

NUCLEAR DATA EVALUATION FOR JENDL **ACTINIDE** FILE
AND HIGH ENERGY DATA FILES

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ABSTRACT

Evaluation of nuclear data for JENDL **Actinide** File and High Energy Files is now in progress. JENDL **Actinide** File will contain evaluated nuclear data for 89 **nuclides** from ^{208}Po to ^{255}Fm including main **fissile** and fertile **nuclides**. The data are evaluated in the neutron energy region from 10^{-5}eV to 20 MeV, by widely using theoretical calculations and systematic of parameters and cross sections. Evaluation of high-energy neutron and charged particle nuclear data has been initiated to make high energy data files of **nuclides** related to an accelerator-driven **spallation** system and a neutron irradiation facility of **ESNIT**. So far, the evaluation of neutron and proton-induced reaction data of ^{27}Al , Pb and ^{209}Bi has been finished up to 1.0 GeV by mainly using the ALICE-F code and associated programs based on **systematics**. This paper describes their evaluation methods and status of these files.

I. INTRODUCTION

Management of high-level radioactive nuclear waste from nuclear reactors is one of the most important research subjects in the nuclear fuel cycle. In 1988, Japan decided to promote the OMEGA (**O**ptions Making Extra **G**ains of Actinides and fission products generated in nuclear fuel cycle) project. One of the possible options of radioactive waste management is to convert the long-lived radioactive **nuclides** to stable or short-lived ones by using nuclear transmutation reactions. Japan Atomic Energy Research Institute (**JAERI**) is investigating two ideas of such nuclear transmutation systems.

In the first process of the both ideas, the high-level radioactive waste is partitioned by chemical processing into 4 groups: minor **actinides** (**Np**, **Pu**, **Am**, **Cm**, etc.), **Tc-Platinum** group (**Tc**, **Ru**, **Rh**, **Pd**, etc.), **Sr-Cs** elements (**Sr** and **Cs**), and other **nuclides** (**Zr**, **Mo**, etc.). A current target of the nuclear transmutation is the minor **actinides** separated from the waste. An actinide burner fast reactor¹ is one of the ideas, in which the minor actinides are mainly transmuted by the fission, capture and **(n,2n)** reactions with a neutron spectrum harder than normal fast reactors. Another idea is an accelerator-driven **spallation** system² with a high intensity and high energy proton accelerator and a **sub-critical** assembly of minor **actinides**. In this system, the transmu-

tation is mainly caused by chain reactions in the sub-critical assembly with source neutrons generated by the **spallation** reactions with target materials such as tungsten, lead and bismuth.

For the actinide burner reactor, in addition to the nuclear data of main **fissile**, fertile and structural material **nuclides**, the data of minor **actinides** are important, in particular, their fission, capture and **(n,2n)** reaction cross sections, the number of neutrons per fission and fission product yield data for the neutrons below 20 MeV. Data of main actinide **nuclides** in available evaluated nuclear data libraries such as JENDL-3, **ENDF/B-VI**, JEF-2, are accurate enough in the energy range below 20 MeV. However, those of the minor **actinides** are neither accurate enough, nor given in the current evaluated nuclear data libraries.

As one of JENDL (Japanese Evaluated Nuclear Data Library) special purpose files, the JENDL **Actinide** File is being made to meet the data requirements for the **actinide** burner reactor, and to provide reliable nuclear data for estimation of amount of **nuclides** generated in the nuclear reactors. The evaluation work for the JENDL **Actinide** File is described in Chapter 11.

The research and development of the accelerator-driven **spallation** system need the proton- and neutron-incident nuclear data in the energy region up to a few GeV for structural material, **spallation** target and **nuclides** to be transmuted. In this energy region, no evaluated nuclear data is available, except for only a few **nuclides**. Necessary data are double differential cross sections (**DDXs**) for the emitted particles, isotope production cross sections and the data related to high energy fission. The **DDXs** are important to calculation of secondary interactions in the materials, particle transportation, shielding design, etc. The isotope production cross sections are necessary to estimate the residual radioactivity.

In addition, the project of Energy Selective Neutron Irradiation Test Facility (**ESNIT**)³ is under way in **JAERI**, which is an **FMIT** type d-Li neutron source. For design of **ESNIT**, the nuclear data of structural materials are required up to 50 MeV because of the high energy tail of d-Li neutrons.

