Fission Cross Section Measurements at Intermediate Energies

Alexander LAPTEV*

Japan Nuclear Cycle Development Institute
Tokai works, Tokai-mura, Naka-gun, Ibaraki-ken 319-1194, Japan
e-mail: laptev@jnc.go.jp

The activity in intermediate energy particle induced fission cross-section measurements of Pu, U isotopes, minor actinides and sub-actinides in PNPI of Russia is reviewed. The neutron-induced fission cross-section measurements are under way in the wide energy range of incident neutrons from 0.5 MeV to 200 MeV at the GNEIS facility. In number of experiments at the GNEIS facility, the neutron-induced fission cross sections were obtained for many nuclei. In another group of experiments the proton-induced fission cross-section have been measured for proton energies ranging from 200 to 1000 MeV at 100 MeV intervals using the proton beam of PNPI synchrocyclotron.

1. Introduction

There is a long-standing need for information about fission reactions of heavy nuclei induced by particles at intermediate energies. Such information is required for many applications, e.g., accelerator-driven transmutation of nuclear waste, energy generation, fundamental physics, etc. To make progress in these areas requires improved accuracy and reliability of the relevant nuclear data, and many of these data needs are not yet fulfilled [1].

The Petersburg Nuclear Physics Institute (PNPI) of Russian Academy of Sciences has a big research program devoted to intermediate energies particles induced fission cross section measurements. The neutron-induced fission cross-section measurements are under way in the wide energy range of incident neutrons from 0.5 MeV to 200 MeV at the GNEIS facility [2]. During the first stage of this research the measurements of fission cross-sections of ^{233}U, ^{238}U, ^{232}Th, ^{237}Np, ^{239}Pu, ^{nat}Pb and ^{209}Bi have been performed [3]. This investigation has been done jointly by PNPI and Khlopin Radium Institute (KRI) in collaboration with Nuclear Data Center of JAERI. Not long ago the new measurements of neutron-induced fission cross-section of ^{240}Pu, ^{243}Am and ^{nat}W have been completed at the GNEIS facility [4,5] jointly by PNPI and KRI in collaboration with LANL. Other research team of PNPI measured the proton-induced fission cross sections of ^{233}U, ^{235}U, ^{238}U, ^{232}Th, ^{237}Np, ^{239}Pu, ^{nat}Pb and ^{209}Bi at the proton beam of PNPI synchrocyclotron energies ranging from 200 to 1000 MeV at 100 MeV intervals [6]. This article presents a brief review of these both neutron- and proton-induced fission cross-section measurements.

2. Neutron-induced fission

2.1. Experimental set-up

At works [3-5] the fission cross-section ratios for investigated nuclei relative to ^{235}U have been measured using the neutron time-of-flight spectrometer GNEIS. This facility is based on the 1 GeV proton synchrocyclotron of PNPI and has average intensity 310^{14} n/s, a burst duration 10 ns and repetition rate up to 50 Hz. The flight path was 48.5 m. The beam is shaped by a system of iron, brass and lead collimators. The measurements were carried out with the use of a sweeping magnet placed at flight path of 30 m. A schematic layout of the experimental set-up is shown in Fig. 1.

The fission reaction rate was measured using two fast parallel plate ionization chambers filled with methane (94-100 %) and CF_4 (6-0 %) mixture working gas at the absolute pressure of 2.5 – 3.5 atm. [3] or pure methane at about 3 atm. [4,5]. Both chambers (for actinides and sub-actinides) had

* On leave from the Petersburg Nuclear Physics Institute, Gatchina, Russia.

several sections. Each section contains one pair of cathode and anode plates spaced by 5 mm. The high quality fissionable materials have been used for actinide targets production. The target foils were 150-560 μg/cm^2 thick and 180 mm in diameter, deposited on one side of 0.05-mm-thick aluminum backings. Also, a weak ^{252}Cf deposit was applied on the actinide foils to match the gains of electronics.

For each nucleus under investigation, the time-of-flight and pulse height spectra were accumulated using the data acquisition system based on a 100-MHz FLASH-ADC in each measuring channel. The start signal was provided by gamma flash-detector, a bare PMT placed in the neutron beam. This signal was also used to control a single-turn deflection of the proton beam to the neutron-producing target.

