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S.V. Nikiforov[1*], D.V. Ananchenko[1], T.V. Shtang[1], A.F. Nikiforov[1], V.M. Lisitsyn[2], M.G. Golkovski[3]

[1]*Ural Federal University, Yekaterinburg, Russia*
[2]*National Research Tomsk Polytechnic University, Tomsk, Russia*
[3]*Budker Institute of Nuclear Physics, SB RAS, Novosibirsk, Russia*
(*Corresponding author's e-mail: s.v.nikiforov@urfu.ru)

Synthesis, luminescent and dosimetric properties of ultrafine oxide ceramics for high-dose dosimetry of ionizing radiation

Thermoluminescent and dosimetric properties of ultrafine alumina and zirconia ceramics exposed to high-dose pulsed electron beam (130 keV, 1.5 kGy per one pulse) were studied. To synthesize ceramics with different sizes of crystallites, the authors sintered nanopowder compacts in air in an electric furnace at T=700-1700 °C, and exposed them to high-energy electrons (1.4 MeV) with high power density. It was found that ceramics annealing at T>1000 °C greatly increases crystallite sizes, which correlates with a significant growth of intensity of thermoluminescent peaks. Maximum thermoluminescent response is present in the ceramics that were obtained with an electron-beam method. This is due to formation of radiation-induced trapping and luminescence centers during synthesis. Analysis of dose dependences of thermoluminescence of the irradiated α-Al_2O_3 и ZrO_2 ceramics showed that these dependences are predominantly sublinear. Unlike their single-crystalline modifications, alumina-based ceramics have anomalous fading, which value increases as crystallites grow in size. The presence of the intensive isolated peak of thermoluminescence and sublinear character of the majority of dose dependences prove usability of the oxide ceramics synthesized in this work for measuring high doses of pulsed electron beams (unities-tens of kGy). Alumina-based ceramics require correction of thermoluminescent response by a fading value.

Keywords: aluminum oxide, zirconium oxide, ultrafine ceramics, thermoluminescence, electron-beam synthesis, crystallite size, dose characteristics, fading.

Introduction

Phosphors based on wide-gap oxide dielectrics (Al_2O_3, MgO, BeO, ZrO_2, etc.) have found applications in different fields of science and engineering. Many of them are used in micro-, nano-, and opto-electronic devices [1–3], in measuring devices for different physical quantities, for instance in scintillators and luminescent dosimeters [4, 5]. Nanostructured modifications of oxide materials, e.g. ultrafine ceramics with 50–500 nm nanoparticles, are a separate class of oxide materials.

Nanostructured phosphors have a number of characteristics which make mechanisms of formation of their luminescent properties under radiation significantly different from those of single-crystalline materials. Due to efficient annihilation of radiation defects on the boundaries of nanoparticles, nanomaterials are more radiation-resistant than the current bulk analogs [6] and retain their characteristics when exposed to intensive radiation. They can be used as dielectric integrated-circuit substrates which are employed at NPPs and in space, and as vacuum windows, lenses, mirrors for plasma diagnostics in fusion energy. Nanostructured ceramics are promising materials for high-dose (over 10 Gy) dosimetry of ionizing radiations, which relies on the effects of thermoluminescence (TL) and optically stimulated luminescence [4]. High doses of ionizing radiations are currently used in radiation technologies and academic research to sterilize food and medical instruments, purify sewage water, in brachytherapy, to modify properties of composite materials, metals and alloys, as well as for spectroscopy of intrinsic and impurity defects in semi-conductors and dielectrics [7].

High radiation resistance results in less effective generation of radiation defects in the exposed ultrafine ceramics with smaller grain size, which significantly affects the fundamental TL characteristics of the materials. The latter include TL response (the intensity of the maximum of TL peak and its light sum under exposure to a "test" dose), its dependence on the dose of ionizing radiation (dose characteristic), and the loss of the accumulated light sum while stored (fading). Possible applications of the material in TL dosimetry depend on these characteristics, since this requires high TL response, dose characteristic that is the closest to the linear one, and low fading. Thus, determining mechanisms of dose effects (formation of TL response in the studied phosphor, its dependence on a dose and storage time) is an important academic and practical task.

The purpose of this work is to synthesize and study the features and mechanisms of the processes of formation of luminescent and dosimetric properties in irradiated ultrafine ceramics based on wide-gap oxide dielectrics.

