THERMOLUMINESCENT DETECTORS FOR SURVEILLANCE STUDIES OF RADIATION EXPOSURE OF THE POPULATION

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Abstract: Luminescent glow occurring in a substance exposed to ionizing radiation (IR) in the process of heating, thermoluminescence (TL) is now an effective method of registration of radiation-absorbed doses. It is important to be aware that the correct absorbed dose when exposed to mixed radiation with unknown characteristics is determined in the material of detector as well as in materials similar in composition (Z eff) and density [1–3]. In this connection, it is expedient to use different types of detectors for solution of different dosimetric problems. This study gives a comparison of the performance characteristics of TLD-K thermoluminescent detectors [4, 5], made of sodium silicate glass ceramic with the characteristics of IR detectors made of luminophors based on lithium fluoride monocrystals containing impurities of titanium and magnesium (TLD –100) [6, 7] and an anion of defective aluminum oxide (TLD–500) [8–11] widely used in thermoluminescence dosimetry. Comparison of a number of parameters that are relevant to the use of detectors in dosimetric monitoring of environment favors TLD-K detectors. The studies were carried out on the territory of the Kemerovo region.

Keywords: Thermoluminescence, detectors, ionizing radiation, absorbed dose, environment

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INTRODUCTION

Different kinds of thermoluminescence detectors (TLDs) can provide determination of absorbed doses in different environments, including tissues of human organism. The advantage of thermoluminescent dosimeters over other ones for determination of absorbed doses of ionizing radiation (IR) lies in high sensitivity of thermoluminescent detectors to different types of radiation, in integral character of dose accumulation, small sizes, and quite low cost price. Based on the above, one can conclude that TLDs can be effectively used for studying the real topography of radiation fields, for medical application of ionizing radiation and for radioecological monitoring of territories while controlling radiation effects in the environment.

In dosimetry, the most widespread thermoluminescent detectors among the staff who work with IR [6-8] are the ones based on LiF. This is due to the affinity between the effective atomic number of LiF on photoemission (Z_{eff} - 8.65) and the effective atomic number of the human soft tissue (Z_{eff} - 7.8). Substances with identical effective index numbers Z have comparable mass attenuation coefficients μ/ρ, where μ is the linear absorption coefficient, cm^{-1}, ρ is the density, g/cm^3. For photon absorption radiation IR is described by the following equation [1, 3, 13]:

$$N = N_0 e^{-\mu l} = N_0 e^{-(\mu/\rho)\rho l} \qquad (1)$$

Where N_0 – is the number of γ-quanta, included in absorber layer, N is the number of γ-quanta, passed through the absorber, g/cm^3, μ/ρ is the mass absorption coefficient, cm^2/g, l is the absorber layer thickness, cm, ρl is the layer thickness, g/cm^2. The mass absorption coefficient values of different substances are close for high-energy radiation and varied widely for low-energy photon radiation. For high-energy radiation, absorption is determined mainly by the absorber thickness, g/cm^2, whereas for low-energy γ-quanta absorber composition is of crucial importance. This is demonstrated in dependence of thermoluminescence on IR energy, especially in the area of photoelectric emission, i.e. in gamma impact energies lower than 200 keV.

Fig. 1 shows the dependence of the mass absorption coefficients on the gamma-quanta energy for some materials [1].

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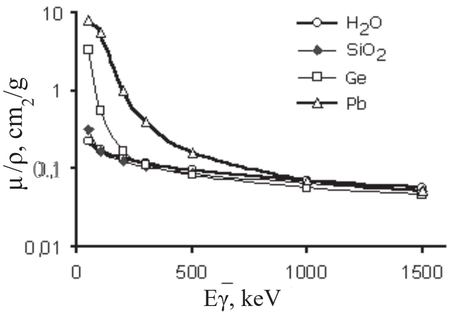

Fig. 1. Dependence of the mass absorption coefficients on the gamma-quanta energy for some materials [1].

