A UNIFIED APPROACH TO DECONVOLUTION RADIATION SPECTRA MEASURED BY RADIOCHROMIC FILMS

by

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A method for the evaluation of energy distribution of a radiation source on the basis of measured space distribution of deposited energy is proposed. The measured data were obtained by using radiochromic films. Mathematical modeling is defined as a Fredholm integral equation inversion problem. Negative solutions were treated as an additional condition expressed through undefined energy group boundaries, caused by virtue of the physical phenomenon of statistical uncertainty. Examples are given of the electron source and neutron radiation field.

Key words: radiation field, radiation source, Fredholm equation, deconvolution, spectrum, radiochromic film, unfolding

INTRODUCTION

Detailed knowledge of depth distribution of energy deposition from various irradiation sources is often required in many areas such as radiation processing, radiotherapy, *etc*. This requires development of a detection system, which can record effects of incoming particles with sufficient sensitivity and spatial resolution, as well as the knowledge of the initial energy spectra of incoming particles.

Radiochromic films are a possible solution for a detection system. It is well known ever since the beginning of the last century that certain dye-cyanide molecules change color when exposed to ultraviolet radiation [1] and later the same effect was found for both gamma and electron radiation [2]. Nowadays, these films are commercially produced for quantitative measurements of photon radiation fields. To obtain depth-dose distribution, stacking of films perpendicular to incoming radiation was proposed [3, 4]. In addition, the latest development of technology such as laser scanners made it possible to determine the optical density of films with spatial resolution of 100 μm. By using these films we were able to perform preliminary measurements of electron fields from pulsed electron sources [5, 6] and to propose a simple method for electron spectra unfolding [7].

It is often difficult to extract energy distribution of incoming particles from the characteristics of the radiation source (except, of course, for gamma sources). Furthermore, direct accurate measurements of energy spectra are often either impossible or cumbersome, so spectra unfolding has to be performed. As far as the existing unfolding methods are concerned, it is known that, when deriving a spectrum from a set of measured data, three groups of effects should be taken into account: *non-uniform efficiency or finite resolution of the apparatus, statistical fluctuations and statistical errors of measurements* [8, 9]. It appears that in all experimental methods modeling is expressed through Fredholm integral equation of the first kind. From the mathematical point of view, this is known as an ill-posed problem [10]. When solving this equation, the least squares method is usually used, as well as some other techniques, in order to avoid unstable results that give negative values for the spectrum calculated. However, such approaches do not take into account that, apart from the above mentioned experimental uncertainties, there are some other uncertainties related to solutions of the problem by using inverse methods in discrete space coordinate.

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For this reason, and in order to give physical meaning to solvability requirements, an effort was made to describe negative solutions as a consequence of an uncertainty with respect to energy boundaries of multigroup (histogram) approximation of Fredholm integral equation [11]. It means that if we select an element of a phase space, then there is no unique answer to the question: which of the phase intervals contribute to the interval phase considered [12]. This phenomenon occurs even in the case when only scattering is taken into account and it is not due to energy or particle loss in the system. As a result, the solvability condition is reduced to a system of linear equations that with an extra condition may be further approximated to the linear programming problem.

Based on such an approach, we are proposing a reasonably simple method for spectra deconvolution that uses radiochromic films to determine space distribution of the absorbed dose and a mathematical method that enables to unfold the energy distribution of incoming particles from such data. This is demonstrated by unfolding of electron spectra from pulsed electron sources and neutron spectra, *i.e.*, in cases where direct measurements are difficult. Some preliminary results on the different aspects of these problems were published in several short reports [5, 6, 7, 13–17]; here we have tried to give an overview.

PULSED ELECTRON BEAM

Radiochromic films and 3D dose distribution

FWT-60 radiochromic films were purchased from Far West Technology Inc., Goleta, CA, USA. The active component of this film is hexahydroxyethylpararosaniline cyanide (HHEPRC), which upon irradiation yields a blue colored product. The intensity of coloration (optical density) can be measured using spectrophotometer in wavelength range of the new spectral line, which originates from the cationic form of molecule. The absorbed dose is a function of the measured absorbance. By definition, the absorbance is $-\log_{10}(I_\lambda / I_{\lambda 0})$, where $I_{\lambda 0}$ and I_λ are the intensities of incoming and transmitted light of chosen wavelength λ. In this analysis the wavelength of spectrophotometric measurements was 633 nm. When radiochromic dosimeters are to be used as secondary standards in a certain radiation field, the dosimeters must be calibrated appropriately in a similar radiation environment. Under circumstances where there is no dose rate dependence (up to 10^{12} Gy/s) and energy dependence (0.1 – 10 MeV) [18, 19], radiochromic films can be calibrated in ^{60}Co gamma-ray field by primary standard dosimeters, such as Fricke dosimeter or calorimeter. An example of the calibration curve of FWT-60 dosimetric film is presented in Fig. 1.