Experimental

Ultrafine ceramics of aluminum and zirconium oxides were studied in the paper. They were produced by using two methods from nanopowders (PLASMOTERM, Moscow, Russia) with 60–120 nm crystallites and impurity concentrations not higher than 0.5 ppm. The first method was based on annealing of the compacts which had been made by cold uniaxial pressing of the nanopowders under 100–120 MPa. The compacts were 5 mm in diameter and 1 mm thick. The compacts were annealed in air in muffle furnace General Therm LHT 04.1800 (S). The annealing temperature was varied within the range of T=700-1700 °C to change the sizes of nanoparticles.

Alongside with the traditional method of sintering ceramics by annealing them in a furnace, another method was used — electron-beam synthesis [8]. This method employs sintering of powders in the field of high-power flux of high-energy electrons (1.4 MeV) for less than 1 s. In the method not only heat processes, but also ionization ones are very important in synthesizing ceramics in a beam of fast electrons. These processes cause electron excitations to split into primary products of radiolysis and reactions between them.

X-ray diffraction analysis of the samples was carried out with Rigaku MiniFlex 600 diffractometer. To excite TL, the samples were exposed to pulsed electron beams (130 keV, 2 ns, 60 A cm^{-2}) from RADAN-EXPERT accelerator. The electron energy was much lower than the threshold energy for defect formation in the oxides (400 keV and higher) [9]. When such exposure is used, only changes in charge state of the present trapping and luminescence centers are observed. The exposure dose was varied by changing a number of pulses during irradiation. One pulse gave the value of the 1.5 kGy absorbed dose. TL curves of the ceramic samples were measured in the 50–400 °C range at linear heating rate of 2 K s^{-1}. FEU-130 photomultiplier tube (the maximum spectral sensitivity at 400–420 nm) was used to register TL.

Results and Discussion

Phase compositions of alumina and zirconia ceramics annealed at different temperatures were determined with X-ray diffraction analysis. The results show that all the samples of Al_2O_3 completely (100 %) consist of alpha-phase [10]. Zirconia ceramics entirely consist of monoclinic phase [11]. In addition, as the annealing temperature grows, the phase compositions of the ceramics do not change.

Scherrer equation was used to find the dependence of the crystallite size on ceramics annealing temperature on the base of analysis of half-width of diffraction reflections. In addition, results of SEM-image analysis were used [10]. The obtained dependences are shown in Figure 1. It can be seen that the crystallite sizes started growing when the annealing temperature increased above 1300 °C in Al_2O_3 samples. In the ceramics annealed at the maximum temperature (1700 °C), a mean grain size could not be determined due to almost complete absence of grain structure in the SEM-image of the sample surface. No significant changes in the crystallite sizes were observed in ZrO_2 ceramics annealed at T=700-1000 °C. Further increasing annealing temperature makes the grain grow to the value of order of 500 nm.

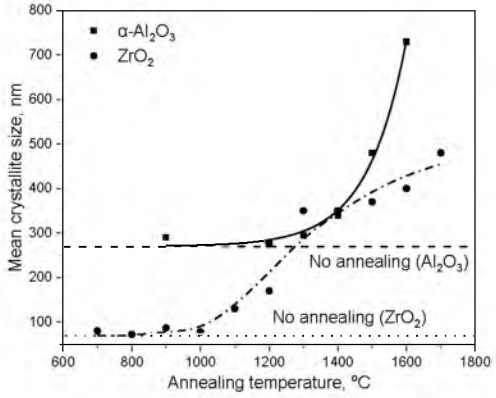

Figure 1. Dependence of the crystallite size in alumina and zirconia ceramics on the annealing temperature

Scherrer equation was also used to find the crystallite size in ZrO_2 ceramics synthesized with the electron-beam method. The resulting value (order of 100 nm) is nearly the same as the size of crystallites in the precursor powder taking into account the measurement error. In the samples synthesized in a flux of fast electrons, in spite of high temperatures, efficiency of crystallization significantly decreases in comparison with that of the samples traditionally annealed in furnace. Due to this fact, the initial size of crystallites is retained. This can be explained by a much shorter period of sintering of nanopowder in a fast electron beam, which is typical of other radiation methods of ceramic synthesis [12].

TL curves of alumina ceramics obtained at different annealing temperatures are shown in Figure 2a. It can be seen that all TL curves feature an isolated peak at 170 °C. It is noteworthy that this peak is close in the temperature position to the dosimetric TL peak in anion-defective α-Al_2O_3 single crystals which are used in TL dosimetry [13].

