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Thermoluminescent dosimetry of mixed nuclear radiation: Radioisotope stands

O.M. Pop[1], V.T. Maslyuk[1], N.I. Svatiuk[1], A.V. Rusyn[2], P.V. Yavorskyi[1], V.I. Roman[1]

[1]*Institute of Electron Physics, Uzhhorod, Ukraine*
[2]*Transcarpathian Cancer Center, Uzhhorod, Ukraine*
[*]*Corresponding author e-mail: pop.ksenja@gmail.com*

Abstract. The study results of dosimeters based on doped lithium fluoride and carbon C-based leucosapphire are presented for use in mixed γ-n fields. The radiation conditions were created using the radioisotope stands based on certified ^{60}Co and Pu–α-Be sources. It is shown that the studied dosimeters are promising for recording the absorbed dose starting from 1 mGy for γ- and mixed γ-n radiation factors. The sensitivity of TLD-100H (LiF:Mg, Cu, P) dosimeters was determined to be several times higher than that of TLD-100 (LiF:Mg, Ti) and Al_2O_3:C. A sensitivity parameter was introduced to quantify the efficiency of dosimetric materials for fixed irradiation conditions. The dependence of luminous intensity on the dose or irradiation time for γ- and mixed γ-n irradiation is shown.

Keywords: radiation stands, γ-n radiation, dosimeters, lithium fluoride, leucosapphire.

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1. Introduction

Personal dosimetry is essential for monitoring treatment prescriptions used in radiation therapy and protecting medical personnel in oncology institutions. Currently, dosimetric support of irradiation procedures is based on using thermally stimulated luminescence (TSL) dosimeters due to their availability, reliability in operation, and long history of use [1–3]. However, this practice should account for the trends in the development of modern radiation therapy, namely the use of new radiation technologies based on beams of the following high-energy nuclear particles: gamma (γ), electron (e), and even proton radiation, as well as beams of radio-active ions, α-particles, π-mesons, or neutrons of different energies [4]. This progress requires additional efforts to ensure and improve the results of radiation therapy to minimize the side effects accompanying the practical use of high-energy nuclear particles. In this case, it is necessary to consider the presence of secondary particles during irradiation, namely neutrons or gamma quanta, which introduce an additional dose not considered during radiation therapy treatment. With an account of the above-mentioned, it is necessary to provide adequate dosimetric support to reduce dose loads on patients and staff. An integral part of radiation therapy is clinical dosimetry, which is necessary for any diagnostic procedure that uses ionizing radiation. The optimization of radiation protection and limitation of radiation doses are essential to prevent overexposure and reduce side health effects. Therefore, research in dosimetry is necessary to improve existing dose monitoring methods and develop new ones, namely the monitoring of high doses [2]. The higher doses monitoring (10^2…10^6 Gy) is relevant in nuclear medicine and for various technological applications of ionizing radiation. It forms new directions for developing TSL dosimetry, outlining additional research to select dosimetric materials, their doping schemes, and calibration methods using intense beams of high-energy nuclear particles. A separate and essential task is the dosimetry of mixed electron, gamma, and neutron radiation. Its development is hampered by the lack of a sufficient number of stands for dosimeter calibration, as well as adequate data on the nature of structural damage in mixed radiation fields.

Therefore, the study of the effect of various types of radiation (gamma and neutron) on the thermoluminescent characteristics of crystalline phosphors, which are the basis of TSL dosimetry, is fascinating.

The main advantages of TSL dosimetry using various crystallophosphates are the following [1–3, 5]: a wide range of measured doses, high sensitivity, long-term storage of absorbed energy (low feeding), the possibility of multiple use of detectors, lack of dependence of readings on environmental parameters, versatility about the registration of various types of radiation, the possibility of full automation of the measurement process and processing of results. Modern TSL dosimetry uses various

semiconductor materials with the sensory properties regulated by doping with various impurities. There is a constant need to search for new materials for these applications, especially for materials with an atomic number (Z_{eff}) close to human biological tissue [6]. These materials can provide information about radiation-induced damage in the human body [6–10]. It is worth noting that the main disadvantages of TSL dosimeters are the limited interval of measured doses due to the influence of radiation defects that are formed during this process and the changing properties of the used materials. It leads to a superlinear dose dependence [1, 2, 11], which requires additional calibration of TSL dosimeters. The above-mentioned indicates the relevance of the comprehensive study of the thermoluminescent characteristics of dosimetric materials of different compositions irradiated on metrological stands using ionizing radiation sources (IRS). These stands can provide fixed and mixed nuclear radiation of various compositions and intensities.