Electron source

A pulsed electron accelerator Febetron 707 (Field Emission Corporation) was used to provide a high intensity pulsed beam of electrons. The nominal maximum energy of electrons is around 2 MeV and the pulse duration about 20 ns full width at half-maximum intensity. Data on energy distribution of electrons were not provided by the manufacturer.

Stacks of FWT-60 dosimetric films (5 × 5 cm), thickness (0.047 ± 0.002) mm, separated by aluminum spacers (5 × 5 cm), thickness (0.07 – 0.21) mm, were irradiated by electron pulse. The electron beam was perpendicular to the stack. The overall thickness of the stack was sufficient for complete absorption of the electron pulse. After the irradiation, the absorbance was measured using scanning He-Ne laser microdensitometer at 633 nm. Results were converted into dose distribution using the calibration curve presented in Fig. 1. The obtained result is the detailed 3D distribution of the deposited energy as a function of penetration depth [20].

Spectra deconvolution

From the experimental point of view, it is known that there is still no device that would enable energy spectra measurements in such a short time interval. Therefore, our experimental method is based on spectra derivation according to depth-dose distribution.

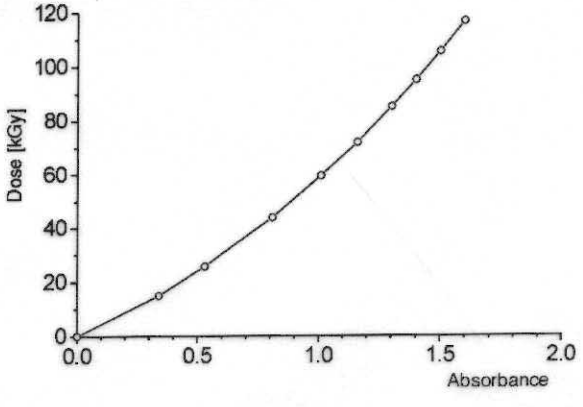

Figure 1. Calibration curve of FWT-60 radiochromic film in ^{60}Co gamma-ray field; Fricke dosimeter and spectrophotometer were used for dose and absorbance measurements, respectively

Theoretical evaluation was performed in two steps. In the first step, a series of depth-dose distribution curves were produced assuming a single energy value of incoming electrons. These curves were then compared to the measured depth-dose distribution from the narrow electron energy range to be included in detailed calculation (E_{min} and E_{max}). In the next step that interval is subdivided into several subintervals (energy groups), with the mean energy as a group representative.

We start from a functional $W(x)$ describing the space dependence of energy deposition caused by electron source $S(E)$:

$$W(x) = \int_{E_{min}}^{E_{max}} S(E)F(E,x)dE \qquad (1)$$

where $F(E, x)$ represents the space-dependent transmission probability of electrons with energy E. Having a set of measured space dependent doses $\{W_m(x_i), i = 1, 2,..., N\}$ the problem is to find the energy distribution function $S(E)$ that meets the positivity condition:

$$\{0 \le S(E) \le 1 \wedge E_{min} \le E \le E_{max}\} \qquad (2)$$

To define a discrete set of energies $\{E_j, j = 1, 2,..., N | E_{min} \le E_j \le E_{max}\}$ that meet the positivity condition [11, 12], a set of monoenergetic sources $\{S(E_j) = S_j \cdot \delta(E-E_j), j = 1,2,...,N\}$ is defined. Then, according to eq. (1), we get:

$$\{F(E_j, x) = W_j(x), j = 1,2,..., N | E_{min} \le E_j \le E_{max}\} \quad (3)$$

where $W_j(x)$ are theoretically obtained space dependent deposited energy distributions of electrons with energy values E_j. The above expression implies that the required transfer matrix is equal to the theoretically obtained monoenergetic electron fluence distributions. Moreover, the inversion problem is now reduced to the best fit with source group values to a family of curves. This is a very practical approach and still not dependent of incoming source entities. Continuing this approach, it has to be recognized that the sequence of W_j can be obtained by using any transport model.

