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ORIGINAL PAPER

Application of LiF:Mg,Cu,P (MCP-N) thermoluminescent detectors (TLD) for experimental verification of radial dose distribution models

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Abstract. In track structure theory, the radial distribution of dose, $D(r)$, around an ion track plays a fundamental role in predicting the response of biological systems and physical detectors after a dose (or fluence) of ions. According to the formulations of $D(r)$, the local dose at radial distances below 1 nm can reach values as high as 10^6 Gy. We propose a new method of verifying experimentally the radial dose distribution around α-particle tracks, using LiF:Mg,Cu,P (MCP-N) thermoluminescent detectors (TLD) which are able to measure γ-ray doses in the kGy range via evaluation of their high-temperature TL glow peak structure over the temperature range of 350–550°C. MCP-N detectors were irradiated with Am-241 α-particles at fluences ranging from 10^7 to 10^{11} particles/cm^2, and by Co-60 γ-ray doses ranging from several Gy up to the MGy. A number N of individual high-temperature TL peaks were analysed in the obtained glow curves by deconvolution, using the GlowFit code. For each of these peaks, an equation relating the intensity, A, of the TL signal obtained after α-particle irradiation and after γ-ray doses, via the dose-frequency function, $f^\alpha(D)$, was written in the form: $A_\alpha^i = \int A_\gamma^i(D) \cdot f^\alpha(D)dD,\ i = 1,..,N$. Using this set of N equations, where A_α^i and $A_\gamma^i(D)$ were known (measured), the single unknown function $f^\alpha(D)$ was unfolded and converted to $D(r)$. Parametric unfolding and the SAND-II iterative code were applied. While we were able to confirm the $1/r^2$ dependence of $D(r)$ in agreement with $D(r)$ expressions, we were unable to conclusively evaluate the dependence of $D(r)$ at intermediate ranges of radial distance r. This preliminary result of our unique experimental approach to determine the radial dose distribution around the path of heavy charged particles in LiF detectors, requires further development.

Key words: heavy charged particles • LiF:Mg,Cu,P (MCP-N) • radial dose distribution • thermoluminescence • unfolding

Introduction

A heavy charged particle (HCP) passing through matter undergoes interactions with the medium by depositing energy through atomic excitations and ionizations. In the latter case secondary charged particles, mainly electrons, are produced. These secondary particles mediate the transfer of energy of the primary particle. The HCP creates a trail of local energy deposition events which is referred to as the radial distribution of dose.

The radial distribution of dose, $D(r)$, is one of the fundamental concepts used to describe tracks of heavy charged particles. It is also applied in several radiobiological models of cell survival [4, 12]. Until now, models based on the $D(r)$ concept were mainly used to predict biological effects and the response of physical detectors after heavy ion irradiation. Most of the analytical formulations of $D(r)$ show that at radial distances below 1 nm from the path of the heavy ion track, the local dose can reach values as high as 10^6 Gy [13, 14], but this has never been verified experimentally. Furthermore, thermoluminescent (TL) detectors have never been used for this purpose, because the γ-ray dose response of most TL detectors typically saturates around 10^3 Gy.

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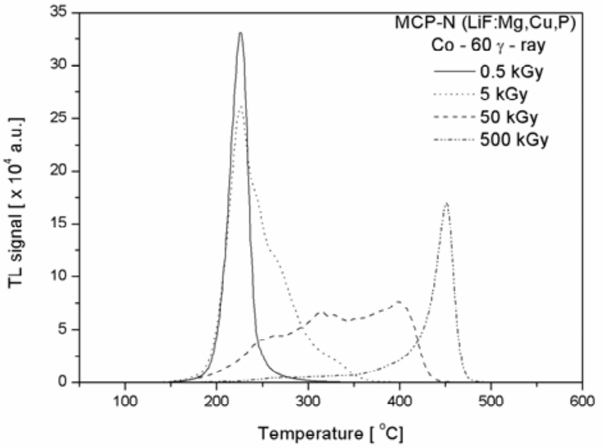


Fig. 1. Non-normalized TL glow curves of MCP-N detectors after irradiation with doses of Co-60 γ-rays.