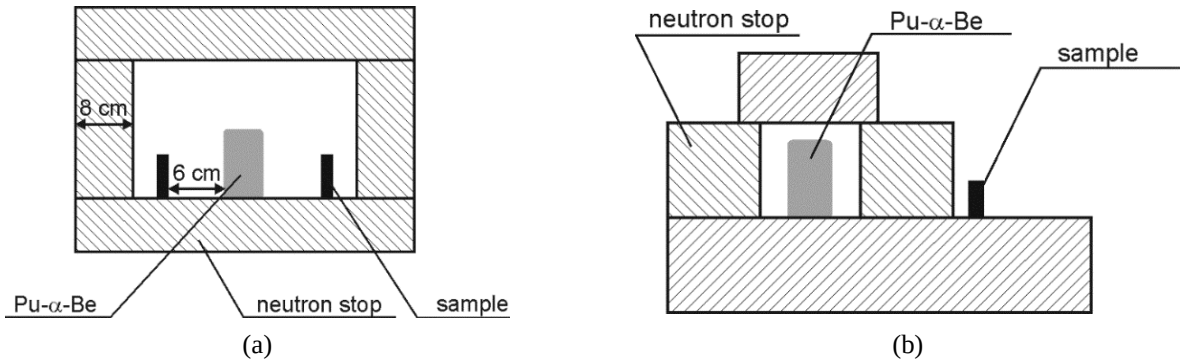
This work presents the results of the study of the thermally stimulated luminescence of dosimetric materials based on doped $A1_2O_3$ and LiF irradiated on radiation stands using Pu–α-Be and ^{60}Co radiation sources.

2. Experimental part

The objects of this study were $A1_2O_3$:C crystals, which are the basis of TSL detectors, namely the DPG, as well as LiF:Mg, Ti detectors DTG or TLD-100 and LiF:Mg, Cu, P detectors NCP or TLD-100H (manufactured in Krakow, Poland), respectively. The irradiation of the studied samples was performed using radiation stands based on ^{60}Co and Pu–α-Be radioisotopes of the IBN-8 brand (No. 088-66) with a neutron flux of $2.73\cdot10^6$ n/s from its surface [13]. This stand allows the study of the effect of mixed and selected ionizing radiation. Additionally, the studied samples were also exposed to 1 mGy and 2 Gy doses of γ-radiation at the therapeutic facility of the Municipal Non-Profit Enterprise "Transcarpathian Cancer Center" of the Transcarpathian Regional Council, Uzhhorod, referred to as the Oncology Center.

The peculiarity of the experimental methodology is illustrated in Fig. 1. The stand is covered with a 5 cm-thick layer of lead to protect personnel from radiation. The created radiation stand allows for the study of dosimetric materials in two modes: directly in the flux of fast and thermal neutrons (0.025…0.5 eV) formed in special deceleration schemes using polyethylene retarders ("neutron-stop" in Fig. 1). The length of the retarder is 6 cm. To study the intensity of thermal and fast neutron radiation, we used the KRAN-1-AN α-n particle radiometer, consisting of four replaceable sensors (thermal, intermediate, fast neutron, and alpha sensors) and a measuring console.

The neutron flux density over the irradiation area was 10^3 neutron·cm^{-2}·s^{-1}. The background radiation intensity was recorded using the radiometer MKS-PM1401K, and was equal to 5.5 mGy/h. This radiation stand allows for solving several tasks: studying the effectiveness of dosimetric materials and recording neutron (*n*) fluxes of various energies and mixed n-γ fluxes. To study the effect of γ-radiation on the dosimetric materials, two experiments were carried out using the radioisotope ^{60}Co with 1173 and 1332 keV lines. The first one was carried out on the stand described in [14], the exposure dose density was 1.32 mGy/h over the irradiation area. This allows for the study of the effects of radiation doses on the TSL characteristics of dosimetric materials starting from 10^{-3} Gy and higher doses. The second series of experiments was carried out at the Oncology Center using the ^{60}Co source with the certified dosimetric monitoring of γ-radiation. The phosphorescence and thermally stimulated luminescence signals were registered with a reader modified for this type of experiment [15]. The FEP-136 photoelectronic multiplier in photon counting mode was used to measure the luminescence intensity. The TSL was excited using the linear heating of the samples to 300 °C. A chromel-aluminum thermocouple was used to control the temperature during heating. TSL measurements were performed immediately after irradiation to reduce the influence of fading.



(a) (b)

Fig. 1. Schematic view of the internal part of the n, γ-, γ-n bench with the Pu-α-Be isotope source of IBN-8 without (*a*) and with (*b*) a "neutron-stop" screen for neutron deceleration.

