

# **INDC International Nuclear Data Committee**

Summary Report

First Research Coordination Meeting on

# **Prompt Fission Neutron Spectra of Major Actinides**

IAEA Headquarters Vienna, Austria

6-9 April 2010

Prepared by

R. Capote Noy IAEA Nuclear Data Section Vienna, Austria

December 2010

IAEA Nuclear Data Section, Vienna International Centre, A-1400 Vienna, Austria

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Produced by the IAEA in Austria

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# Abstract

A summary is given of the First Research Coordination Meeting on *Prompt Fission Neutron Spectra of Actinides*. Experimental data and modelling methods on prompt fission neutron spectra were reviewed. The programme to compile and evaluate prompt fission spectra including uncertainty information over the neutron energy range from thermal to 20 MeV was proposed. Validation of the resulting data against integral critical assembly and dosimetry data is foreseen. Detailed coordinated research proposals have been agreed. Summary reports of technical presentations at the meeting are given. The resulting work plan of the Coordinated Research Programme is summarized, along with actions and deadlines.

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#### 1. Introduction

The energy spectrum of prompt neutrons emitted in fission plays an important role in many applications in nuclear science. In particular, accurate predictions of nuclear criticality using neutron transport codes are dependent on the underlying nuclear data, especially the fission spectrum. The high sensitivity of calculated quantities to fission data has been recently emphasized by researchers in many groups around the world who are working on conventional as well as advanced reactors, and non-proliferation applications.

While the accuracy of fission cross sections and neutron multiplicities (nubar) in the relevant energy range have been steadily improved, we are faced with the situation that existing measured prompt fission neutron spectra (PFNS) are in many cases discrepant, and that different PFNS theoretical models give differing predictions.

In November 2008, a Consultants' Meeting (CM) was organised in Vienna to review and discuss the adequacy and quality of the recommended prompt fission neutron spectra to be found in existing nuclear data applications libraries<sup>1</sup>. These prompt fission neutron spectra were judged to be inadequate. Therefore, the meeting participants strongly recommended initiating a new IAEA Coordinated Research Project (CRP) on prompt fission neutron spectra evaluations. The proposed goal was to determine the prompt fission neutron spectra and covariance matrices for actinides in the energy range from thermal to 20 MeV, including validation against integral critical assembly (k-eff) and dosimetry data. The following nuclei were recommended for study in the following order of priority: - major actinides <sup>235,238</sup>U and <sup>239</sup>Pu;

- <sup>232</sup>Th and <sup>233</sup>U of relevance to the Th-U fuel cycle; \_
- minor actinides such as <sup>237</sup>Np, <sup>241</sup>Am, <sup>242m</sup>Am, <sup>240</sup>Pu and <sup>245</sup>Cm.

The Coordinated Research Project (CRP) named "Prompt Fission Neutron Spectra for Actinides' began in 2010. Its first Research Coordination Meeting (RCM) was held at IAEA Headquarters, Vienna, Austria from 6 to 9 April 2010 and was attended by twelve CRP participants. The IAEA was represented by R. Forrest, N. Otsuka and R. Capote, who served as Scientific Secretary, F.-J. Hambsch (EC-JRC-IRMM) was elected Chairman of the meeting and P. Talou from (LANL, USA) agreed to act as rapporteur. The approved Agenda is attached as Appendix 1 and the list of participants and their affiliations as Appendix 2.

Following the recommendations of the CM prior to this meeting, the assignment of tasks was discussed by e-mail between the participants and Scientific Secretary. Primary aims of this meeting were to discuss scientific and technical matters related to the subject, coordinate related tasks, and to assess assigned responsibilities and deadlines.

The Director of the IAEA Division of Physical and Chemical Sciences, N. Ramamoorthy, welcomed the participants and emphasized the significance of their role in the improvement of the prompt fission neutron data for reactor applications. R. Capote (IAEA-NDS Project Officer of the CRP) summarized the research objectives and expected outputs of the CRP as outlined in the preceding CM. The following outcomes are expected from the proposed research project:

<sup>&</sup>lt;sup>1</sup> R. Capote, V. Maslov, E. Bauge. T. Ohsawa, A. Vorobyev, M.B. Chadwick and S. Oberstedt, Summary Report of Consultants' Meeting on Prompt Fission Neutron Spectra of Major Actinides, INDC(NDS)-0541 (IAEA, Vienna, Austria, January 2009)

- Make available new evaluations of prompt fission neutron spectra and covariance matrices for actinides in the energy range from thermal to 20 MeV, including validation against integral critical assembly (k-eff) and dosimetry data on the web.
- Publication of a comprehensive technical document in the IAEA TRS series.

Summary reports of presentations by meeting participants including relevant figures are attached as Appendix C.

The actions to be undertaken prior to the next RCM to be held in winter 2011 were agreed together with their relative time-schedule and deadlines (default deadline for all actions is the next RCM, if not otherwise stated). The assigned actions together with deadlines and recommendations as agreed by all CRP participants are summarized below.

