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CREATING REFERENCE NEUTRON FIELDS FOR CALIBRATION OF NEUTRON SPECTROMETERS AND COMPUTING THEIR SPECTRA

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We consider the current state of metrological support for neutron spectrometry. We propose a method for creating reference neutron fields with energy spectra of different shapes from radioisotope sources, with the aim of designing a test setup for verification of real-time neutron spectrometers we have developed.

Keywords: *neutron radiation, spectrometry, metrological support, reference neutron fields.*

Introduction. In [1], we worked up a proof-of-concept for designing a multidetector real-time neutron spectrometer with computer-assisted retrieval of the spectra of the measured fluxes, using a pretrained neural network. It essentially involves coprocessing the signals obtained from several neutron detectors with different spectral characteristics, together spanning the entire energy range of the neutron fluxes to be measured. In [2], we described methods for obtaining neutron detectors with different spectral characteristics and gave the results of their calculations using macroscopic and microscopic approaches, where the latter were realized using the GEANT4 program library. However, commercialization of neutron spectrometers is not possible without developing their metrological support, the current state of which is unsatisfactory not only for real-time neutron spectrometers but also for neutron spectrometry in general: standard neutron sources with certified energy spectrum are not available, there are no certified procedures for verification of neutron spectrometers, standards for neutron spectrometers and methods for their verification. Even the activation analysis method widely used in nuclear engineering, for which metrological support has been the focus of the most attention [3], does not have satisfactory metrological support since its most important step (retrieval of the energy spectrum from the measured values of the induced activities for a set of indicators bombarded by the neutron flux under study) is not metrologically supported and is done by purely computational means. In this case, existing computer programs for retrieval (deconvolution or unfolding) of the spectrum require using additional *a priori* information about the shape of the spectrum, since mathematically this problem is ill-posed, i.e., allows for many solutions. Such problems in obtaining a unique solution require regularization, which also involves drawing on additional information. In [3], two approaches to solving this problem are considered.

The first approach is based on a matrix solution and requires a sufficiently large number of indicators in the set with diverse spectral characteristics, the maxima of which correspond to different regions of the spectrum for the neutron fluxes to be measured. Only the most general requirements are imposed on the shape of the function for the envelope of the retrieved spectrum: positivity over the entire energy range, continuity, smoothness. However, actually these conditions proved to be insufficient for obtaining a unique solution, due to constraints on the diversity of spectral characteristics for the indicators used. Therefore it was necessary to assign a shape for the prototype spectrum. This group can include the programs RDMM, SPECTRA, CRISTALL BALL, RFSP mentioned in [3]. In [4], a FORTRAN program is proposed that was developed based on the principle of maximum entropy and the maximum likelihood method. It showed satisfactory characteristics for many spectra of different shapes, but the authors admitted that uniqueness of the solution obtained was not proven. In [5], the PRIP and GRAVEL algorithms, used for retrieval of spectra from the results of activation and multisphere methods for measuring neutron spectra, were compared and their robustness and high accuracy were proven. Detector responses were determined computationally using the GEANT4 software. The measurements were made with monoenergetic spectra

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with energies 8 MeV and 12 MeV, but the quantitative accuracy characteristics of the algorithms were not given; only the convergence characteristics were indicated.

The second group includes algorithms initially based on an iterative procedure for distortion of the *a priori* specified spectrum to match results of the neutron activation measurements. The comparative analysis done at VNIIFTRI [All-Russian Research Institute of Physicotechnical and Electronics Measurements] for these methods showed that the algorithms in the second group provide greater robustness and less dependence on correctly assigning the *a priori* spectrum, but nevertheless the dependence on *a priori* assigned characteristics of the spectrum to be retrieved is quite substantial for algorithms in both groups. Therefore in order to make it easier to solve this problem, at VNIIFTRI libraries of classified spectra, BKS-1 and BKS-2, were prepared, which were used in the computer programs developed at the Institute for retrieval of the spectrum: PROSPEKT-1 and PROSPEKT-2 (the procedure for their application is given in the guideline MI 1806-87). However, the question of the uncertainties in retrieval of the spectrum remains unanswered. We can only estimate the statistical uncertainty in the characteristics of the retrieved spectrum with mandatory variation of the initial data (measured values of the induced activity of the indicators), which determines not so much the uncertainties in the method as the robustness of the solution obtained.

