INTERMEDIATE ENERGY NEUTRON-INDUCED FISSION CROSS-SECTIONS FOR PROSPECTIVE NEUTRON PRODUCTION TARGET IN ADS

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Abstract

Up-to-date status is considered of the experimental database on neutron-induced fission cross-sections of tantalum, tungsten, lead, mercury, gold, and bismuth nuclei in the neutron energy range from the fission threshold to 175 MeV. The perspective of creating a more complete database is discussed, including (n,f) cross-sections for separated isotopes of lead and tungsten.

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1. Introduction

Intermediate-energy fission data are of interest not only for fundamental physics, but also for applied nuclear research. First of all it is connected with problems of accelerator driven systems (ADS) for power production and transmutation of long-lived radioactive waste (see e.g. [1]). Such elements as Ta, W, Hg, Pb and Bi are either already used as neutron producing target materials or considered as potential candidates. Since fission and spallation reactions at intermediate energies are the main reaction channels of neutron interactions with heavy nuclei, they have the most practical significance, but are poorly investigated. Fission reaction contributes to the generation of the neutron field in the target-blanket assembly, as well as to the production of radionuclides and chemically toxic products in the target. For relatively light nuclei, such as Pb and Bi with fission cross-section of only a few percent of the total reaction cross-section, the residual activity of the fission products with high energy release and, often, long half-lives, is expected to be significant. It is estimated that the contribution of the fission products to the overall residual activity of a lead target irradiated by 1.6-GeV protons may be as much as 10-15% for cooling times of about one year [2]. At present, theoretical description of sub-actinide nuclei fission cannot match the practical needs. For example, the ^{nat}W(p,f) cross-section predicted by the LAHET code was found to be about 20 times lower than the experimental results [3]. If one bears in mind the insufficient predictive power of available nuclear reaction models, especially with respect to fission, it is possible that the real residual activity may be significantly different.

Intermediate-energy fission data are also of interest for nuclear standards, and particle beam monitoring required for other applications as neutron cancer therapy, shielding of accelerators, cosmic studies, thermonuclear synthesis etc. Furthermore, due to the insensitivity to low energy neutrons, the ^{209}Bi(n,f) cross-section has been approved by IAEA as a secondary standard for neutron flux determination at intermediate energies [4].

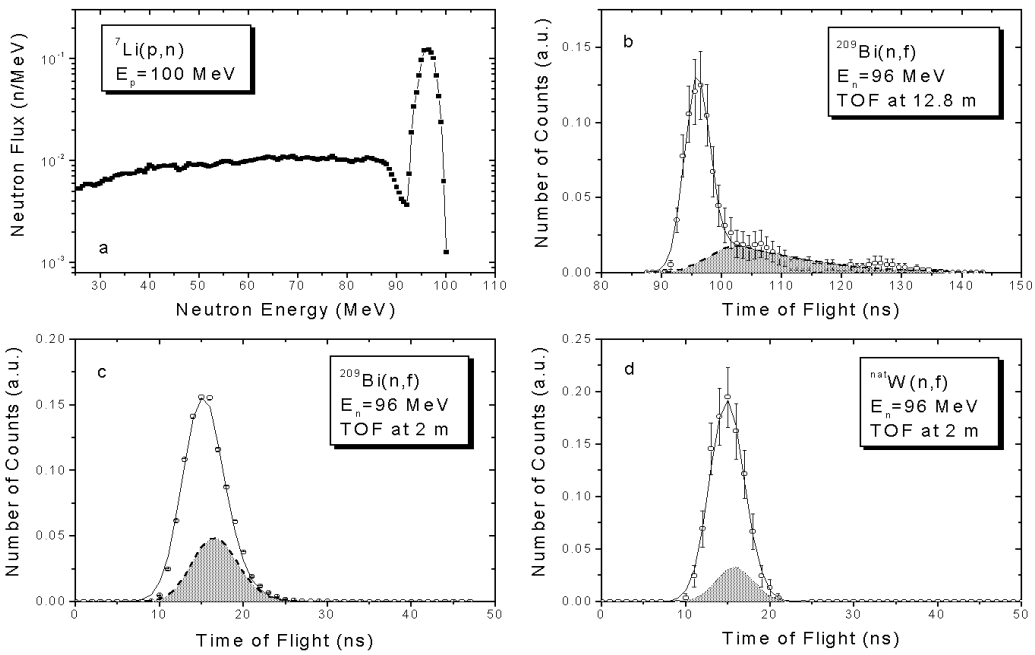
As a response to the outlined needs, V.G. Khlopin Radium Institute and Uppsala University perform a joint program of (n,f) cross-section measurements for sub-actinides in the energy region between 20 and 180 MeV. Earlier, results for the absolute and relative (n,f) cross-sections of ^{238}U, ^{209}Bi and ^{208}Pb have been published [5-7]. In the framework of ISTC project #540 the measurements for ^{208}Pb and ^{209}Bi were continued with better experimental conditions, including new measurements on ^{181}Ta, ^{nat}W, ^{nat}Hg, ^{197}Au and ^{nat}Pb. The measurements on gold were included because of their methodical and theoretical importance. Preliminary results for the above listed sub-actinides have been published recently [8-11]. In the present work the results of further processing of the data are given and the status of the ^{209}Bi (n,f) cross-section standard is discussed.