# 2. Experimental data

- 1. Establish web site with upload capabilities [Capote] (05/2010)
- 2. Upload presentations of 1<sup>st</sup> meeting [Capote]
- 3. Establish a list of experimental data sets, with numerical values, to be used in this CRP [coordinated by Hambsch]
  - a. Upload <sup>235</sup>U, <sup>239</sup>PuStaples data, original and corrected [Kornilov] (06/2010)
  - b. Upload <sup>235</sup>U Johansson data, original and corrected [Kornilov] (06/2010)
  - c. Include new data sets from IRMM [Kornilov] (06/2010)
  - d. Include new data from PNPI [Vorobyev] (06/2010)
  - e. Upload Starostov data, original and corrected [Maslov, Pronyaev] (06/2010)
  - f.  $^{232}$ Th and 238U data at ~2-3 MeV [Ganesan]
  - g. <sup>239</sup>Pu data at 1-50 MeV; Th232? [Granier]
  - h. New <sup>238</sup>U and <sup>237</sup>Np data at 6-7 MeV? [Granier]
- 4. Establish list of available experimental data of PFNS for all actinide isotopes of relevance to advanced reactors [Ganesan]
- 5. Preliminary estimates of uncertainties, and documentation (check list) of sources of uncertainties [Hambsch, Vorobyev] (12/2010)
- 6. Inter-comparisons between Kornilov and Vorobyev data sets for new thermal measurements on <sup>235</sup>U [Kornilov, Vorobyev] (06/2011)
- 7. Provide numerical values for <sup>252</sup>Cf standard spectrum (Mannhart), 2 versions (pointwise, and smoothed) [Capote] (06/2010)
- Provide updated dosimetry reaction evaluations, including covariance matrices, for ~60 reactions. Provide technical reports INDC(NDS)-0526 and 0546. [Capote] (12/2010)
- 9. Provide recommended experimental values for spectrum average cross sections in <sup>235</sup>U and <sup>252</sup>Cf spectra [Capote] (12/2010)
- 10. Compile Indian published data on PFNS into EXFOR [Ganesan]
- 11. Clean-up EXFOR entries for PFNS, and study improvements over existing format to include additional, multi-parametric data such as <nu>(A,TKE) [IAEA NDS]

# 3. Theory & Modeling

[Ohsawa, Tudora, Maslov, Talou, Shu, Granier]

- 1. Perform sensitivity studies on model parameters for all models used in the CRP.
- 2. Document all input parameters (TKE, fission probabilities, optical model, preequilibrium, anisotropy, ...) entering in model calculations, and provide ranges of reasonable values.
- 3. Cases studied: 7 and 14 MeV for <sup>238</sup>U, <sup>239</sup>Pu and <sup>232</sup>Th; thermal and 0.5 MeV for <sup>235</sup>U and <sup>239</sup>Pu.

- 4. Provide tables of numerical values of calculated PFNS, from 1 keV to 20 MeV outgoing energies. (09/2011)
- 5. Calculate spectrum-averaged cross sections (for provided <sup>235</sup>U thermal PFNS) for selected reactions. [Capote]

# 4. Benchmarking

- 1. Provide several energy group structures to be used for processing. [Manturov] (06/2010)
- Provide <sup>239</sup>Pu, <sup>240</sup>Pu PFNS (MF5, MT18) and PFNS covariance matrices (MF35, MT18) and uncertainty on <Eout> at 0.5 MeV [Talou] (06/2010)
- 3. Provide n(0.5 MeV) + <sup>239</sup>Pu and <sup>235</sup>U PFNS covariance matrix and uncertainty on <Eout> using Kornilov model [Kodeli] (06/2010)
- 4. Process available covariance matrices with NJOY [Kodeli] (08/2010)
- Provide <sup>235</sup>U PFNS covariance matrices (MF35) and uncertainty on <Eout> at 0.5 MeV. [Talou]
- 6. Establish list of benchmarks to be used (a few thermal, most fast) [Kodeli, Manturov]
- 7. Perform sensitivity calculations for all selected benchmarks [Kodeli, Manturov]
- 8. Collect available <sup>237</sup>Np criticality benchmarks [Manturov, Ganesan, Talou] (12/2010)
- 9. Provide new evaluation of <sup>237</sup>Np PFNS [Maslov] (06/2010)
- 10. Perform sensitivity calculation using Maslov's <sup>237</sup>Np PFNS evaluation [Manturov, Kodeli, Ganesan]

# 5. Conclusions

Presentations and discussions during the meeting showed a good start of the work needed for the CRP. Further extensive work needs to be done in the next 15 months so that the necessary progress can be achieved before the next RCM. A truly co-ordinated programme of work was agreed among the participants, leading to several additional actions to be undertaken. Issues related to the creation of a consistent set of experimental data as well as the physics to be considered in employed models were extensively debated. The expected output of the CRP is going to be an updated set of PFNS evaluations for major and selected minor actinides.



# 1<sup>st</sup> Research Coordination Meeting on "Prompt Fission Neutron Spectra of Actinides"

IAEA Headquarters, Vienna, Austria 6 – 9 April 2010 Meeting Room A0742

# AGENDA

# Tuesday, 6 April

08:30 - 09:30	Registration (IAEA Registration desk, Gate 1)				
09:30 - 10:00	Opening Session				
	Welcoming address – N. Ramamoorthy, Director NAPC				
	Introductory Remarks – Roberto Capote Noy				
	Election of Chairman and Rapporteur				
	Adoption of Agenda				
10:00 - 10:45	Administrative and Financial Matters related to participants, Coffee break				
10:45 - 12:30	Session 1: Discussion of Research Proposals and expected outputs				
	Experimental data:				
	T Granier, A Vorobyev, N Kornilov, F-J Hambsch				
10 00 11 00	<b>•</b> •				

# 12:30 – 14:00 Lunch

 14:00 – 18:00 Session 1 (cont'd): Discussion of Research Proposals and expected outputs *Coffee break as needed* 
 Experimental data (continued) T Granier, A Vorobyev, N Kornilov, F-J Hambsch, other contributors

# Wednesday, 7 April

09:00 - 12:30	Session 2: Discussion of Research Proposals and expected outputs					
	Modelling and uncertainty estimates:					
	T Ohsawa, V Maslov, A Tudora, P Talou, Shu Nengchuan					
12:30 - 14:00	Lunch					
14:00 - 18:00	Session 2 (cont'd): Discussion of Research Proposals and expected outputs					
	Coffee break as needed					
	Modelling and uncertainty estimates:					
	T Ohsawa, V Maslov, A Tudora, P Talou, Shu Nengchuan, other contributors					
19:00	Dinner at a restaurant in the city					

# Thursday, 8 April

09:00 - 12:30	Session 3: Discussion of Research Proposals and expected outputs				
	Coffee break as needed				
	Benchmark performance:				
	S Ganesan, G Manturov, I Kodeli, other contributors				