A similar situation arose with metrological support of the multisphere Bonner method, which is rather popular in Europe and the USA [6–8]. Besides the same problem with determination of the uncertainties in the computed retrieval of the spectrum from the measured integrated flux densities with each of the moderator spheres, an additional and no less complicated problem involves estimating the uncertainties in determination of the spectral characteristics of the detector together with each moderator sphere, without knowing which it is not possible to retrieve the spectrum.

We can objectively estimate the uncertainties in retrieval of the measured spectrum by any measurement means only by measuring neutron fluxes with an exactly known spectrum. And if this measurement means is certified as a measuring instrument, then it must be verified in neutron fields with the most different shapes of the spectrum in the specified energy range. Therefore the metrological basis for neutron spectrometers should be standard sources, certified not only with respect to activity and total neutron fluence, but also with respect to the energy spectrum, and the shapes of their spectra should span as much as possible the diversity of shapes of spectra for real neutron fields which will be encountered in measurement practice.

In this paper, we propose obtaining reference neutron fields with spectra of different shapes in test setups, in which the primary radioisotope neutron sources are placed in a collimating system, the channel of which can be covered by moderator disks made from hydrogen-containing material of different thicknesses. We present the results on optimization of the design for such a setup, which would let us incorporate this system into current neutron test setups used for verification of neutron radiometers and dosimeters, and also spectra of reference neutron fields obtained on such a setup, computed using the GEANT4 program library. We selected the GEANT4 software because of its universality, accessibility (free access) and regular updates, although the MCNPX software is considered as the most accurate for calculations of neutron interactions. The special comparison of the calculation results using MCNPX and GEANT4 done in [9] showed agreement between them within 3–5% when estimating the detector response in the energy range from 0.025 eV to 20 MeV and within 1% for energies from 100 eV to 5 MeV. In order to estimate the accuracy of these programs, in [9] a comparison was done for simulations of the spectrum of a ^{252}Cf source from the ^{252}Cf ISO-8529 standard. For the MCNPX software, the discrepancy was 2.2–2.5%, for GEANT4 5.3–6.5 %. Thus using GEANT4 for calculations of the spectra of reference neutron fields ensures an accuracy that is sufficient in practice and lets us calculate the spectra not only of the neutron component but also the gamma components of mixed radiation.

Creating Reference Neutron Fields with Spectra of Different Shapes. The most convenient, robust, and suitable for application in practical test setups are radioisotope sources of neutron radiation. They are mass produced and all together span a sufficiently wide range of energies. Of all the (α,*n*) sources, the ^{239}Pu–α–Be source has the longest half-life (24,360 years), and hence high long-term stable neutron yield when a sufficient quantity of them escape ($1.8 \cdot 10^5$ neutrons/s per gram), a low level of accompanying gamma radiation (~3 photons per neutron), and a well-studied energy spectrum (average energy of the neutron radiation $\overline{E}$ = 4.5 MeV, the maximum of the distribution corresponds to the energy E_{max} = 10.7 MeV). It has been successfully combined with a spontaneous fission source based on the ^{252}Cf isotope with half-life 82 years for neutron decay and 2646 years for alpha decay. This source has an exceptionally high specific neutron yield: 10 neutrons/s per mg, which lets us literally create point sources. It has a smooth and simple shape for the spectrum with average energy 1.9 MeV, maximum E_{max} ~ 0.7 MeV, and practically linear fall-off as the energy increases from the spectral maximum. The accompanying gamma radiation in this case is also small: ~3 gamma photons per emitted neutron.

There are standard sources of neutron radiation of lower energies, using (γ,n)-reactions: ^{24}Na–γ–Be with average energy 0.8 MeV, ^{226}Ra–γ–Be with 0.3 MeV, ^{24}Na–γ–D with 0.2 MeV, ^{124}Sb–γ–Be with average energy 24 keV, which are approved by the State Standard GOST 8.355-79 [10] for verification of neutron radiometers within UKPN-1 setups and their analogs. However, sources with (γ,n)-reactions give substantially stronger accompanying gamma radiation, which requires use of detectors which are not sensitive to gamma radiation or taking special measures to separate the detector output pulses generated by neutrons and gamma photons, which present a serious problem since it is not possible to separate their pulses according to pulse amplitude or duration. Therefore, in order to obtain low-energy neutron fields, it is better to use californium and (α,n)-sources with neutron moderators. As the neutron moderators, we can use hydrogen-containing or other neutron moderators of different thicknesses, while cadmium or boron filters let us additionally vary the shape of the spectra for the created neutron fields.