The prospects for creating of more complete database are considered, including (n,f) cross-sections for separated isotopes of lead and tungsten, specifically for ^{208}Pb and ^{184}W , which cross-sections are included in the High Priority Request List of nuclear data for the nucleon energy region up to 200 MeV [12]. These data are needed for the development of adequate nuclear fission models, as well as computer codes for ADS. The needs of fission cross-section measurements for the above mentioned nuclides are stressed, not only with neutrons, but also with protons, in the same projectile energy region. Comparison of proton- and neutron-induced fission cross-sections [13], carried out on a common physical basis [14], gives added credence to the experimental database.

2. Up-to-date status of the (n,f) cross-sections of sub-actinides

The (n,f) cross-sections of ^{181}Ta, ^{nat}W, ^{nat}Hg, ^{197}Au, ^{208}Pb and ^{nat}Pb published in [9-11] have been obtained with the assumption that the fraction of the total fission events induced by full energy neutrons is equal for the studied nuclides and for the monitor reaction ^{209}Bi(n,f). In this work we have calculated the fraction of the total fission events induced by full energy neutrons for each reaction under study. For this purpose the TOF spectra of fission events have been simulated using the experimental neutron spectra [15-19] and the given time parameters of the proton beam. The final cross-sections and the fractions of the full energy fission events have been obtained as a result of an iteration procedure with the initial cross-sections taken from [4,9]. The results of the calculations carried out for the ^{209}Bi(n,f) and ^{nat}W(n,f) reactions at a peak neutron energy of 96 MeV are shown in Figure 1 together with the neutron spectrum from the ^{7}Li(p,n) reaction measured by Nakao *et al*. [15] at the similar incident proton energy.

Figure 1. **(a) Neutron spectrum from ^{7}Li(p,n) reaction at 100 MeV proton energy [15]; (b) and (c) TOF spectra of ^{209}Bi(n,f) fission events at 12.8 m and 2 m flight distances, correspondingly; (d) TOF spectrum of ^{nat}W(n,f) fission events at 2 m flight distance. Open dots in (b), (c), and (d) are experimental TOF spectra. Solid curves are calculated TOF spectra. Filled areas under dashed curves are calculated fractions of fissions induced by low energy tail neutrons.**