However, noting the high-temperature structure of TL glow curves of MCP detectors, observed by Bilski *et al.* in 2008 [1], we wished to explore the possibility of using these detectors to verify the analytical $D(r)$ formulae over the whole range of doses and radial distances.

The high-sensitive MCP-N-type TL detectors are based on lithium fluoride doped by magnesium, copper and phosphorus (LiF:Mg,Cu,P). MCP-N detectors can be used to measure doses of ionizing radiation ranging from a few microgray (μGy) up to at about 1 kGy where response saturation occurs. Over this dose range, the response of MCP-N is quite linear, and in its glow curve the shape of the main dosimetric peak, with its maximum around 220°C, remains practically unchanged [9]. After exposure to very high doses of γ-radiation above about 1 kGy up to 1 MGy, a high temperature glow peak structure appears [1, 8]. For doses up to about 50 kGy, the structure occurs predominantly at temperatures between 250°C and 450°C. For doses up to 1 MGy, the peak structure continues to grow and gradually shifts towards higher temperatures, up to 500°C. TL glow curves of MCP-N detectors after irradiation with Co-60 γ-ray doses of 0.5, 5, 50 and 500 kGy, as measured by Obryk [7], are plotted in Fig. 1. For the purpose of this work, it should be noted that MCP-N detectors are known to measure doses of α-particles with low efficiency, typically 0.04–0.06, relative to γ-ray doses [3].

In this work we make an attempt to link the experimental data from irradiations of MCP-N detectors with Co-60 γ-rays and Am-241 α-particles over a wide range of doses and fluences, to propose an innovative method of exploiting the high-dose range capability of the MCP-N detector to verify experimentally the expressions describing the radial dose distribution, $D(r)$. To our knowledge, this is the first attempt at such a verification of $D(r)$ formulae in solid-state LiF-based TL detectors.

Materials and methods

The approach

It is assumed that the TL glow curve of MCP-N detectors after a dose D of γ-rays, $I^\gamma(D)$, can be deconvoluted into N individual peaks $i = 1,\ldots, N$:

$$I^\gamma(D) = \sum_{i=1}^{N} A_i^\gamma(D) \cdot I_i^\gamma \tag{1}$$

where $A_i^\gamma(D)$ is the γ-ray dose (D)-dependent peak amplitude and I_i^γ is the TL peak of unit amplitude described by the first-order kinetics. It is also assumed that the TL glow curve, $I^\alpha(F)$, after α-particle irradiation, can be deconvoluted using the same set of individual peaks as for γ-rays:

$$I_i^\alpha = I_i^\gamma = I_i \tag{2}$$

differing only in their amplitudes, A_i (Fig. 3). Then:

$$I^\alpha(F) = \sum_{i=1}^{N} A_i^\alpha(F) \cdot I_i \tag{3}$$

where F is the fluence of α-particles. If F is small enough for the probability of overlapping of α-particle tracks to be negligible (is close to zero), then the peak amplitude is proportional to F.

$$I^\alpha(F) = c\, F \sum_{i=1}^{N} A_i^\alpha \cdot I_i \tag{4}$$

where c is the proportionality constant. The dose distribution in the α-particle track is strongly non-uniform and the local dose (point dose) can vary significantly, (see equations of radial dose distribution by Katz [4], Waligórski *et al.* [13] or Zhang *et al.* [14]), from $D_{\min}$ to $D_{\max}$, even reaching doses of MGy range. The response of the detector, irradiated with α-particles, A_i^α is equal to the integrated response of the detector (as a function of dose), measured for the irradiation with γ-rays, $A_i^\gamma(D)$ weighted over a dose frequency distribution function, $f^\alpha(D)$.

$$A_i^\alpha = \int_{D\min}^{D\max} A_i^\gamma(D)\, f^\alpha(D)\, dD, \quad i = 1,\ldots, N \tag{5}$$

where $f^\alpha(D)$ is the frequency distribution of dose D within a single track of an α-particle. Having measured A_i^α and $A_i^\gamma(D)$ the $f^\alpha(D)$ function can be unfolded and later converted to the radial dose distribution $D(r)$, using the following formula:

$$\int_{D\min}^{D_{\max}} D \cdot f^\alpha(D)\, dD = \frac{2\pi}{r_\delta^2} \int_0^{r_\delta} r \cdot D(r)\, dr \tag{6}$$