We can insert eq. (3) into eq. (1) and applying it to a given measured data $W_m(x)$ we get:

$$W_m(x_i) = \sum_{j=1}^{N} F(E_j, x_i) \int_{E_{j-1}}^{E_j} S(E)dE =$$

$$= \sum_{j=1}^{N} S_j W_j(x_i), \quad i = 1,2,...,N \qquad (4)$$

Thus, a set of linear algebraic equations is obtained. By solving the eq. (4), the energy source distribution can be obtained. However, an arbitrary selected energy interval within the energy boundaries (E_{min}, E_{max}) often gives negative values of S_j. The problem of the existence of negative values can be related to statistical uncertainties that may be the consequence of Boltzmann H-theorem. Namely, in scattering theory of linearized Boltzmann transport operator, a connection can be established between the Boltzmann H-theorem of statistical irreversibility and irreversibility of transport process (cf. ref. [11] and [12]). As a consequence of this phenomenon, a statistical uncertainty appears in regard to the phase space contribution due to the statistical nature of the scattering process.

The positivity conditions increase our task do derive N amplitudes of the source S_j from eq. (4) to a problem do derive N additional variables, the energy representatives of energy groups $\{E_{j-1}, E_j, j = 1,2, ..., N\}$ in such a way as to give positive S_j. In the absence of explicit expressions that give the required energy points [11], and in order to demonstrate the efficiency of the proposed convolution model, the problem was modeled by the linear programming method. Such a task can be solved by applying any linear programming routine.

Moreover, in order to make the use of the proposed deconvolution model easier, a semiempirical model of electron transmission is used (EDEPOS code [21]) for the derivation of multigroup transmission coefficients (Table 1). The obtained energy spectrum of the electron source is shown in Fig. 2. Spatial electron dose distributions calculated for discrete incoming electron energies appearing in eq. (4), and a comparison with the calculated (energy spectrum as on Fig. 2) and measured Febetron electrons distributions are given in Fig. 3.

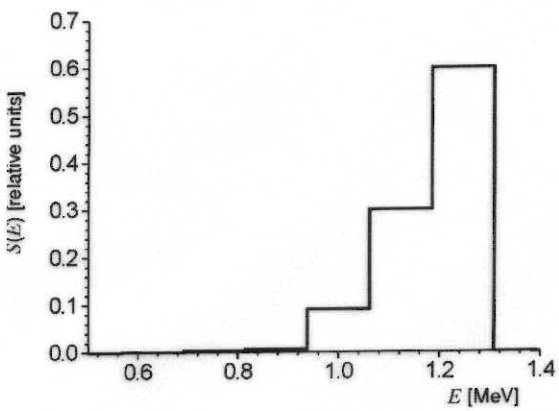

Figure 2. Calculated energy spectrum for the FEBETRON electron source

Table 1. Spatial distribution of energy deposition $W(x)$

E_j [MeV]→ ↓x [g/cm^2]	$W_j(x)$ [relative units] 0.75	0.80	0.85	0.90	0.95	1.00	1.05	1.10	1.15	1.20	1.25	1.30	1.35	$W_c(x)$ [rel. un.]	$W_m(x)$ [rel. un.]
.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
.057	.800	.819	.836	.850	.862	.872	.881	.889	.896	.902	.907	.912	.917	.898	.814
.114	.527	.571	.610	.643	.672	.698	.720	.740	.757	.773	.787	.799	.810	.764	.581
.171	.276	.329	.378	.424	.466	.503	.538	.569	.597	.622	.645	.665	.684	.608	.442
.228	.108	.150	.194	.238	.281	.322	.362	.399	.434	.466	.496	.523	.549	.451	.321
.285	.025	.048	.077	.109	.143	.179	.216	.252	.288	.322	.355	.386	.415	.310	.247
.315	.009	.020	.040	.064	.092	.123	.155	.189	.222	.255	.288	.319	.350	.246	.230
.345	.003	.007	.017	.034	.055	.079	.107	.136	.166	.197	.228	.259	.289	.190	.172
.365	.001	.004	.009	.019	.036	.057	.080	.106	.134	.163	.192	.222	.251	.157	.144
.422	.000	.000	.001	.004	.008	.016	.029	.046	.065	.086	.110	.134	.160	.085	.098
.479	.000	.000	.000	.001	.001	.003	.007	.014	.024	.038	.054	.072	.092	.040	.056
.536	.000	.000	.000	.000	.000	.001	.002	.003	.007	.012	.021	.033	.046	.016	.028
.570	.000	.000	.000	.000	.000	.000	.001	.001	.003	.006	.010	.017	.028	.008	.000

