NEUTRON-INDUCED FISSION CROSS SECTIONS OF ^{232}Th, ^{233}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{nat}Pb AND ^{209}Bi RELATIVE TO ^{235}U IN THE ENERGY RANGE 1–200 MeV

O.A.Shcherbakov, A.B.Laptev, G.A.Petrov, A.S.Vorobyev, A.Yu.Donets [1], A.V.Evdokimov [1], A.V.Fomichev [1], S.M.Soloviev [1], T.Fukahori [2], A.Hasegawa [2], V.M.Maslov [3], Yu.V.Tuboltsev [4]

[1] *V.G. Khlopin Radium Institute, 2-nd Murinski Ave. 28, 194021, St. Petersburg, Russia*
[2] *Nuclear Data Center/JAERI, Tokai-mura, Naka-gun, Ibaraki-ken, 319–1195, Japan*
[3] *Radiation Physics and Chemistry Institute, 220109, Minsk-Sosny, Belarus*
[4] *A.F.Ioffe Physical-Technical Institute, Polytekhnicheskaya, 26, St. Petersburg, Russia*

1. Introduction

Among the applications of the fission data for actinides above 20 MeV, the most important are accelerator-driven transmutation of waste reactor materials and energy production, accelerator-based conversion of weapon plutonium and production of tritium, accelerator and spaceship shielding, radiation therapy, *etc*. A need in information about neutron-induced fission of lead and bismuth at intermediate energies also exists mainly due to probable use of these metals as structural materials in the neutron-producing targets of high-current proton accelerators of new generation. Besides, neutron-induced fission cross sections of Pb and Bi are very convenient as standards in the intermediate energy range because they have thresholds at 25–40 MeV which eliminates the influence of low energy neutrons.

During the last decade, measurements of neutron-induced fission cross sections in the energy range 1–200 MeV with continuous spectrum of neutrons have been systematically performed at the GNEIS facility [1–17]. The target nuclei ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{nat}Pb and ^{209}Bi have been chosen for present investigation according to the Nuclear Data Request List for new JENDL High Energy File (JENDL/HE-3.3). At the moment of starting present measurements, besides of our earlier data [2, 3], there existed only one experimental data set obtained at Los Alamos for ^{232}Th, ^{233}U, ^{234}U, ^{236}U, ^{238}U, ^{237}Np, ^{239}Pu relative to ^{235}U in the energy range 1–400 MeV (P.W.Lisowski *et al., Proc. Int. Conf. Nucl. Data for Sci. and Tech., Mito, Japan, 30 May–*3 *June 1988*, p. 97 (1988*)*; *Proc. Int. Conf. Nucl. Data for Sci. and Tech., Julich, Germany, 13–17 May 1991,* p. 732 (1992)). These data have been published as graphs only with exception of ^{235}U and ^{238}U published as recommended data by A.Carlson *et al.* (see "Summary Report of a Consultants' Meeting, Austria", 2–6 December 1996, INDC(NDC)-368, IAEA, Vienna, 1997, p. 23). Later on, an experimental data set for ^{239}Pu, ^{240}Pu, ^{242}Pu and ^{244}Pu obtained at LANL in the energy range 0.5–400 MeV (up to 62 MeV for ^{239}Pu) has been published – see P.Staples and K.Morley, Nucl. Sci. Eng. **129**, 14 (1998).

As for Pb and Bi, besides our data, the experimental data base on neutron-induced fission cross sections is very scarce and can be roughly divided into two groups: a few results obtained in isolated energy points (*e.g*., references in A.V.Prokofiev *et al.,* LANL Report LA-UR-98-0418, Los Alamos, 1998) and the only one data set obtained at Los Alamos using a continuous neutron spectrum up to 400 MeV (P.Staples *et al*., Bull. Am. Phys. Soc. **40,** 962 (1995); P.Staples, private communication, 2000).

The description of experimental technique and data analysis are presented in this report together with the final results which are shown in comparison with the data of other authors and theoretical calculations.

2. Experiment

The fission cross-section ratios relative to ^{235}U have been measured using the neutron time-of-flight spectrometer GNEIS [1] based on the PNPI 1 GeV proton synchrocyclotron. A water-cooled lead target situated inside the vacuum chamber of the accelerator is used as a pulsed spallation neutron source with an average intensity of 3×10^{14} n/s, a burst duration of 10 ns and a repetition rate up to 50 Hz. The measurements were performed using the bare source target, a 48.5 m flight path and neutron beam contained in the evacuated flight tubes (Fig. 1). A set of iron, brass and lead collimators provides the beam diameter of 18 cm at the fission chambers location. The measurements were carried out with the use of the "sweeping" magnet to remove charged particles from the neutron beam.