The radial dose distribution $D(r)$ is defined as a point dose D at the radial distance r from the track axis. It is assumed here that ion paths are straight lines. $D_{\max}$ is defined for $r = 0$, while $D_{\min}$ corresponds to the maximum range of δ-rays, r_δ, as shown in Fig. 2 r_δ is a function of the α-particle energy, since the α-particle

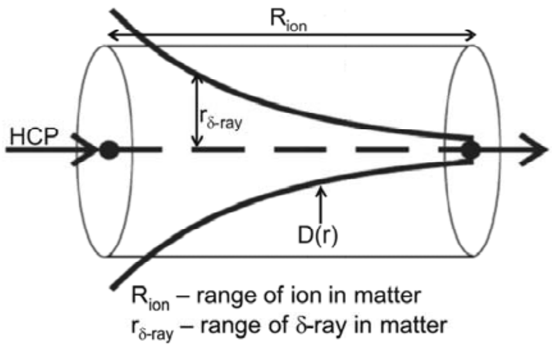


Fig. 2. Track structure of a heavy charged particle.

slows down along its path in the detector. Therefore, in order to use Eq. (6) to calculate the $D(r)$ distribution, additional integration over the energy of the α-particle is required. Since such calculations are complicated, as a result of this work the $f^\alpha(D)$ function, evaluated from Eq. (5) and the corresponding function resulting from model calculations, using a given $D(r)$ formula, will be compared.

Unfolding methods

Since a finite number of discrete observables, A_i^α cannot, in general, define a continuous function $f^\alpha(D)$, such a system of equations is, in most cases, undefined and does not have a unique solution. Therefore, in the unfolding procedure to yield $f^\alpha(D)$, some additional assumptions as to the expected response function should be applied, e.g. non-negativity, continuity, smoothness, monotonicity, etc. As the values of A_i^α are known with a certain experimental error, δA_i^α Eq. (5) can be rewritten in the following form:

$$(7) \qquad A_i^\alpha \pm \delta A_i^\alpha = \int_{D'} A_i^\gamma(D)\, f^\alpha(D)dD \qquad i = 1, ..., N$$

Due to the above-mentioned assumptions on the behaviour of the function $f^\alpha(D)$, the system can become overdeterminated which justifies looking for approximate solutions, e.g. in the least squares sense.

The quality of the unfolded function can be verified by introducing additional criteria. Let us assume that A^α_{calc} is the effect calculated for a particular response function. Minimizing the following quantity:

$$(8) \qquad Q = \left(\frac{A^\alpha_{\mathrm{exp}} - A^\alpha_{\mathrm{calc}}}{A^\alpha_{\mathrm{calc}}} \right)^2$$

can constitute a criterion for finding the best matching function.

Unfolding with discrete intervals

There is a broad group of numerical methods that can be used to evaluate the $f^\alpha(D)$ function from Eq. (7). Here, two methods are of particular interest.

In the least squares method, requiring matrix inversion, the response function is determined by solving Eq. (7) in the least square sense. This procedure can be continued iteratively until a satisfactory solution is obtained. The advantage of this method is that the covariance matrix corresponding to the unfolded function gives some information on the error involved. This method, however, does not exclude negative solutions and tends to oscillate. Therefore, for practical calculations, some additional assumptions such as smoothness, non-negativity, etc. need to be used.

In iterative methods, a starting function, f^0, is iteratively modified until an acceptable fitting error is reached. The most widely used computer code for iterative unfolding is SAND-II [6]. The deconvolution process starts by setting up an initial function, f^0. With this function effects A_0^α are calculated and compared with experimental values. If the quality of the fit (Eq. (8)) is not good enough then the initial function must be corrected. The iteration process stops if the assumed condition is fulfilled or if the number of iterations exceeds a fixed limit.

