FRANK LABORATORY OF NEUTRON PHYSICS

In 2013, the Frank Laboratory of Neutron Physics' scientific programme was aimed at obtaining new results under four research themes of the JINR Plan for Scientific Research and International Scientific and Technical Cooperation: in condensed matter physics ("Investigations of Nanosystems and Novel Materials by Neutron Scattering Methods", 04-4-1069-2009/2014, headed by V. L. Aksenov, A. M. Balagurov and D. P. Kozlenko); in neutron nuclear physics ("Investigations in the Field of Nuclear Physics with Neutrons", 03-4-1104-2011/2013, headed by V. N. Shvetsov and Yu. N. Kopatch); in development of the FLNP basic facilities ("Development of the IBR-2M Reactor with a Complex of Cryogenic Moderators of Neutrons", 04-4-1105-2011/2013, headed by A. V. Belushkin and A. V. Vinogradov); in development of the IBR-2 spectrometers and computation complex ("Novel Development and Creation of Equipment for the IBR-2M Spectrometers Complex", 04-4-1075-2009/2014, headed by V. I. Prikhodko and S. A. Kulikov).

CONDENSED MATTER PHYSICS

The greater part of experimental research was carried out on the spectrometers of the modernized IBR-2 reactor.

In 2013, within the framework of the User Programme, 195 proposals for conducting experiments were received from 17 different countries. The received proposals covered a broad spectrum of neutron research in physics (41%), materials science (22%), chemistry, geosciences, biology and applied sciences (constituting the rest 37%). Of the received proposals, 158 were admitted for realization.

Scientific Results. The crystal and magnetic structure of multiferroic $RbFe(MoO_4)_2$ has been studied [1]. In this compound a spontaneous electric polarization occurs due to the fact that the inversion symmetry of the crystal structure is broken because of the occurrence of noncollinear antiferromagnetic ordering. In addition, Fe magnetic moments in the trigonal structure of $RbFe(MoO_4)_2$ (space group P-3m1) form a two-dimensional magnetic triangular lattice, where magnetic coupling between the magnetic planes is 25 times weaker than the in-plane coupling. With increasing pressure a structural phase transition to the monoclinic C2/c phase with a phase coexistence in a wide pressure range was observed. The antiferromagnetic (AFM) symmetry for the trigonal phase is characterized by a propagation vector $q = (1/3, 0, k_z)$. With a rise in pressure, an increase in the k_z value from 0.45 to 0.48 and in the Néel temperature with a pressure coefficient of 0.09 GPa^{-1} was observed. No evidence of the formation of the magnetic ordering in the high-pressure monoclinic phase was found down to a temperature of 1.5 K.

On the HRFD diffractometer the investigations of electrodes in lithium-ion accumulators [2] have continued. The experimental data have made it possible to follow more closely the stages of Li intercalation into graphite with the successive formation of several LiC_n phases and a reversible transition $LiFePO_4 \leftrightarrow FePO_4$. The comparison of charging/discharging processes in batteries with a cathode of pure LFP and LFP containing $\sim 1\%$ vanadium (LFPV) has demonstrated that in the latter case a significantly greater fraction of the anode material undergoes a transition into a final LiC_6

phase because of a smaller relative mass of graphite as compared to fluorine ferrophosphate. The analysis of changes in the microstructure of a vanadium-doped cathode has shown a significant increase in the degree of structure imperfection, which correlates with the best electrochemical properties of LFPV as compared to LFP.

On the basis of the small-angle neutron scattering analysis, a continuous spatial transition of the carbon state from crystalline diamond (sp^3-hybridization) inside the particle to a graphite-like state (sp^2-hybridization) at DND surface has been suggested. Such a transition makes it possible to combine the experimentally observed shift in the mean scattering length density of DND as compared to pure diamond (which is indicative of the presence of a non-diamond component in the DND structure) and the diffusive character of the particle surface, which can be deduced from the deviation from Porod's law [3]. The proposed profile is of a simple power-law type and due to a number of specific features explains a homogeneous decrease in the total scattering intensity at the contrast variation. A spherical "core-shell" representation of DND particles used previously, which gives a reasonable thickness of a non-diamond shell of about 0.5 nm, can be considered as an approximation to the continuous density profile reflecting naturally the diamond–graphite transition in terms of the averaged scattering length density. Along with it, this profile naturally suggests that non-diamond transitional bonds (presumably sp^{2+x}-bonds) are mainly concentrated close to the particle surface. It also allows one to directly determine the parameters of the particle size distribution function.