12:30 - 14:00 Lunch

14:00 – 18:00 Session 4: Coordination of required work, outputs, formatting Coffee break as needed

# Friday, 9 April

**09:00 - 12:30 Drafting of the Summary Report of the Meeting** *Coffee break as needed* 

12:30 - 14:00 Lunch

14:00 – 17:30 Review and Approval of the Summary Report Closing of the Meeting



# First Research Coordination Meeting on "Prompt Fission Neutron Spectra of Actinides"

IAEA Headquarters, Vienna, Austria 6-9 April 2010

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# 1. Experimental and theoretical investigations on prompt neutron spectra and their angular distributions for selected major and minor actinides<sup>2</sup>

R. K. Choudhury, A. Saxena, B. K. Nayak, Devesh Raj, Anek Kumar, H. Naik and S. Ganesan Bhabha Atomic Research Centre, Trombay, Mumbai-400085, INDIA

**Introduction:** The present write-up briefly discusses the BARC Research Proposal and expected outputs under the above-mentioned project as part of new IAEA-CRP: "Prompt fission neutron spectra (PFNS) of actinide nuclei" by the IAEA-NDS.

**Perspectives on the importance of this CRP to Indian programme:** The Indian nuclear energy programme and growth scenario are described in the official website: <u>www.dae.gov.in</u>. BARC/DAE is in the process of initiating the formation of a strong and sustainable Nuclear Data Physics Centre of India (NDPCI). An overview of progress and the current status of NDPCI are available in Ref. [1] and are not repeated here to save space. BARC considers this IAEA-CRP as important in the light of our current perspectives on nuclear data physics activities in India.

The uncertainties affect the design parameters of thermal, fast, fusion-fission hybrids and accelerator driven systems reactor designs. For instance, the uncertainty in criticality due to uncertainties in PFNS alone can affect criticality by about 50 pcm to a few hundred pcm (1 pcm =  $10^{-5} \Delta k/k$ ) depending upon the reactor spectra. Uncertainties in the knowledge of the PFNS nuclear data in the higher energy region result in larger uncertainties in each of the predicted activation rates caused by reactions with high neutron threshold energies. The PFNS data have larger uncertainties in the lower (less than about 0.1 MeV) and higher (greater than about 4.0 MeV) energy regions. The uncertainty in PFNS (as the initial fission neutron source) influences considerably the uncertainty in the estimate of the high energy tail of the reactor spectra above the (n, 2n) threshold, and in this way, influences the uncertainty in the estimated production rate, for instance, of <sup>232</sup>U, in thorium fuel cycle. The data available on PFNS in EXFOR data needs clean-up and improvements. The data in EXFOR for PFNS and other associated fission physics data are needed to be coded into EXFOR for applications in testing the fission physics model and nuclear data evaluations. There is a strong need to benchmark the PFNS data against available experimental integral benchmarks using error propagation methods employing covariance methodology.

Basic research facilities in BARC for use in this CRP: It may be noted that BARC has a long tradition of interest and experimental research programme in advanced fission physics studies. For example, the BARC team in the early sixties had performed [2, 3] several interesting and new studies in neutron induced fission of <sup>235</sup>U. In the nineties, the experimental work on fission physics was continued and, for instance, reported [4] in an IAEA Meeting. Presently, BARC is in final stages of setting up a new and advanced experimental facility for PFNS related work. This facility includes advanced neutron detectors and fission fragment detectors for carrying out high resolution neutron spectrum measurements using NE213 type detectors and silicon strip detectors. BARC has expertise in indigenous electronics modules and time-of-flight methodology. BARC has accelerator facilities (e.g., Pelletron [5] and FOTIA [6]) providing Li (p,n) based neutron source. Research reactors based (e.g., DHRUVA) neutron source facilities are in use for several applications. BARC also has some actinide targets, such as, <sup>232</sup>Th, <sup>238</sup>U, <sup>235</sup>U, <sup>233</sup>U and some minor actinide targets such as <sup>237</sup>Np, <sup>240</sup>Pu, <sup>241</sup>Am etc. **Proposal:** The proposed research activities to be performed under the IAEA-CRP over the next 3 to 4 years at BARC include conducting a detailed survey of available experimental and evaluated data of PFNS, surveying and digesting available theoretical studies on PNS, performing experiments with thermal as well as with a few MeV neutrons for selected actinide targets including <sup>232</sup>Th, <sup>238</sup>U, <sup>235</sup>U and <sup>233</sup>U and performing of sensitivity studies using available integral critical assembly and dosimetry data. As a first step, we have agreed to carry out measurements of PFNS for <sup>232</sup>Th and to compare with available data at 3 MeV, relative to <sup>235</sup>U. At BARC, the PFNS measurements will be carried out as ratio measurements with respect to the well established PFNS standard of <sup>252</sup>Cf, as accepted by the

<sup>&</sup>lt;sup>2</sup> Presented by S. Ganesan in the First Research Coordination meeting on "Prompt Fission Neutron Spectra of Actinides", 6-9 April 2010, IAEA Headquarters, Vienna, Austria. email: <u>ganesan@barc.gov.in</u>; Chief Scientific Investigator: R. K. Choudhury; email: <u>rkc@barc.gov.in</u>

IAEA. This proposed BARC nuclear data physics activity would complement other proposed programmes of various other countries, where extensive measurements of <sup>235</sup>U have been undertaken. BARC's contributions will also include performing calculations of inter-comparison of available and evaluated PFNS in ENDF/B files, such as, ENDF/B-VII.0, JENDL-4.0 etc., of various countries and performing our own reactor benchmark sensitivity studies to assess the influence of uncertainties in PFNS on reactor integral parameters using covariance matrix error propagation methodologies both for U-Pu and Th-U fuel cycles. BARC will also explore the possibility to compile and place in the IAEA-EXFOR database, as soon as possible, the missing numerical nuclear physics data of PFNS generated in early sixties [2, 3] and later by experiments conducted by BARC.