W_j – for monoenergetic source at energy E_j (calculated using EDEPOS code [21])
W_c – for source spectrum as on Fig. 2 (calculated using EDEPOS code [21])
W_m – measured at Febetron accelerator

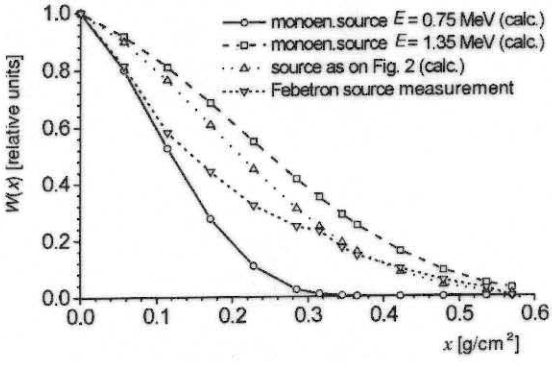

Figure 3. Calculated and measured energy depositions as a function of penetration depth

NEUTRON SOURCES

Radiochromic films sensitive to neutron and alpha radiation

It might appear that radiochromic films cannot be used for neutron detection. However, neutrons can be detected through nuclear reaction if the films contain an element that converts neutrons into charged particles by nuclear reaction. Thus, the method used for electron spectra unfolding can in principle be used for neutron spectra unfolding as well. Therefore, we performed neutron measurements by manufacturing radiochromic films from a mixture of boron acid (H_3BO_3) and radiochromic dye HHEPRC (0.15 mol each) dissolved in polyvinyl-butyral, utilizing $^{10}B(n, \alpha)^7Li$ nuclear reaction. The film thickness was 0.1 mm, *i. e.*, approximately equal to the mean free path of thermal neutrons (0.025 eV) in such media. The total energy released per absorbed neutron is 2.79 MeV, whereas α-particle carries 1.78 MeV. The range of 1.78 MeV α-particle in film is around 5 μm, *i. e.*, much lower than the thickness of the film, so total energy of α-particles and ^7Li created in a particular film will be deposited on that film. Due to relatively low energy required for the formation of blue colored products from HHEPRC (around 3 eV per molecule), around $9 \cdot 10^5$ such molecules per absorbed neutron can be expected. Such arrangements can enable high sensitivity, that is important if available thermal neutron fluxes are low. Preliminary experiment, carried out in the thermal neutron field of the RB nuclear reactor in VINČA Institute of Nuclear Sciences showed that films exhibit substantial exposure dependent changes of optical density when exposed to a rather low neutron flux (around 10^6 cm^{-2}s^{-1}) for irradiation time ranging from 15 to 45 minutes [17]. Absorbance was measured at 603 nm, a wavelength of maximal absorbance of blue colored product of HHEPRC, in order to maximize sensitivity.

Neutron measurement

To estimate the possibility to use radiochromic films for neutron detection, a trial stock of three films was made on. Thermal neutron field was the RB reactor at VINČA Institute with a flux 10^6 cm^{-2}s^{-1} per 1 W of the fission reactor power.

Using a standard dose to absorbance curve $D(x_j) = f(A)$ calibrated with Fricke dosimeter, a set of measured doses has been obtained $D_m(x_i)$. On the other hand, since the reactor spectrum of incoming neutrons is known [22], we were able to evaluate the corresponding theoretically expected doses $D_t(x_i)$ according to the following expression:

$$D_t(x) = \frac{tE_\alpha \int_x^{x+\Delta} \Psi(x)\Sigma dx}{\rho R_\alpha} \approx \frac{tE_\alpha \Delta \Psi(x)\Sigma}{\rho R_\alpha} \quad (5)$$

where t is the irradiation time, E_α – energy of alpha particles, $\Delta = 0.1$ mm – the thickness of films, Σ – macroscopic neutron absorption cross section for boron in mixture, ρ – density of films and R_α – corresponding alpha particle range. The nominator in this expression represents the energy density deposited by the produced alpha particles around point x, while the denominator represents the corresponding mass. Comparing them revealed a large discrepancy, shown in Table 2.