At the REMUR spectrometer the magnetic state of the layer nanostructure Ta(10 nm)/V(150 nm)/$Fe_{0.7}V_{0.3}$(1 nm)/V(1.2 nm)/$Fe_{0.7}V_{0.3}$/Nb(150 nm)/Si composed of ferromagnetic and superconducting layers has been studied by polarized neutron reflectometry. Three phenomena were supposed to take place in this nanostructure. The first one was assumed to be an antiferromagnetic ordering of the pair of $Fe_{0.7}V_{0.3}$(1 nm) layers in an external magnetic field. The second effect was supposed to concern the magnetization of the superconducting pair by ferromagnetic layers, which would result in magnetization of the superconducting layer. Finally, the third phenomenon was presumed to be the formation of a domain structure with small domain sizes and zero mean magnetization. During the measurements the temperature and magnetic field strength were varied in the ranges of 1.3–110 K and 30 Oe – 9.5 kOe, respectively. The neutron scattering with a maximum at a temperature of 8 K (which is below the temperature of superconducting transition in the niobium layer) was observed in the range of 1.3–10 K. The obtained experimental data are indicative of the existence (in a certain temperature range below the superconducting transition temperature) of a domain lattice phase where a rotation of the magnetization vector takes place and which is characterized by two directions. The latter is testified by a strong scattering, which cannot be explained only by the scattering in the vertical direction. The absence of neutron scattering in the second direction suggests that the lattice constant in this direction lies in the range below one thousand angströms. These observations are the first direct experimental evidence of a cryptoferromagnetic phase in superconducting ferromagnetics, which is the appearance of an antiferromagnetic ordering at the scale of superconducting coherent length (size of a superconducting pair). At the same time, the magnetic period of the cryptoferromagnetic state was found to be an order of 1000 times less than the size of usual domains in ferromagnetics (microns).

A process of spontaneous phospholipid vesicle formation in the presence of calcium ions has been studied by small-angle neutron scattering (Fig. 1). For the first time, the behavior of intermembrane distance in the transition region has been considered in detail for the membranes in both liquid and gel phases. It has been shown that the transition of the system from the bound to the unbound state in both phases has a continuous character, which is rather unusual for gel phases. The earlier theoretical studies on gel phases suggested that on addition of calcium ions to lipid multilayer membranes there should be a sharp transition of membranes from the bound to the unbound state, since there are no undulations in the gel phase. The investigations performed have shown that there is a significant contribution of undulation forces to membrane interactions. The critical calcium ion concentrations at which the studied transition takes place in gel (0.3 mM) and liquid (0.4 mM) phases have been obtained together with the direct determination of the affinity constants for calcium ions with respect to lipid membranes (22 M^{-1} and 24 M^{-1} in gel and liquid phases, respectively).

When operating nuclear facilities, a surveillance programme of witness specimens positioned at the inner wall of the reactor cavity serves as an important source of information on the changes in the properties of vessel steels, which tend to worsen as a result of neutron irradiation. It is necessary to control the level of residual stresses after welding in the reconstituted witness specimens. On the FSD diffractometer, experiments have been carried out to study the distribution of residual stresses in witness specimens that develop after electron-beam welding (EBW) and laser beam welding (LBW). The experimental results have shown that the level of residual stresses for an LBW sample is much higher than for an EBW sample and ranges to 550 MPa in the weld region. This supports the well-known fact that among all methods the application of electron-beam welding results in the lowest level of residual stresses in welds. This is most probably due to a low heat input of the EBW process (4–5 times lower than, for example, in arc welding) which significantly reduces the deformation of a final product. In addition, the diffraction peak broadening was used to determine the level of residual