# **References:**

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- 5. 14 MV BARC-TIFR Pelletron Accelerator located at TIFR, Mumbai <u>http://www.tifr.res.in/~pell/</u>
- 6. P. Singh et al., "Status report on the folded tandem ion accelerator at BARC," Pramana, Vol. 59, 739, 2002.

## 2. Studies of the Prompt Fission Neutron Spectra: EC/JRC-IRMM

#### F.-J. Hambsch

## 1) Prompt fission neutron spectrum

The results of the prompt fission neutron spectrum (PFNS) measurements of  $^{235}$ U(n,f) at 100K incident neutron energy were presented. The experiment took place at the Budapest Nuclear Research Reactor. The motivation of this measurement was to verify the existing literature data and to understand the contradiction between experimental data and model calculations based on the so-called Los Alamos model of Madland and Nix [1]. As the PFNS is a very important spectrum entering into reactor neutronics calculations this discrepancy has been identified as of high priority to be resolved. Already within the OECD's Nuclear Energy Agency (NEA) Working Party of Evaluation Cooperation (WPEC) it had been emphasized as crucial and a subgroup (SG9) coordinated by D. G. Madland summarized already in 2003 the current situation [2]: "No calculated thermal spectrum has been found that simultaneously reproduces either of the two modern thermal differential measurements and the set of measured integral cross sections to within an acceptable level .... A new and highly accurate measurement of the prompt fission neutron spectrum for the reaction n(thermal) +  $^{235}$ U should be undertaken as soon as possible".

This conclusion and the importance of this spectrum is also the reason why the IAEA has now initiated a new Coordinated Research Project on Prompt Fission Neutron Spectra of Actinides [3].

The results of the experiments performed at the Budapest Nuclear Research Reactor have now been published [4] and I summarize here only the conclusion of this work.

Based on the present measurement the following can be concluded:

- 1. The measurements with three independent detectors are in excellent agreement. No dependence on the emission angle relative to the neutron beam is observed.
- 2. Our new result is in very good agreement with literature data at thermal energy, and with some data measured at 0.5 MeV incident neutron energy. However, the bulk of experimental data at 0.5 MeV does not agree with the thermal PFNS.
- 3. The PFNS at thermal energy did not confirm the model calculations based on the assumption that fission neutrons are emitted from fully accelerated fragments. An improved model incorporating scission neutron emission describes all experimental data at thermal energy within the error bars in the energy range from 0.1 12 MeV.
- 4. The thermal microscopic PFNS can neither describe the integral experiments nor benchmark results  $(K_{eff})$ .

It is unlikely, that this contradiction may be attributed to uncertainties of the integral data or threshold reaction cross sections. Therefore, new experimental efforts should be made to eventually find the origin of the disagreement.

#### 2) Integral cross section measurements using activation

In view of the fact that the present experimental PFNS can not describe integral cross section measurements, a new experiment using the neutron activation method has been performed very recently at the Budapest Nuclear Research Reactor. A common feature of the integral experiments compared to the differential measurements is the large amount of fissile material in massive <sup>235</sup>U samples used for the generation of the fast neutron spectrum. The large amount of fissile material may however lead to a change in the shape of the observed PFNS in those samples. Thus, this effect could provide an explanation for the difference between microscopic and macroscopic results for PFNS estimation. Therefore, in order to find the true PFNS, an experiment has been performed using the neutron activation method applying small samples and in similar experimental conditions as in previous differential measurements to eliminate systematic uncertainties as much as possible. To create a new set of activation data, we used the newly established measurement technique (DONA detector), recently published in [5]. The DONA method is based on measuring the neutron activity induced in a series of small metal disks. After exposure to a neutron field, the radionuclides produced in the discs are detected using high resolution gamma–ray spectrometry and, the neutron spectrum is

obtained using a spectrum unfolding technique. The detector discussed here, however, is aimed for the measurement of much lower fluxes which is facilitated by using thicker disks and low-background gamma-ray detectors for measuring the decay of the activation products. The detector activation device consists of a set of small metal discs placed in a circular plastic holder with a diameter of 120 mm. For this study the following metal discs might be selected: Ti, Ni, Fe, Co, In, Mg, Al, Zr and Au. These isotopes will cover neutron energies from 0.3 MeV – 20 MeV. The detector had been validated previously in a well-characterized <sup>252</sup>Cf neutron reference field [6]. So far, the analysis of the data revealed a severe problem with counting statistics. Much less activity was found on the different disks as anticipated. The data analysis is still in progress, but it might be, that the results are not up to expectations.

## 3) Correlations of prompt neutron emission with fission fragments in <sup>252</sup>Cf(SF)

The motivation for this experiment lies in the fact that from theoretical modelling of prompt fission neutron emission the experimentally observed drop in the neutron multiplicity as a function of total kinetic energy (TKE) cannot be reproduced [7]. Data acquisition has been performed applying a double Frisch grid ionisation chamber and a NE213 equivalent neutron detector along the fission axis. In addition, the availability of digital signal acquisition will add a new quality to the data analysis. As a preliminary first result we could clarify the close to linear behaviour of the prompt neutron multiplicity as a function of TKE down to 140 MeV. The reduction in neutron multiplicity at these TKE values observed in literature must be related to experimental shortcomings.

Also the slope of this dependence could be determined to about 8.5 MeV/neutron, a value in accordance with neutron binding energy considerations. Detailed data analysis is still in progress.

#### References:

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## 3. Evaluation and Use of the Prompt Fission Neutron Spectrum and Spectra Covariance Matrices in Criticality and Shielding

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<u>Summary</u>: Evaluation and use of PFNS and spectra covariance matrices for <sup>235</sup>U, <sup>238</sup>U and <sup>239</sup>Pu is described. The constrained sensitivity method was used for the calculation of fission spectra sensitivity.

## 1 Differences between ENDF/B-VII, Watt and Kornilov model spectra

The following PFNS formulations were considered in the study:

- Watt distribution
- Kornilov spectra
- ENDF/B-VII: Madland-Nix model

New parameters were determined for the Kornilov PFNS based on the measurement performed at the VENUS-3 and pressure vessel surveillance dosimetry (see Table below).