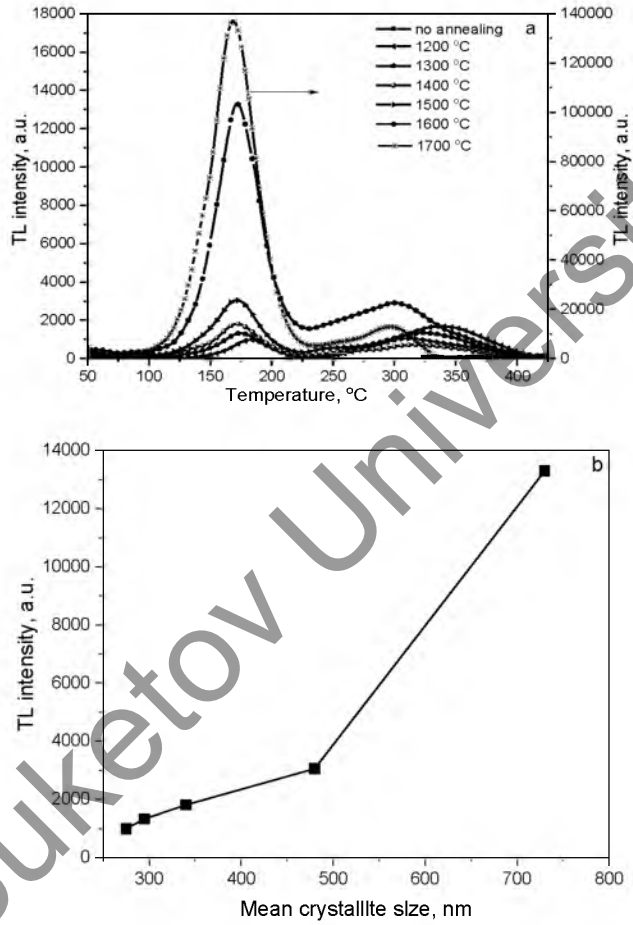

Figure 2. TL curves of α-Al2O3 ceramics annealed at different temperatures and exposed to a pulsed electron beam with 15 kGy dose (a) and dependence of TL peak intensity at 170 oC on the crystallite size (b)

Figure 2b shows dependence of TL peak intensity at 170 °C on the crystallite size. As it can be seen, growing grain size increases TL intensity. The most intensive rise of the intensity is observed when the crystallite are larger than 500 nm.

Figure 3a features TL curves of ZrO_2 ceramics exposed to a test dose of a pulsed electron beam (15 kGy). TL peak at 120 °C is seen in all the samples. A less intensive peak at 210 °C is also found in the ceramics synthesized with the electron-beam method. Figure 3b shows a dependence of the TL maximum intensity in the peak at 120 °C on the crystallite size. It can be seen that growing grain size increases TL intensity like in the alumina-based ceramics. The fastest growth is observed when the crystallites are bigger than 350 nm. The maximum TL intensity in the 120 °C peak is found in the ceramics synthesized with the electron-beam method. This can be due to formation of radiation defects during the synthesis. The defects are either charge carrier traps or luminescence centers.

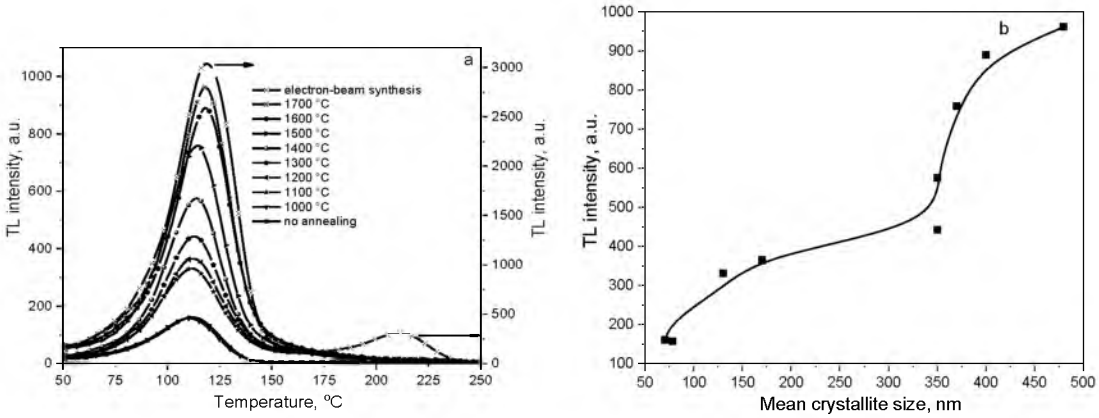

Figure 3. TL curves of ZrO_2 ceramics synthesized under different conditions, after exposure to a test dose of a pulsed electron beam (15 kGy) (a) and dependence of TL peak intensity at 120 °C on the crystallite size (b)