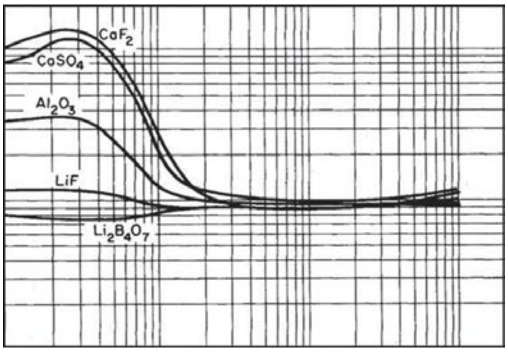

Fig. 2. Energy dependences of the thermoluminescence yield on a logarithmic scale (with reference to ^{60}Co) for different types of detectors [14].

Fig. 2 shows the energy dependences of the thermoluminescence yield on a logarithmic scale with reference to ^{60}Co for detectors based on different luminophors. The X-axis is the energy; the Y-axis is the relative yield of TL-$I_E/I_{Co\text{-}60}$

Despite the undoubted merit (equivalence of human soft tissues), detectors made of LiF-based materials, used in personal dosimetry, have a number of significant disadvantages (spread in sensitivity within the lot, a number of thermoluminescence peaks, a complex multi-step heating mode, dose dependence superlinearity at doses of 0.1 Gy, hygroscopicity) [6, 7].

Currently, the range of materials used to produce these detectors, has expanded considerably. Depending on the dosimetry problem, it is expedient to apply the most appropriate type of detector (luminophor). In addition to LiF, fundamentally different classes of substances are currently considered as radiation-sensitive environments, the most important of which are wide-band oxide materials such as Al_2O_3, BeO, MgO, SiO_2, and others [8–11, 13].

If you want to study the topography of radiation fields, used, for example, for exposure of semiconductor or ceramic components, it is advisable to use detectors with effective atomic number and element composition as close as possible to the irradiated material [14, 15].

If a luminophor has multiple traps in different depths (E_i), TL curve will have some peaks. Various thermoluminescence peaks may have different yield of luminescence, i.e., different probability of photon emission upon recombination, different luminescence spectral composition, which may lead to different luminescence registration efficiencies depending on the spectral sensitivity of the photodetector used. In this regard luminophors having single peak thermoluminescence curve, for example, α-Al_2O_3, SiO_2 (Fig. 3) have clear advantages over luminophors with a complex thermoluminescence curve, for example, LiF: Mg, Ti [13] (Fig. 4).

However, single-peaked shape of the thermoluminescence curve does not indicate that recombination after the charge carriers release from a trap is carried out at one of the luminescence center, but indicates only that the charge carriers are released from the trap of a certain depth (E_i). The luminescence spectrum may have some bands associated with the recombination of carriers at different impurity or structural defects - luminescence centers.

Experimental Part. Comparison of characteristics of different types of detectors

Thermoluminescent method of dosimetry is used in 90% of all cases of individual radiation control of personnel working in contact with ionization sources worldwide. The advantages of thermoluminescent detectors (TLDs) are: high sensitivity to different types of radiation, integral accumulation of radiation doze, small dimensions and relatively low cost. Due to the small size of TLD, these detectors can be used to detect real topography of ionization fields, including medical application of ionization radiation, as well as for radioecology monitoring of areas exposed to radiation.

It is necessary to use exactly the same dosimetric equipment and experimental conditions, and to compare results between the different types of detectors, to be

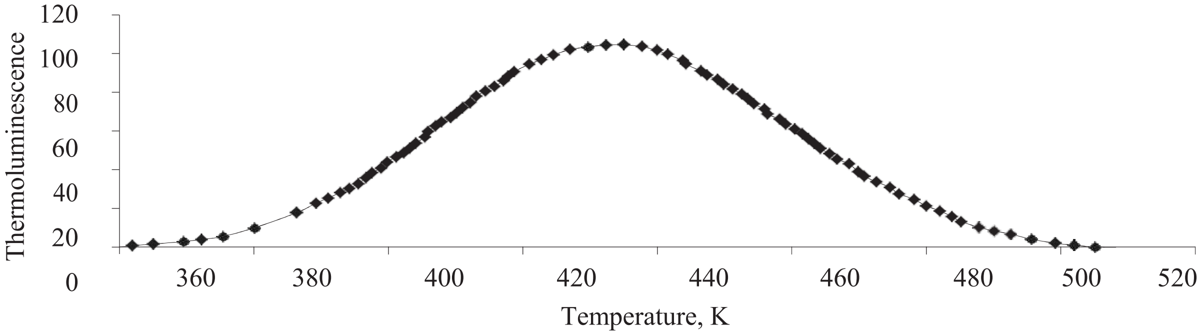

Fig. 3. Thermoluminescence curve of TLD-K detector (SiO_2) at a heating rate of 2°C/sec (a).