High energy data files are being planned to provide the data for **ESNIT** and the accelerator-driven **spallation** system. The file for **ESNIT** will contain the neutron-induced reaction data up to

50 MeV. The other file for the accelerator-driven spallation system provides the neutron- and proton-induced data of the above-mentioned quantities of all the kinds of nuclides needed such as structural materials and candidates of spallation targets in the energy region up to 1.5 GeV. Chapter 111 is devoted to description of the high-energy data files.

II. EVALUATION FOR JENDL ACTINIDE FILE

The JENDL Actinide File will contain the evaluated data for 89 nuclides from ^{208}Tl to ^{255}Fm , in the neutron energy region from 10^{-5} eV to 20 MeV. The nuclides were selected on the basis of the following considerations:

- 1) nuclides from Th to Es with a half-life longer than 1 day.
- 2) nuclides on the 4 major decay paths of actinides, with a half-life longer than 1 day.
- 3) nuclides whose data are already stored in JENDL-3.
- 4) some exceptional nuclides.

Table 1 lists the 89 nuclides to be stored in the JENDL Actinide File. The nuclides having high priority in the actinide burner reactor are marked with "A" or "B". "A" indicates major actinides, and "B" important minor actinides for design of the actinide burner reactor. The data for 57 nuclides marked with "J3" exist in JENDL-3⁴. Data of the nuclides with "x" are not given in JENDL-3, ENDF/B-VI nor JEF-2. The evaluation of the minor actinides for JENDL-3 has been reported elsewhere.^{5,6} Data for ^{237}U , ^{236}Np and ^{238}Np have been evaluated recently. Other evaluation will be completed by about 1997.

The data of the 7 nuclides with "A" will be taken from JENDL-3.2, which is the second revision of JENDL-3, without any modification. Those for the 13 nuclides of "B" will be based on JENDL-3.2 and some modifications will be made if needed. For the other minor actinides, the data of JENDL-3.2 will be taken or new evaluation work will be made.

In the following sections, the evaluation work made so far for minor actinides is summarized, and as examples, the recently performed evaluation for ^{237}U , ^{236}Np and ^{238}Np is described.

A. Parameter for Theoretical Calculation

For the minor actinides, theoretical calculation is more important for the evaluation work because experimental data are so scarce. Theoretical calculation of cross sections and angular distributions of secondary neutrons were performed with a spherical optical and statistical model code CASTHY⁷ for most of minor actinides.

Optical Potential Parameters

Potential parameters most commonly used are as follows:

	Set 1	Set 2	Set 3
V	43.4-0.107E	$V_0-0.05E$	41.0-0.05E
W_s	6.95 -0.339E	6.5+0.15E	6.4+0.15√E
	+0.0531E ²		
V_o	7.0	7.0	7.0
r, r_o	1.282	1.32	1.31
r_s	1.290	1.38	1.38
a, a_o	0.60	0.47	0.47
b	0.5	0.47	0.47

Their units are MeV for energy and fm for length. The parameters (Set 1) were determined so as to reproduce the total cross section of ^{241}Am ⁸, and used to nuclides heavier than Am. Some alternatives were adopted for several nuclides. The parameters (Set 2) determined in Ref. 9, and those (Set 3) by Ohsawa and Ohta¹⁰ were used for U to Pu and nuclides lighter than Pa, respectively. The constant term of real potential (V_0) of Set 2 is determined for each nucleus so that the calculation might reproduce the s-wave strength function estimated from experiments or systematic.

Level Density Parameters

A composite level density formula of Gilbert and Cameron¹¹ was adopted. The parameters were adopted from Ref. 10 or determined so as to be consistent with average resonance level spacing¹² and the number of low-lying excited levels¹³.

Level Scheme

Nuclear level scheme for inelastic scattering of each nucleus was determined on the basis of ENSDF¹³ or the Nuclear Data Sheets.