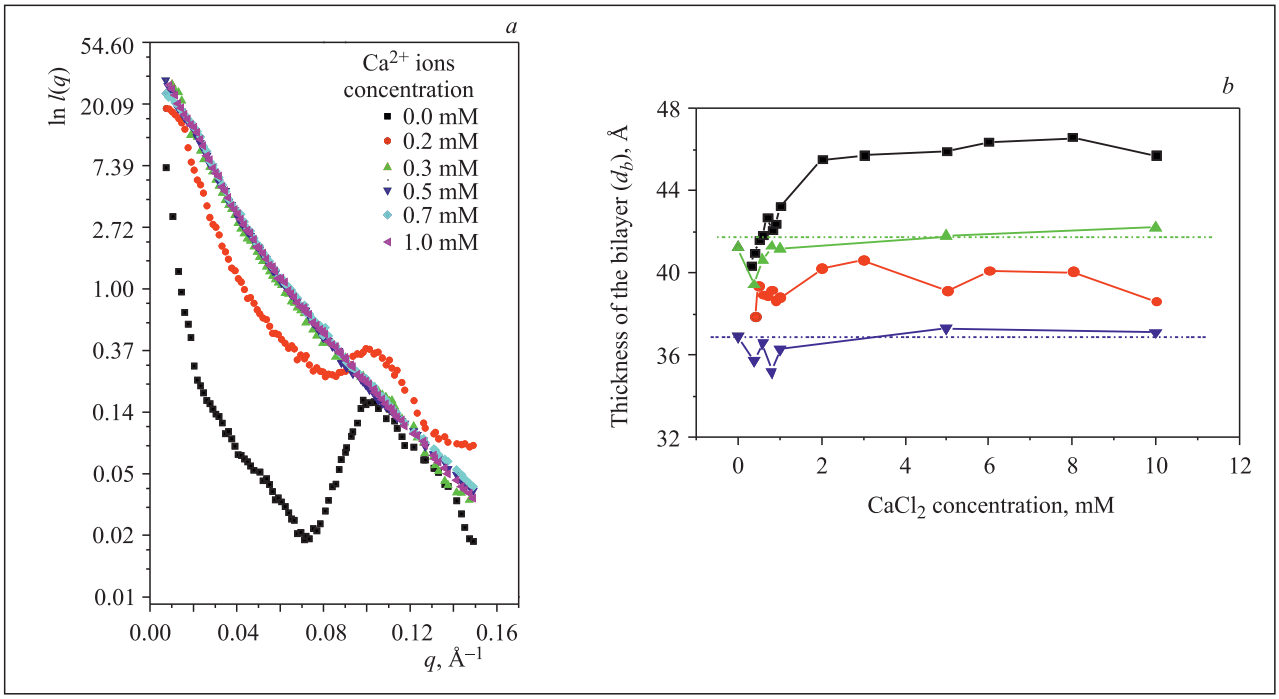

Fig. 1. *a*) Small-angle neutron scattering curves from multilayer DMPC membranes (1% wt) in the water/$CaCl_2$ solution for molar concentrations of Ca^{2+} ions: 0.0, 0.2, 0.3, 0.5, 0.7 1.0 mM. *b*) Concentration dependence of the bilayer thickness: for unilamellar vesicles prepared by extrusion: ▲ — at $T = 15\,^{\circ}$C; ▼ — at $T = 55\,^{\circ}$C and for spontaneously formed unilamellar vesicles: ■ — at $T = 15\,^{\circ}$C; ● — at $T = 55\,^{\circ}$C

microstrains, which directly characterizes the density of dislocations in a material being studied. The microstrain in the EBW specimen amounts to $3.5 \cdot 10^{-3}$ and is slightly higher in the LBW specimen — $4.5 \cdot 10^{-3}$. This effect is accompanied by a considerable (~ 2.5 times) increase in microhardness in weld seam regions. The observed increase in microhardness is likely to be the result of the formation of martensite (or martensite-bainite) structure in welds and heat-affected zones.

Instrument Development. Work to develop and test sample environment devices for the new DN-6 diffractometer has been carried out. High-pressure cells with diamond anvils with an operating range up to 15 GPa (culet diameter of 0.8 mm) and 50 GPa (culet diameter of 0.5 mm) have been purchased. The first experiments have demonstrated a possibility of their successful application in experiments with DN-6.