3. Results and discussion

Irradiation with γ-quanta of ^{60}Co was performed with 10^{-3}…2 Gy doses. Both the TSL spectra and dosimetric parameters, particularly the area of their characteristic peaks, were investigated. Fig. 2 shows the TSL curves of these dosimeters irradiated with a single dose of gamma radiation of 9 mGy at the IEP stand of the NAS of Ukraine containing the radioisotope ^{60}Co. The irradiation was carried out for 6 hours and 40 minutes, and the TSL spectra were immediately obtained. The data are presented in linear (a) and logarithmic scale (b), demonstrating the similarity of TSL spectra of TLD-100 and TLD-100H dosimeters based on lithium fluoride.

As can be seen from Fig. 2, for the DPH dosimeter, the characteristic peak used in dosimetric studies is located at 170 °C for the TLD-100, one intense emission peak can also be distinguished, which is promising for TSL dosimetry at 250 °C; less intense TSL peaks are observed (Fig. 2b) at temperatures of 120, 170 °C.

For TLD-100H dosimeters, which also use a lithium fluoride matrix (LiF:Mg, Cu, P), the above low-temperature peaks and an intense peak near 225 °C are observed. Studies have shown that for each of these dosimeters for the range of absorbed doses of 10^{-3} to 2 Gy, the temperature position of the characteristic peaks does not change significantly, but their intensities may differ. The sensitivities of these dosimeters are also different. As shown in Fig. 2, in TLD-100H samples, under these irradiation conditions, the absorbed dose of 9 mGy, the light sum of the peak at 225 °C, is 4 times higher than for peaks at 120 and 170 °C. In particular, as can be seen from Fig. 2, the sensitivity of TLD-100H was 2.5 times higher compared to TLD-100 and DPH, which agrees with the literature data [1, 3].

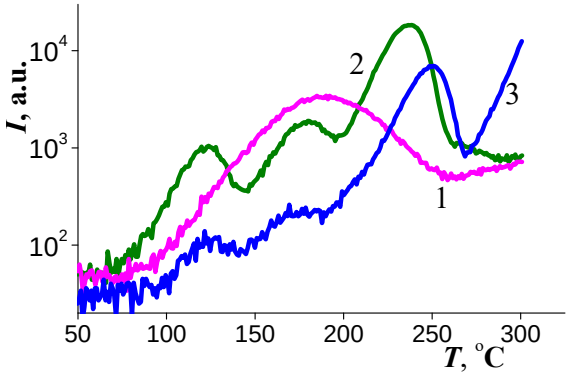


Fig. 2. Curves of thermally stimulated luminescence of samples *1* – $A1_2O_3$:C (DPH), *2* – LiF:Mg, Ti (TLD-100) and *3* – LiF: Mg, Cu, P (TLD-100H), irradiated on a radioisotope ^{60}Co stand with the dose 9 mGy. For interpretation of the colors in the figures, the reader is referred to the web version of this article.