Table 2. Expected D_t and measured D_m as the function of penetration depth x

x [mm]	$D_t(x_i)$ [kGy]	$D_m(x_i)$ [kGy]
0	2.16	0.86
0.1	2.04	0.83
0.2	1.89	0.79

It is necessary to remember that, when exposed to ionizing radiation, HHEPRC exhibits blue colored product only if the cationic form of molecule is formed, *i. e.*, when a specific bond is broken. The quantitative assessment of the effect was related to the appearance of the blue colored product when the dye is exposed to gamma rays or electrons. However, the quantitative relations between the measured optical density changes of films and the absorbed doses, as assessed when standard gamma ray sources (*e. g.*, ^{60}Co) are used, do not necessarily apply when films are exposed to high LET particles. For example, it was reported that sensitivity of Fricke dosimeter was nearly four times lower for alpha particle with energy about 2 MeV then for ^{60}Co source [23]. Having this in mind, our results are not unexpected.

From the physical point of view this phenomenon can be proved by using an approximate expression for MeV energy alpha particles [24]:

$$D_\alpha \cong \frac{0.02\Psi_\alpha \bar{E} t \cdot 10^6}{R_\alpha \rho} \quad (6)$$

where D_α is the absorbed dose [kGy], Ψ_α – flux [cm^{-2}min^{-1}], $\bar{E}$ – mean energy [MeV], t – exposition time [min], R_α – range [cm], and ρ – density of the mixture [g/cm^3]. Calculations for $D_\alpha = D_m$ bring to $\bar{E} \approx 720$ keV instead of 1.78 MeV belonging to boron (n, α) reaction. Nevertheless, there is a chance that the use radiochromic films technique might become a powerful experimental method for radiation fields spectra analysis.

The experimental method described above can be applied in case of fusion neutron fields too [17]. In this case neutrons are supposed to income perpendicularly to the 1 cm light water moderating layer. Moderation is necessary to produce epithermal neutrons where the (n, α) reaction rate is significant.

Neutron spectra deconvolution

In the case of stacked radiochromic films, a half space is supposed to be placed in $x \geq 0$, exposed to a thermal neutron source at a boundary $x = 0$ with an arbitrary energy distribution. Also, it is assumed that the medium is filled with boron acid, where the neutron-alpha particle reaction with $1/v$ dependence is dominant. A given sequence of points is also assumed with measured neutron fluence $\{x_i, \Psi_i, i = 1, 2, ..., N\}$ with respect to the space coordinate, obtained according to unfolded experimental data.

As in the case of electrons, eq. (1), we start from a general Fredholm integral equation:

$$\Psi(x) = \int_{E_{\min}}^{E_{\max}} G(E,x)q(E)dE \quad (7)$$

which is a relation between the energy distribution of the incoming particles $q(E)$ and measured fluence space distribution $\Psi(x)$.

Since in the case of boron the scattering is negligible compared to absorption cross section which behaves as $1/v$, eq. (7) reduces to analytical form:

$$\Psi(x) = \int_0^\infty e^{-x/v} q(v)dv, \; x, v \geq 0 \quad (8)$$

where energy parameter E is expressed by speed v.

It has been proven [7] that the inverse solution of eq. (8) is the inverse Laplace transformation:

$$q(v) = \frac{1}{2\pi v^2 i} \int_{\sigma-i\infty}^{\sigma+i\infty} e^{s/v} \Psi(s)ds \quad (9)$$

The proof is obvious and will be omitted here. The next examples can serve as a practical illustration of the irreversibility problem.

Example 1. The measured fluence is of the form

$$\Psi(x) = e^{-x/\ell}, \; x \geq 0 \quad (10)$$

Inserting (10) into (9) we get:

$$q(v) = \frac{1}{2\pi v^2 i} \int_{-i\infty}^{+i\infty} e^{s(1/v - 1/\ell)} ds =$$

$$= \frac{1}{2\pi v^2} \int_{-\infty}^{+\infty} e^{it(1/v - 1/\ell)} dt =$$

$$= v^{-2} \delta\left(\frac{v-\ell}{v\ell}\right) = \delta(v-\ell) \quad (11)$$

That means a monoenergetic source is obtained. This example is important because of the usual exponential form of the fluence.