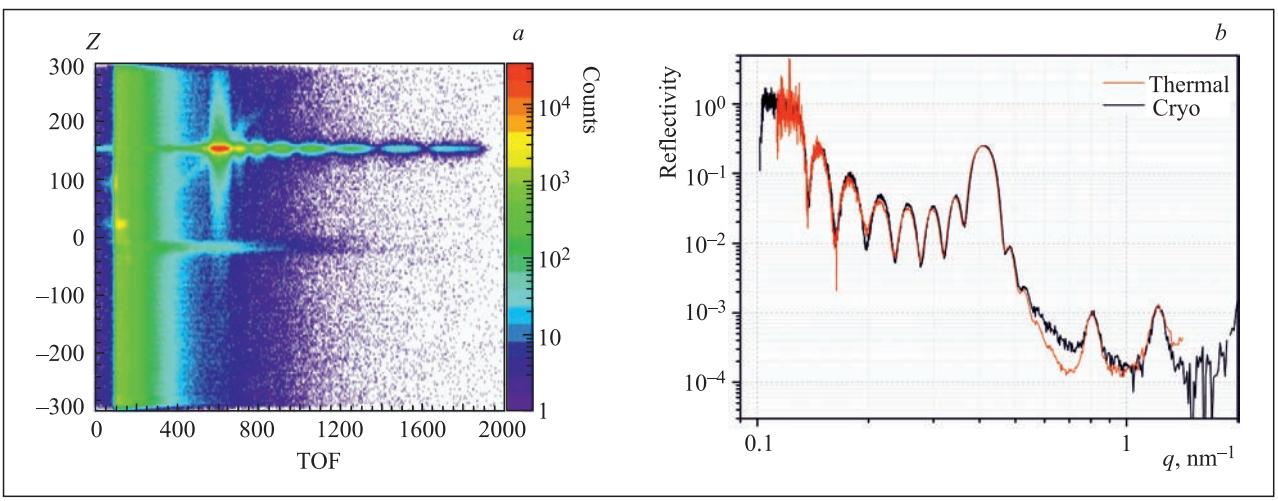

Fig. 2. *a*) 2D spectrum of non-polarized neutron beam reflected from a layer structure [Ni(8.4 nm)Ti(7 nm)]×8/Floatglass (MIRROTRON Ltd., Hungary) obtained on the GRAINS reflectometer in a cryogenic operating mode of the moderator; data are represented in coordinates Z (detector channel width 0.35 mm) — Time-Of-Flight (channel width 32 μs). *b*) Reflectivity curves for the same system measured in thermal and cryogenic operating modes of the moderator

The operation of the first-stage of the GRAINS reflectometer has started. Beam profiles have been measured and optimized for different configurations of the reflectometer elements. The experimental estimations of the total flux of non-polarized thermal neutrons (wavelength above 0.05 nm) after deflector have been made in thermal ($2\cdot10^6$ cm$^{-2}\cdot$s^{-1}) and cold ($1\cdot10^6$ cm$^{-2}\cdot$s^{-1}) operating modes of the moderator. Time-of-flight spectra have been optimized over the fast neutron background. The first reflectivity curves for standard systems have been obtained in two operating modes of the moderator (Fig. 2). During the start-up the first experiment to study the oxidation effect on the structure of thin titanium films on a glass substrate has been carried out in the framework of the development of new coatings for neutron optical devices.

The development and construction of a prototype of a radiography spectrometer on beam 14 continued. A vacuum collimation system has been manufactured and installed on the beam. A CCD-camera-based imaging system has been produced and tested on beam 12.

NEUTRON NUCLEAR PHYSICS

In 2013, at FLNP the scientific activity in the field of neutron nuclear physics was carried out in the following traditional directions: investigations of time and space parity violation processes in neutron-nuclear interactions; studies of the fission process; experimental and theoretical investigations of fundamental properties of the neutron; gamma spectroscopy of neutron-nuclear interactions; atomic nuclear structure, obtaining of new data for reactor applications and for nuclear astrophysics; experiments with ultracold neutrons. In 2013, the IREN facility operated for physical experiments for about 1050 h.

A mobile reconfigurable gamma-spectrometer system nGamma has been developed and tested. The system is intended for studying nuclear reactions with the emission of gamma rays induced by neutrons of various energies. In the initial (minimum, test) configuration it consists of 24 NaI(Tl) gamma-ray detectors mounted on two rings. The energy and time characteristics of individual sections of the system have been determined experimentally. By using the system, the energy dependence of neutron flux density has been measured at a distance of 60 m from a neutron-generating target of the IREN pulsed neutron source.

A 12-detector (2 modules of 6 NaI(Tl) crystals each) gamma-spectrometer system "Romashka", designed to study resonance radiative capture (and fission) of nuclei by neutrons, has been assembled and tested on beam 4 of the IREN facility.