The effect of TL response drop with decreasing crystallite size had been observed earlier by other authors [14–16]. Ref. [17], which reviews literature data, shows that the reported effect is a common characteristic for many TL materials. Physical nature of the effect of dropping TL yield with decreasing crystallite size has not been well grounded yet and requires further studies. Increasing possibility of non-radiative transitions when nanoparticles lessen in size due to, for instance, localized transitions of electrons and holes between trapping and luminescence centers may be one of the reasons for the reported effect. Other possible causes of TL intensity drop may include decreasing concentration of traps, growing of the surface-to-volume ratio with decreasing crystallite size [17].

We studied dose dependences of TL intensity of dosimetric peaks of the ceramics (at 170 °C for aluminum oxide and at 120 °C for zirconium oxide). The obtained dependences were approximated with linear functions in different ranges of the changing dose. Slope angles of the dependences were values of non-linearity coefficient *k*. Approximation results are given in Table.

Table

Approximation results of dose dependences of TL peaks in the ceramics synthesized under different conditions

No.	Type of samples	Dose range, kGy	Non-linearity coefficient
1	Al_2O_3 (1700 °C)	1.5-15	1.0
2	Al_2O_3 (1700 °C)	15-75	0.3
3	Al_2O_3 (1600 °C)	1.5-150	0.49
4	Al_2O_3 (1500 °C)	1.5-15	0.62
5	Al_2O_3 (1500 °C)	15-150	0.22
6	Al_2O_3 (1400 °C)	1.5-7.5	0.58
7	Al_2O_3 (1400 °C)	7.5-45	0.34
8	ZrO_2 (electron-beam synthesis)	1.5-7.5	0.84
9	ZrO_2 (1700 °C)	1.5-15	0.79
10	ZrO_2 (1600 °C)	1.5-15	0.51
11	ZrO_2 (1500 °C)	1.5-15	0.49
12	ZrO_2 (1400 °C)	1.5-7.5	0.89

It can be seen that the majority of dose dependences are sublinear ($k<1$), the only exception is Al_2O_3 ceramics synthesized at 1700 °C. In these ceramics, TL dependence on the dose is close to a linear one ($k=1$) in the 1.5–15 kGy range. The maximum range of the registered doses in which k=const is found in the alumina samples that were synthesized at 1600 °C. In these samples $k=0.49$ when the dose is changed by two orders of magnitude (from 1.5 to 150 kGy). It is noteworthy that according to the data from Table, there is not any noticeable correlation of non-linear coefficients and crystallite sizes in the ceramics under study. Sublinear character of dose dependences may be due to the competition in trapping charge carriers between different defects [18, 19]. The presence of such defects is more likely in nanostructured materials in comparison with bulk analogs.

In addition, we studied TL fading in alumina and zirconia ceramics. The samples were irradiated at room temperature with a 15 kGy dose and kept at this temperature in darkness for a set period of time. After this, the remaining TL was measured. The results show that unlike single-crystalline aluminum oxide, the samples of Al_2O_3 ceramics have significant fading. Maximum losses of dosimetric information (up to 60 % for one hour) are observed in the samples with the largest size of the grains annealed at 1600 °C. When the annealing temperature and nanoparticle size decrease, fading becomes less pronounced. At T=1400 °C, its value does not exceed 30 % per hour (Fig. 4).

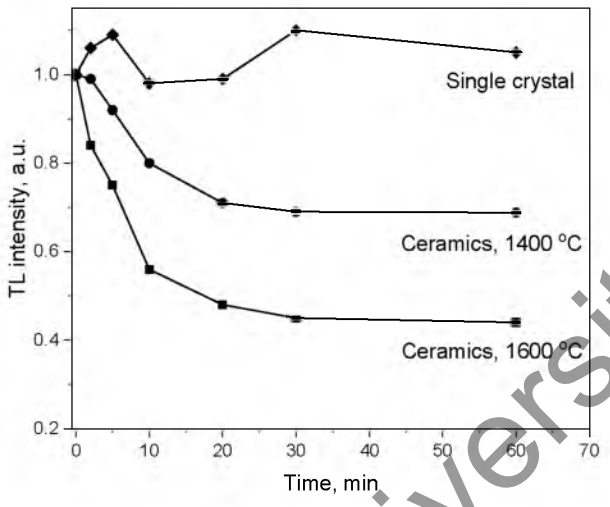

Figure 4. Fading of TL peak at 170 °C of α-Al_2O_3 ceramics synthesized by annealing at different temperatures