The disadvantage of the discrete interval method is in its difficult error analysis. The number of modalities used to investigate a given end-point does not usually exceed 10 and is much smaller than the number of discrete intervals (typically chosen between 10 and 20 per decade). Equation (7) is therefore undefined and does not have a unique solution. Therefore, significance analysis can be performed only with some prior knowledge. Even if the error band of the unfolded function is known, the quantitative comparison of two solutions depends on initial guesses, types of regularizations, etc.

Radial dose distribution

For the purpose of this study, the Geiss formula for the radial distribution of dose [2] has been chosen as a guess function in the unfolding procedure. Geiss's $D(r)$ formula is based on a parameterization of the Monte Carlo calculations, performed by Krämer *et al.* [5]. The model assumes that in the area of the "core" of heavy charged particles, i.e. at radial distances, r, below 0.1 nm from ion's path, the dose has a constant value, C. The second assumption is that the range of delta electrons, r_δ, is proportional to their energy, to the power 3/2. The analytical representation of Geiss formula and the energy-range relationship for delta-ray electrons are as follows:

$$(9) \qquad D(r) = \begin{cases} C & \text{for } r \le r \\ C\dfrac{r_0^2}{r^2} & \text{for } r_0 < r \le r_\delta \end{cases} \qquad r_\delta(E) \propto \sqrt{E^3}$$

The value of the constant C (which is 4.72 Gy for 4.8 MeV α-particles) is determined from normalization of the distribution to the particles' total stopping power value:

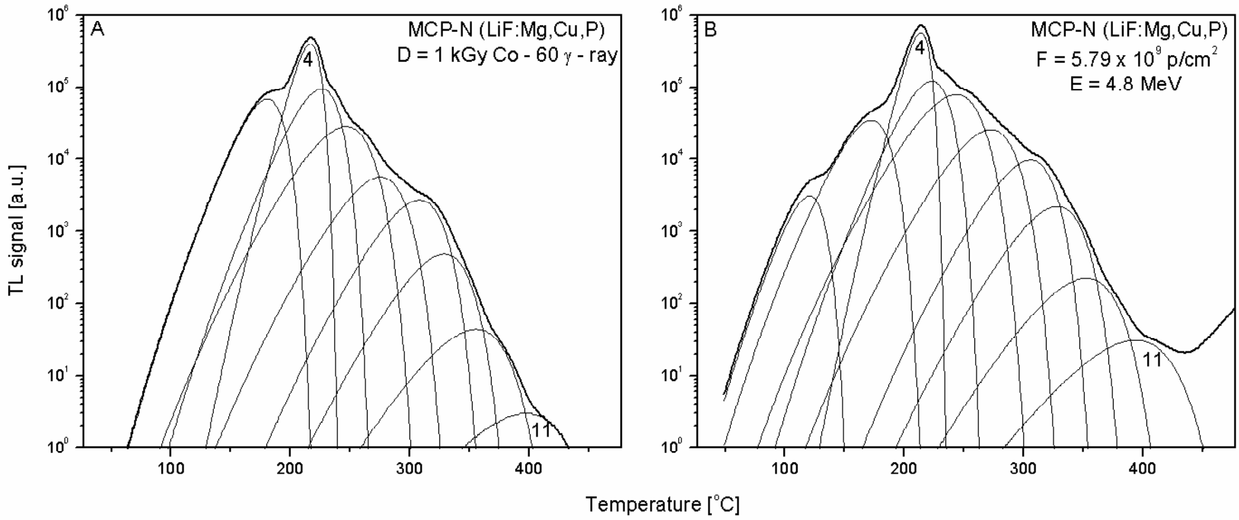
$$(10) \qquad 2\pi \int_0^{r_\delta} r \cdot D(r)dr = \frac{1}{\rho}\frac{dE}{dx}$$

where ρ is the density of the medium (LiF).