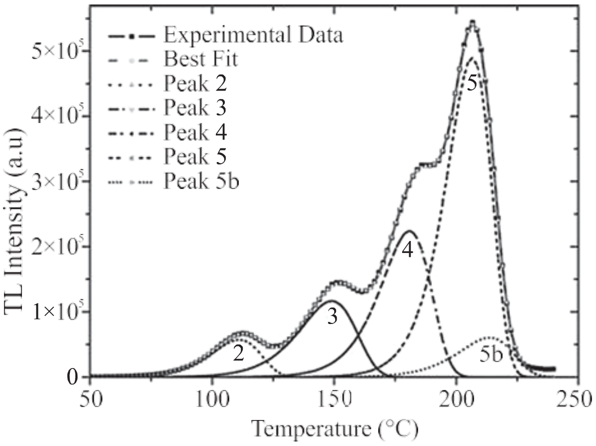

Fig. 4. Thermoluminescence curve of LiF: Mg, Ti under irradiation by ^{90}Sr/^{90}Y, dose 0.1 Gy, heating rate 1°C/sec (b).

able to measure the advantages and disadvantages of different types of detectors.

This work has used a modified dosimetric installation DTU-01M.

The dosimetric characteristics of various thermoluminescent detectors (made from various materials by differentmanufacturers) weretestedandcomparedduring experiment. A range of detectors was tested, including LiF (USA), LiF (Stavropol, Russian Federation), LiF (China), TLD-100 (LiF, Irkutsk, Russian Federation), TLD-K (SiO_2 Kemerovo, Russian Federation), TLD-500 (Al_2O_3) (Sverdlovsk, Russian Federation), Ca_3F, $CaSO_4$ and detectors based on the silicon oxide mixed with nanodispersed diamonds (TLD-KAS (SiO_2, C)). TLD-KAS is a synthesized test prototype based on a heterogeneous material composed of a luminophor with high luminescence yield, and diamond nanoparticles (varying the percentage of diamond nanoparticles results in variation of Zeff of the detector). A study of dosimetric characteristics was carried out. The test of the experimental material has demonstrated that the addition of diamond particles significantly increases the sensitivity range of the detector, which is important for industrial dosimetry.

During the experimental work, the dimensions and volume of detectors were calculated. Slope coefficients of linear dependence on the radiation dose, and sensitivity of detectors and materials were calculated with reference to the DTG-04 detector (relative coefficients taking into account the volume of detector).

Table 1 shows the main parameters (appearance, dimensions, peak temperature) of the detectors examined. Table 2 shows relative characteristics of different types of detectors based on different materials (LiF, Al_2O_3, SiO_2 CaF_2 $CaSO_4$).

Table 3 summarizes some characteristics of the detectors and conditions of their annealing and measurements (experimental and literature data).

Table 4 shows literature data for sensitivity of some LiF-based detectors under various irradiation conditions

TLD-K detectors have relatively wide detection range.

Table 1. Main parameters of the detectors used

Detector type	Diameter, mm	Thickness, mm	Volume, mm^3	Peak temperature, °C	Appearance
LiF (USA)	4.66	0.73	12.37	207	Non-transparent polycrystalline disk
LiF (Stavropol, Russian Federation)	4.41	0.95	14.6	200	Non-transparent polycrystalline disk
LiF (China)	4.58	0.75	12.2	200	Non-transparent polycrystalline disk
LiF (Irkutsk, Russian Federation)	4.57	0.94	15.3	203	Transparent crystalline disk
TLD-K(SiO_2)	2.78	0.51	3.9	132	Semi transparent amorphous square
TLD–500 (Al_2O_3)	5.04	0.85	17.0	171	Transparent crystalline disk
CaF_2	5.01	1.01	20.0	346	Non-transparent polycrystalline disk
$CaSO_4$	5.08	1.02	20.7	215	Non-transparent polycrystalline disk
TLD–KAS (SiO_2, C)	4.23	0.9	12.5	168	Non-transparent polycrystalline disk