Fission averaged detector cross-sections (VENUS-3)									
Detector	Measured	New		Orig.	Watt		JENDL-3.3		ENDF/B-
		Kornilov		Kornilov					VII.0
$^{27}$ Al(n, $\alpha$ ) [mb]	0.706	0.699	±	0.989	0.742	±	0.732	±	0.746
		11.1%			11.1%		14.1%		
$^{58}$ Ni(n,p) [b]	0.1085	0.1056	±	0.1147	0.1085	±	0.1072	±	0.1075
		3.0%			4.0%		5.2%		
$^{115}$ In(n,n')[b]	0.1903	0.1851	±	0.1895	0.1871	±	0.1883	±	0.1881
		1.2%			1.7%		2.5%		

## 2 Data Formats for Cross Section Covariances in Evaluated Data Files

PFNS covariance matrices can be in principle provided in two ENDF formats:

- MF=35: covariance of energy distribution of secondary particles; data can be processed using NJOY-ERRORR (former ERRORJ). Processing is currently restricted to MT=18 only;
- MF=30: Covariances obtained from parameter covariances and sensitivities (no processing available, for linear models)

#### **3** Constructing fission spectra covariance matrices

Two methods were applied to construct the covariance matrices relative to the Watt and Kornilov PFNS formulations:

#### - Analytic Method (File-30 Formalism)

For the Watt distribution the covariance matrix of the parameters a and b given in the form of the (2 x

2) matrix:	1	
δa/a=1.2%	$P = \begin{bmatrix} 1 \end{bmatrix}$	U
$\delta b/b = 5.9\%$	$ \Lambda  = 0$	1

Using the File-30 formalism, the uncertainty in an integral parameter (such as k-eff) is calculated directly from the above  $(2 \times 2)$  matrix and the sensitivity coefficients. This method can be used only in case of (reasonably) linear systems.

#### - Monte Carlo Method

MC method can be applied both to linear and non-linear models. This method was used to produce covariances for <sup>235</sup>U, <sup>238</sup>U and <sup>239</sup>Pu fission spectra based on the Watt and Kornilov models. The covariance matrices based on Watt model were successfully validated against an analytical approach restricted to linear approximation.

#### 4 Normalisation applied to sensitivity coefficients

An alternative procedure for the calculation of the PFNS sensitivity coefficients, called constrained sensitivity method (discussed during WPEC-SG-26), was implemented in the SAGEP and SUSD3D codes. The virtue of the method is to consistently re-normalise (correct if needed) the prompt fission spectra covariance matrices. This option is useful in cases where the fission spectra covariance matrices do not comply with the ENDF-6 Format Manual rules.

The constrained sensitivity method and the new covariance matrices were tested on sets of thermal (KRITZ) and fast (SNEAK) critical experiments. For thermal systems the uncertainties in k-eff due to the fission spectra uncertainties were found to be rather low (of the order of ~10 - 30 pcm). On the other hand these uncertainties were substantially higher (~200 – 300 pcm) for MOX and fast systems.

The differences in the k-eff calculated using different PFNS evaluations were found to be reasonably consistent with the uncertainties calculated using the available PFNS covariance matrices.

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# 4. <sup>235</sup>U Prompt Fission Neutron Spectra - experimental mistakes or lack of understanding? N. Kornilov

#### Introduction

 $^{235}$ U is the most important isotope for various nuclear applications. The Prompt Fission Neutron Spectrum (PFNS) from the  $^{235}$ U(n,f) reaction has been investigated in many experiments at different incident neutron energies from thermal to the fast region. The different types of experiments microscopic (differential) and macroscopic (integral average cross section at thermal energy and K<sub>eff</sub> experiments) are applied for estimation of the PFNS shape. The analysis and evaluation of the microscopic data measured in the past century in the energy range 0-5 MeV can be found in Ref. [1] (see also [2, 3]). The general conclusion is that the PFNS can be described reasonably well in the frame of traditional theoretical model [2] or with its modification [1] or with semi-empirical systematic [3].

At the same time a continuing conflict still exists at thermal neutron energy. Microscopic experimental PFNS can not describe macroscopic data. The subgroup of NEA OECD [4] analyzing all available experimental data concluded: "...no calculated thermal spectrum has been found that simultaneously reproduces either of the two modern thermal differential measurement and the set of measured integral cross-sections to within an acceptable level". I would like to highlight that this is a long standing conflict. It exists from the first [5] to the last [6] publications and is not resolved yet. The traditional arguments used by evaluators "this is due to experimental mistakes of the microscopic data" may construct non physical "zig-zag" dependence of the average energy of the PFNS [7] only. This conclusion does not agree with previous analysis of the microscopic data [8] and recent experiments [6].

The experimental facts which can not be explained in the frame of traditional theoretical model are collected and discussed below.

#### 1. Mechanism of neutron emission and PFNS shape. Problem #1.

The experimental data at the thermal point [6, 9] are in perfect agreement and show another PFNS than is predicted by the LANL model even with corrected level density parameter: a=A/10.2. The difference of the spectrum shape is rather small ~5% but is out of the experimental uncertainties and the shapes of the experimental and calculated spectra are different. However the incorporation of the scission neutron emission (so named "3 sources model") increases the agreement very much ( $\chi^2=0.64$  instead of 3.5).

# 2. The dependence on incident neutron energy between thermal and 0.5 MeV. Problem #2.

Any theoretical model predict rather small dependence of the PFNS shape versus input energy. Average energies at the thermal point and 0.5 MeV input neutron energy are  $\langle E_{th} \rangle = 2.031$  MeV and  $\langle E_{05} \rangle = 2.045$  MeV according to LANL model calculations. If you will normalize the PFNS to the corresponding Maxwellian you may compare the experimental and calculated result at these two energies where we have a lot of experimental spectra. The difference between ratios should be  $\langle 1.3 \rangle$  in the energy range  $\langle 10 \rangle$  MeV if our theoretical models are correct.

