 mm. A schematic structure of the paraffin block comprising a source of ^{239}Pu-Be is presented in the Figure 1.



^{239}Pu-Be – plutonium and beryllium.

Fig. 1. Paraffin block containing ^{239}Pu-Be neutron source

The formation of neutrons in the source takes place in 2 stages. In the first one, the ^{239}Pu isotope decays by emitting α particles. In the second one, the α particles bombard the ^{9}Be that forms part of the source.

This leads to a nuclear reaction. The isotope ^{12}C and a neutron are products of this reaction. Since the energy of this reaction is distributed between the resulting isotope ^{12}C and the neutron, depending on the excited state of formed isotope ^{12}C – the neutron gets the rest of the energy.

At the same time, the excitation of the carbon isotope depends on the energy of incident particles α which may also lose energy within the source, which means that the neutrons produced in the reaction are not monoenergetic.

The neutrons produced directly from the ^{239}Pu-Be source are fast neutrons with energies contained in the range of 1–11 MeV. Paraffin wax fills the block, acting as a moderator. As a result of the slowdown, we receive the neutrons that remain in thermal equilibrium with the moderator, which at temperature of 293 K corresponds to the energy of neutrons at about 0.025 eV. The role of the moderator is not just to slow down the neutrons to thermal energy; it also prevents an immediate escape from the area surrounding the source. In the slowed down neutrons energy spectrum, there is a small number of neutrons with energies higher than thermal; those energies are called resonances (0.5–1 keV).

Flux density of neutrons in a block of paraffin containing a ^{239}Pu-Be source

Thermal neutron flux density in a block of paraffin moderator was specified as part of the unpublished work [7]. This chapter presents an idea of measuring the flux density of neutrons emitted from a ^{239}Pu-Be source by using a surface-barrier detector (solid state detector). The diameter of the active surface of the detector was 25 mm. Registration efficiency of α particles and tritons reached 100%. During the measurements 2 targets were used as well, composed of lithium fluoride (LiF) spread on aluminum substrate with a thickness of 200 μm.

The detector with a target was installed in a vacuum chamber, allowing for the removal of air. The pressure in the chamber was 5×10^{-2} mm Hg (after pumping the air out). The energetic calibration of the spectrometer path was performed by using a source of ^{234}U (uranium) and ^{238}U. Measurements in the paraffin block for targets containing the isotope ^{6}Li were realized for 6 selected measurement points, inside the activation path in which a chamber with the target and the detector were placed. The positions of points were measured from the edge of the paraffin block. At each of the points, there were 3 measurements performed, with the target with and without a cadmium shield and also to determine the background for an aluminum substrate of targets with/without a cadmium shield.

Measurements were also made to determine the contribution of resonance neutrons. The vacuum chamber was surrounded by a cover of a metal cadmium sheet and the measurements were made with the target and without the target. Three control measurements were performed as well for a target containing 3.65% of isotope ^{6}Li without a cadmium cover at the point closest to the source. A correction was made for the particles' solid angle of detection. For this purpose, numerical methods were used to count the usable solid angle of detection for given geometrical parameters.

It was assumed that the emission of particles from the target was isotropic and also that the density of the target over the entire surface was isotropic. The correctness of the method of calculating the solid angle was confirmed experimentally by changing the distance of the target – detector. The difference between the number of registrations with the results obtained by calculation does not exceed 2.1%. The flux of thermal neutrons (Φ) has been determined on the basis of the relationship:

$$\Phi = \frac{N}{n_j \sigma \omega t} \quad (1)$$

where:
N – the number of registrations of tritons or α particles,
n_j – the number of nuclei in the target ^{6}Li,
σ – reaction ^{6}Li(n, α)^{3}T cross section,
ω – correction to the particles solid angle of detection,
t – time of measurement.

Calibration TL detectors by using ^{239}Pu-Be source

The idea [8] was based on the determination of dose equivalent rate (H), which is given by:

$$H = Kw_R \quad (2)$$

where:
$K = \Phi d_k$ – the average kerma rate,
w_R – weighting factor for neutrons with energies corresponding to the energy of thermal neutrons and is 2.5,
d_K – fluence-kerma conversion factor is 3.73×10^{-9} rad×cm^2/neutron for ^{239}Pu-Be source [9].

The empirically determined function describing the density of thermal neutron flux depending on the dis-

tance from the source allowed to specify the value of flux density of thermal neutrons in the paraffin block at a distance of 38 cm from the edge of the paraffin block, which means the nearest point of the source. This, in turn, allows to determine the values of kerma rate and dose equivalent rate and finally to assess the time length of the detectors' exposure at the source.

Exposure of calibration

For the calibration process, the 14 MTS-6 and MTS-7 detectors were placed in a paraffin block within 2 cm from the source of ^{239}Pu-Be for 72 h, 144 h, 240 h and finally 480 h. The background was included individually for type MTS-6 and MTS-7 detectors.

RESULTS

The calculated value of the thermal neutron flux density was 1817.5 neutrons/cm^2/s and the average value of kerma rate determined on this basis amounted to 244 μGy/h, and the dose equivalent rate stood at 610 μSv/h. The calculated value allowed for the assessment of the length of time of exposure of the detectors directly in the paraffin block.