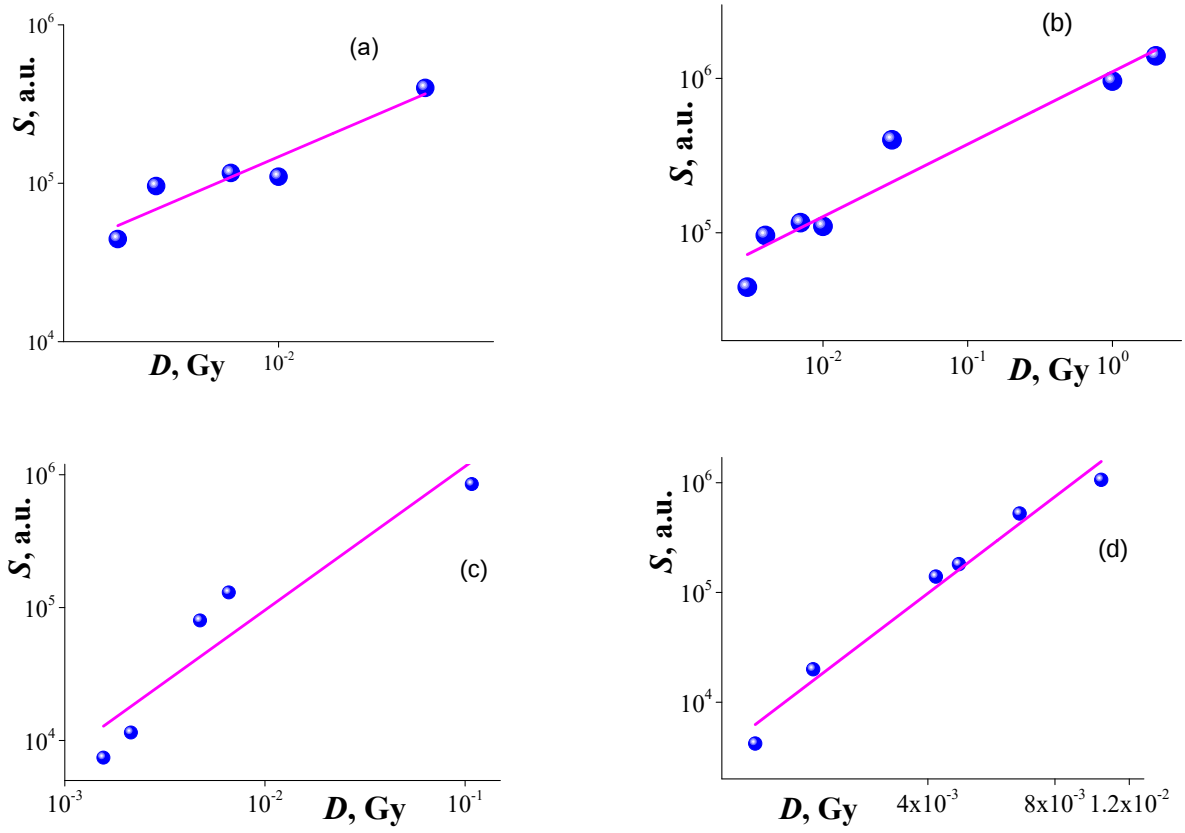


Fig. 3. Dose dependence of the luminosity of the characteristic peaks of thermal emission for the studied dosimeters: TLD-100, peak at 250 °C (*a*), the region of small (0…0.1 mGy), doses 0…2 Gy (*b*); DPH, peak at 170 °C (*c*); TLD-100H, peak at 225 °C (*d*).

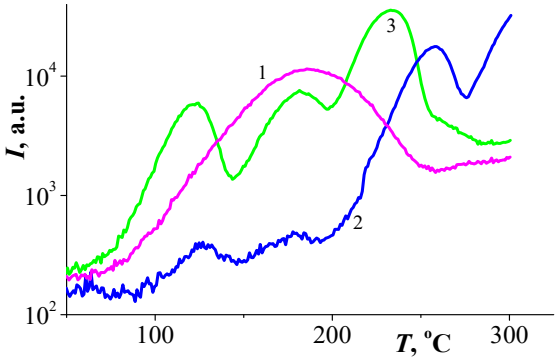


Fig. 4. Curves of thermally stimulated luminescence of $A1_2O_3$:C (*1*), LiF:Mg, Ti (*2*) and LiF:Mg, Cu, P (*3*) samples irradiated with a mixed gamma neutron field of a Pu–α-Be source. Irradiation time 166 hours.

Fig. 3 shows the results of studying the dose dependence of the luminescence characteristics, luminous intensity *S*, of the materials under study at the ^{60}Co radioisotope stand of the IEP of the National Academy of Sciences of Ukraine and the therapeutic ^{60}Co unit of the Oncology Center. Studying the dependences of the areas under the characteristic peaks of the TSL curves of the investigated samples on the radiation dose, *i.e.*, their luminosity rather than the peak values (Fig. 2), is a standard method of practical dosimetry. For the TLD-100 dosimeter, the data of such dependences are presented for the TSL peak at T = 250 °C, the intervals of both small 1…10 mGy (Fig. 3a) and larger doses, from 1 mGy to 2 Gy (Fig. 3b), were investigated. The results shown in Figs 3c and 3d relate to similar dependences of the DPH (peak T = 170 °C) and TLD-100H (peak T = 225 °C) dosimeters, respectively. As can be seen, the data shown in Fig. 3 indicates a linear dose-effect relationship for the studied dosimeters. Entering the sensitivity parameter of the dosimeters $\Delta S/\Delta D$ from the experimental data of Fig. 3, it can be established that: $(\Delta S/\Delta D)_{\text{TLD100H}}$ = 9.31 imp./μGy, $(\Delta S/\Delta D)_{\text{TLD100}}$ = 7.88 imp./μGy, $(\Delta S/\Delta D)_{\text{DPH}}$ = 3.631 imp./μGy. This confirms the hierarchy of TSL peak intensities of these dosimeters in isodose studies, Fig. 2.

As can be seen, the same temperature position of the characteristic peaks of TSL spectra is preserved as when irradiated at 60 °C (Fig. 1). However, there is a change in their intensities: for TLD-100H, the intensities of low-temperature peaks (120, 170 °C) increase by 2-3 times relative to the dosimetric peak at 225 °C compared to those obtained at the ^{60}Co bench. This can be attributed to the abolition of the interaction of γ-n and γ-radiation factors with the substance and to different irradiation times at Pu–α-Be and ^{60}Co stands. A comparison of Figs 1 and 4 shows that the sensitivity of TLD-100H to the registration of mixed γ-n rather than γ-radiation increases significantly compared to TLD-100 and DPH dosimeters.