However, the direct comparison between thermal and 0.5 MeV experimental data does not confirm this theoretical prediction. The difference for Johansson's data [11] is ~5% however it requires to reduce the level density parameter from a=A/10.2 at the thermal point to a=A/11 at 0.5 MeV or to reduce the share of the low energy component from  $\omega$ =0.26 to  $\omega$ =0.08 to achieve agreement between calculated and experimental results. The difference between Staples's data [12] and the thermal spectra is ~15 % at ~6 MeV energy of fission neutrons. It is important to highlight that the small difference between thermal and 0.5 MeV data as it should be according to the theoretical estimation, and the biggest difference (Staples and IRMM data) was supported by independent experimental result [6, 11, 12, 13]. Is it experimental mistakes which are reproduced with this high accuracy?

#### 3. Angular effect and left-right anisotropy. Problem #3.

An angular dependence of the PFN emission at 0.5 MeV input neutron energy was measured in IRMM during 2006-08. The first result was submitted to ND2007 [14]. The measurements were repeated 3 times with 3 detectors placed at different angles on the left and right side relative to the proton beam. Authors of Ref. [13] analyzing the experimental procedure concluded that this angular effect can not be explained by known experimental mistake. At the same time no correlation between experimental parameters and the observed effect was found.

#### 4. Integral and differential data. Problem #4.

The new thermal spectrum [6] which is in very good agreement with the old experimental results of Ref. [9] cancels any doubts concerning experimental mistakes of the microscopic data as a source of the disagreement with macroscopic data. However, this result intensified the contradiction only. The ratio of calculated cross sections to experimental ones R=C/E for ENDF/B-VII (a=A/11) and the "3 sources model" are very different. The experimental data and reaction cross sections were taken from Ref. [15], and IRDF-2002. The reaction cross sections were verified with the <sup>252</sup>Cf results. The average ratios are  $< R_{Cf} >= 0.995 \pm 0.004$ ,  $< R_{ENDF} >= 0.998 \pm 0.009$ ,  $< R_{3sources} >= 0.938 \pm 0.010$ .

The PFNS which agrees with differential data can not describe the integral results, and ENDF/B-VII data which was fitted to only one experimental spectrum at 0.5 MeV is very good. This spectrum contradicts to the incident energy dependence (see problem #2). The level density should be changed very much (~8% when the excitation energy changes with ~2% only) to describe Johansson data with the LANL model. There is no reasonable physical explanation for this fact. At the same time, it is not clear why the low energy component of the scission neutron (SCN) spectrum should be changed in whatever integral experiment set up.

#### Conclusion

The first problem is most simple to solve. We should incorporate the SCN emission in the model and realize additional experiments to investigate the properties of the scission neutron emission for different isotopes. Improved theoretical models are very important, too.

Problems #2, 3 are more difficult. The incorporation of the SCN emission is not enough. One may conclude that a factor exists which has a rather strong influence on the PFNS shape and asymmetry effects but was not fixed in the experimental investigations at 0.5 MeV input neutron energy.

All experiments which results were used were made with <sup>7</sup>Li(p,n) reaction as a neutron source and pulsed mode. One may assume that this factor is the neutron polarization. We should take into account the possible proton polarization also due to the pulsed mode of the accelerators (chopper, bunching high voltages, analyzing and switching magnets). In the preparation stage of any PFNS experiment it was assumed that this factor is not important or by definition should be equal to zero. If this explanation is true, the transmission mechanism of the information from the incident neutron to the secondary fission neutron should be found. The only possibility might be scission neutron emission, a fast process without formation of the *compound nucleus*. This may provide the link between the incident neutron and the secondary fission neutron. So, for a real clarification of this effect we need new experiments with polarized thermal neutron beams. When we will confirm and verify this effect new theoretical model should be developed.

The most difficult to understand is problem #4. There are no realistic ideas for solving it. May be this will come after new experimental efforts mentioned above.

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# 5. CONSYST/ABBN Data Code System and RUSFOND Nuclear Data Library Gennady Manturov (IPPE, Obninsk)

Designing of neutronics characteristics of fast reactor cores and fuel cycle requires using certified, qualified sets of codes and constants. The calculation codes should be related to the modern state of computational techniques. The used constants should be adequate to the most reliable evaluations, adopted in modern libraries of evaluated nuclear data.

The last ABBN version, appeared in 1990-93, was 299-grouped and quite differed from the previous ones. The ABBN-93 [2] is based on a library of evaluated nuclear data files FOND -2.2 [1]. In 1995 the ABBN-93 was certificated as a recommended data. For treating the ABBN-93 data a special code system CONSYST/ABBN was developed [3].

The main features of the ABBN-93 Group Data Set:

- □ The ABBN-93 was prepared from the FOND-2.2 data library (<u>Files Of evaluated Nuclear</u> <u>D</u>ata), which contains selected data from JENDL-3, BROND-2, ENDF/B-VI, and JEF-2 libraries.
- □ ABBN-93 presents group constants for 299 neutron energy groups from the thermal up to 20 MeV, and for 127 photon groups from 0 and up to 11 MeV.
- Resonance self-shielding effects are taken into account by using Bondarenko self-shielding ffactors.
- □ Nikolaev's subgroup parameters are also included to 299-group constants sets (they can be used in cases when the spatial dependence of group constants is essential)
- □ Effects of neutron thermalization are accounted by P0 and P1 73-grouped thermal scattering matrices in the energy region below 4.65 eV as function of the neutron gas temperature.
- □ The ABBN-93 group constants are presented as in the form of formatted tables which can be easily viewed so in binary form. Both data sets (and their combination) can be used in the calculations.

In 2005 were started and in 2006 finished a work on creation the first version of new Russian library of evaluated nuclear data files RUSFOND.