MTS-6 and MTS-7 detectors were placed in a paraffin block containing the source of ^{239}Pu-Be for the time of 72 h, 144 h, 240 h and 480 h. After this time, the detectors were removed and read.

The Table 1 includes the calculated neutron flux density (Φ) as a function of the distance (L) from the edge of the paraffin block. Finally, the flux density (Φ (L)) of the thermal neutrons as a function of distance from the edge of the block (L) is described by the relationship:

$$\Phi(L) = 148 \times \exp(0.066 \times L) \quad (3)$$

The total uncertainty of the thermal neutrons' flux density assessment does not exceed 3.5%.

The Table 2 contains the exposure time of detectors and calculated values of dose equivalent rate.

As a result of the exposure and reading of detectors, a calibration curve was obtained, as presented in the Figure 2.

The calibration curve describes the relationship:

$$H = 6.80 \times 10^{-7} \times N \quad (4)$$

where:
H – dose equivalent,
N – impulse.

Table 1. Calculated neutron flux density (Φ) as a function of the distance (L) from the edge of the paraffin block containing a ^{239}Pu-Be source

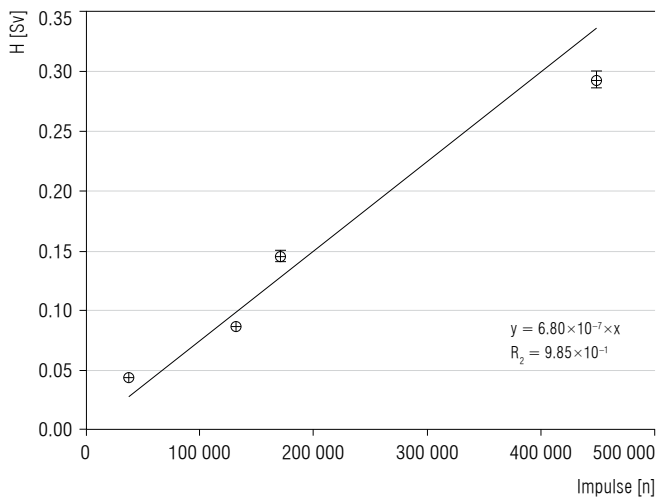
Distance [cm]	Neutron flux density [neutrons/cm^2/s] (M±SD)	
	calculated based on registered tritons	calculated based on recorded α particles
39.7	1 597±45	1 563±27
33.6	1 285±41	1 249±24
27.6	933±19	908±28
21.6	641±16	634±11
15.6	415±13	417±12
10.3	285±8	284±9

^{239}Pu-Be – plutonium and beryllium.

Table 2. Calculated dose equivalent rates and the assessed thermoluminescent detectors (TLD) exposure time in the paraffin block containing a ^{239}Pu-Be source

Dose equivalent rate [Sv/h]	Detectors exposure time [h]
0.044±0.001	72
0.087±0.002	144
0.146±0.004	240
0.293±0.007	480

^{239}Pu-Be – plutonium and beryllium.



^{239}Pu-Be – plutonium and beryllium.

Fig. 2. Calibration curve for MTS-6 thermoluminescent detector describing the relationship between the dose equivalent (H) and impulses received after detector's readout

The slope, combined with uncertainty, is (6.80± 0.42)×10^{-7} Sv/impulse.

DISCUSSION

The purpose of this work is to present the idea that enables the calibration process of thermoluminescent detectors that record thermal neutrons by using a ^{239}Pu-Be source that emits neutrons in the energy range 1–11 MeV. The subject of thermoluminescent detector calibration in the neutron radiation field is difficult. The following issues pose a problem in this case:

- in practice, we do not have access to "convenient to use" natural neutron sources. Typically, the mechanism of production is associated with spontaneous fission or nuclear reactions and this means that there are neutrons in a wide energy spectrum of thermal energy (0.025 eV) to the high energy (tens of MeV),
- neutrons are always present together with photons, biological effects of which are different from the action of neutrons,
- calibration in units of equivalent dose requires the application of an appropriate phantom,
- high cross section of ^{6}Li and the low one of ^{7}Li impacts the absorption of thermal neutrons,
- source ^{239}Pu-Be emits neutrons with the energy range 1–11 MeV.

Most often, as it may be seen in publications from recent years, even simulation methods [10–12] are used in this regard.

Since the original objective of the authors was to use the secondary standard – Bonner sphere (and on the basis of its indications – to determine the doses of neutron radiation, which should be used for exposing the TLD). Unfortunately, the size of the paraffin block does not allow to perform measurements by using Bonner sphere. Therefore the method, that was used, was based on the experimental determination of the flux density of thermal neutrons in a paraffin block and also used a flux to kerma conversion coefficient that allowed for the determination of dose values used to exposed the detectors.

The purpose of the study described in the publication has been achieved.

CONCLUSIONS

The calibration procedure of MTS-6 detectors was performed in units of the equivalent dose using the ^{239}Pu-Be neutron source. The information about the flux density of thermal neutrons designated for different distances between the source and the detector and a flux to kerma conversion coefficient was used, which allowed for the determination of dose values used in respect of exposed detectors. The carrying out of the calibration procedures of the detectors forced the selection of detectors for the uniformity of their responses.

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