Table 1 Nuclides in JENDL Actinide File

nuclide	status	nuclide	status	nuclide	status
^{208}Tl	x	^{210}Pb	x	^{210}Bi	x
^{210}Po	x	^{222}Rn	x	^{223}Ra	J3
^{224}Ra	J3	^{225}Ra	J3	^{226}Ra	J3
^{228}Ra	x	^{225}Ac	J3	^{226}Ac	J3
^{227}Ac	J3	^{227}Th	J3	^{228}Th	J3
^{229}Th	J3	^{230}Th	J3	^{231}Th	x
^{232}Th	J3,A	^{233}Th	J3	^{234}Th	J3
^{229}Pa	x	^{230}Pa	x	^{231}Pa	J3
^{232}Pa	J3	^{233}Pa	J3	^{230}U	x
^{231}U	x	^{232}U	J3	^{233}U	J3,A
^{234}U	J3	^{235}U	J3,A	^{236}U	J3
^{237}U	New	^{238}U	J3,A	^{234}Np	x
^{235}Pu	x	^{236}Np	x	^{237}Np	J3,B
^{238}Pu	New,B	^{239}Np	J3	^{236}Pu	J3
^{237}Pu		^{238}Pu	J3,B	^{239}Pu	J3,A
^{240}Pu	J3,A	^{241}Pu	J3,A	^{242}Pu	J3,B
^{244}Pu		^{246}Pu	x	^{247}Pu	x
^{241}Am	J3,B	^{242}Am	J3,B	$^{242\text{m}}\text{Am}$	J3,B
^{243}Am	J3,B	^{244}Am	J3	$^{244\text{m}}\text{Am}$	J3
^{240}Cm	x	^{241}Cm	J3	^{242}Cm	J3,B
^{243}Cm	J3,B	^{244}Cm	J3,B	^{245}Cm	J3,B
^{246}Cm	J3,B	^{247}Cm	J3	^{248}Cm	J3
^{249}Cm	J3	^{250}Cm	J3	^{245}Bk	x
^{246}Bk	x	^{247}Bk	x	^{248}Bk	x
^{249}Bk	J3	^{250}Bk	J3	^{246}Cf	x
^{248}Cf	x	^{249}Cf	J3	^{250}Cf	J3
^{251}Cf	J3	^{252}Cf	J3	^{253}Cf	J3
^{254}Cf	J3	^{251}Es	x	^{252}Es	x
^{253}Es		^{254}Es	J3	$^{254\text{m}}\text{Es}$	x
^{255}Es	J3	^{255}Fm	J3		

J3: data exist in JENDL-3, New: new evaluation for JENDL Actinide File has been completed, x: data are not existing in JENDL-3, ENDF/B-VI nor JEF-2, A: major actinide, B: important for the actinide burner reactor.

B. Resonance Parameters

In the low energy region, resolved resonance parameters were given on the basis of available experimental results. The parameters were adjusted so as to reproduce the thermal cross sections and resonance integral. Unresolved resonance parameters were determined with ASREP¹⁴ by fitting the fission and capture cross sections up to the energy around 30 keV.

C. Cross Sections

The fission cross sections were evaluated mainly on the basis of experimental data if available. If no experimental data were available, some systematics¹⁵ were used to estimate the cross section in the MeV region.

The (n,2n), (n,3n) and (n,4n) reaction cross sections for the minor actinides have not been measured. They were calculated with the simple evaporation model of Pearlstein¹⁶ or Segev and Caner.¹⁷

Calculation of the total, elastic and inelastic scattering and capture cross sections was made with CASTHY⁷. The fission and (n,Xn) reaction cross sections were considered as competing processes to the decay of the compound nucleus. The γ -ray strength functions were determined with normalizing the capture cross section to the measured one in the 100-keV region.

D. Other Quantities

Angular distributions of elastically and inelastically scattered neutrons were also calculated with CASTHY.

Howerton's semi-empirical formula¹⁸ was adopted for the number of prompt neutrons per fission, and Tuttle's systematics¹⁹ for delayed neutrons. If the measured data were available, they were adopted.