The conditions of this experiment allow us to regulate the ratio of fast and thermal neutrons by introducing a “neutron stop” screen made of polystyrene.

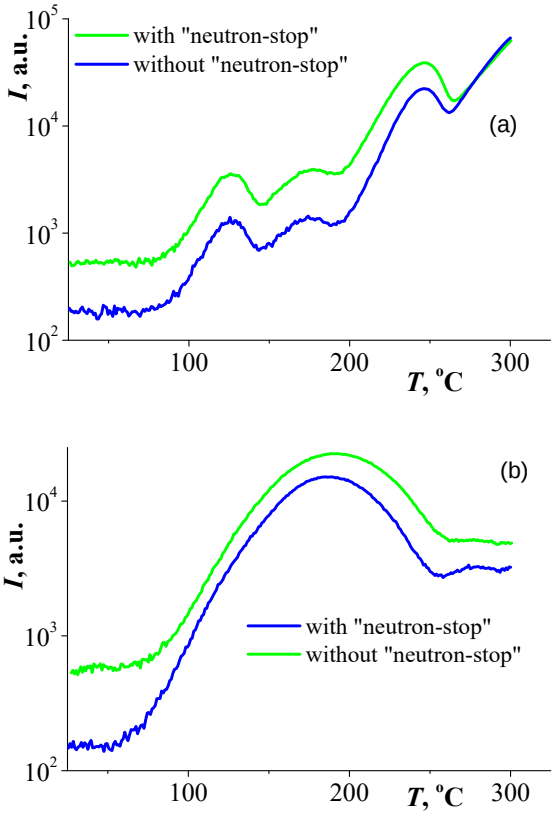


Fig. 5. Curves of thermally stimulated luminescence of TLD-100H samples (*a*) and DPH (*b*), irradiated with a mixed gamma-neutron field of a Pu–α-Be stand without (*a*) and with (*b*) the presence of a “neutron-stop” screen for neutron deceleration.

Such a shield does not affect the intensity of the accompanying γ-radiation of the Pu–α-Be stand, but it slows down fast neutrons. Fig. 5 shows the results of studying the TSL spectra of TLD-100H and DPH samples irradiated on the Pu–α-Be bench without and using a neutron retarder.

As can be seen from Fig. 5, the thermal illumination curves of the TLD-100H (LiF:Mg, Ti) and DPH (Al_2O_3:C) dosimeters without a neutron retarder, “neutron stop”, have a higher illumination intensity. This may indicate a more significant influence of fast neutrons on formation of the absorbed radiation dose in the studied materials than thermal energies. Fig. 6 shows the dependence of the luminous intensity *S* of the studied materials on the time of irradiation with the gamma neutron field of the Pu–α-Be stand without the presence of a neutron stop shield.

It can be seen that, as in the case of γ-quantum irradiation of the ^{60}Co stand, a linear dependence of the peak area on the irradiation time is observed, analogous to the absorbed dose. Fig. 6d shows the same results for TLD-100H obtained without and using the neutron retarder “neutron stop.” As in Fig. 5, it can be seen that the presence of a shield made of neutron stop blocks reduces the intensity of TSL illumination. We note the possibility of adjusting the parameters of radiation