In 2013, the adjustment of the experimental setup AURA for measuring the energy dependence of angular anisotropy of slow neutrons scattered by noble gases in order to determine the (n,e)-scattering length was in progress. At present, the AURA setup is placed on a 15-m flight path of beam 2 of the IREN facility.

The activities carried out in cooperation with the Czech Technical University in Prague on the application of pixel silicon detectors for detecting charged particles emitted in fission are in progress. In 2013, the measurements of ternary spontaneous fission of ^{252}Cf using TimePix detectors were carried out at FLNP in collaboration with the Technical University in Prague. The $\Delta E{-}E$ technique, which allows charge identification of light charged particles, was used to identify ternary particles. A thin silicon detector (12 μm) was used as a ΔE-detector and a pixel detector TimePix with a 300-μm-thick sensor layer as an E-detector.

In 2013, a setup for precision measurements of prompt fission neutron multiplicity depending on the mass distributions of fission fragments and their total kinetic energy was designed and constructed.

The experimental and theoretical investigations of the (n,p), (n,α) reactions induced by fast neutrons continued. The experiments are carried out at the Van de Graaff accelerators EG-5 at JINR FLNP (Dubna, Russia) and EG-4.5 of the Institute of Heavy Ion Physics of Peking University (Beijing, China). Data on the neutron reactions with the emission of charged particles induced by fast neutrons are of much interest for studying the mechanisms of nuclear reactions and atomic nuclear structure. In addition, these data are of importance in choosing engineering materials and in performing calculations in the development of new facilities for nuclear power engineering. At the end of 2013, the measurements of the ^{66}Zn$\,(n,\alpha)\,^{63}$Ni and ^{144}Sm$\,(n,\alpha)\,^{141}$Nd reactions at $E_n = 4$ MeV were conducted, thus completing a series of measurements that started a year ago. The measurements of the ^{54}Fe$\,(n,\alpha)\,^{51}$Cr reaction were also carried out at $E_n = 5.5$ and 6.5 MeV. The energy spectra of charged particles were obtained and the data treatment is in progress. The data treatment for the measurements of the ^{57}Fe$\,(n,\alpha)\,^{54}$Cr and ^{63}Cu$\,(n,\alpha)\,^{60}$Co reactions at $E_n \sim 4.0{-}6.5$ MeV has been completed. A comparison with the available library estimates and with the data obtained by other authors has been performed (Figs. 3 and 4). The analysis reveals a significant discrepancy between the estimates given by different nuclear data libraries, while no experimental data are available for ^{57}Fe isotope. The available data for ^{63}Cu from two rather old measurements in the range of several MeV show a considerable discrepancy.

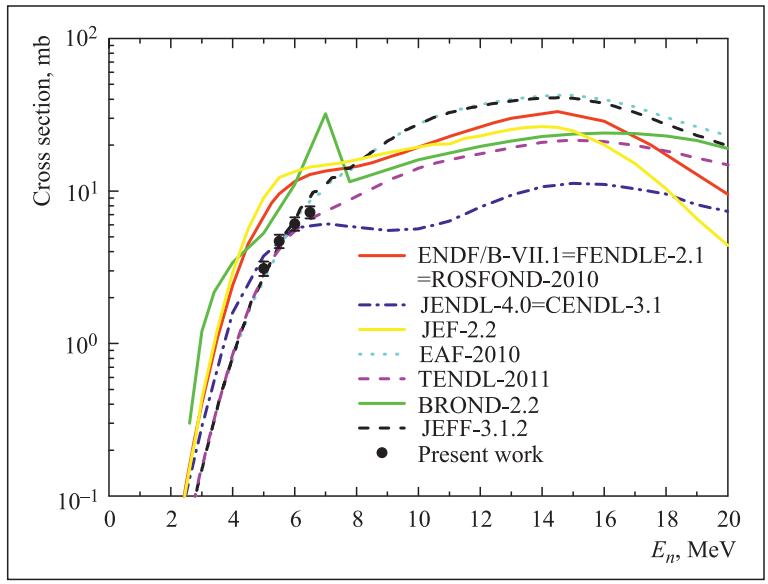

Fig. 3. The obtained cross sections of ^{57}Fe (n, α) ^{54}Cr in comparison with the available library estimates and with the data obtained by other authors