The fission reaction rate was measured using simultaneously two fast parallel plate ionization chambers filled with methane (94–100%) and CF_4 (6–0%) mixture working gas at an absolute pressure of 2.5–3.5 atm. The first (actinide) chamber consisted of 6 sections, every one containing a pair of cathode and anode

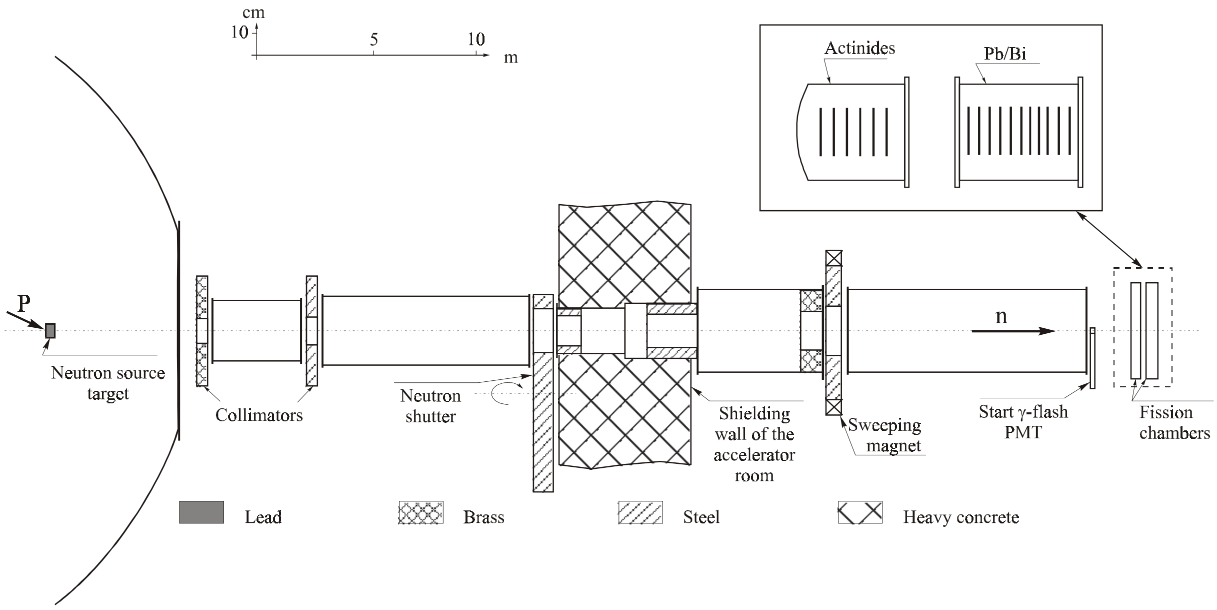

Fig. 1. The scheme of the 48.5 m flight path used for fission cross-section measurements

plates spaced by 5 mm. The second chamber contained 10 foils with lead or bismuth targets. A painting technique has been used for actinide targets production. The Pb and Bi targets have been produced from high purity metal using evaporation in vacuum. All targets were 180 mm in diameter, deposited on one side of 0.05 mm thick Al backings. The actinide and Pb/Bi targets were 150–560 μg/cm^2 and 260–520 μg/cm^2 thick, respectively. Both chambers contained 1 or 2 targets of 99.992% enriched ^{235}U 150–260 μg/cm^2 thick. A weak ^{252}Cf deposit was applied on actinide targets to match the gains of electronics. The foils were oriented perpendicular to the neutron beam, all target deposits facing away from the neutron source.

The time-of-flight and pulse height spectra were accumulated in a “multi-stop” mode with zero dead time using the data acquisition system based on a 100-MHz FLASH-ADC in each measuring channel. The start signal was provided by a gamma flash detector – the bare PMT placed in the neutron beam 2 m upstream of the fission chambers. This signal was also used to control a one-turn disposal of proton beam to the neutron-producing target. The “time-of-flight *vs* neutron energy” calibration with an accuracy 0.025–0.1% has been made for every target foil using a relativistic formula and position of the carbon and lead total cross-section resonances (Fig. 2). The graphite and lead filters, as well as the natural structure of the spectrum of fast neutrons, were used for this purpose. Also shown in the insert to Fig. 2 is a weak gamma-flash peak observed in the TOF-spectra and used as a true time-zero.