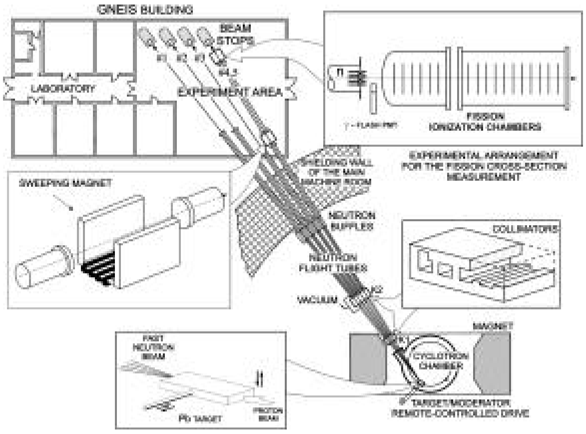

Fig. 1 Schematic layout of the GNEIS facility and experimental arrangement for fission cross-section measurements.

2.2. Data processing

Identification of start (gamma flash) and stop (fission) signals on the background of electronic noise, alpha- and pile-up events in works [3-5] has been made by a method of digital filtering [7]. The raw data reduction included composing pulse height and TOF information into a 2-dimensional matrix consisting of 512 TOF channels by 128 amplitude channels. The example of this matrix in case of ^{240}Pu is shown in Fig. 2. The average pulse height spectrum in case of ^{nat}W and its fission and background components are presented in Fig. 3.

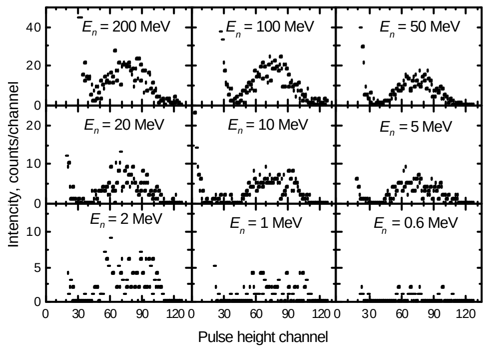

Fig. 2 Pulse height spectra of ^{240}Pu measured at different neutron energies.

The "*time-of-flight vs neutron energy*" calibration was made with an accuracy 0.03 % using the positions of lead total cross section resonances and the weak gamma-flash peak observed in the TOF-spectra from which a true time-zero was derived (v. Fig. 4).

In the data reduction process, the fission event counting rates were corrected for (1) background events, (2) the neutron flux attenuation for investigated and reference nuclei, (3) fragment losses in the targets due to finite deposit thickness, and (4) neutron momentum transfer and angular anisotropy of fission fragments. The attenuation correction was calculated using the neutron total cross-sections from ENDF/B-VI. A correction for the energy dependent linear-momentum and angular-momentum effects were calculated following a method of G. Carlson [8]. 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 200 mm distance between them.

The fission cross-section ratios for investigated nuclei and the reference nucleus ^{235}U obtained in the "shape" measurements have been normalized using the following methods:

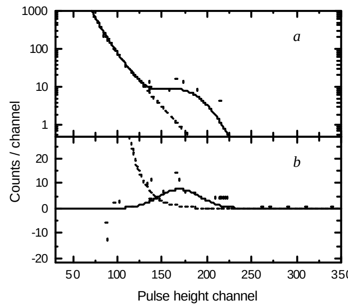

Fig. 3 (a) Measured pulse height spectrum for one foil of ^{nat}W; the solid curve denotes fission fragments and the dash curve shows background events; (b) pulse height spectrum for fission fragments after background subtraction; the background is shown again as the dashed curve.

1) calculation of the target thickness and detection efficiency ratios;
2) normalization to the recommended data;
3) threshold cross-section method [9].

All three methods were used, but the best accuracy was obtained using the threshold method for threshold isotopes ^{238}U and ^{232}Th. For this purpose, a part of the measurements [3] has been done using mixed targets. The fission cross-section ratio normalization for ^{233}U, ^{237}Np and ^{239}Pu in [3] has been done in the 1.75-4.0 MeV energy interval using the data of JENDL-3.2 file. In [5] the fission cross-section ratios for investigated actinide nuclei ^{240}Pu, ^{243}Am and the reference nucleus ^{235}U obtained in the "shape" measurements have been normalized using the target thicknesses. An accuracy of last normalization was 5%.