Irradiation and read-out

All measurements for this study were performed using MCP-N (LiF:Mg,Cu,P) TL detectors, produced at the Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences (IFJ PAN, Kraków, Poland). Data from earlier irradiations [7] with Co-60 γ-rays in the dose range from 0.5 to 1000 kGy, at the KAERI (South Korea) and Co-60 γ-rays in the dose range from 1 to 300 kGy, at the TU, Delft (Netherlands) were used. Irradiation with α-particles was performed using an Am-241 source of surface fluence rate 1.71×10^4 particles·cm^{-2}·s^{-1}, as verified with CR-39 track-etched detectors in measurements performed at the IFJ PAN.

The MCP-N detectors were 4.5 mm in diameter, 0.9 mm thickness and the entire detector surface (from one side) was uniformly irradiated by α-particles. The irradiation time varied from 1.2×10^2 s to 5.2×10^6 s, which led to fluences ranging from 2×10^6 to 8.9×10^{10}



	Peak	2	3	4	5	6	7	8	9	10	11
	Amplitude [a.u.]	0	68839,2	394254,6	95152,4	28473,1	5660,2	2731	482	44	3
Panel A	Temperature [C]	126	180	218	227	247	276	310	331	357	399
--	Energy [eV]	1,14	1,12	2,56	1,63	1,14	1,07	1,11	1,09	1,06	1,48
--	Amplitude [a.u.]	3089,6	34714,8	570403,3	122005,1	79645,4	25866,6	9789,1	2223,8	225	31
Panel B	Temperature [C]	126	178	220	229	249	278	312	334	358	400
	Energy [eV]	1,14	1,12	2,56	1,63	1,14	1,07	1,11	1,09	1,06	1,48

Fig. 3. Comparison of deconvolution results of MCP-N TL glow curves after irradiation with γ-rays (panel A) and α-particles (panel B). High local doses, within a single α-particle track, cause the appearance of high-temperature peaks, characteristic of doses exceeding 100 kGy of γ-rays.

particles·cm^{-2} (i.e. average doses from about 0.3 Gy to about 15 kGy in the irradiated volume – a layer of 18.54 μm thickness).

Read-out was performed using a RA'94 manual TLD reader with a platinum heater, a bialkali photomultiplier tube and a BG12 filter, at a heating rate of 2°C per second up to 550°C. In all measurements the volumetric flow rate of argon was about 40 l/h. All experiments were performed using virgin detectors and after their read-out to 550°C the detectors were no longer used. All TL glow curves were deconvoluted into N single peaks using the GlowFit code [11], assuming first-order kinetics.

Results and discussion

It was assumed that in the read-out TL glow curves of MCP-N detectors irradiated by α-particles and by γ-rays same individual peaks were observed, differing only in amplitude, as shown in Fig. 3. All measured TL glow curves were normalised to read-out at the temperature of 220°C and deconvoluted into individual peaks using the GlowFit code [11]. The results of deconvolution of all TL glow curves, after irradiation with γ-rays, $A_i^\gamma(D)$ are presented in Fig. 4.

To avoid the effect of overlapping α-particle tracks [10], deconvolution was performed for TL glow curves obtained after the fluence of 5.9 × 10^9 α-particles/cm^2. The measured values of $A_i^\gamma(D)$ and A_i^α were used as input data for the unfolding code.

Application of the iterative unfolding method requires a prior knowledge of the guess function. For the monoenergetic α-particles, the $f^\alpha(D)$ is related to $D(r)$ as follows (see Appendix):

$$f^\alpha(D) = \frac{\pi r_0^2 C}{D(r)^2} \tag{11}$$

The $f^\alpha(D)$ function for slowed-down α-particles was calculated numerically from the initial energy of 4.8 MeV. The non-normalized $f^\alpha(D)$ function, used as the initial guess function for the iterative unfolding procedure, is shown in Fig. 5. As described above, the iterative unfolding algorithm was applied to solve Eq. (7). Calculations involved 10 runs and 1000 iterations per each run (values set arbitrarily). For each run, the values of A_i^α were randomized from a Gaussian distribution with an average value, μ, equal to A_i^α and a standard deviation value, σ, equal to δA_i^α $0.5\delta A_i^\alpha$ and 0.33 δA_i^α. The best result was obtained for the parameter $\sigma = \delta A_i^\alpha$ and this result is presented in Fig. 5