Evaporation spectra were given for the neutrons due to inelastic scattering to the continuum region and (n,Xn) reactions. The nuclear temperature was estimated from the level density parameters. For fission neutrons, evaporation spectra were also adopted with temperature mainly based on the systematic by Smith et al.²⁰

E. Evaluation of Uranium-237 Data

Resonance parameters from 46 to 200 eV were determined from the fission area data measured by McNally et al.²¹ They measured the ²³⁷U fission cross sections by using neutrons from an underground nuclear explosion (Pommard). Below 46 eV, hypothetical levels were generated by assuming level spacing of 3.5 eV²¹, the s-wave strength function of 1×10^{-4} , Γ_γ of 35 meV and Γ_f of 4 meV obtained from the data of McNally et al.²¹ Parameters of a negative level and low-lying levels were adjusted so as to reproduce the thermal fission cross section of about 2 b²², the absorption cross section of 478 ± 160 b²³ and capture resonance integral of 1200 ± 200 b²⁴. An effective scattering radius of 9.38 fm was obtained from the optical model calculation. Unresolved resonance parameters were determined on the basis of cross sections up to 30 keV.

The fission cross section in the MeV region was based on a calculation with a simple formula by Bychkov et al.²⁵, experimental data by Cramer and Britt²⁶ and systematic by Behrens and Howerton.¹⁵ The fission cross section from 200 eV to 100

keV was interpolated from 2.5 b at 200 eV and 0.6 b at 100 keV.

The (n,2n) and (n,3n) reaction cross sections were calculated with the formula by Segev and Caner¹⁷ by considering the fission cross section determined above. The (n,4n) reaction was ignored. Therefore the present result of the (n,3n) cross section above 17 MeV could be too large.

The total, elastic and inelastic scattering and capture cross sections were calculated with CASTHY. The optical potential of Set 2 was adopted with VO of 40.5 MeV that reproduces the s-wave strength function of 1×10^{-4} . The average level spacing of 3.5 eV²¹ and Γ_γ of 35 meV were assumed. The fission, (n,2n) and (n,3n) reaction cross sections described above were taken into account as competing processes to the decay of compound nucleus of ²³⁸U.

The fission cross section is compared in Fig. 1 with ENDF/B-VI.²⁷ Below 200 eV, the cross sections in the figure are mean values in suitable energy intervals. The cross section is largely discrepant in the resonance region because the present evaluation adopted the experimental results of McNally et al., whereas ENDF/B-VI are not based on the experimental data but on hypothetical resonances. According to our experience, the fission cross sections measured with underground nuclear explosions are often to be too large. The data of McNally et al. also might be too large but we have no other experimental data for this cross section. The other cross sections are shown in Fig. 2.

The thermal cross sections and resonance integral are listed in Table 2. Large discrepancies are found in the resonance integral between the present results and ENDF/B-VI.

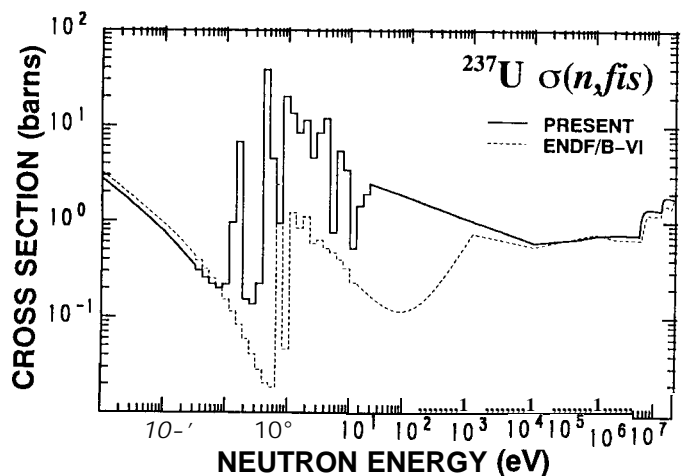


Fig. 1 The fission cross section of ²³⁷U. Data below 200 eV are mean values in energy bins.

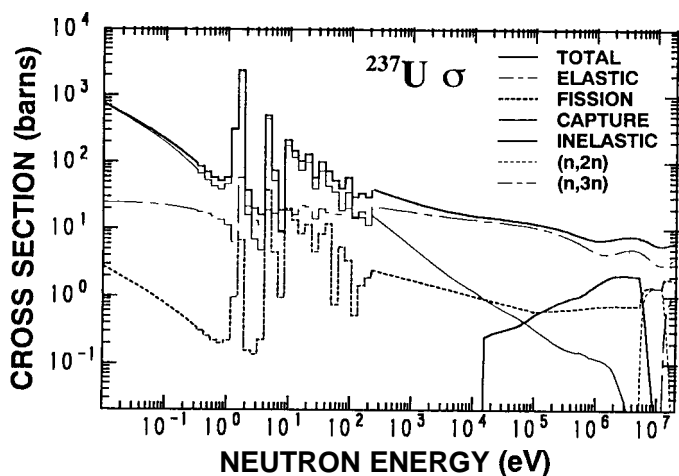


Fig. 2 Cross sections of ^{237}U . Data below 200 eV are mean values in energy bins.