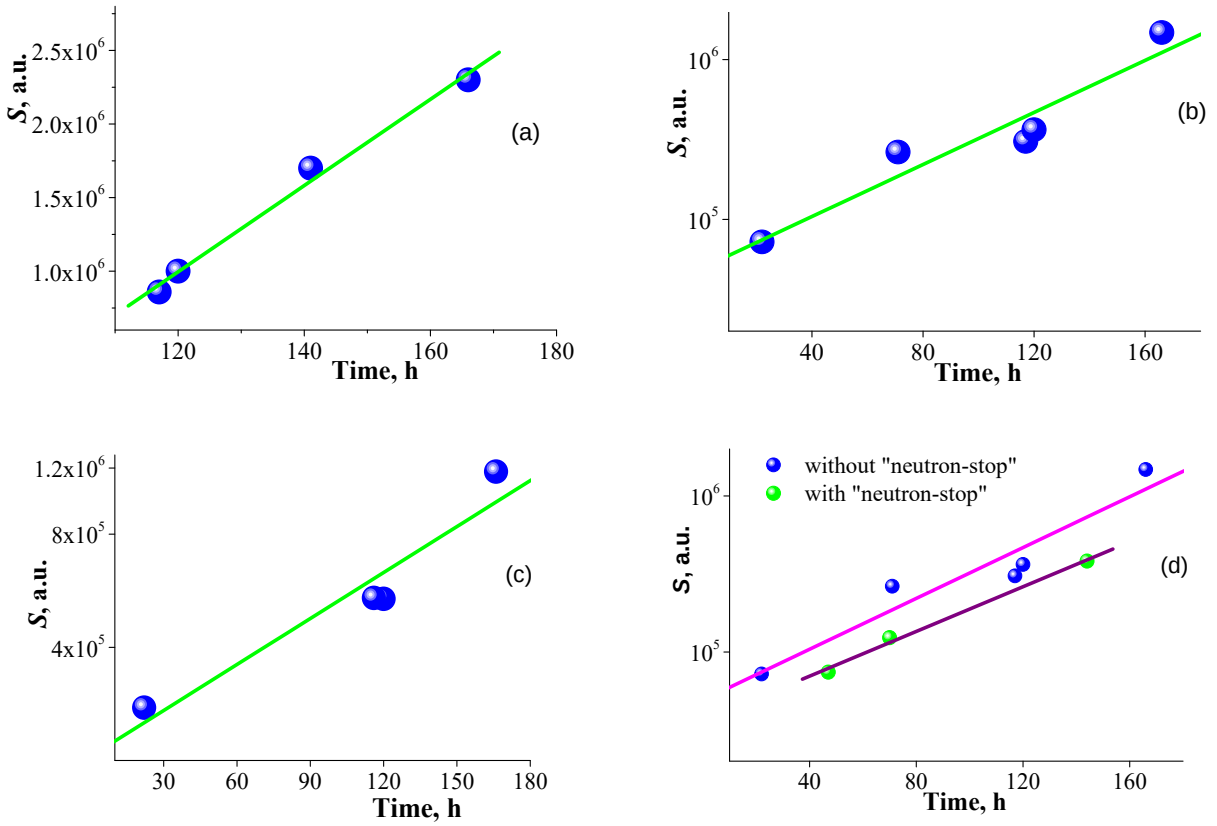


Fig. 6. Dose dependence of the luminosity of the characteristic thermal emission peaks for the studied dosimeters irradiated on the Pu–α-Be stand. Data for: DPH, peak at 170 °C (*a*); TLD-100H, peak at 225 °C (*b*); TLD-100, peak at 250 °C (*c*). TLD-100H, without and with the presence of a neutron moderator (*d*).

factors of the Pu–α-Be stand, particularly the ratio of γ- and n-components, as well as the number of thermal and fast neutrons of irradiation. Such a stand can be used to calibrate TSL dosimeters for operation in radiation fields of mixed nuclear radiation. The characteristics of TSL radiation are determined by the scheme of energy levels introduced into the conduction zone by doping the dosimeter matrix. The experimental data for TSL, obtained in Figs. 3 and 5, allow us to estimate the values of energy levels and kinetic parameters that determine luminescent phenomena in dosimetric materials. This practice is widespread in the modern theory of dosimetry. It involves [16–18] deconvolution of the experimental TSL dependences by a set of Gaussians and approximating the latter by theoretical dependences obtained from model calculations at fixed energy and frequency parameters. As a rule, the OTOR (one trap, one recombination) model is used, which takes into account the kinetics of the "zone-trap" transitions with intensity k_1, thermal excitation, "trap-zone," respectively, k_2 and recombination intensity with light emission k_3. Calculations in such a model are based on the use of various assumptions, for example, about the first- or second-order kinetics of electron-hole annihilation [19–21] under stimulated luminescence. In [17, 22], a method that is free from such approximations is proposed, which more adequately considers the physics of processes in the OTOR model and allows us to go beyond the one-level approximation. It is based on the scaling method, simplifying the calculation procedure and enabling us to

Table. Values of the energy and kinetic parameters responsible for the peak-like features of the spectrum of TSL LiF:Mg, Cu, P, Fig. 1, obtained by the Lumini package.

	k_1, $cm^3 \cdot s^{-1}$	k_2, $cm^3 \cdot s^{-1}$	k_3, $cm^3 \cdot s^{-1}$	E, eV	FOM, %
Peak 1	$1.2 \cdot 10^{17}$	$5 \cdot 10^{19}$	$3.3 \cdot 10^{15}$	1.6	0.25
Peak 2	$5 \cdot 10^{16}$	$8.2 \cdot 10^{19}$	$3.3 \cdot 10^{15}$	1.83	0.36
Peak 3	$1.8 \cdot 10^{18}$	$6 \cdot 10^{19}$	$3.3 \cdot 10^{15}$	2.1	0.089