Examples of both decreasing fading when nanoparticle sizes grow and increasing one are available in literature [15, 16]. The effect of quantum tunneling may be a reason for anomalous TL fading in the samples [20]. In [20], computer simulation of the effect showed that both decrease and increase in intensity of tunneling recombination with growing grain size is possible at certain parameters of the nanomaterial (nanocrystal radius, tunneling length, initial mean distance between electrons and positive ions). Moreover, the intensity rise may serve as a reason for TL fading growth when crystallites become bigger in size, as it was observed in our experiments (Fig. 4).

Unlike alumina ceramics, noticeable decrease in TL intensity in the peak at 120 °C is not observed when the exposed ceramic ZrO_2 samples are kept for one hour. While its value accidentally changes within 10–15 %, which is comparable with an exposure dose error. The absence of anomalous fading may imply an insignificant role of quantum-mechanical tunneling and localized transitions of charge carriers in the formation of TL at 120 °C in the studied ZrO_2 ceramics.

Conclusions

In this work, samples of α-Al_2O_3 and monoclinic ZrO_2 ceramics were obtained by static annealing of nanopowder compacts. The samples were synthesized in air in a 700–1700 °C temperature range. Scanning electron microscopy and X-ray diffraction analysis were used to find that high-temperature annealing does not affect phase composition. However, it results in a significant growth of crystallite sizes. In ZrO_2 ceramics synthesized with the electron-beam method, the nanoparticle size is almost the same as the size of crystallites in the initial nanopowder. It was revealed that intensities of TL peaks grow with increasing grain size in the studied samples.

Dose dependences of TL in the samples of the ultrafine ceramics are predominantly sublinear. For aluminum oxide ceramics, unlike single crystals, the effect of anomalous TL fading which is presumably due to the presence of tunneling recombination and localized transitions typical of nanostructured materials was determined. Moreover, the value of anomalous TL fading increases with growing crystallite sizes.

The obtained sublinear dose dependences of TL show that the synthesized oxide ceramics are promising materials for TL dosimetry of high-dose (1–100 kGy) pulsed electron beams, which are used in radiation technologies and academic research. For aluminum oxide ceramics, due to significant TL fading, dosimetric

information should be read out immediately after the end of exposure or special methods for correction of the readings from TL detectors should be used.

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21

С.В. Никифоров, Д.В. Ананченко, Т.В. Штанг, А.Ф. Никифоров,
В.М. Лисицин, М.Г. Голковский

Иондаушы сәулеленудің жоғары дозалы дозиметриясына арналған аса жұқа оксидті керамиканың синтезі, люминесцентті және дозиметриялық қасиеттері

Жоғары дозалы импульсті электронды сәулемен (130 кВ, импульсқа 1,5 кГр) сәулеленген алюминий және цирконий оксидтеріне негізделген ультра жұқа керамиканың термолюминесценттік және дозиметрлік қасиеттері зерттелді. Кристаллит өлшемдері әртүрлі керамикаларды синтездеу үшін наноұнтақтардан жасалған компакттарды ауада T = 700-1700 °C температурада электр пешінде агломерациялау, сондай-ақ оларды жоғары қуат тығыздығы бар жоғары энергиялы электрондармен (1,4 МэВ) сәулелендіру қолданылды. Керамиканың T>1000 °C күйдіруі термолюминесценттік шыңдардың қарқындылығының айтарлықтай өсуімен корреляциялық кристаллиттер мөлшерінің айтарлықтай ұлғаюына әкелетіні анықталды. Электрондық сәуле әдісімен өндірілген керамика максималды термолюминесценттік реакцияға ие, бұл синтез кезінде радиацияның әсерінен түсіру және люминесценция орталықтарының пайда болуымен байланысты. Сәулеленген a-Al_2O_3 және ZrO_2 керамикасының термолюминесценциясының дозаға тәуелділіктерін талдау олардың басым сублинарлық сипатын анықтады. Алюминий оксиді негізіндегі керамика үшін, монокристалды модификациядан айырмашылығы, кристаллит өлшемі ұлғайған сайын шамасы арта түсетін аномальды термолюминесцентті фединғтің болуы анықталды. Интенсивті оқшауланған термолюминесценция шыңының болуы және көптеген дозаға тәуелділіктердің субсызықтық сипаты импульстік электронды сәулелердің жоғары дозаларын (бірнешеден ондаған кГр) өлшеу үшін осы жұмыста синтезделген оксид керамикасының болашағын дәлелдейді. Бұл жағдайда алюминий оксиді негізіндегі керамика үшін фединг мөлшеріне термолюминесцентті реакцияны түзету қажет.