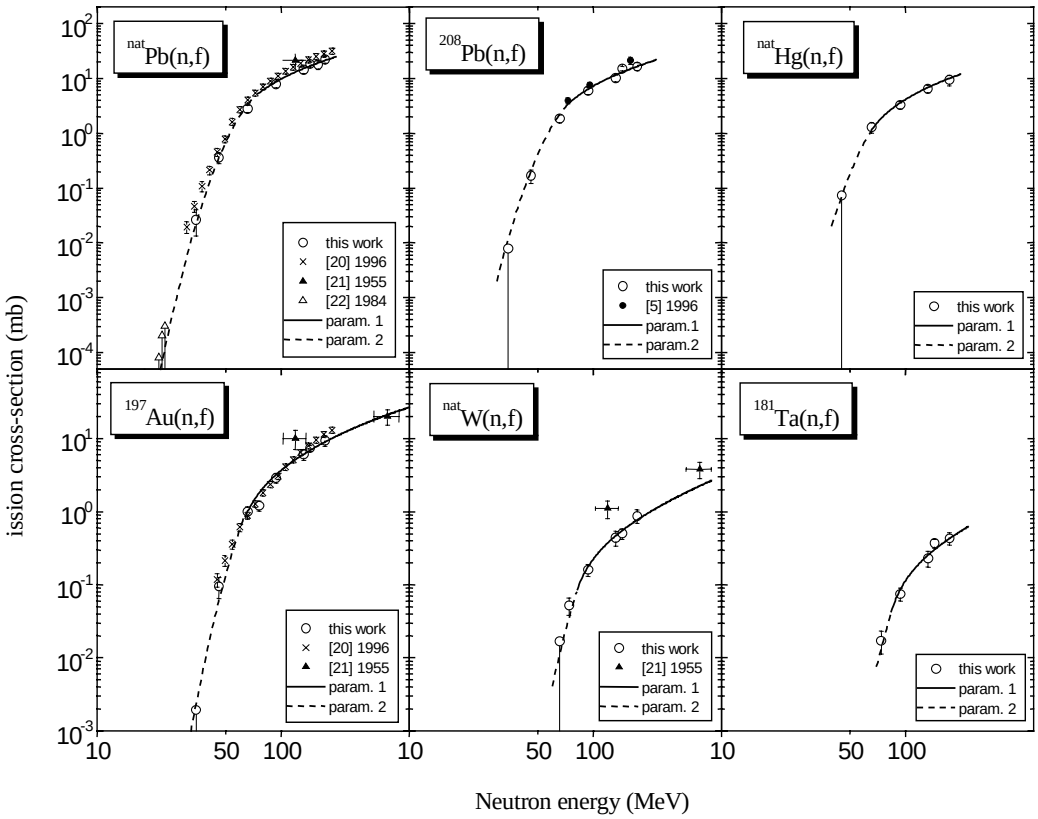
The results for (n,f) cross-section ratios are given in Table 1. The absolute (n,f) cross-sections obtained using the ^{209}Bi(n,f) cross-section as a standard [4] are shown in Figure 2 together with fits according to Equations 1) and 2) below and our previously reported data for ^{208}Pb [6].

Table 1. **Relative neutron-induced cross-sections**

E_n	**(n.f) cross-section ratio**					
MeV	$^{nat}Pb/^{209}Bi$	$^{208}Pb/^{209}Bi$	$^{nat}Hg/^{209}Bi$	$^{197}Au/^{209}Bi$	$^{nat}W/^{209}Bi$	$^{181}Ta/^{209}Bi$
35	0.166 ± 0.079	<0.051	–	<0.012	–	–
45	0.191 ± 0.012	0.069 ± 0.005	<0.036	0.048 ± 0.013	–	–
66	0.292 ± 0.026	0.185 ± 0.017	0.127 ± 0.026	0.099 ± 0.017	<0.0017	–
75	0.305 ± 0.026	0.205 ± 0.019	0.099 ± 0.021	0.099 ± 0.010	0.0040 ± 0.0007	0.0013 ± 0.0004
94	0.379 ± 0.031	0.288 ± 0.026	0.158 ± 0.017	0.139 ± 0.017	0.0075 ± 0.0014	0.0036 ± 0.0006
96	0.391 ± 0.036	0.305 ± 0.028	–	–	0.0089 ± 0.0022	0.0037 ± 0.0012
133	0.429 ± 0.037	0.305 ± 0.029	0.199 ± 0.019	0.184 ± 0.022	0.0134 ± 0.0026	0.0071 ± 0.0014
144	0.505 ± 0.045	0.418 ± 0.037	0.158 ± 0.019	0.205 ± 0.025	0.0140 ± 0.0025	0.0101 ± 0.0017
160	0.422 ± 0.062	–	–	–	–	–
174	0.526 ± 0.042	0.412 ± 0.034	0.215 ± 0.040	0.211 ± 0.020	0.0210 ± 0.0027	0.0096 ± 0.0013