Table 2 Thermal cross sections and resonance integral of ^{237}U

2200m/sec a(b)	σ_{cap}	σ_{fis}	σ_{ela}	σ_{tot}
present	452.4	1.70	24.39	478.5
ENDF/B-VI	476.0	2.00	9.13	487.1
Resonance integral (b)				
present	1080	σ_{fis}		48.7
ENDF/B-VI	311	9.22		

F. Evaluation of Neptunium-236 and -238 Data

No resolved resonance parameters have been measured for these two nuclides. Since the level spacings (0.11 eV for ^{236}Np , 0.29 eV for ^{238}Np) estimated from their level densities are very small compared with average fission widths (about 0.4 eV²⁸ for ^{236}Np), strong overlapping of resonances can be expected. Therefore no resonance parameters were given to these two isotopes.

The fission cross section of ^{236}Np was based on the data measured by Val'skiy et al.²⁸ below 20 keV. The capture cross section in this energy region was determined by assuming the capture-to-fission ratio of 0.253 that was estimated from the CASTHY calculation at 20 keV. The fission cross section of ^{238}Np was assumed to be in the form of $1/v$ and 2070 b²⁹ at 0.0253 eV. The capture cross section was calculated with CASTHY.

The fission and (n,Xn) reaction cross sections above 20 keV were estimated with the same method as ^{237}U . Other cross sections were calculated with CASTHY by using the same optical potential parameters ^{237}U . The γ -ray strength functions were obtained from Γ_{γ} of 35 meV and the above-mentioned level spacing.

The thermal cross sections and resonance integral are listed in Table 3. The cross sections of ^{236}Np and ^{238}Np are shown in Figs. 3 and 4.

Table 3 Thermal cross sections and resonance integral of ^{236}Np and ^{238}Np

2200m/sec σ (b)	^{236}Np		^{238}Np	
	σ_{cap}	σ_{fis}	σ_{cap}	σ_{fis}
present	701	2770	450	2070
ENDF/B-VI			202.8	2026.9
Res. integ(b)				
present	σ_{cap}	σ_{fis}	σ_{cap}	σ_{fis}
ENDF/B-VI	259	1030	100	898

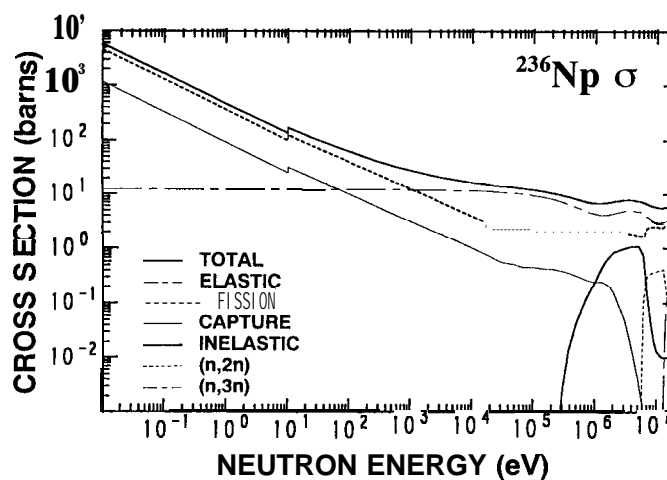


Fig. 3 Cross sections of ^{236}Np .