Table 2. Comparative characteristics of detectors

Type	Material	Diameter	Thickness	V, mm^3	Relative sensitivity coefficient (with reference to DTG-04)	Relative coefficient (with reference to DTG-04)
USA	LiF	4.7	0.7	12	0.3	0.4
Stavropol	LiF	4.4	1.0	15	0.3	0.3
China	LiF	4.7	0.7	13	0.3	0.3
DTG–04 Irkutsk	LiF	4.6	0.9	15	1.0	1.0
Kemerovo TLD–K	SiO_2	2.9	0.5	4	0.8	2.7
Ekaterinburg TLD–500	Al_2O_3	5.0	1.0	19	32	29
CaF_2	CaF_2	5.0	1.0	20	6	5
Tartu $CaSO_4$	$CaSO_4$	5.1	1.0	21	13	10
TLD-KAS	SiO_2 +C	4.2	0.9	13	0.03	0.03

Table 3. Characteristics of the detectors and conditions of their annealing and measurements (experimental and literature data)

Detector type	Material	Annealing conditions	Measurement conditions	Dose equivalent mSv	Repeatability, %	Within-lot variation %
TLD–100	LiF - Mg, Ti	60 min at T 400°C	Prior heating to 100°C	0.042±0.008	4	23
TLD–400	LiF - Mg, Ti	60 min at T 400°C	Prior heating to 100°C	0.040±0.004	5	27
DTG–4	LiF - Mg, Ti	60 min at T 400°C	Prior heating to 100°C	0.039±0.004	4	20
TLD–1011	LiF - Mg, Cu, P	10 min at T 240°C	Prior heating to 60°C, 10 sec	0.004±0.002	6	24
TLD–500K	Intrinsic defects	Annealing at 400°C	Linear heating	<0.004	4	27
TLD–K	Intrinsic defects	Annealing at 400°C	Linear heating	<0.004	3	7

Table 4. Comparison of sensitivity of detectors (literature and experimental data)

Type of detector	Relative sensitivity ^{60}Co 10 mm of absorber layer	Relative sensitivity ^{60}Co	Relative sensitivity Cs-137 10 mm of absorber layer	Relative sensitivity
TLD–100	0.40±0.02	0.40±0.02	0.41±0.02	0.41±0.03
TLD–400		1.01±0.03		1.0±0.04
DTG–4	1±0.03	1.03±0.04	1±0.04	0.97±0.03
TLD–1011	5.8±0.2	6.2±0.3	4.8±0.2	5.2±0.3

Fig. 5 shows the dose dependence of TLD-K detector on a logarithmic scale. The Co-60 MXP-20 source was used. Fig. shows that detector reliably measures doses in the range of up to 1 kGy.

The slopes of the dose linear accumulation, i.e. sensitivity to photons of different energies are different. Exposure to gamma rays in MXP–20 (^{60}Co source) has shown that the thermoluminescence intensity dose dependence maintains its linearity in the range of up to 1 kGy (Fig. 6).

LiF-based detectors show deviation from the linearity at values near 0.1 Gy. TLD–500 detectors features a wider linearity range, but it is limited to 10Gy maximum. Thus, in the measurement of high doses, only TLD-K detectors allow high-dose measurements typical for radiation technologies.