We have studied methods for creating reference neutron fields with energy spectra of different shapes from radioisotope sources, with the aim of creating a prototype test setup for verification of neutron spectrometers that we developed [11–13]. In [11], we studied the method of transformation of the spectrum for a model radioisotope neutron source using a "thermal insert" (a truncated cone made from hydrogen-containing material, mounted between the source and the detector), recommended in the State Standard GOST 8.355-79. For this purpose, a polyethylene neutron moderator was made in the form of a truncated cone of height 420 mm and end face diameters 65 mm and 150 mm, divided into 9 parts (disks): 8 of them 50-mm thick each and one of them (the one with the smallest diameter) of thickness 20 mm. Using four guides, any number of its sections can be assembled, which lets us control the total thickness of the moderator cone from 20 mm to 420 mm. As the detector, we used a three-channel BDKS-05S detection module for an MKS-03S neutron radiometer/dosimeter. This detection module is a polyethylene sphere of diameter 220 mm, in which five gas-discharge neutron detectors based on helium-3 were embedded at different depths. The detector located at the center of the sphere forms a fast-neutron measurement channel. At some distance from it (but in another diametral plane), two other detectors were symmetrically positioned, forming a measurement channel for intermediate neutrons, and the remaining two detectors, positioned symmetrically but close to the periphery of the sphere, form a thermal neutron channel.

The spectra of the fluxes arriving at the detector, without the cone and with a successive increase in the number of cone sections, were computed using the GEANT4 program library. In this case, we took into account scattered neutrons repeatedly reflected from the walls, floor, and ceiling of the room and reaching the detector. We measured the integrated neutron flux density detected by the detector. The calculation and measurements were done for distances between source and detection module of 45 cm and 100 cm.

The calculation results for the spectra of the neutron fields obtained for different thicknesses of the moderator cone show that as the cone thickness changes, mainly the high-energy component of the spectrum changes. No appreciable increase was observed in the fraction of intermediate and thermal neutrons in the neutron flux detected by the detector, for any thicknesses of the moderator cone. With an increase in the thickness of the moderator cone, only the flux incident on the detector is decreased, as a result of its intensive scattering in the moderator cone, and the shape of the spectrum changes insignificantly. A significant part (up to 1/3) of this flux is made up of scattered neutrons, repeatedly reflected from the walls, floor, and ceiling of the room, and it is these neutrons that form the thermal and intermediate part of the spectrum. Direct neutrons, passing through the cone and reaching the detector, contain practically no neutrons of thermal and intermediate energies. Such a situation leads to a dependence of the measurement results on the room geometry and the location of the source and detector in the room, which is undesirable. In order to minimize the influence of scattered neutrons and to improve diversity in the shapes of the reference fields, in [12] we considered the possibility of shielding the source and detector with shielding made from hydrogen-containing material. The measurement geometry is shown in Fig. 1.

The neutron fluxes were simulated for thicknesses of the source and detector shielding equal to 10, 20, 40, 80, 150, 200, and 300 mm. With an increase in the source shielding thickness, the neutron flux for neutrons reaching the detector increases, as a result of their reflection toward the detector. The steepest increase occurs for shielding thicknesses up to 40 mm, then it slows down and practically stops for thicknesses >150 mm. Computation of the spectra obtained in this case shows that the flux increases as a result of neutrons reflected by the source shielding, which lost a substantial fraction of their energy upon interaction with the shielding, and this is apparent in the increase in the fraction of thermal and intermediate neutrons as the shielding plates become thicker. The high-energy component of the spectrum in this case changes insignificantly. A change in the thickness of the moderator cone affects the high-energy part of the spectrum (the fast and intermediate neutron regions). The thermal peak decreases, but these changes are proportional to changes in the integrated flux reaching the detector, but the shape of the thermal region of the spectrum does not change. Figure 2 shows