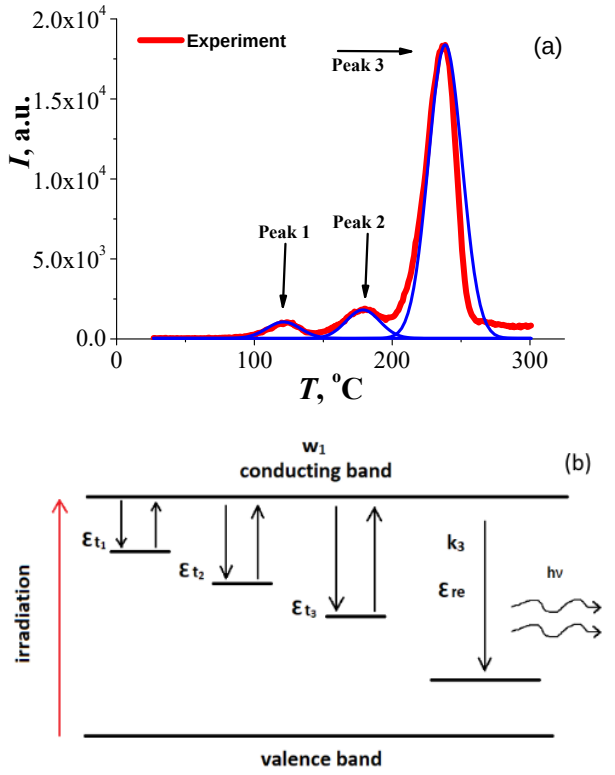


Fig. 7. (a) TSL spectrum of the TLD 100H dosimeter irradiated with a dose of 9 mGy after the deconvolution procedure. The results of deconvolution of peaks are highlighted in the figure by blue lines; (b) energy diagram of such a dosimeter, including three types of traps with different energies ε_{t1}–ε_{t3}, thermal activation of which leads to the appearance of peaks 1–3.

offer the Lumini package for *ab initio* studies of the model energy and kinetic parameters. Fig. 7 shows the result of deconvolution of the TSL spectra of the TLD 100H dosimeter by a set of Gaussians and the scheme of kinetic transitions in a model containing a set of three OTORs. The energy scheme of such a model contains three sets of traps with energies ε_{t1}–ε_{t3}, the thermal activation of which leads to the appearance of peaks in Fig. 7a).

As seen from Table, the *ab initio* method allows us to better determine the kinetics of the processes that form the TSL spectra and to establish the values of those parameters that are ignored when using various approximations.

4. Conclusions

A wide selection of dosimetric materials and research methods ensured the comprehensive nature of the studies. The results obtained indicate the possibility of using dosimeters based on doped lithium fluoride and leucosapphire, Al_2O_3:C, for dosimetric studies of γ- and mixed γ-n radiation fields. Such radiation conditions were created based on radioisotope stands using ^{60}Co and Pu–α-Be sources, the characteristics of which can be metrologically certified. It is shown that such dosimeters, widely used in medical and sanitary control, are promising for recording the absorbed dose starting from 1 mGy for γ- and mixed γ-n radiation factors. Moreover, the sensitivity of the TLD-100H (LiF:Mg, Cu, P) dosimeter was several times higher than that of the TLD-100 (LiF:Mg, Ti) and Al_2O_3:C (DPH). The linear dependence of the luminous intensity, *i.e.*, the integrated flux of the illumination of the temperature peaks of the TSL spectra of the studied samples on the dose or irradiation time for γ- and mixed γ-n irradiation, was established. However, these data indicate only the linearity of the dose-effect relationship; for the practical use of dosimeters to record the conditions of exposure to nuclear factors of different nature, a procedure for their calibration is necessary. The radiation stands used in this work allow for their metrological certification for electron, gamma, and neutron irradiation factors. A separate task is to create metrological stands-simulators of mixed nuclear radiation. In this case, it is possible to regulate the ratio of nuclear factors of different natures and for neutron fluxes to change their energy spectrum from thermal to high energies. It is also shown that the TSL structure of the peaks of dosimeters irradiated on ^{60}Co and Pu–α-Be stands does not differ qualitatively. Still, their intensity values are consistent with absorbed radiation doses of different natures. To quantify the effectiveness of dosimetric materials for fixed irradiation conditions, the sensitivity parameter was introduced in the work, the values of which satisfy the inequalities: $(\Delta S/\Delta D)_{TLD100H} > (\Delta S/\Delta D)_{TLD100} > (\Delta S/\Delta D)_{DPH}$. This confirms the hierarchy of intensities of characteristic peaks of TSL spectra of these dosimeters in isodose studies.