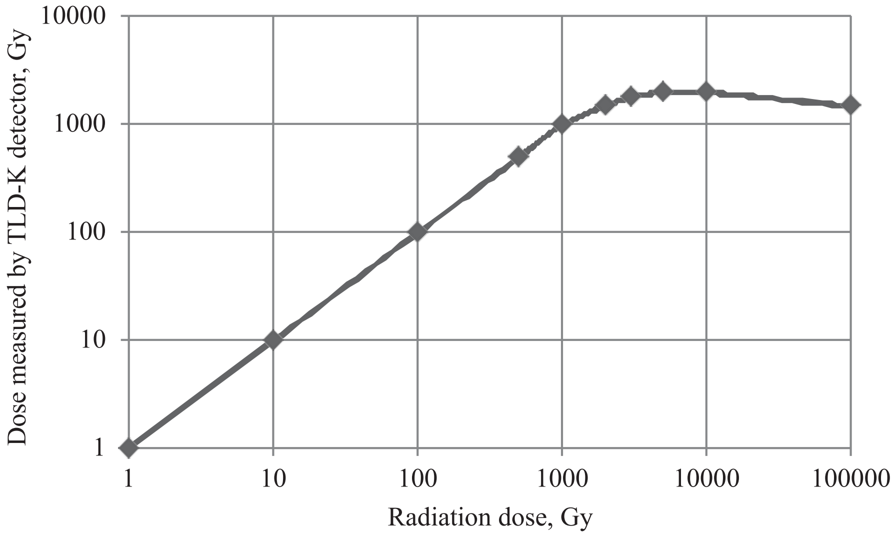

Fig. 5. Dependence of thermoluminescence yield on radiation dose a large dose range.

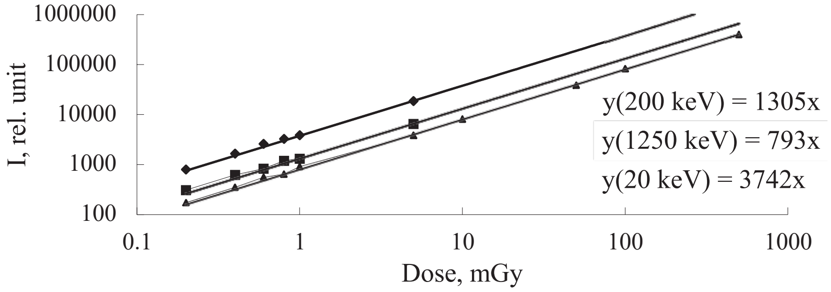

Fig. 6. Dependence of thermoluminescence intensity on radiation dose during photon bombardment (in the energy range of photons of 0.2-1000 mGy).

Practical dosimetry. Monitoring

A great potential for application of TLD-K detectors is determined by the adequacy of determination of the absorbed dose rate in soils, sediments, Quaternary sediments, archaeological pottery and other quartziferous materials. It is currently used in the method thermoluminescent dating of archaeological finds and Quaternary sediments, as well as in appraisal survey and surveillance studies in geology and medicine [16-23]. Due to the high uniformity of characteristics, lack of light sensitivity, low cost, and chemical stability, detectors are also used for the territorial radioecological monitoring and large-scale individual dosimetric control of population by measuring the maximum absorbed dose in the bone tissue.

Surveillance studies require a quantitative characteristic of the negative impact of human activity on the characteristics measured in the process of surveillance. The major question is: "What is the contribution of man-made factor in the results obtained?" The direct answer is monitoring before and after certain activities. However, precedently, the monitoring was not carried out prior to the beginning of industrial activity. Therefore, analysis of the results uses "plausible reasonings" based on a comparison of data for similar regions, theoretical estimates, intuitive notions, etc. It is therefore quite natural that the results are often not sufficiently reliable, and sometimes just wrong. Therefore, attempts to develop methods of environmental monitoring results processing, allowing to properly quantitative estimate of contribution of man-made factor in the results obtained. The fundamental possibility of solution is related to the following. Distribution of parameters in the absence of man-made factor is well described by the logarithmically normal distribution. The influence of man-made factor usually leads to distortion of this distribution. Thus, deviation value allow to get a conceptual idea of the role of man-made factor.

Fig.s 7 and 8 shows the dose distribution during monitoring measurements.

Gaussian is the natural radiation background caused by space radiation and radiation of naturally distributed radionuclides (without human influence). The most probable average value for Kemerovo Region for the monitoring period of five years and approximately 3,000 measurements is 0.3 ± 0.03cGy. The average values of natural background vary slightly for North (0.28) and South (0.32) of the region.

The influence of man-made of other factors (e.g, elevated concentrations of radon) leads to distortion of the distribution (Fig. 8).