Figure 2 shows also the earlier data of other authors [20-22]. In order to make a comparison for available absolute ^{197}Au and $^{nat}Pb(n,f)$ cross-sections, the $^{nat}Pb/^{235}U$ and $^{197}Au/^{235}U$ ratios measured at LANSCE [20] were multiplied by the standard $^{235}U(n,f)$ cross-section [4]. Early measurements of Reut *et al.*, Goldanskiy *et al.*, and Dzhelepov *et al.*, [21] were made using neutrons from the Cu(d,n) reaction. Neutrons produced by that reaction have a broader spectrum, as indicated by the horizontal error bars in Figure 2. Nevertheless, there is a qualitative agreement between those and more recent data. Vorotnikov [22] performed unique studies of (n,f) cross-sections for sub-actinides in the vicinity of the fission barrier (about 20-25 MeV) using a deuterium-tritium neutron source and solid state nuclear track detectors for the fission fragment detection.

Figure 2. **The ^{181}Ta(n,f), ^{nat}W(n,f), ^{nat}Hg(n,f), ^{197}Au(n,f), ^{208}Pb(n,f) and ^{nat}Pb(n,f) cross-sections. Solid and dashed curves are the fits made with the use the formulae 1, 2 (see the text below)**



The available experimental data on the ^{nat}Pb(n,f) and ^{197}Au(n,f) cross-sections [20] and the data of the present work are in qualitative agreement. There are, however, systematic discrepancies between these data sets in the neutron energy regions below about 50 MeV and above about 100 MeV for ^{197}Au(n,f) and practically in the whole energy region for ^{nat}Pb(n,f). This fact, together with the comparison for ^{197}Au/^{209}Bi and ^{nat}Pb/^{209}Bi cross-section ratios performed in [10], leads to a suggestion that some background may not have been taken fully into account in the LANSCE measurements of the ^{nat}Pb(n,f) and ^{197}Au(n,f) cross-sections. The low-energy ^{nat}Pb(n,f) data of the present work are compatible with the upper limits of the cross-section obtained by Vorotnikov [22]. The old data of Reut *et al.*, Goldanskiy *et al.*, and Dzhelepov *et al.*, [21] for ^{nat}Pb(n,f), ^{197}Au(n,f), and ^{nat}W(n,f) cross-sections at a neutron energy of 120 MeV are considerably (two-three times) higher than our data in the same energy region.

The fits in Figure 2 were made using the same formulae that have been used for parameterization of the ^{209}Bi(n,f) cross-section [4]:

$$\sigma_{nf} = p_{11} \cdot (1 - \exp(p_{12} \cdot (E_n - p_{13}))) \tag{1}$$

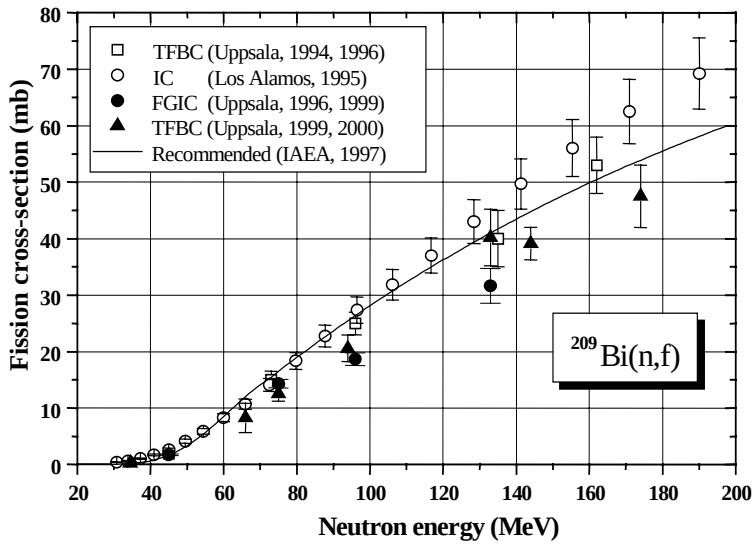
above about 70 MeV [23], and

$$\sigma_{nf} = \exp(p_{21} + p_{22} \cdot \ln E_n + p_{23} \cdot \ln^2 E_n) \tag{2}$$