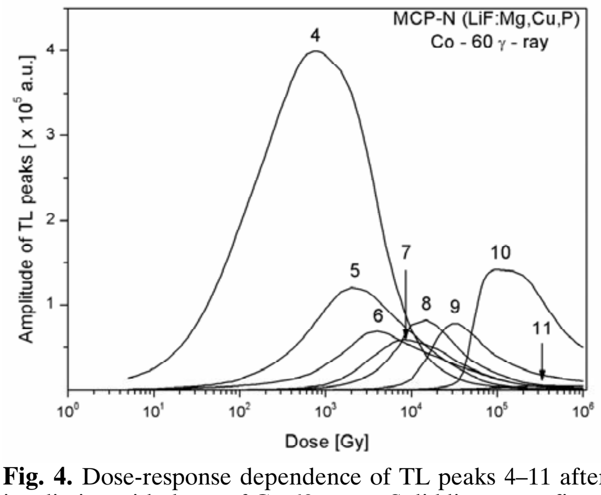


Fig. 4. Dose-response dependence of TL peaks 4–11 after irradiation with doses of Co-60 γ-rays. Solid lines were fitted using the Lagrange interpolation method.

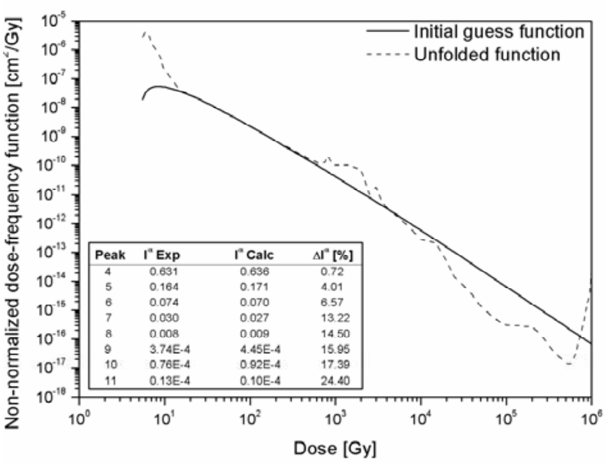


Fig. 5. Comparison between the unfolded dose-frequency function and the function resulting from model calculations using the Geiss formula, Eq. (9).

together with a comparison of experimental and model data. With reduced range of random values, the algorithm was not able to fit the response function properly. If wider range of σ values were used, the individual results were more divergent, but their average value fitted better the model predictions. This may result from the underestimation of the uncertainties of A_i^α values, especially for peaks located at the highest temperatures. The curve obtained for $\sigma = \delta A_i^\alpha$ is consistent with the approximate model curve in the dose range up to 10^4 Gy. For higher doses, the unfolded function does not fit the model prediction. There may be at least two reasons for these discrepancies: poor experimental data coverage and uncertainty of experimental data in this dose range. The second possible reason is especially difficult to resolve because of the complicated relationship between the dose, intensity and position of the "B" peak [7] and the instability of the TL reader during high-dose read-outs. Our results seem to support this hypothesis: comparing experimental and calculated data (Fig. 5) one may observe that for peaks located at higher temperatures, corresponding to higher doses of radiation, the relative percentage error increases up to the value of about 25% for the highest-temperature peak. In turn, discrepancies over the lowest dose regions might result from exclusion from the calculations of low-temperature peaks corresponding to the lowest doses measured.

Conclusions

A new method of experimental verification of radial dose distribution formulae using MCP-N thermoluminescent detectors has been proposed. The method is based on unfolding of the dose-frequency function, a weighting factor between the peak intensities from irradiations with α-particles and γ-rays. The method was tested using α-particles of initial energy 4.8 MeV stopping in the detector. Our initial results demonstrate conformity with the assumed $D(r)$ dependence over the dose range up to 10^4 Gy. The measured $f^\alpha(D)$ overestimates model calculations at the lowest doses and underestimates them at the doses exceeding 10^4 Gy. The discrepancies over the low dose range may result from excluding the low-temperature peaks from our calculations, while discrepancies over the higher dose range might result from uncertainties in the experimental data. However, our analysis showed that the new method gave a consistent result within a 25% range of uncertainty. Conversion of the unfolded function to the radial dose distribution function is complicated because the particle slowing-down process has to be considered. The problem would be much simpler if the energy stored in the detector were deposited by monoenergetic particles. Work on developing ultra-thin MCP-N detectors for this purpose is under way.