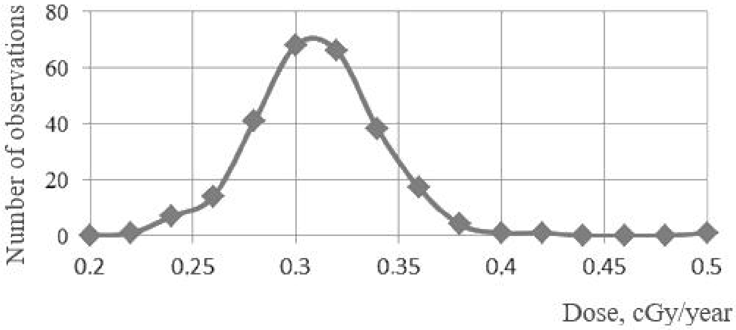

Fig. 7. An example of the radiation exposure distribution at the territorial monitoring in Kuzbass (natural background in the absence of man-made impact).

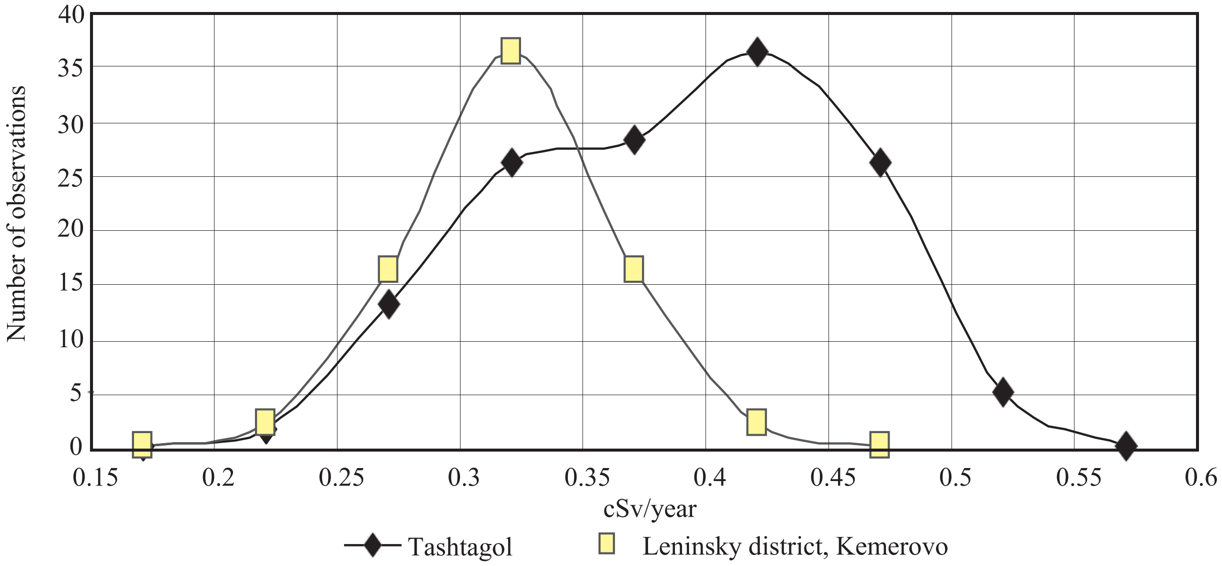

Fig. 8. An example of the radiation exposure distribution at the territorial monitoring in Kuzbass (background with additional exposure).

Radiation monitoring in propagation of the accident at the Fukushima nuclear power plant

TLD-K detector registers the absorbed dose of mixed radiation with a lower limit of 100 µGy. When exposed to normal background this dose is accumulated for 12 days. After two-day dose accumulation (March 14 and 15), the absorbed dose was 200 ± 120 µGy.

After four-day dose accumulation (from March 14 to 17), the absorbed dose was 170 ± 90 µGy, i.e. exponential decay was observed (see Fig. 9).

Thus, on March 13 and 14, the maximum absorbed dose of mixed radiation in Kemerovo was 10 times higher than the normal background radiation exposure. This indicates that at the beginning of the accident propagation a radioactive cloud moved towards the Far East, China, and Siberia at a speed ensuring its passage over Kuzbass 2 days after the earthquake and tsunami.

Medicine

Currently, medical procedures and treatment modes involving the use of radioactivity are the main contributors to the man-made dose of radiation received by humans. Unlike other sources of radiation dose received by the population, this type of radiation exposure can and should be controlled with the aim of reduction or optimization taking into account the "risk- benefit" effect. This involves determination of radiation exposure to patients and staff, as well as workspace dosimetry, study of dose distribution over the radiation field by comparing the effect of using various devices and various treatments. TLD-K detectors are well-suited to solve the above problems.