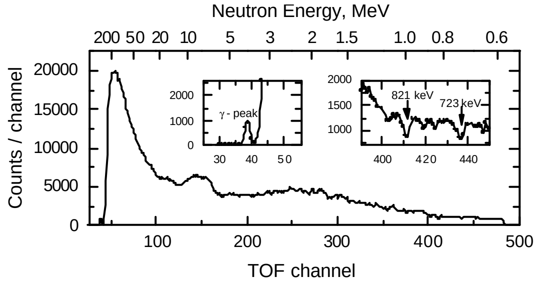

Fig. 4 Time-of-flight spectra (10 ns channel width) obtained for ^{235}U target after background subtraction. Inserts show TOF-spectra in regions of gamma-flash and lead resonances in more detail.

An absolute normalization of the measured cross-section ratios for sub-actinides ^{nat}Pb, ^{209}Bi in [3] and ^{nat}W, ^{209}Bi in [5] has been done using the thickness of the targets and detection efficiencies.

Finally, the normalized fission cross-section ratios have been converted to the cross-sections using the fission cross-section of ^{235}U from JENDL-3.2 [10] below 20 MeV and the recommended data of A. Carlson *et al.* [11] above 20 MeV.

2.3. Results

The results of measurements [3] and [4,5] are shown in Figs. 5,6 (for actinides) and Figs. 7,8 (for sub-actinides) in comparison with other experimental data and some systematics and theoretical calculations. The error bars represent the statistical errors only (one standard deviation). These