References

1. Bilski P, Obryk B, Olko P, Mandowska E, Mandowski A, Kim JL (2008) Characteristics of LiF:Mg,Cu,P thermoluminescence at ultra-high dose range. Radiat Meas 43:315–318
2. Geiss OB, Kramer M, Kraft G (1998) Efficiency of thermoluminescent detectors to heavy charged particles. Nucl Instrum Methods Phys Res B 142:592–598
3. Horowitz YS, Stern N (1990) Relative thermoluminescent efficiency of LiF:Mg,Cu,P to alpha particles: theory and experiment. Radiat Prot Dosim 33:287–290
4. Katz R (1978) Track structure theory in radiobiology and in radiation protection. Nucl Track Detect 2:1–28
5. Krämer M (1995) Calculations of heavy-ion track structure. Nucl Instrum Methods Phys Res B 105:14–20
6. McElroy WN, Berg S, Crockett T, Hawkins RG (1967) A computer automated iterative method for neutron flux spectra determination by foil activation. AFWL-TR-67-41
7. Obryk B (2010) Development of a method of high dose measurement using highly sensitive LiF:Mg,Cu,P detectors. PhD thesis, Institute of Nuclear Physics PAN, Kraków. IFJ Report no. 2045/D (in Polish). http://www.ifj.edu.pl/publ/reports/2010/2045.pdf
8. Obryk B, Bilski P, Budzanowski M *et al*. (2009) Development of a method for passive measurement of radiation doses at ultra-high dose range. IEEE Trans Nucl Sci 56:3759–3763
9. Olko P (1996) Microdosimetric interpretation of thermoluminescence efficiency of LiF:Mg,Cu,P (MCP-N) detectors for weakly and densely ionizing radiations. Radiat Prot Dosim 65:151–158
10. Olko P, Bilski P, Gieszczyk W, Grzanka L, Obryk B (2011) Microdosimetric analysis of response of LiF:Mg,Cu,P (MCP-N) TL detectors for alpha-particle and ultra-high doses of gamma-rays. Radiat Meas 46;12:1349–1352
11. Puchalska M, Bilski P (2006) GlowFit – a new tool for thermoluminescence glow-curve deconvolution. Radiat Meas 41:659–664
12. Scholz M, Kraft G (1996) Track structure and the calculation of biological effects of heavy charged particles. Adv Space Res 18:5–14
13. Waligórski MPR, Hamm RN, Katz R *et al*. (1986) The radial distribution of dose around the path of a heavy ion in liquid water. Nucl Tracks Radiat Meas 11;6:309–319
14. Zhang C, Dunn DE, Katz R (1985) Radial distribution of dose and cross-sections for the inactivation of dry enzymes and viruses. Radiat Prot Dosim 13:215–218

Appendix

Derivation of the guess function $f^{\alpha}(D)$, Eq. (11).

The element of cross-sectional area, dS, irradiated by a given dose, D, can be calculated using the following formula:

$$dS = 2\pi r dr = \left| \begin{array}{c} D(r) = C\frac{r_0^2}{r^2} \quad r = r_0\sqrt{\frac{C}{D(r)}} \\ dr = -\frac{r_0}{2}\sqrt{\frac{C}{D(r)^3}}dD \end{array} \right| = -\pi r_0\sqrt{\frac{C}{D(r)}}r_0\sqrt{\frac{C}{D(r)^3}}dD = -\pi r_0^2 C\frac{dD}{D(r)^2}$$

$$\frac{dS}{dD} = -\frac{\pi r_0^2 C}{D(r)^2}$$

This function has exactly the same physical meaning as the $f^{\alpha}(D)$ function:

$$f^{\alpha}(D) = -\frac{dS}{dD}$$

The minus sign results from the fact that the derivative decreases over the whole range of doses.