An example of using TLD-K detectors to control the radiation dose during certain X-ray surgical procedures (coronary angiography, stenting) is shown in Fig. 11. The energy dependence of the detector sensitivity is similar to the bone tissue energy dependence, therefore, this detector measures the maximum absorbed dose in the bone tissue with no corrections.

Dosimetry is conducted directly during fluoroscopy or certain X-ray surgical procedures ensuring the real working conditions during the procedure.

According to preliminary information on the work of a physician during certain X-ray surgical procedures (coronary angiography, stenting), based on ~ 200 days a year and an average daily absorbed dose of ~ 100 mSv, medical staff receives an annual radiation exposure of 20 mSv, which is lower than the dose limit for Group A professionals. Hand doses are higher. Thyroid dose of patients was very high; ~ 6mSv per surgical procedure, and gonad dose was 60 µSv, i.e. 100 times less (Fig. 11).

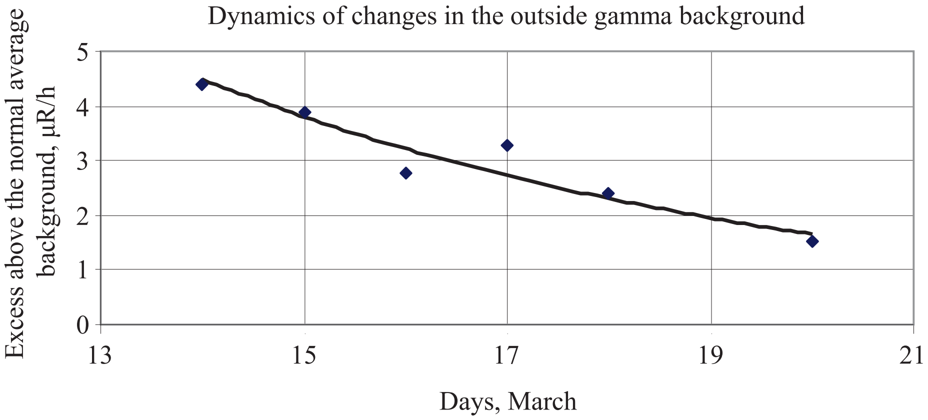

Fig. 9. Dynamics of change in the radiation background measured by TLD-K detectors.

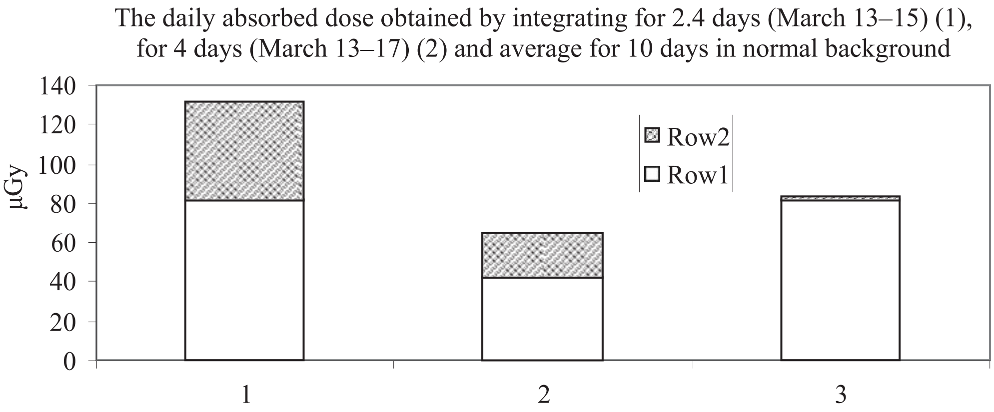

Fig. 10. Doses registered by TLD-K detectors.