The obtained TSL spectra of the studied dosimeters can be used to assess the structure of their energy levels and parameters that determine the kinetics of luminescent phenomena. A common practice in recent years is the development of computerized deconvolution of light curves (CGLC) [19], which is based on the spectral decomposition of experimental dependences of TSL spectra and the use of various approximations in modeling. In this work, we demonstrate the possibility of supplementing the CGLC method with a scaling procedure that allows for *ab initio* modeling of the thermoluminescent characteristics of dosimetric materials for arbitrary energy levels and kinetic parameters. For the first time, the Lumini computer package [17, 22] was used for such purposes, which allowed us to establish the values of energy and kinetic parameters responsible for the peak-like features of LiF:Mg, Cu, P.

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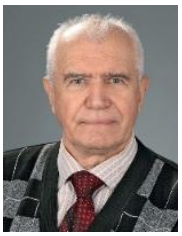
Authors' contributions

Pop O.M.: investigation, data curation (partially), writing – original draft.
Maslyuk V.T.: conceptualization, methodology, writing – review & editing.
Svatiuk N.I.: investigation, resources.
Rusyn A.V.: methodology, validation.
Yavorskyi P.V.: investigation, data curation.
Roman V.I.: investigation, resources.

Authors and CV

Oksana M. Pop, PhD in Physics and Mathematics (nuclear, elementary particle, and high energy physics), senior researcher at the Institute of Electron Physics, NAS of Ukraine. She is the author of over 80 publications and 5 patents. Her scientific interests include solid-state radiation physics, nuclear medicine, and nuclear radiation dosimetry. http://orcid.org/0000-0002-5690-1030



Volodymyr T. Maslyuk, Doctor of Sciences in Physics and Mathematics, Head of the department of photonuclear processes at the Institute of Electron Physics, NAS of Ukraine. Authored over 450 publications. The areas of his scientific interests include solid-state radiation physics, the theory of stability and fission of atomic nuclei, nuclear medicine, and radioecology.
E-mail: volodymyr.maslyuk@gmail.com,
http://orcid.org/0000-0002-5933-8394

Andriy V. Rusyn, Doctor of Medical Science, Head of the Transcarpathian Regional Clinical Oncological Centre. Authored over 117 publications. The areas of his scientific interests include radiotherapy, solid-state radiation physics, rectal cancer, and chemoradiotherapy. E-mail: arus27373@gmail.com, https://orcid.org/0000-0001-7886-9521

Natalia I. Svatiuk, PhD in Technical Sciences (environmental safety), Senior Researcher at the Institute of Electron Physics, NAS of Ukraine. She is the author of over 85 publications and 5 patents. Her scientific interests include environmental safety, bioactive materials, and nuclear medicine.
E-mail: svatiuknatalia@gmail.com,
https://orcid.org/0000-0001-9022-9252

Petro V. Yavorskyi, PhD student, Institute of Electron Physics, NAS of Ukraine. His research interests include nuclear radiation dosimetry, computer modeling, and the development of a mathematical package for finding dosimetric characteristics.
E-mail: petro0798@gmail.com,
https://orcid.org/0009-0008-3316-4382

Victoria I. Roman, PhD in Physics and Mathematics, Senior Research Associate, Head of the Electron Processes Department and the Elementary Interaction at the Institute of Electron Physics, NAS of Ukraine. She is the author of over 92 publications. Her scientific interests include solid-state radiation physics, autoionization, and resonance phenomena.
E-mail: viktoriyaroman11@gmail.com,
https://orcid.org/0000-0003-2499-8357

Термолюмінесцентна дозиметрія змішаного ядерного випромінювання: радіоізотопні стенди

О.М. Поп, В.Т. Маслюк, Н.І. Сватюк, А.В. Русин, П.В. Яворський, В.І. Роман

Анотація. Наведено результати дослідження дозиметрів на основі легованого фториду літію та лейкосапфіру на основі вуглецю С для використання в змішаних γ-n-полях. Радіаційні умови створювали радіоізотопні стенди на основі джерел ^{60}Co та Pu–α-Be. Показано, що досліджувані дозиметри є перспективними для реєстрації поглиненої дози, починаючи з 1 мГр для γ- та змішаного γ-n факторів випромінювання. Чутливість дозиметрів ТЛД-100Н (LiF:Mg, Cu, P) в кілька разів перевищує чутливість дозиметрів ТЛД-100 (LiF:Mg, Ti) і Al_2O_3:C. Для кількісної оцінки ефективності дозиметричних матеріалів для фіксованих умов опромінення введено параметр чутливості. Показано залежність сили світла від дози або часу опромінення для γ- та змішаного γ-n опромінення.

Ключові слова: радіаційні стенди, γ-n випромінювання, дозиметри, фтористий літій, лейкосапфір.