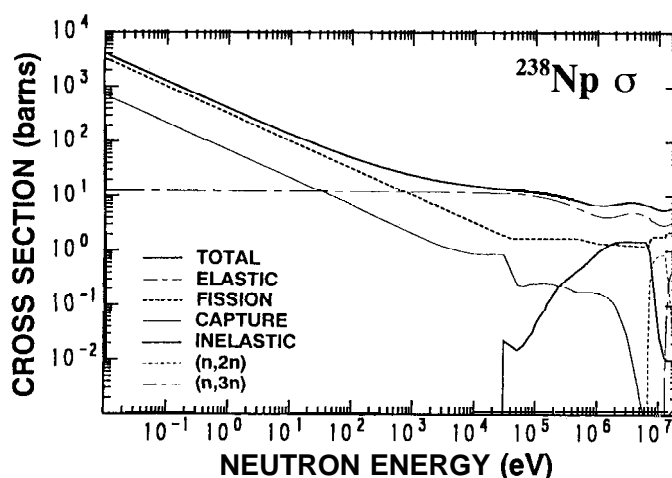


Fig. 4 Cross sections of ^{238}Np .

G. Future Work for Class B Nuclides

Among the class B nuclides, ^{237}Np , ^{238}Pu , ^{241}Am and ^{243}Am are the main fissile nuclides which sustain the chain reactions in the actinide burner reactor. Hence more accurate data are required for the fission and capture cross sections, ν -values and delayed neutron data. Particularly, the inventories of ^{237}Np and

²³⁸Pu are large, and their inelastic scattering cross sections affect the reactor spectrum. The direct inelastic scattering should be taken into account for them.

IH. EVALUATION FOR HIGH ENERGY DATA FILES

Two high energy data files will be produced; one includes the neutron nuclear data in the energy range up to 50 MeV and another file the neutron- and proton-induced reaction data up to 1.5 GeV. The file up to 50 MeV will supply the neutron data for the ESNIT project of JAERI. In this energy range, the evaluation method similar to the evaluation below 20 MeV can be adopted. The latter file up to 1.5 GeV is planned to support the design of the accelerator-driven spallation system. The data provided in this file are those of structural materials of the system, candidates of the spallation targets (W, Pb, Bi), and nuclides to be transmuted. These two files will be compiled in the ENDF-6 format. In this Chapter, the typical method of the nuclear data evaluation for the file up to 1.5 GeV is explained using the results of evaluations for ²⁷Al, Pb and ²⁰⁹Bi up to 1.0 GeV.

A. Total and Reaction Cross Sections

Total and total reaction cross sections are basic quantities of nuclear data evaluation. They can be obtained by optical model calculation with a global potential derived by Pearlstein or estimated from Pearlstein's systematic.³⁰ The latter was mainly used in the evaluation of Al, Pb and Bi. The evaluated result of neutron total cross section for ²⁰⁸Pb with Pearlstein's systematic is compared with experimental data in Fig. 5. The evaluated result reproduces the overall trend of the experimental data well.

B. Isotope Production Cross Sections

The isotope production cross sections are the total residual nuclide production cross sections integrated over all channels of reactions. They were mainly evaluated by using a multi-step compound decay model code with precompound correction, ALICE-F,³¹ which is a modified version of ALICE/89.³² The main improvements from ALICE/89 are the following

- 1) The preequilibrium cluster particle emission is considered for d, t, ³He and α with Iwamoto-Harada cluster formation model^{33,34} in order to reproduce spectra in the high energy region of the emitted cluster particles.
- 2) The latest mass formula derived by Tachibana et al.³⁵ is added for binding energy calculation.
- 3) The γ-ray incident reaction can be treated.

The calculated result of the ²⁷Al(p,x)²⁴Na reaction cross section is shown in Fig. 6 with the experimental data. Although the calculation was performed with the default parameters of ALICE-F, the result follows the trend of experimental data.

C. Double Differential Particle Emission Cross Sections

The DDXS were basically evaluated with the semi-empirical formulas which were derived by Pearlstein for neutrons³⁶ and by Kalbach for charged particles.³⁷ Kalbach's formula needs angle-integrated particle spectra and the ratio of the component for multi-step direct process to that for total emission. They were mainly calculated by the ALICE-F code.