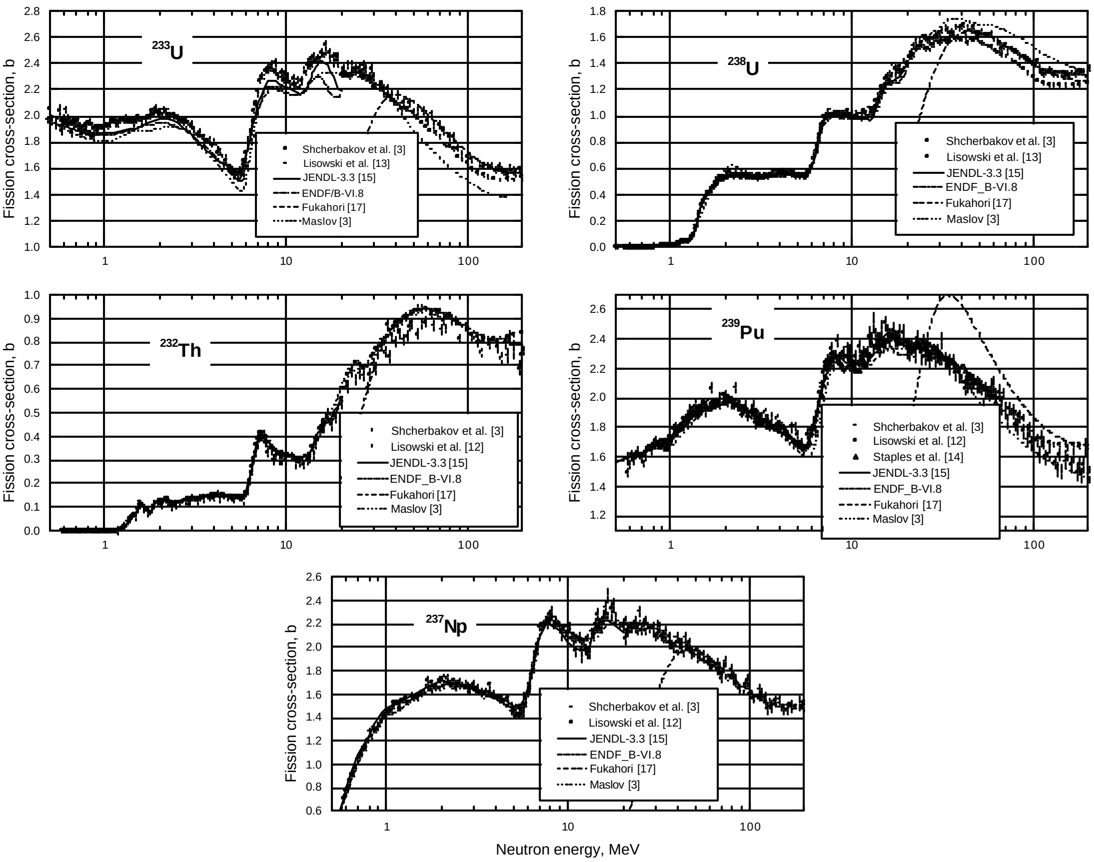

Fig. 5 Fission cross section of ^{233}U, ^{238}U, ^{232}Th, ^{239}Pu and ^{237}Np measured in [3] in the energy range 1-200 MeV.

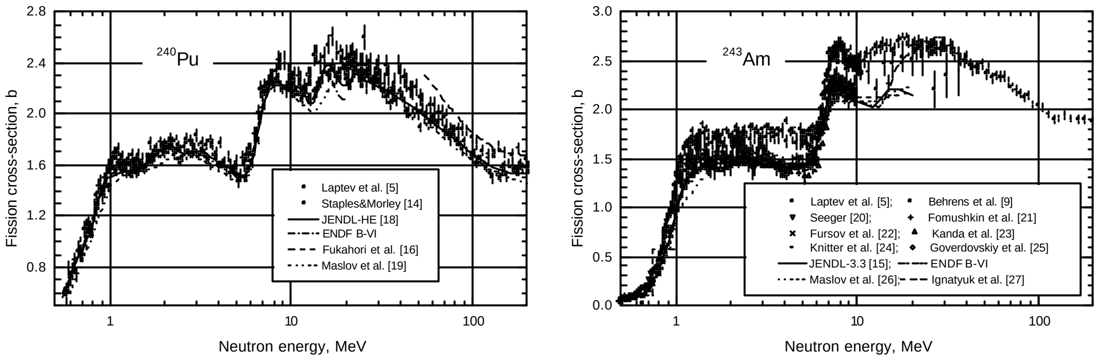

Fig. 6 Fission cross section of ^{240}Pu and ^{243}Am measured in [4,5] in the energy range 1-200 MeV.

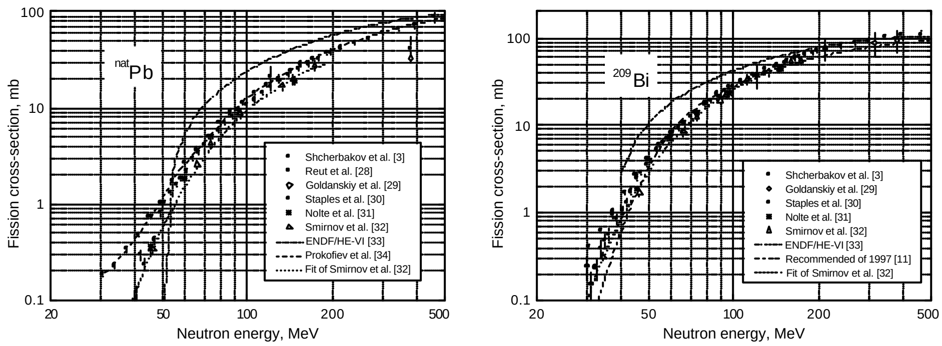

Fig. 7 Fission cross section of ^{nat}Pb and ^{209}Bi measured in [3] in the energy range up to 200 MeV.

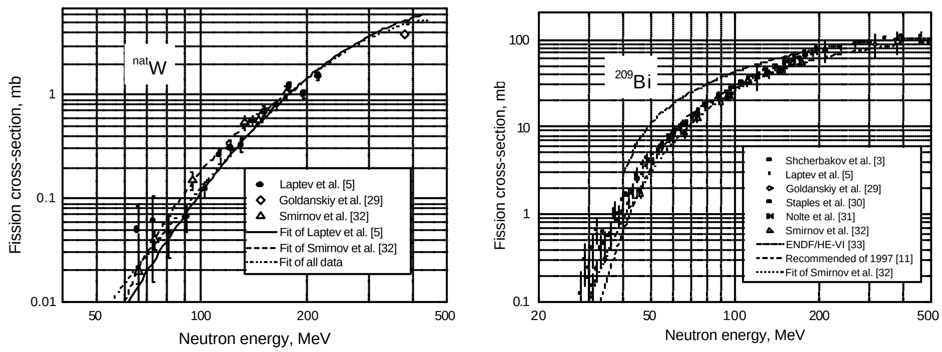

Fig. 8 Fission cross section of ^{nat}W and ^{209}Bi measured in [5] in the energy range up to 200 MeV.