below about 70 MeV [24] , where σ_{nf} is the (n,f) fission cross-section (mb), E_n is the neutron energy (MeV) and p_{ij} are variable parameters. The parameters p_{ii} as functions of the parameter Z^2/A of the compound nuclei are available upon request.

Taking into account the uncertainties coming from the experimental technique and the cross-section standard we can state that the accuracy of the presented data is about 20% for most nuclei in the energy range above 75 MeV and 30-50% below 75 MeV. As the neutron energy and the target atomic number are decreased, the uncertainty is increased.

Figure 3. **Experimental data on the ^{209}Bi(n,f) cross-sections**



As it was mentioned above, the present status of the experimental data on the ^{209}Bi(n,f) cross-section is of particular interest, because it is a new standard in the energy region above 50 MeV. All experimental data available in the energy range of interest are given in Figure 3 along with the ^{209}Bi(n,f) cross-section parameterization from [4]. It is obvious that there is a discrepancy between the recommended parameterization and some experimental results published more recently [8,9]. Specifically, our data obtained at the neutron energies 96 MeV and 133 MeV (solid circles) are systematically lower than the recommended curve and do not fall into the confidence interval (10%) stated in [4]. To understand whether (or not) this discrepancy will eventually result in a recommendation to change the standard, a closer look at the quality of the experimental data must first be undertaken. The data under consideration have been obtained with the use of three fission fragment detectors: thin-film breakdown counters (TFBC), a conventional parallel plate ionization chamber (IC), and a Frisch-gridded ionization chamber (FGIC).

The TFBCs are insensitive to the background radiation and offer excellent timing characteristics [25]. Previous data obtained with the use of the TFBC [5,6] and the FGIC [7], together with earlier data [21,22] have been used as a basis for the parameterization, recommended as a standard [4]. However, recently it was found out that the decomposition procedures applied in [5,6,11] to the TOF spectra of fission events lacked accuracy at some neutron energy points. We suppose, that a more sophisticated background approximation (see e.g. [7]) has to be used to extract from TOF spectra a number of fission events induced by “peak” neutrons. Solid triangles in Figure 3 show our data from [9,11] corrected due to a more accurate decomposition procedure as well as data

obtained more recently. It is seen that these data deviate systematically from the parameterization [4] and become closer to our data obtained with the FGIC [8,9].

An ionization chamber (IC) is ideally suitable for (n,f) cross-section measurements at the currently available high-energy neutron beams, because this device offers nearly 100% detection efficiency with no limitations on the fissile target dimensions. It should be noted, however, that the energy spectrum of fission fragments is contaminated by light charged particles arising in upstream material from energetic neutrons. This background makes the determination of the fission fragment yield difficult. As the incident neutron energy increases, the situation becomes more complicated (particularly for sub-actinides) due to larger overlap between fission and background spectra. We suppose that data obtained with the simple ionization chamber [20] may be subject, especially at high neutron energies, to some systematic errors caused by the background problems.

The FGIC not only incorporates the main advantages of conventional parallel plate ionization chambers, but also offers some extra ones for (n,f) cross-section measurements. The key advantage is that FGIC allows discrimination against background charged particles [8]. The principle of so called angular discrimination lies in the fact that fission fragments and light charged particles give different combinations of anode and grid signals, and thus may be separated from each other by off-line processing. Taking also into account that an accurate decomposition procedure has been applied to the TOF spectra in [8,9], we consider the data obtained with the FGIC as the most reliable at present.

All aforesaid gives some grounds to expect changes of the standard in the future, but in the present work all data on (n,f) cross-sections for sub-actinides are given relative to the old standard.