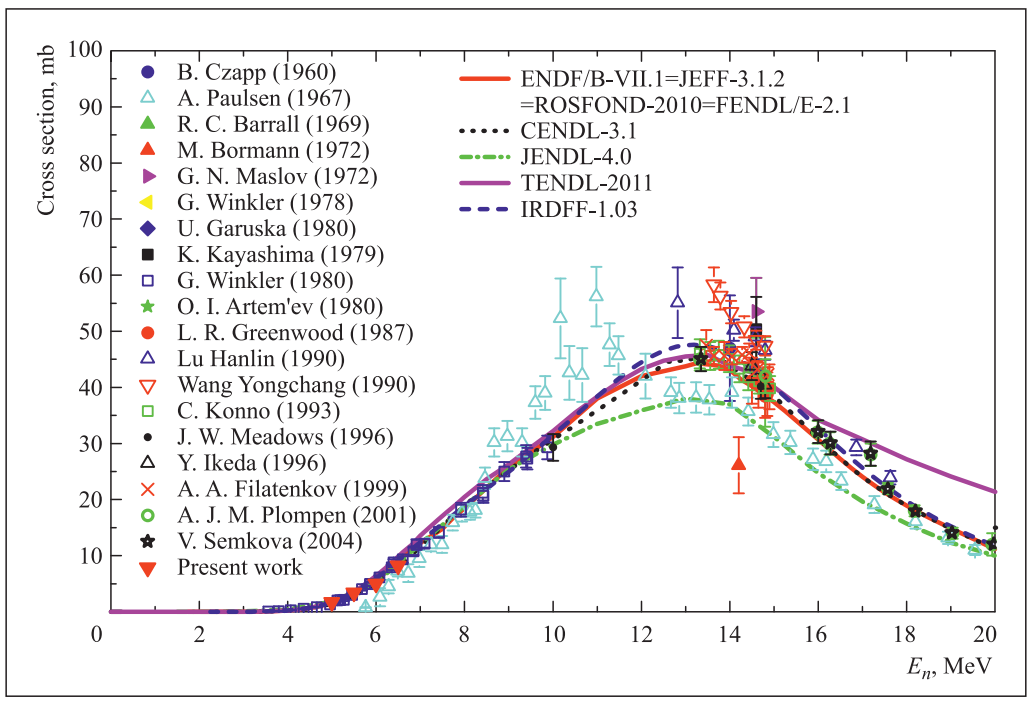

Fig. 4. The obtained cross sections of ^{63}Cu (n, α) ^{60}Co in comparison with the available data and estimates

Test measurements have been carried out to test the idea of using a cavity of solid methane for producing a cold neutron flux at the end of a neutron guide with thermal neutrons. The measurements were done in 2013 on the DIN-2PI instrument on beam 2 of the IBR-2 reactor. The results show that one can obtain the neutron spectrum close to that from the cold reactor source by using the thermal neutron beam inside the methane cavity. The albedo of solid methane for cold neutrons is close to the calculated value. Thus, the idea of a helium UCN source inside a cold cavity at the end of a neutron guide with thermal neutrons appears feasible. The next step in the development of the given idea is to construct a prototype of the source to test the solutions for a number of technical problems.

In 2013, the radiation tests of scintillators and megatile samples of the CMS setup (CERN) were performed. It was necessary for the optimization of the conditions for future experiments. The neutron spectrum at IREN is close to that at the CMS hadron calorimeter. The samples were irradiated by the total fluence of 10^{12} cm^{-2}. Then during three weeks the

induced activity was measured at two distances from the sample.

The work on the active moss-transplant biomonitoring of airborne trace elements made it possible to study the air pollution in the centre of Belgrade, Serbia, as well as in Greater Thriasion Plain, Attica, one of the most ecologically unsafe regions of Greece.

The results of complex investigations of air pollution using mosses and lichens as well as of water ecosystem using mollusks and oysters near a growing port in Cape Town (Saldanha Bay, the Atlantic Ocean near the West coast of the Republic of South Africa) have aroused considerable interest among environmental specialists of the RSA and willingness to cooperate in this research area.

In cooperation with the Western Cape University (South Africa), the NAA study of coal fly ash from the Matla coal power station in the Mpumalanga Province in South Africa has been conducted.

The elemental composition of microbiological samples and the efficiency of accumulation of zinc and other metals by Spirulina biomass were determined using the NAA technique on the IBR-2 reactor. This study was awarded with a gold medal at the V European Exhibition of Creativity and Innovation EUROINVENT 2013, Iaşi, Romania, in the category "PhD research project".