uncertainties are about 1-2 % at neutron energies above 1 MeV and that of sub-actinide nuclei ^{nat}Pb and ^{209}Bi varies from 12-20 % at 40 MeV to 1-2 % at 200 MeV but in case of ^{nat}W it varies from 19 % at 100 MeV to 7 % at 200 MeV.

There is a some disagreement between data of [3] and that of Lisowski *et al.* [12,13] for ^{233}U, ^{238}U, ^{239}Pu and ^{232}Th above 20 MeV while both data sets are consistent for ^{237}Np. In general, the comparison of data of [3] with results of other authors shows that in the overlapping energy regions (below 20 MeV) data of [3] are in reasonable agreement with evaluated data. The data of systematics [16,17] 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 calculations of Maslov in [3] show satisfactory description of the measured fission cross-sections.

For ^{240}Pu (Fig. 6) of [5], there is good agreement between these data and those of Staples and Morley [14] up to 70 MeV, but there are disagreements in the energy range of 70-200 MeV. Some other small differences are within stated errors. On the opinion of authors of [5], most of the differences are in normalization rather than shape. Such differences vanish, except for short ranges 1.5-3 and 7-8 MeV, after normalization of both data sets at neutron energy 14 MeV. Smooth curves present cross section values from JENDL High Energy [18] and ENDF B-VI libraries, theoretical calculations of Maslov *et al.* [19] (Hauser-Feshbach statistical model) and systematics of Fukahori *et al.* [16]. All these evaluations show a rough agreement with data of [5] for ^{240}Pu in the full energy range under investigation. But there are some visible disagreements in detailed data description. For example, the evaluation of JENDL-HE library [18] and the result of calculation [19] lie below data of [5] at the energy range above 40 MeV but the systematic [16] lies above these data.

For ^{243}Am (Fig. 6) of [5], a comparison of these data with other data sets shows good agreement of the present data with that of Behrens *et al.* [9] and Goverdovskiy *et al.* [25]. There are no available previous data for the fission cross section of ^{243}Am above ~40 MeV, these data have been obtained for the first time. In case of ^{243}Am there are significant disagreements between previous data. The cross section values from the libraries JENDL-3.3 [15] and ENDF B-VI, theoretical calculations of Maslov *et al.* [26] and evaluation of Ignatyuk *et al.* [27] correspond to other data sets rather than [5]. Normalization of data for ^{243}Am of [5] to libraries' 14 MeV value could not make agreement of all experimental data sets but withdraws disagreements with evaluations.

Above 50 MeV the data for ^{nat}Pb of [3] (Fig. 7) are in a good agreement with that of Staples *et al.* [30], other data sets and the parameterization of Prokofiev *et al.* [34] based on the measurements with ^{nat}Pb and ^{208}Pb, including the data of Staples *et al.* [30] and other known at that moment experimental data carried out mainly in separate energy points. The ENDF/HE-VI data based on the systematics of Fukahori *et al.* [33] for ^{208}Pb lies much higher than all experimental data.

In the case of ^{209}Bi (Fig. 7), an agreement of the data [3] with that of other authors is good within the stated uncertainties. There is a noticeable discrepancy between data [3], as well as the data of Staples *et al.* [30], and the parameterization [11] at neutron energies below ~50 MeV and above ~120 MeV. The ENDF/HE-VI data [33] lies up to 2 times higher than all experimental data below ~150 MeV, but above this energy the tendency to agreement can be seen.