D. Fission Cross Section

For heavier elements than silver, the fission cross section is

not negligible in the energy region above a few hundred MeV. The proton-induced fission cross section was obtained by the following empirical formula derived by Fukahori:³⁸

$$\sigma_{fis}[mb] = PI \times (1 - \exp(-P2 \times (E_p - P3))), \quad (1)$$

where

$$\begin{aligned} PI &= Y \times \exp(0.575637 \times X - 1.72680), \\ P2 &= Y \times \exp(-0.456190 \times X + 1.52102), \\ P3 &= Y \times \exp(0.549152 \times X - 0.194530) \times 0.001, \end{aligned} \quad (2)$$

and $X = Z^2/A$, $Y = A^{2/3}$, E_p is the incident proton energy in MeV, Z and A the atomic and mass numbers of the target nuclides. For the neutron-induced fission, the cross section is estimated to be a half of Eq. (1) as a result of comparison with several sets of experimental data. The calculated result of proton-induced fission cross section for ²⁰⁹Bi is compared with the experimental data in Fig. 7.

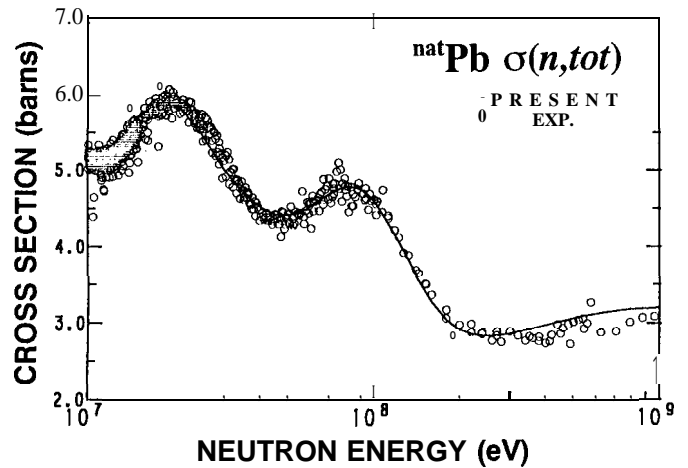


Fig. 5 The total cross section of ²⁰⁸Pb.

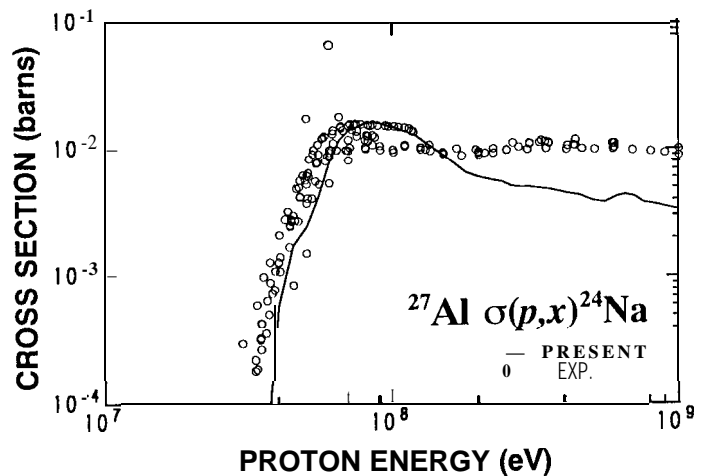


Fig. 6 The ²⁷Al(p,x)²⁴Na reaction cross section.

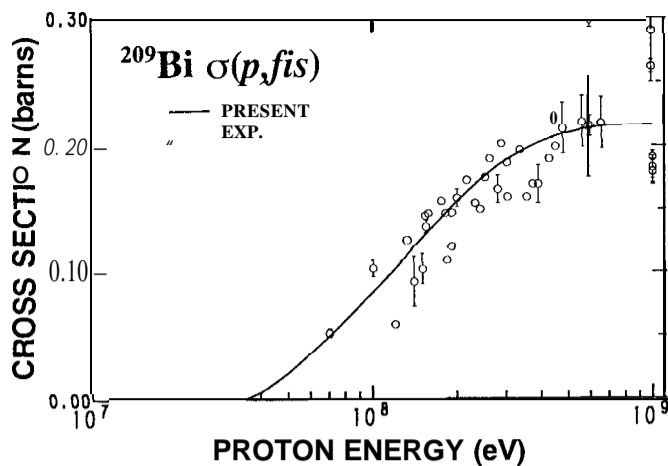


Fig. 7 The fission cross section of ^{209}Bi .