THE IBR-2 PULSED REACTOR

The IBR-2 research nuclear facility is operated under Rostechnadzor license No. ГН-03-108-2614 of 27 April 2012.

In 2013, in accordance with the license requirements, the specialized organizations, in cooperation with the IBR-2 personnel, performed the scheduled work on the technical evaluation and assessment of the remaining life of the technological reactor equipment. The activities to prolong the service life of the equipment of the IBR-2 safety-related systems have been completed.

Since January 2013 regular IBR-2 cycles of physical experiments have been carried out at a power of 2 MW with the CM-202 moderator operating either in the water or cryogenic mode depending on the schedule of the physical start-up of the cold moderator.

From September 13 to 19, a fresh fuel assembly was loaded into the IBR-2 reactor core and the reactor was brought to criticality in a steady-state operation mode followed by an assessment of the efficiency of the loaded fuel assembly and of the integrated efficiency of the regulating units of the control and safety system. The reactor was turned on to a power of 250 kW followed by an assessment of the efficiency of the loaded fuel assembly at pulsed criticality.

The table presents data on the IBR-2 operation for physics experiments in 2013.

In 2013, in accordance with the contract with the JSC "Dose" the dosimetry equipment for the stationary radiation monitoring system (RMS) of IBR-2 was delivered. Its installation and adjustment began.

Data on the IBR-2 operation for physics experiments

No. cycle	Period	Moderator type	Reactor operation for physics experiments, h
1	January 22–30	Cryogenic	176
2	February 11–22	Water	260
3	March 18–29	Cryogenic	264
4	April 9–20	Water	242
5	May 21 – June 6	Water	281
6	September 23 – October 4	Water	262
7	October 14–22	Water	169
8	October 24 – November 1	Cryogenic	174
9	November 11–18	Water	170
10	November 21–23	Cryogenic	44
11	November 29 – December 13	Water	330
12	December 17–26	Cryogenic	206
		Total:	2578

NOVEL DEVELOPMENT AND CONSTRUCTION OF EQUIPMENT FOR THE IBR-2 SPECTROMETER COMPLEX

In January 2013, during the last and longest operation cycle of the moderator at a reactor power of 2 MW, research activities were carried out in the framework of the CM-202 commissioning programme. All in all, during the start-up period there were six CM operation cycles with fresh loadings of beads composed of a frozen mixture of mesitylene and m-xylene with the duration of the cycles ranging from several to 178 hours. As a result of the analysis of the CM-202 operation in the specified cycles, the key questions, which are important for ensuring efficient and long-term operation of the moderator, were answered:

• Time of loading beads into the moderator chamber — minimum 4 h. Loading proceeds without jams and noticeable defragmentation of beads at a gas flow rate of 1.2–1.5 g/s and a temperature of 80–85 K.

• Hydraulic resistance of the contour and parameters of a gas blower ensure a helium flow rate of 6 g/s.

• A KGU-700/15 cryogenic refrigerator cools beads in the CM-202 chamber at a reactor power of 2 MW down to an average temperature of 32–33 K (design value is 23–25 K).

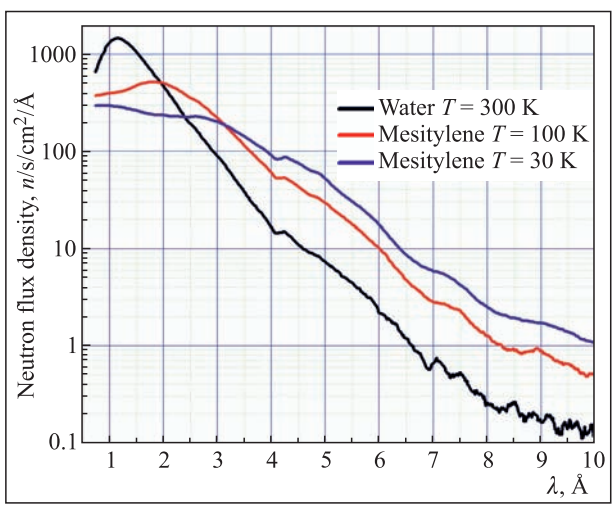

Fig. 5. The differential neutron flux density at the location of a PSD detector for an empty chamber (black line) and for the filled one at temperatures of 100 and 30 K

• Gain factor for cold neutrons with wavelengths of 8–10 Å is 13–14 (Fig. 5), design value is up to 20 at 20 K. Degradation in the cold neutron (6–10 Å) flux for 350 MW · h is no more than 5–7%; the flux of neutrons with shorter wavelengths increases with a radiation dose.