There is generally good agreement between data [5] and those of Refs. [29,32] for ^{nat}W (Fig. 8), except for a possible discrepancy in the 90-100 MeV region. In the work [5] the neutron-induced fission cross-section of ^{nat}W has been measured for the first time with a "white" neutron source. Smooth curves in Fig. 8 present fit of the data [5], fit of ref. [32] and fit of all data sets. Comparison (Fig. 8) of fission cross section of ^{209}Bi measured in the experiment [5] with that obtained in previous measurement [3] shows a very good agreement.

3. Proton-induced fission

The proton-induced fission cross sections of the ^{233}U, ^{235}U, ^{238}U, ^{232}Th, ^{237}Np, ^{239}Pu, ^{nat}Pb and ^{209}Bi have been measured at the proton beam of PNPI synchrocyclotron energies ranging from 200 to 1000 MeV at 100 MeV intervals [6]. The measurement method is based on the registration in coincidence of both fission fragments by two parallel plate avalanche counters (PPAC's) located at very close distance from a target. To obtain the proton beams with different energies, the method of degrading of the initial 1000 MeV proton beam by copper degraders has been used.

The 100 % detection efficiency of fission fragments at a large solid angle acceptance (10 sr) has been achieved in the work [6]. The investigated targets have been produced by method of vacuum

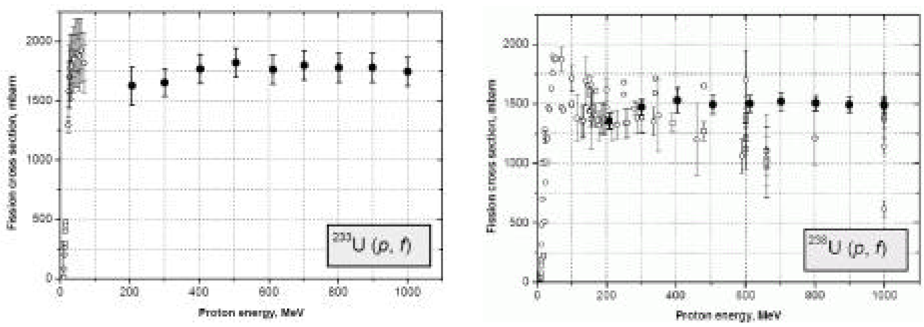

Fig. 9 Fission cross section of ^{233}U and ^{238}U measured in [6] in the energy range 200-1000 MeV. Black circles are the results of [6] and open ones are previous data taken from reviews [35,36].

evaporation of fissile material. The thicknesses of targets were in the range from 100 to 450 μg/cm^2. The proton beam monitoring has been done at low beam intensity (~10^5 p/s) by direct count of scintillation telescope counters, and at high beam intensity (~10^7 p/s) by registration of *pp*-elastic scattering on the CH_2 target. The results [6] on energy dependence of total fission cross-sections of ^{233}U and ^{238}U targets are presented in Fig. 9. A statistical accuracy of the measured fission cross sections [6] is better than 1.5% and the total one is better than 10%. The cross sections for proton induced fission of ^{233}U in the energy range 200-1000 MeV was obtained in [6] for the first time. According to opinion of authors [6], their results for ^{238}U in the energy range from 300 to 900 MeV do not agree with a majority of the early-obtained data.