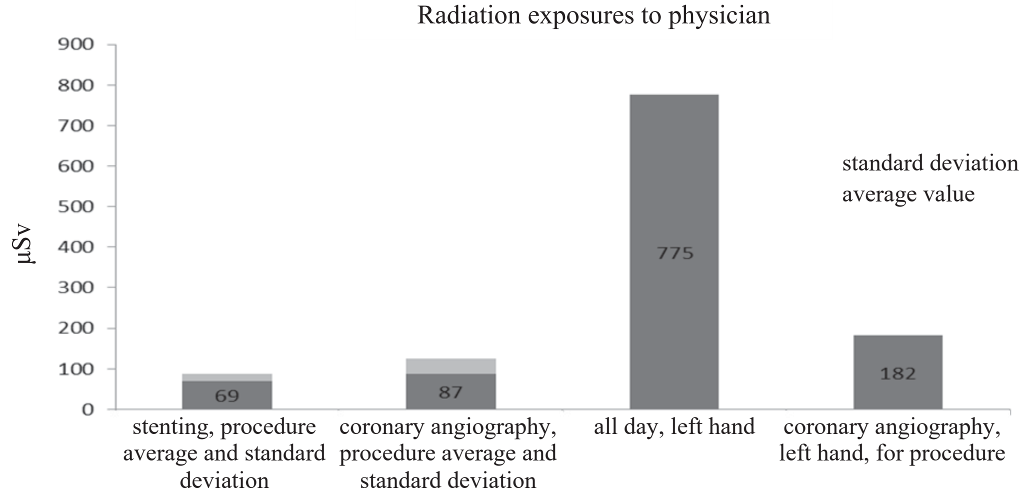

Fig. 11. Radiation doses during certain X-ray surgical procedures (coronary angiography, stenting).

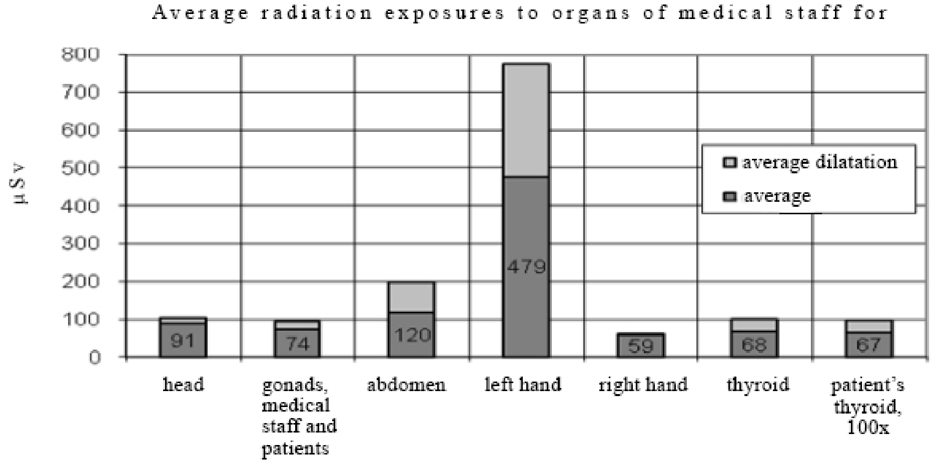

Fig. 12. Average radiation exposures to organs of medical staff for working day and to patients' thyroid.

CONCLUSION

The main advantages of TLD-K detectors (amorphous SiO_2) over similar devices is high homogeneity of thermoluminescent properties within the lot of detectors due to their physical and chemical structure - amorphous material. Therefore detectors grading by sensitivity necessary for all TLDs of monocrystalline material is not required. Thus, reliability of dosimetry increases and labor costs reduce.

Detection limit for small doses is less and more reliable than for LiF-based detectors, currently used for personnel monitoring.

Registration limits (linear region) are higher than that of all existing domestic and foreign TLD detectors, which is of crue importance for technological dosimetry.

Lack of sensitivity increases the reliability of the dosimetry.

The detectors have a high chemical resistance to aggressive media, high mechanical strength, are not soluble in water and therefore can be used in vivo.

Cost price of TLD-K (based on amorphous SiO2) is less than the cost of monocrystal detectors of domestic and foreign production.

The developed TLD-KAS will allow to vary the effective atomic number of the detector (Z_{eff}), varying the nanodiamond content and tailoring it to Z_{eff} of certain materials or human tissues.

This causes the preference for use of these detectors for the purposes of large-scale monitoring and process dosimetry compared to existing types of detectors.

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