E. New Evaluation Methods for High Energy Files

The examples of data evaluation in the intermediate energy region outlined above is mainly based on the **phenomenological** models. However, these methods are not free from a lot of adjustable parameters, and therefore reliability of their results is not certain, especially when they are applied to the energy region where the parameters have not been examined. Moreover, they do not consider the final channels including n-meson and other elementary particles, **and such exotic reactions as multi-fragmentation are not treated explicitly. In order to solve** these problems, new techniques, based on more microscopic description of the nuclear process, will be investigated and developed which are going to readopted in the evaluation for the file up to 1.5 GeV.

For example, the nucleon total, elastic and reaction cross sections are known to be well described by the relativistic impulse approximation (RIA)³⁹ in the energy region above around 100 MeV. Figure 8 gives the total and reaction cross sections of Pb, where the solid curves give the predictions with RIA. The RIA was proved to be a powerful tool to give a consistent description of the total, elastic (including angular distribution) and reaction cross sections in this energy region. The reaction cross section calculated in this way can be then used to normalize the results obtained by other methods, such as those described below.

The reaction part of the cross section can be recalculated by a microscopic simulation method such as the cascade model⁴⁰ and Quantum Molecular Dynamics (QMD).⁴¹ These methods are considered to be vital tools in calculating the cross section in the intermediate energy region because of their flexibility in including N-Nonelastic channels, fission, dynamical fragmentation, and so on. In this approach, an extension of the NMTC/JAERI code⁴² is being planned; 1) it will be rewritten to a full cascade program, 2) cross sections of the basic nucleon-nucleon reaction will be updated by taking account of the present phase-shift analysis, and 3) number of inelastic channels will be increased. Next, the effects of the nuclear mean field will be taking account according to the concept of QMD; a nucleon will be expressed by a Gaussian wave packet. Between two nucleon-nucleon collision, it will move according to the Newtonian equation. In this way,

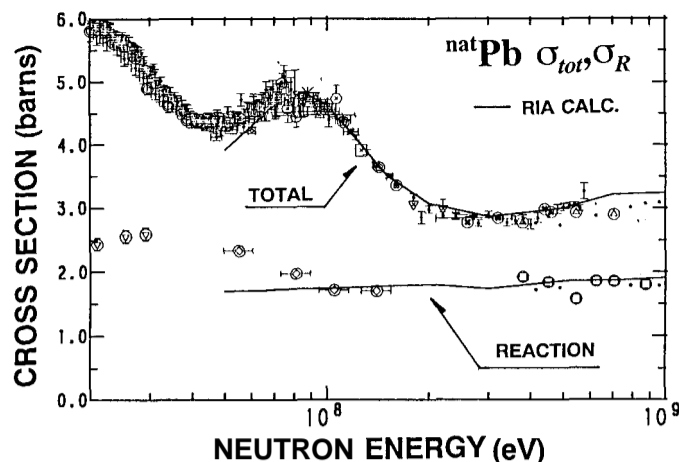


Fig. 8 The Pb total and reaction cross sections calculated with RIA.

the nucleon motion and nuclear mean field can be calculated in the self-consistent manner. By normalizing the event number versus total history of QMD to the reaction cross section calculated by RIA, the reaction can be described in a fully microscopic way. This approach will be applied in the evaluation work for the high energy file up to 1.5 GeV.

IV. CONCLUDING REMARKS

The present status and plans of the JENDL Actinide File and high energy files were shortly described. The JENDL Actinide File will provide the neutron-induced reaction data for 89 nuclides from ^{208}Tl to ^{253}Fm in the energy range from 10^5eV to 20 MeV. The evaluation for 61 nuclides out of 89 has been finished. We have to make new evaluations for 28 nuclides. For lack of experimental data for these minor actinides, the evaluation of large part of the data will be based on calculations. Advanced methods will be investigated for this. Furthermore, some modifications of existing evaluated data will be also made especially for important nuclides.

The high energy files will store the data needed for ESNIT and for the accelerator-driven spallation system. Examples of typical evaluation method and results for the file up to 1.5 GeV were shown in this paper. The evaluation for the high energy data files will be made with the method described in Sections A to D of Chapter III. Reliability of these methods were confirmed. However, for most of nuclides, available experimental data are not enough for the evaluation and for checking reliability of the evaluation methods. Therefore more careful theoretical calculations are desirable. The method given in Section E of Chapter III will be also applied to the evaluation.

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