References

[1] M. Salvatores, *J. Nucl. Sci. Tech.*, Suppl. 2, 4 (2002).
[2] N.K. Abrosimov, G.Z. Borukhovich, A.B. Laptev *et al.*, *Nucl. Instr. Meth.* **A242**, 121 (1985).
[3] O. Shcherbakov, A. Donets, A. Evdokimov *et al.*, *J. Nucl. Sci. Tech.*, Suppl. 2, 230 (2002).
[4] A.B. Laptev, A.Yu. Donets, A.V. Fomichev *et al.*, *Nucl. Phys.* **A 734S**, E45 (2004).
[5] A.B. Laptev, A.Yu. Donets, V.N. Dushin *et al.*, Report at the *Int. Conf. on Nucl. Data for Sci. and Tech. (ND2004)*, Santa Fe, New Mexico, USA, Sept. 26-Oct. 1, 2004.
[6] A. Kotov, Y. Gavrikov, L Vaishnene *et al.*, Report at the *XVI Int. Workshop on Physics of Nuclear Fission*, IPPE, Obninsk, Russia, October 7-10, 2003.
[7] D.D. Burgess, R.J. Tervo, *Nucl. Instr. Meth.* **214**, 431 (1983).
[8] G.W. Carlson, *Nucl. Instr. Meth.* **119,** 97 (1974).
[9] J.W. Behrens, J.C. Browne, *Nucl. Sci. Eng.* **77**, 444 (1981).
[10] T. Nakagawa, S. Shibata, S. Chiba *et al.*, *J. Nucl. Sci. Tech.* **32**, 1259 (1995).
[11] A.D. Carlson, S. Chiba, F.-J. Hambsch *et al.*, IAEA Report INDC(NDS)-368. Vienna, 1997.
[12] P.W. Lisowski, J.L. Ullmann, S.J. Balestrini *et al.*, *Proc. of Int. Conf. on Nucl. Data for Sci. and Tech.*, Mito, Japan, May 30-June 3, 1988, p.97.
[13] P.W. Lisowski, A. Gavron, W.E. Parker *et al.*, *Proc. of Int. Conf. on Nucl. Data for Sci. and Tech.*, Julich, Germany, May 13-17, 1991. Ed. S.M. Qaim, Springer-Verlag, 1992, p.732.
[14] P. Staples, K. Morley, *Nucl. Sci. Eng.* **129**, 149 (1998).
[15] K. Shibata, T. Kawano, T. Nakagawa *et al.*, *J. Nucl. Sci. Tech.* **39**, 1125 (2002).
[16] T. Fukahori, S. Pearlstein, Report BNL-45200, 1991.
[17] T. Fukahori, *private communication*, 1999.
[18] JENDL High Energy Data File 2004. Numerical data were obtained from T. Fukahori, 2004.
[19] V. Maslov, Yu. Porodzinskij, M. Baba, A. Hasegawa, *J. Nucl. Sci. Tech.*, Suppl. 2, 80 (2002).
[20] P.A. Seeger, LANL Report LA-4420. Los Alamos, 1970.
[21] E.F. Fomushkin, G.F. Novoselov, Ju.I. Vinogravdov *et al.*, *Ydernie Konstanti* **3/57**, 17 (1984).
[22] B.I. Fursov, E.Ju. Baranov, M.P. Klemyshev *et al.*, *Atomnaya Energiya* **59**, 339 (1985).
[23] K. Kanda, H. Imaruoka, H. Terayama *et al.*, *J. Nucl. Sci. Tech.* **24**, 423 (1987).
[24] H.-H. Knitter, C. Budtz-Jorgensen, *Nucl. Sci. Eng.* **99**, 1 (1988).
[25] A.A. Goverdovskiy, A.K. Gordyushin, B.D. Kuz'minov *et al.*, *Atomn. Energ.* **67**, 30 (1989).
[26] V.M. Maslov, E.Sh. Sukhovitskij *et al.*, IAEA Report INDC(BLR)-006. Vienna, 1996.
[27] A.V. Ignatyuk, A.I. Blokhin, V.P. Lunev *et al.*, *VANT, Ser. Jad. Konst.*, issue 1, 25 (1999).
[28] A.A. Reut et al., Report of the Institute of Physics Problems, 1950.
[29] V.I. Goldanskiy, E.Z. Tarumov, V.S. Penkina, *Dokl. Akad. Nauk* **101**, 1027 (1955).
[30] P. Staples *et al.*, *Bull. Am. Phys. Soc.* **40**, 962 (1995).
[31] R. Nolte, M.S. Allie, P.J. Binns *et al.*, *J. Nucl. Sci. Tech.*, Suppl. 2, 311 (2002).
[32] A.N. Smirnov, V.P. Eismont, N.P. Filatov *et al.*, Submitted to *Phys. Rev.* **C** (2004).
[33] T. Fukahori, S. Pearlstein, IAEA Report INDC(NDS)-245. Vienna, 1991.
[34] A.V. Prokofiev, S.G. Mashnik, A.J. Sierk, LANL Report LA-UR-98-0418. Los Alamos,1998.
[35] A.I. Obukhov, *Phys. Particl. Nucl.* **32**, 162 (2001).
[36] A.V. Prokofiev, *Nucl. Instr. Meth*. **A 463**, 557 (2001).