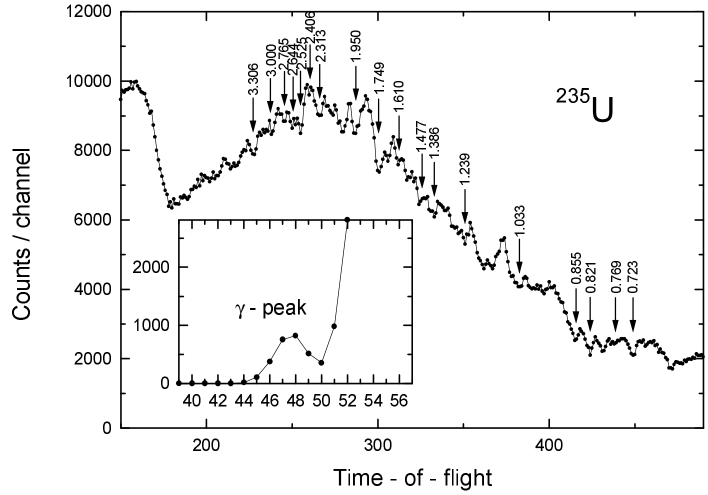

Fig. 2. Time-of-flight spectrum (10 ns channel width) for ^{235}U target. Positions of the dips due to resonances in total cross section of lead are shown by arrows (in MeV)

The initial stage of the raw data reduction included the following procedures:

1) digital processing of the data from FLASH-ADCs, including timing and determination of the pulse height, identification of start and stop signals on the background of electronic noise, alpha and pile-up events;

2) re-arranging of the data into 3 arrays: total pulse height spectrum (Fig. 3), total TOF-spectrum and two-dimensional matrix consisting of 512 TOF × 128 amplitude channels.

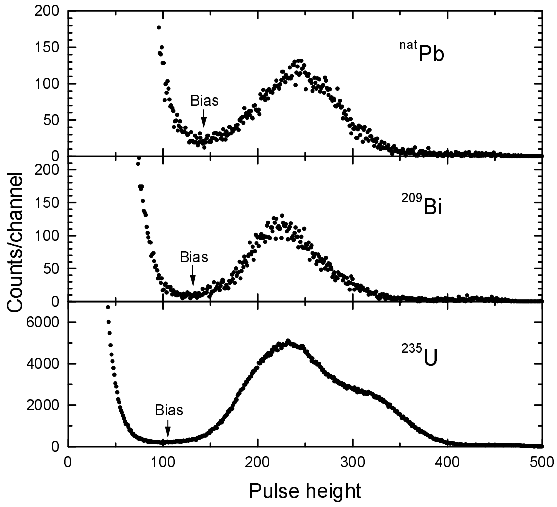

Fig. 3. Pulse height spectra observed for ^{nat}Pb, ^{209}Bi and ^{235}U targets in neutron energy range 25–200 MeV (Pb, Bi) and 0.5–200 MeV (^{235}U)

3. Data processing

In the data reduction process, the fission event counting rates were corrected for: 1) time independent background caused by alpha particle pile-up, spontaneous fission and electonic noise; 2) time dependent background caused by low-energy wrap-around neutrons and also by non-fission events due to high energy interactions of neutrons with target and backing foil materials; 3) attenuation of the neutron flux due to different flight path length, as well as absorption and scattering in the working gas and backing foil material; 4) fragment losses in the targets, neutron momentum transfer and angular anisotropy of fission fragments.

The attenuation correction was calculated using a simple assumption that the neutron is lost after first interaction with nuclei of the gas and structural materials of the fission chamber. The correctness and accuracy of this approach was tested by comparison of the TOF-spectra obtained for two ^{235}U targets placed in the chamber at 40–300 mm distance between them. A correction for the energy dependent linear-momentum and angular-momentum effects were calculated following a method of G.W.Carlson (Nucl. Instr. Meth. **119**, 97 (1974)).

The fission cross section ratios for ^{232}Th, ^{233}U, ^{238}U, ^{237}Np and ^{239}Pu and the reference nucleus ^{235}U obtained in the "shape" measurements have been normalized using the following methods: 1) calculation of the target thickness and detection efficiency ratios (only this method has been used for Pb and Bi); 2) normalization to recommended data; 3) threshold cross-section method (J.W.Behrens and J.C. Browne, Nucl. Sci. Eng. **77**, 444 (1981)). To use the last method, a part of the measurements has been done using "^{235}U-mixed" targets for threshold isotopes ^{232}Th, ^{238}U, ^{237}Np, and "^{238}U-mixed" targets for fissile isotopes ^{233}U and ^{239}Pu. It means that for the first three targets ^{235}U was used as a standard, while ^{238}U served as an intermediate normalization standard for the last two targets.