The main features of the RUSFOND nuclear data library:

- **RUSFOND** Russian library of files of evaluated nuclear data
- Created on the order of the Russian Ministry for Science and Education in 2005 -2006.
- □ Contains evaluated nuclear data for all stable nuclides and all radio nuclides with T1/2 > 1 day.
- □ The most reliable files were selected from FOND-2.2, BROND-3, JENDL-3.3, ENDF/B-VII, JEFF-3.1 and EAF-2003 (for radio nuclides).
- □ The selection was based on cross-checking of the evaluated data one with another and comparison with experimental data from EXFOR. Preliminary neutron data validation through the integral experiments was also made during the selection of the data. As the result of this, different modifications were included in the selected files for many nuclides.
- □ It includes files for 654 materials and 20 files of different lows for scatterings in the thermal region

Three directions in developing of codes for fast reactor neutronics calculations can be stated: (1) discrete codes, (2) based on Monte-Carlo, (3) used synthesis methods.

Codes, which are used in the design calculations, mostly solve the Boltzman transport equation in diffusion approximation, as: TRIGEX, JARFR, GEFEST, FACT-BR, SYNTES.

Codes, which are based on Monte-Carlo method, were developed during many years. Nowadays they have additional impulse in interest due to fast developing of the computational technique. Among Russian codes, as MMKFK, American codes KENO and MCNP are now widely used. Recently, based on MMKFK and KENO a hybrid complex MMKKENO [3] was developed.

For the shielding calculations as well as for determining diffusion-transport corrections codes TWODANT and DORT-TORT are used. For the depletion and kinetic calculations CARE and ORIGEN codes are used.

The main feature of the all mentioned codes is that they use one, same, unique constants data base ABBN-93 with the code CONSYST for generation of effective cross-sections. The scheme of interrelation of constants and codes used for fast reactor calculations shown in Fig.1.



Fig.1. Scheme of interrelation of constants and codes used for fast reactor calculations.

Experimental Data Bases Used for Validation:

- ICSBEP NEA/OECD, benchmark experiments on criticality safety
- IRPhEP NEA/OECD, reactor physics benchmark experiments
- BFS-IPPE, FR mock-up experiments
- SINBAD NEA/OECD, shielding benchmarks

Benchmark	MCNP RUSFOND	MCNP ABBN-RF	%	(C/E-1)*100,% ABBN-93	(C/E-1)*100,% ABBN-RF	
SCHERZO-556	0.99765	0.99722	-0.06	0.31	-0.27	
GODIVA	0.99939	0.99997	-0.09	-0.07	-0.06	
TOPSY	1.00030	0.99967	-0.06	0.50	-0.02	
FLATTOP25	1.00181	1.00089	-0.04	0.30	0.09	
BIG-10	0.99778	0.99721	0.06	0.52	0.26	
239PU JEZEBEL	0.99911	0.99922	0.01	-0.26	-0.10	
240PU JEZEBEL	1.00092	1.00051	-0.04	0.09	0.08	
POPSY	0.99946	0.99881	-0.07	0.20	-0.01	
FLATTOP-PU	1.00073	0.99948	-0.12	-0.17	-0.01	

Some Calculation Results with RUSFOND Data Library:

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# 6. <sup>235</sup>U(n,f) prompt fission neutron spectra V.M. Maslov

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## **ON BEHALF OF THE COLLABORATION**

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A new prompt fission neutron spectrum matrix for the  $n+^{235}U$  system is proposed. The thermal neutron spectrum in this matrix describes the newest data of JRC IRMM by Hambsch et al. (2009) [1]. The longstanding problem of inconsistency of integral thermal data testing and differential prompt fission neutron spectra data (PFNS) seems to be solved. It was mostly due to rather poor fits of differential PFNS data in major data libraries (see Fig. 1).



The older measured database is updated here using modern standards like spontaneous fission neutron spectra of <sup>252</sup>Cf(sf). That largely removes the inconsistency of older thermal neutron-induced PFNS data with the newest data. A phenomenological approach, developed by Kornilov et al. (1999) [2], for the first-chance fission and extended for the emissive fission domain by Maslov et al. (2005) [3] is normalized at  $E_{th}$  (see Fig. 2) to predict the PFNS average energy  $\langle E \rangle$  and the PFNS shape up to 20 MeV. In the first-chance and emissive fission domain evaluated PFNS are consistent with the  $\langle E \rangle$  data by Ethvignot et al. (2005) [4] (see Fig. 3).



Fig.3. Average energy of PFNS of  $^{235}$ U(n, f). HEU-MET-FAST, TRIPOLI, Morillon et al.



Fig.4. HEU-MET-FAST benchmark C/E values for  $K_{eff}$  calculated replacing the  $^{235}$ U PFNS.



Fig.5. LCT benchmarks C/E values for K<sub>eff</sub>, calculated with <sup>235</sup>U PFNS being replaced.

A compiled ENDF-6 formatted file of the <sup>235</sup>U(n,f) PFNS largely removes the inconsistencies of the evaluated differential PFNS with integral data benchmarks. Fast integral critical experiments like GODIVA or Flattop benchmarks are reproduced with the same accuracy as with the PFNS of the major data libraries (see Fig. 4). That reveals a rather delicate compensation effect, since present and previous PFNS shapes are drastically different from each other. Thermal assembly benchmarking reveals positive biases in k<sub>eff</sub>, which might be attributed to the influence of a soft energy tail of the present PFNS. For some of Valduc's LCT benchmarks biases in k<sub>eff</sub> are less than 20 pcm. The positive bias in LCT benchmarks might be removed using the <sup>238</sup>U PFNS and secondary neutron spectra, calculated in the same approach, as that used for <sup>235</sup>U.

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#### 7. Physics-based Calculation of the Prompt Fission Neutron Spectrum

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#### 1. Introduction

The basic idea meant by "physics-based" implies that the calculation model of the PFNS should be grounded on the present knowledge of fission physics and be consistent with it. Thus the present author keeps away from "ad hoc fitting" or "fitting-for-the-sake-of-fit", which simply obscures the physics involved in the fission process. The problems discussed in this paper include the possibility of scission neutrons, angular anisotropy, and neutron emission during acceleration, analyzed on the basis of the multimodal Madland-Nix (MN) model.