Кілт сөздер: алюминий оксиді, цирконий оксиді, ультра жұқа керамика, термолюминесценция, электронды сәуленің синтезі, кристаллит мөлшері, доза сипаттамалары, фединг.

С.В. Никифоров, Д.В. Ананченко, Т.В. Штанг, А.Ф. Никифоров,
В.М. Лисицин, М.Г. Голковский

Синтез, люминесцентные и дозиметрические свойства сверхтонкой оксидной керамики для высокодозной дозиметрии ионизирующего излучения

Исследованы термолюминесцентные и дозиметрические свойства ультрадисперсных керамик на основе оксидов алюминия и циркония, облученных высокодозным импульсным электронным пучком (130 кэВ, 1,5 кГр на один импульс). Для синтеза керамик с различным размером кристаллитов использовалось спекание компактов, изготовленных из нанопорошков, на воздухе в электрической печи при T=700–1700 оС, а также их облучение высокоэнергетическими электронами (1,4 МэВ) с высокой плотностью мощности. Установлено, что отжиг керамик при T>1000 °С приводит к существенному росту размера кристаллитов, что коррелирует со значительным увеличением интенсивности термолюминесцентных пиков. Максимальным термолюминесцентным откликом обладают керамики, полученные электронно-лучевым методом, что связано с образованием радиационно-индуцированных центров захвата и свечения при синтезе. Анализ дозовых зависимостей термолюминесценции облученных керамик α-Al_2O_3 и ZrO_2 выявил их преимущественно сублинейный характер. Для керамик на основе оксида алюминия, в отличие от монокристаллической модификации, установлено наличие аномального термолюминесцентного фединга, величина которого увеличивается с ростом размера кристаллитов. Наличие интенсивного изолированного пика термолюминесценции и сублинейный характер большинства дозовых зависимостей доказывают перспективность синтезированных в настоящей работе оксидных керамик для измерения высоких доз импульсных электронных пучков (единиц — десятки кГр). При этом для керамик на основе оксида алюминия требуется коррекция термолюминесцентного отклика на величину фединга.

Ключевые слова: оксид алюминия, оксид циркония, сверхтонкая керамика, термолюминесценция, электронно-лучевой синтез, размер кристаллитов, дозовые характеристики, фединг.

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Information about the authors

Sergey Nikiforov (corresponding author) — Doctor of physical and mathematical sciences, Head of Chair, Department of Physical Techniques and Devices for Quality Control, Ural Federal University, Mira street 21, 620002, Yekaterinburg, Russia; *e-mail: s.v.nikiforov@urfu.ru*. https://orcid.org/0000-0001-7797-8267

Daria Ananchenko — Senior lecturer, Junior researcher, Department of Physical Techniques and Devices for Quality Control, Ural Federal University, Mira street 21, 620002, Yekaterinburg, Russia; *e-mail: d.v.ananchenko@urfu.ru.* https://orcid.org/0000-0002-1998-8405

Tatiana Shtang — Candidate of physical and mathematical sciences, Associate professor, Department of Physical Techniques and Devices for Quality Control, Ural Federal University, Mira street 21, 620002, Yekaterinburg, Russia; *e-mail: t.v.shtang@urfu.ru.* https://orcid.org/0000-0002-6454-4870

Alexander Nikiforov — Doctor of chemical sciences, Professor, Department of Radiochemistry and Applied Ecology, Ural Federal University, Mira street 21, 620002, Yekaterinburg, Russia; *e-mail*: *a.f nikiforov@urfu.ru*

Victor Lisitsyn — Doctor of physical and mathematical sciences, Professor, Department of Materials Science, Engineering School, National Research Tomsk Polytechnic University, Lenin Avenue, 30, 634034, Tomsk, Russia; *e-mail: lisitsyn@tpu.ru*. https://orcid.org/0000-0002-2075-4796

Michael Golkovski — Candidate of physical and mathematical sciences, Senior research fellow, Budker Institute of Nuclear Physics, SB RAS, Academician Lavrentiev Avenue, 11, 630090, Novosibirsk, Russia; *e-mail: golkovski@mail.ru*. https://orcid.org/0000-0003-4399-444X