Example 2. The measured fluence is of the form

$$\Psi(x) = \mathrm{e}^{-x/\ell} \sum_{k=0}^{N} a_k x^k \qquad (12)$$

Then solution of eq. (9) is:

$$q(v) = (2\pi v^2)^{-1} \sum_{k=0}^{N} (v\ell)^{k+1} a_k \delta^{(k)}(v-\ell) \qquad (13)$$

When applying this expression, the delta function derivations operate as:

$$\int f(x)\delta^{(k)}(x)dx = (-1)^k f^{(k)}(0)(x) \qquad (14)$$

However, the most efficient approximation of the measured data might be the one shown in the next example.

Example 3. The measured fluence is of the form

$$\Psi_k(x) = \sum_{k=1}^{N} a_k \mathrm{e}^{-x/\ell_k}, \ \ell_k \geq 0 \qquad (15)$$

In the same way as before we get:

$$q(v) = \sum_{k=1}^{N} a_k \delta(v-\ell_k) \qquad (16)$$

Suppose now the sequence $\{\ell_k\}, k = 1,2,\ldots,N$ is ordered and let $E_k = (\ell_{k-1} + \ell_k)/2$. Then, performing integration we get a histogram (multigroup representation):

$$\bar{q}_k = \int_{\ell_{k-1}}^{\ell_k} q(v)dv = a_k, \ k = 1,2,\ldots,N \qquad (17)$$

The problem is how to realize the approximation (15). It appears in this case that, although there is no scattering reaction, the uncertainty of choosing exponential parameters still remains. One realization of approximation (15) is discussed in [7] and will be omitted at this point.

CONCLUDING REMARKS AND DISCUSSION

Unfolding the energy distribution of a radiation source on the basis of measured space distribution of deposited energy in a sample exposed to radiation appears to be an extremely complex task from both the experimental and the mathematical point of view. A thorough treatment of the problem has to include an adequate experimental set-up and a method of spectra deconvolution. The underlying motive to perform this study, however, is the fact that radiochromic films when used in stacked configurations, due to their small thickness, enable a superb spatial resolution of spatial distribution of energy (dose). The possibility of manufacturing films that can suit the needs of detecting any form of radiation can be instrumental in providing a simple and inexpensive system that enables dose measurements from a variety of radiation sources. The critical point in using radiochromic films for unfolding an energy distribution of a radiation source is the knowledge of the relationship between the energy deposited by incoming radiation and changes in optical density of films. The response of films to low LET radiation (photons and electrons) can be easily calibrated by using standard sources (*e. g.*, ^{60}Co) and standard dosimeters (*e. g.*, Fricke dosimeter). However, calibration of the response of films to the high LET radiation (light ions and neutron induced radiation) requires an additional effort, which, having in mind the usefulness and versatility of radiochromic films, may be well justified.

Spectra deconvolution based on space fluence distribution can also be successfully realized in the classic way (the method of linear programming), but in that case there is no unfolding as in other methods. In that sense, the section on electrons, which was done at the beginning of our dealing with these problems, should serve as an illustration of the use of the existing methods and is suitable for experimentators.

This experimental and theoretical method of deriving a source energy spectrum on the basis of the measured energy deposited on the films is significant in the first place because of the use of radiation for human needs, for investigation of materials, *etc.*

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ОБЈЕДИЊЕН ПРИСТУП ЗА ОДРЕЂИВАЊЕ РАДИЈАЦИОНИХ СПЕКТАРА МЕРЕНИХ РАДИОХРОМНИМ ФИЛМОВИМА

Предложена је метода за евалуацију енергетског спектра извора радијационог поља, заснована на измереној просторној расподели депоноване енергије. Мерени подаци добијени су коришћењем радиохромних филмова. Математичко моделовање је дефинисано као инверзни проблем Фредхолмове интегралне једначине. Негативна решења разматрана су као додатни услов неодређености граница енергетских група који је уведен услед физичког феномена статистичке неодређености. Дати су примери електронског извора и неутронског поља зрачења.