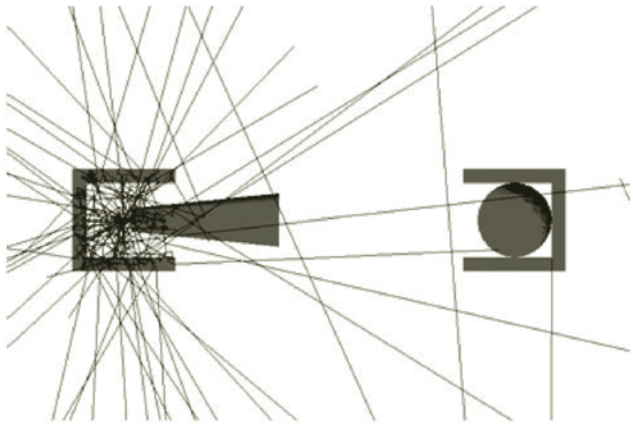

Fig. 1. Measurement geometry and neutron tracks when shielding is present for the source and detector with a moderator cone (distance between source and detector: 100 cm).

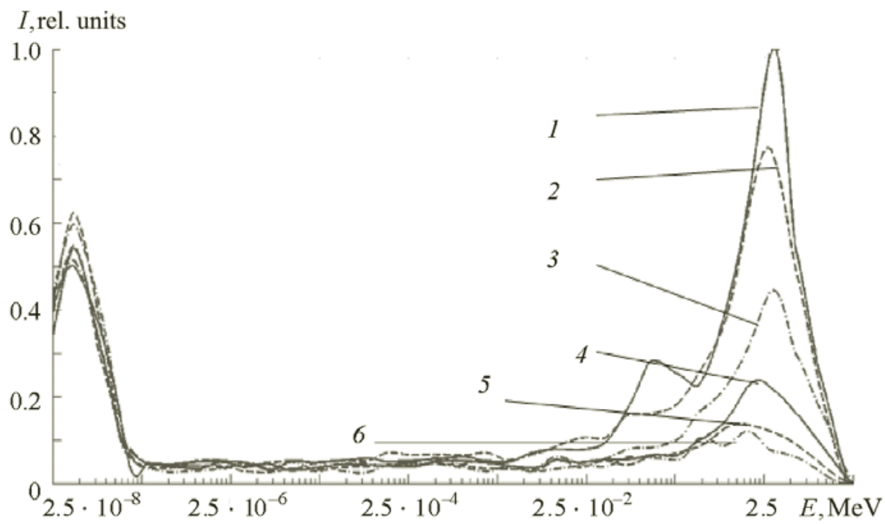

Fig. 2. Neutron flux spectra for neutrons reaching the detector, for moderator cone thicknesses 0 (1), 20 (2), 70 (3), 120 (4), 170 (5), and 220 (6) mm when using 4-cm thick shielding for the source and detector.

the simulation results for the case of constant shielding thickness of 40 mm and different thicknesses of the moderator cone (from 20 mm to 420 mm).

Then we searched for the optimal design of the shielding system, suitable for incorporation into existing test benches for verification of neutron radiometers and dosimeters. Using simulation modeling and the GEANT4 program library, we studied several different designs for the shielding devices. The best design involved using a polyethylene reflector with a conical recess, at the center of which the source should be placed, in contact with a thick polyethylene tube, in the cavity of which polyethylene disks of different thicknesses can be mounted. Based on the simulation results, we constructed a prototype shielding system which can be incorporated into test setups.

The results of the calculations for the spectra of the neutron fields created on this setup are presented in Fig. 3. As was expected, the shielding system substantially increases the thermal component, and also increases the intermediate component by a factor of 1.5 on the average, while the high-energy part of the spectrum does not change significantly (curve 1). Using moderator disks strongly distorts specifically the high-energy part of the spectrum, and distorts the intermediate and thermal components significantly more weakly. Even for a total thickness of the disks equal to 80 mm, the maximum in the thermal peak is comparable with the maximum for fast neutrons, while this stops with further increase in the thickness of the disks. It was specifically on this setup that we conducted laboratory tests on a prototype model for our multidetector