• Discharge of the spent liquid proceeds rather quickly; the initial solution viscosity increases no more than 10 times after operating for 7.3 days.

• Filling of the chamber and subsequent discharge of mesitylene have no effect on the reactivity of the IBR-2 reactor.

The modernization of the detector system for the DN-12 diffractometer for investigations of microsamples at high pressures, as well as of the automation systems for the Fourier diffractometers, has been completed: HRFD (6 control channels) and FSD (12 channels) were modernized, and a new system was developed and constructed for the GRAINS spectrometer (26 channels).

In 2013, a prototype of a scintillation counter of the ASTRA detector for the FSD diffractometer was manufactured and tested at a test stand with a source. The construction of a section consisting of four scintillation counters of the detector ASTRA is in progress.

Seven sets of digital and analog MPD-32 units for data acquisition and accumulation systems for the IBR-2 spectrometers have been manufactured and adjusted. The DAQ systems assembled from these units have been put into operation on the YuMO, DN-12 spectrometers mentioned earlier, and also the systems for the FSD and HRFD diffractometers have been completed and are in the adjustment stage.

In 2013, a new universal graphical user interface (GUI) on the basis of PyQt and matplotlib (introduced on the YuMO, NERA-PR, SKAT, and REMUR spectrometers) was developed. Operation library for reflectometers (REMUR, REFLEX, and GRAINS) and programmes for visualization (SpectraViewer) and adjustment (ICE) were improved on demand of the users. Over the past year, a new significantly improved version of the system for remote monitoring of parameters and control of spectrometers (WebSonix) was prepared, which is now in trial operation on the SKAT and YuMO spectrometers.

CONFERENCES AND MEETINGS

In 2013, two scientific schools for advanced training of young scientists were organized at the Frank Laboratory of Neutron Physics: the V International Neutron School for Young Scientists and Students "Modern Neutron Diffraction Studies: Interdisciplinary Research of Nanosystems and Materials" (October 28 – November 1, Dubna) and the IV International Scientific School for Young Scientists and Students "Instruments and Methods of Experimental Nuclear Physics. Electronics and Automatics of Experimental Facilities"

(November 5–8, Dubna). These Schools were dedicated to the fundamental and applied aspects of research in the fields of neutron physics, condensed-matter physics, and materials science. These Schools were attended by students, postgraduates and young specialists from Russia and nine JINR Member States.

On May 13–17, the 3rd Research Coordinated Meeting (RCM-3) related to the IAEA coordinated research project "Development, Characterization and Testing of Materials of Relevance to Nuclear Energy Sector Using Neutron Beams" was held in Dubna.

On May 20–25, the XXI International Seminar on Interaction of Neutrons with Nuclei (ISINN-XXI) was held in Alushta. The Seminar was held under the banner of the 50th anniversary of neutron activation analysis at JINR.

On August 18–21, FLNP in collaboration with the Federal Ministry of Education and Research of Germany (BMBF) organized a meeting "Instrument Development on Long Pulse Neutron Sources". The event was aimed at discussing current trends in the development of facilities for neutron scattering on the sources operated in pulse mode.

On November 11–14, Tula (Russia) hosted the II International Conference "Multiscale Modeling of Structures, Composition of Matter, Nanostructured Materials and Nanotechnologies" dedicated to the memory of Professor A. N. Nikitin, who used to work at FLNP. The Frank Laboratory of Neutron Physics was the co-organizer of this event.

On November 25–27, the Joint JINR–Romania International School on Small-Angle Neutron Scattering and Complementary Methods of Research of "Smart" Materials was held in West University of Timisoara. The School was held in the framework of the TIM 2013 Physics Conference.

REFERENCES

1. *Kozlenko D. P. et al.* // Phys. Rev. B. 2013. V. 87. P. 014112-1–6.
2. *Bobrikov I. A., Balagurov A. M.* // JINR News. 2013. No. 3. P. 19.
3. *Karpinsky D. V. et al.* // J. Appl. Phys. 2013. V. 113. P. 187218.