Finally, the normalized fission cross-section ratios have been converted to the cross-sections using fission cross section of ^{235}U from JENDL-3.2 (T.Nakagawa *et al.*, J. Nucl. Sci. Tech. **32**, 1259 (1995)) below 20 MeV and the recommended data of A.Carlson *et al.* above 20 MeV.

4. Results and discussion

The results of present measurements are shown in Figs. 4, 5 in comparison with experimental data of P.Lisowski *et al.* (for actinides), P.Staples and K.Morley and some others (for Pb and Bi), systematics of T.Fukahori and S.Perlstein (Report BNL-45200, 1991) and theoretical calculations. The error bars

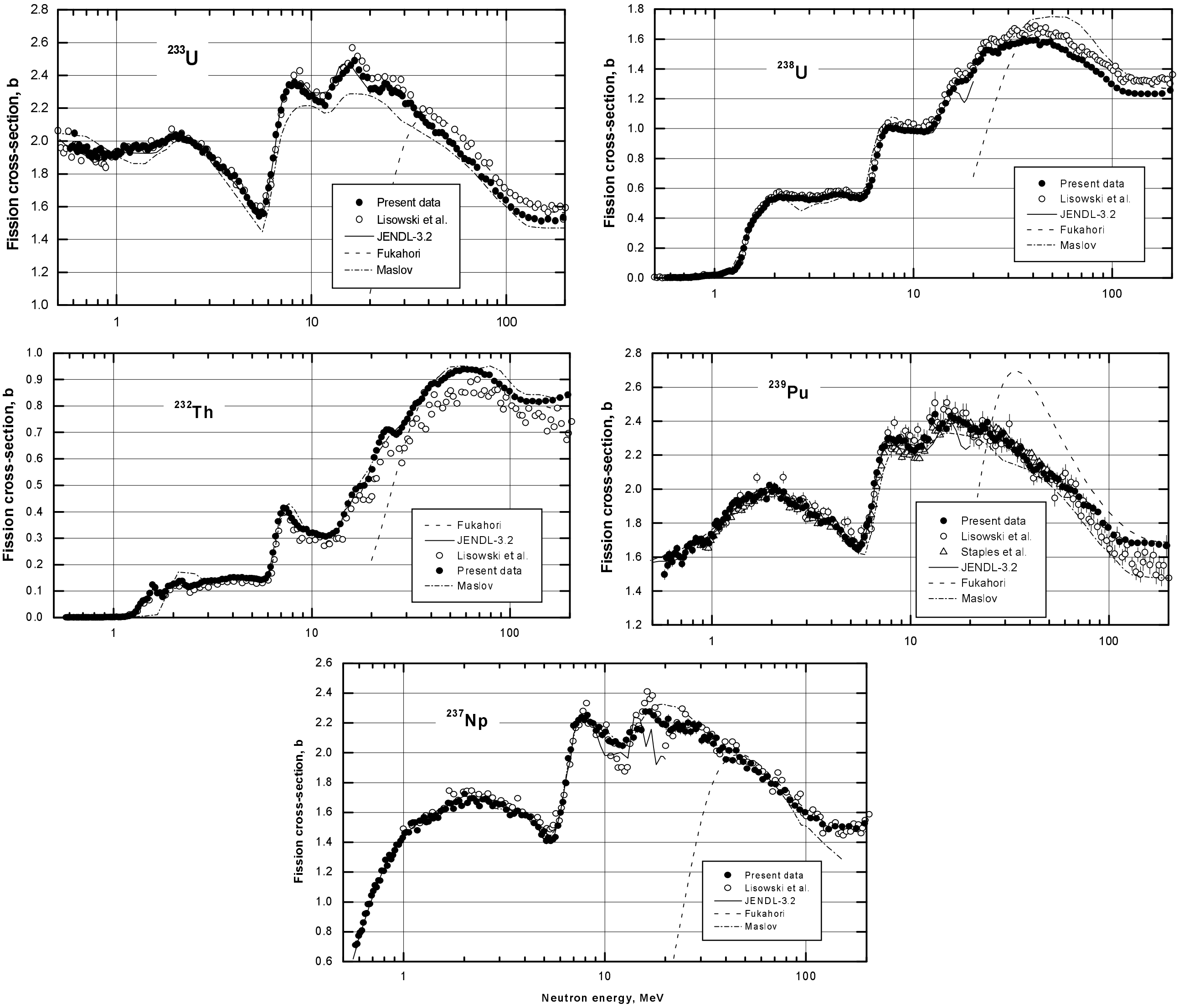

Fig. 4. Fission cross section of ^{232}Th, ^{233}U, ^{238}U, ^{237}Np and ^{239}Pu in the energy range up to 200 MeV