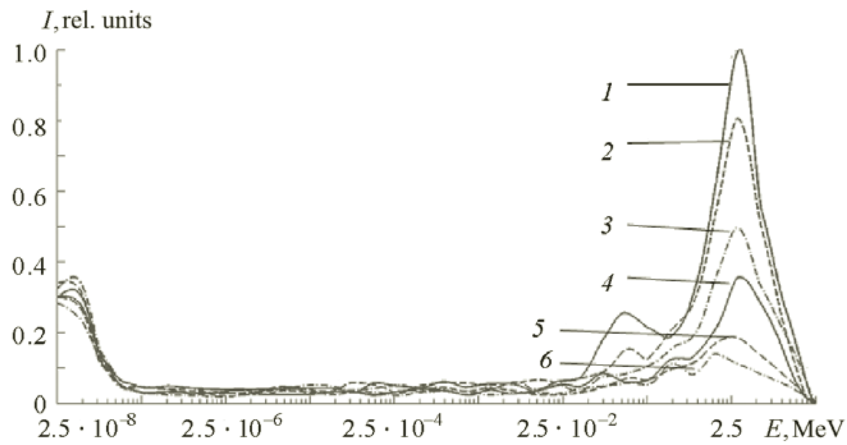

Fig. 3. Energy spectra of neutron fluxes reaching the detector, with shielding of the source and moderator disks of thickness 0 (1), 20 (2), 50 (3), 80 (4), 130 (5), and 180 (6) mm.

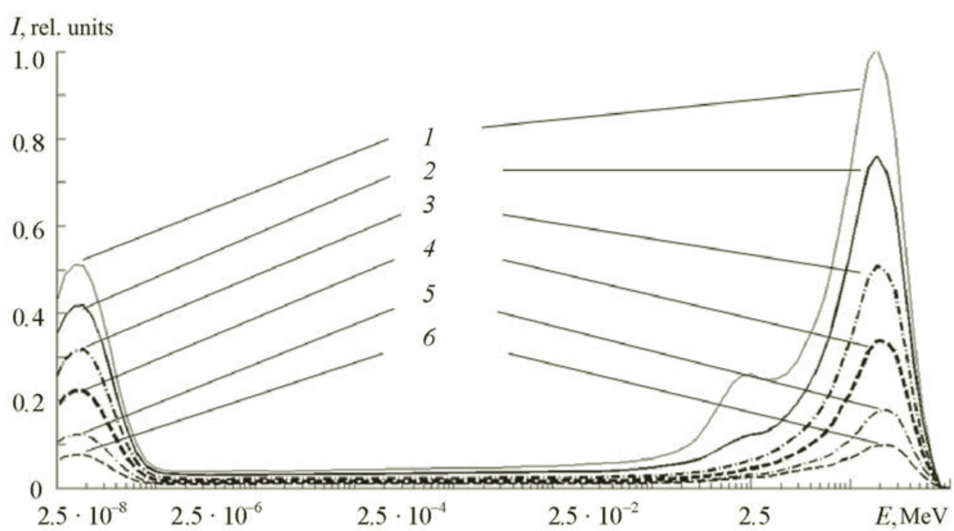

Fig. 4. Shapes of the spectra for some reference neutron fields, obtained on the IL-1M2 test setup for moderator disk thicknesses 0 (1), 20 (2), 50 (3), 80 (4), 130 (5), and 180 (6) mm.

real-time neutron spectrometer described in [1]. They confirmed the effectiveness of the concept that was the basis for this spectrometer, and the advisability of developing a commercial verification test setup making it possible to create reference neutron fields with energy spectra of different shapes, suitable both for verification of our neutron spectrometers and for calibration of any neutron radiometers and dosimeters in fields with spectra of different shapes.

The developed setup IL-1M2 differs from the prototype in the greater thickness of the shielding tube (30 mm), the presence of cadmium shielding covering the outer surface of the neutron reflector and collimating tube, the presence of detachable cadmium shielding covering the output hole of the collimating tube. These changes help increase the diversity of the spectra for the created neutron fields. Figure 4 shows the shapes of the computed spectra of the neutron fields obtained with a plutonium–beryllium source. Even more diversity in the shapes of the spectra for the neutron fields created on such a setup can be achieved as a result of successive use of two neutron sources: a plutonium–beryllium source and a californium source, which are characterized by quite different shapes of the spectra for the emitted neutrons. When a cadmium filter is placed on the end of the tube, the thermal component of the flux decreases ten-fold and is determined only by the scattered neutrons passing through the walls of the shielding tube and repeatedly reflected from the walls, floor, and ceiling of the room.