3. Prospects for advancement of the existing experimental data base

Further development of the experimental techniques is needed to improve the quality of the data. This refers both to the characteristics of neutron beam and to the fission fragment detectors.

To increase the number of fissile targets to be irradiated simultaneously, the new FGIC has been designed and manufactured at the KRI within the framework of ISTC project #1309. The chamber consists of seven units. Each unit constitutes a twin Frisch-gridded ionization chamber with a common cathode. By this means 14 different targets may be irradiated simultaneously.

Due to the insensitivity of TFBCs to light ionizing particles it is possible to carry out the (p,f) and (n,f) cross-section measurements under comparable geometrical conditions. Experiments can be done with a broad proton beam passing through the target and the TFBC [26]. This makes it possible to reduce the uncertainty in the comparison analysis of data on the proton and neutron-induced fission cross-sections.

Combined analysis of (n,f) and (p,f) cross-sections is of special interest for studies of the fission process. The quantitative comparison of these cross-sections carried out in [13] revealed the following empirical dependence of the cross-section ratios on the Z^2/A parameter of the target nucleus:

$$\sigma_{pf}/\sigma_{nf} = \exp\,[k(37- Z^2/A)] \qquad (3)$$

where k is a function of energy ($k>0$ for $Z^2/A\leq 37$, and $k = 0$ for $Z^2/A>37$). This dependence was explained in terms of the fissility of the nucleus ($P_f = \sigma_{pf}/\sigma_{in}$) which is defined by the fission and evaporation widths: $P_f = \Gamma_f/\Gamma_f + \Gamma_n + \ldots$

Since for sub-actinides $\Gamma_f/\Gamma_n << 1$, one can obtain that:

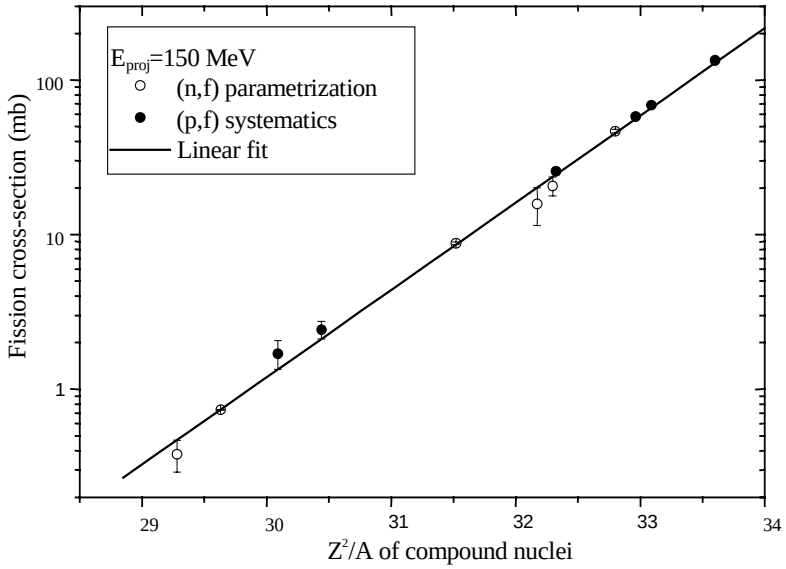
$$P_f \cong \exp(-(B_f\,(Z^2/A) - B_n)/\,T\,), \tag{4}$$

where B_f and B_n are the fission barrier and the neutron binding energy, respectively, and T is the nuclear temperature. For $P_{pf} = P_{nf}$ it is easy to verify that:

$$\sigma_{pf}\,(Z,A,E_p) = (\sigma_{in})_p/(\sigma_{in})_n\;\sigma_{nf}\,(Z+1,A,E_p), \tag{5}$$

provided that $E_p = E_n + B_n - B_p$. This relation is clearly seen in Figure 4, where both cross-sections are given (at 150 MeV) as functions of the Z^2/A parameter of the compound nucleus. One can see that all points follow a straight line giving added credence to the experimental data.

Figure 4. **The (p,f) and (n,f) cross-sections of sub-actinides nuclei vs Z^2/A parameter of compound nucleus**



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