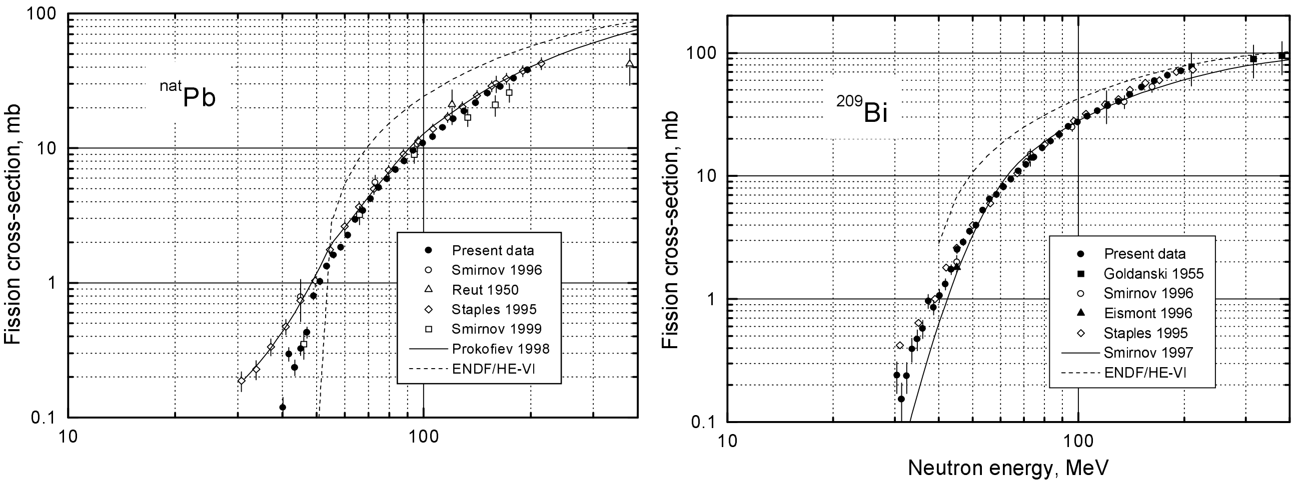

Fig. 5. Fission cross section ^{nat}Pb and ^{209}Bi in the energy range up to 200 MeV

represent the statistical errors only (one standard deviation). These uncertainties are less than 1% above ~ 1 MeV for actinides, and for lead and bismuth are as much as 4−7%, 2−3% and 1.2−1.6% at 60 MeV, 100 MeV and 200 MeV, respectively.

There is a large disagreement between present data and those of P.Lisowski *et al*. for ^{232}Th, ^{233}U, ^{238}U and ^{239}Pu above 20 MeV while for ^{237}Np both data sets are not in contradiction. In general, the comparison of our data with results of other authors shows that in the overlapping energy regions (below 20 MeV) our data are in a reasonable agreement with evaluated data. The data of systematics of T.Fukahori and S.Perlstein could be compared with experimental data only above 50 MeV and show a very strong disagreement with experiment in the case of ^{239}Pu.

The statistical model description of the measured fission cross sections up to 150 MeV showed that fission dynamics effects are not evident in the total fission cross section of ^{232}Th, ^{233}U, ^{238}U, ^{237}Np and ^{239}Pu. A supposition was made that collective mode damping is independent on the deformation of nuclei. The most challenging feature of the experimental data is a dependence of the observed fission cross section on the fissility of a target nuclide up to 150 MeV that needs some other modification of the statistical model approach.

Above 50 MeV, the present data for ^{nat}Pb are in a good agreement with of P.Staples *et al*. and with the parameterization of A.V.Prokofiev *et al.* based on the measurements with ^{nat}Pb and ^{208}Pb, including the data of P.Staples *et al.* and other known experimental data obtained mainly in separate energy points. The ENDF/HE-VI data based on the systematics of T.Fukahori and S.Perlstein for ^{208}Pb lie much higher than all experimental data. It should be noted that, while comparing ^{nat}Pb and ^{208}Pb data, the latter were multiplied by 1.43, as it was done in the paper of A.V.Prokofiev *et al*. where this coefficient was verified experimentally with an accuracy ± 10% in the energy range 45−160 MeV.

In the case of ^{209}Bi, an agreement of the present data with those of other authors is good within the stated uncertainties. There is a noticeable discrepancy between our data, as well as the data of P.Staples *et al.*, and the parameterization at neutron energies below ~ 50 MeV and above ~ 120 MeV. The ENDF/HE-VI data lies up to 2 times higher than all experimental data below ~ 150 MeV, but above this energy the tendency to agreement can be seen.

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