Since the measurement geometry using this setup is far from an open geometry with a point source, the inverse square law is not followed here. Therefore, in order to make the measurements in different measurement ranges of the instruments to be verified, we need to compute the spectra of the reference neutron fields beforehand for different distances between the neutron source and the detector of the instrument to be verified. In this case, the constraints on the distances connected with following the inverse square law are dropped, which lets us obtain a broader range of integrated flux densities from the same source. As a result of the change in the relationship between the direct neutrons passing through the collimator channel (with or without moderator disks) and the scattered neutrons incident on the detector that have passed through the walls of the collimating system and have been repeatedly reflected from the walls, floor, and ceiling of the room, as the distance changes between the neutron source and the detector we will see a change not only in the flux density but also in the shape of the spectrum (largely in the thermal part of the energy range). Therefore the spectra of the reference fluxes should be computed beforehand for all the distances to be specified.

Conclusions. We have shown that the current state of metrological support for neutron spectrometry is unsatisfactory. We propose designing test setups for neutron spectrometers by creating reference neutron fields with energy spectra of different shapes from existing radioisotope neutron sources, by placing the source in a collimating system, in the channel of which moderator disks of different thicknesses can be inserted. Using mathematical simulation, we have searched for the optimal design of such a test setup. The spectra of the reference neutron fields obtained on this setup, with a plutonium–beryllium source and moderator disks of different thicknesses, were computed using the GEANT4 program library. In order to obtain greater diversity in the shapes of the spectra on this setup for the reference neutron fields, we suggest successive use of different primary neutron sources, including a spontaneous fission source based on the ^{252}Cf isotope, the spectrum of which is quite different from the spectrum of the plutonium–beryllium source. In order to verify instruments in different measurement ranges, we suggest changing the distances between the neutron source and the detector of the instrument to be verified. Since the inverse square law is not followed for this measurement geometry, we need to compute the spectra of the reference fluxes beforehand for all the distances to be specified.

REFERENCES

1. V. E. Dreyzin, A. A. Grimov, and D. I. Logvinov, *Zh. Prikl. Spektrosk.*, **83**, No. 3, 462–468 (2016) [V. E. Dreyzin, A. A. Grimov, and D. I. Logvinov, *J. Appl. Spectrosc.*, **83**, 454–459 (2016) (English translation)].
2. V. E. Dreyzin, D. I. Logvinov, and A. A. Grimov, *Zh. Prikl. Spektrosk.*, **83**, No. 6, 953–958 (2016) [V. E. Dreyzin, D. I. Logvinov, and A. A. Grimov, *J. Appl. Spectrosc.*, **83**, 1001–1006 (2016) (English translation)].
3. Yu. I. Bregadze, É. K. Stepanov, and V. P. Yaryna, *Applied Metrology of Ionizing Radiation* [in Russian], Énergoatomizdat, Moscow (1990).
4. Sh. Itoh and T. Tsunoda, *J. Nucl. Sci. Technol.*, **26**, 833–843 (1989).
5. G.-Y. Wang, R. Han, J.-L. Liu, X.-P. Ouyang, J.-Ch. He, and J.-Y. Yan, *Radiat. Detect. Technol. Method.*, **1**, 11 (2017).
6. D. J. Thomas and A. V. Alevra, *Nucl. Instrum. Method. Phys. Res. A*, **476**, 12–20 (2002).
7. M. Matske, *Nucl. Instrum. Method. Phys. Res. A*, **476**, 230–241 (2002).
8. B. R. L. Siebert, *Nucl. Instrum. Method. Phys. Res. A*, **476**, 256–262 (2002).
9. B. M. van der Ende, J. Atanachovich, A. Erlandson, and G. Bentoumi, *Nucl. Instrum. Method. Phys. Res. A*, **820**, 40–47 (2016).
10. GOST 8.355-79 "*Neutron Radiometers. Testing Methods and Means*."
11. V. E. Dreyzin, D. I. Logvinov, and I. V. Polishchuk, *Izv. Yugo-Zapad. Gos. Univ.*, No. 5, Pt. 2, 24–29 (2012).
12. V. E. Dreyzin, D. I. Logvinov, and I. V. Polishchuk, *Izv. Yugo-Zapad. Gos. Univ.*, No. 6, 130–136 (2012).
13. V. E. Dreyzin, I. V. Polishchuk, and D. I. Logvinov, *ANRI*, No. 4(71), 2–12 (2012).