#### 2. The PFNS in the Energy Region E < 0.5 MeV

It has been known that there are differences between measured data of PFNS by Johansson *et al.*<sup>1)</sup> and Starostov and Nefedov<sup>2)</sup> in the region E<0.5 MeV for <sup>235</sup>U( $n_{th}$ ,f). At present, it is not known which is correct, but anyway it is of physical interest to examine the effects of scission neutrons (SCN), anisotropic neutron emission in the CM-system, and neutron emission during acceleration of fragments.

(a) Scission neutrons: Scission neutrons have been studied over several decades, but data and views are dispersed, their estimated fraction distributing from  $1.1\%^{3}$ ,  $3\%^{4}$  and  $5\%^{5}$ . The author attempted to consider a contribution of SCN to the multimodal MN model calculation in a phenomenological way, *i.e.*, by adding SCN component with some assumed fraction and nuclear temperatures of Weisskopf-Ewing shape.

**Fig.1** Multimodal MN model calculation plus 3% of scission neutrons with different temperatures.

As can be seen from Fig. 1, 3% of SCN with nuclear temperature 0.3 to 0.5 MeV would be enough to fill the gap; 5% is too much. Kornilov-Maslov's claim<sup>6)</sup> of finding 25% of SCN is just a make-up to make the fit beautiful, because it is simply based on fitting to Starostov-Nefedov's data with a combination of antiquated Watt functions and two SCN terms. SCN should be studied from physics point of view, and should



not be treated as a convenient tool for fitting.

(b) Anisotropic neutron emission: It is easy to incorporate the angular anisotropy of emitted neutrons in the CM system of the fragment into the frame of the multimodal MN theory. The results are shown in Fig. 2 for different values of the anisotropy parameter defined as  $b=W(0^{\circ})/W(90^{\circ}) - 1$ . It can be seen that the angular anisotropy enhances the low-energy ( $E_n < 0.6$  MeV) and high-energy ( $E_n > 4$  MeV) wings of the spectrum and diminishes the intermediate part. Thus this may account for some portion of the discrepancy in the low-energy wing



(c) Neutron emission during acceleration : It has been suggested that some fraction of neutrons are emitted during, rather than after, the acceleration of the fission fragments. This possibility was examined, results of which are shown in **Figs. 3** and 4 for  $^{239}$ Pu(n<sub>th</sub>,f).

**Fig.3** Fragment acceleration (full line) and decay by neutron emission (dashed line) for four FFs from S1- and S2-modes for  $^{239}$ Pu(n<sub>th</sub>,f).

It can be seen that, (1) the time scale of acceleration of FF does not depend on the fission mode, while the decaying time is strongly dependent on the fission mode, and (2) there is a fair chance of neutron emission during acceleration for S2-mode. From Fig. 4, we see that this effect tends to raise the low-energy part of the spectrum.

St 1 HF St1 LF St2 HF St2 LF 1) 1 0.5 E/B 1.0E-22 1.0E-21 1.0E-20 1.0E-18 1.0E-17 n [s] 1.0E-16 1.0E-15 1.0E-14 1.0E-19 Tim 1.4 Full Acceleration During Acceleration D.Abramson(1977)





3. Application of Multimodal Madland-Nix Model to Bimodal Fission

It is known that the fragment mass distribution for Fm-isotopes changes suddenly from asymmetric to symmetric at  $^{258}$ Fm (N=158) (**Fig. 5**). This phenomenon is known as *bimodal fission*. It is interesting to ask what would be the impact of the sudden switchover to symmetric fission, on the PFNS.

A series of calculation have been done for Fm-isotopes and the results are compared (**Fig. 6**). The spectra are observed to get harder in the increasing order of mass number for Fm-256, -257 and -258. For Fm-259, the spectrum suddenly gets much softer. This change in PFNS reflects the change in the branching ratio at the bifurcation point to the super-short (SS) mode; sudden softening of the spectra for Fm-259 is accounted for by complete switchover to the SS mode, in which most of the released energy goes to TKE and the two spherical fragments are left cold, due to the shell effect around A=132. This exemplifies the importance of considering fission modal changes in calculating the PFNS.

**Fig.5** Systematic change in the fragment mass distributions for Fm-isotopes.

**Fig.6** Calculated PFNS for Fm-isotopes. Relevant fission mode branching ratios are also shown.



- 4. Questions about "three-neutron-source model" Kornilov and Maslov proposed "three-neutron-source model". However, there are several questionable points in the model.
  - 1) There are no physically convincing reasons for asserting that Watt formula with the fixed functional form and a single nuclear temperature is a better representation of PFNS than the Madland-Nix model.
  - 2) The "three-neutron-source model" is essentially a *fitting-for-the-sake-of-fit* model, with more than ten adjustable parameters which have only poor physical meaning.
  - 3) "25% of SCN", claimed in their model, is obtained simply by fitting their model to the measured data of Starostov and Nefedov<sup>2</sup>). The present author has shown that same degree of fitting to the mentioned measured data can be obtained by assuming only 3% of SCN using the multimodal MN model. This strongly suggests that the "25% of SCN" is a makeup, or, at least, just one of the adjustable parameters needed to get a better fit to the measured data.
  - 4) Curiously enough, the "three-neutron-source model" transformed itself into "single-neutronsource model" *without SCN-term* in the integral verification<sup>7)</sup>. Such an inconsistency reduces the confidence and reliability to the model itself.

Historically, the study of PFNS started with fitting the spectra with fixed functions such as Maxwellian and Watt functions or combinations thereof. With the progress of understanding of the fission mechanism, abreast with the progress of physics of large-amplitude deformation of the nucleus and dissipation of the excitation energy, the study of PFNS has advanced to physical modelling of the whole fission process leading to the emission of prompt neutrons. Examples for this involve Madland-

Nix model, complex cascade emission model, Hauser-Feshbach-type model, Monte Carlo model, *etc*. The idea behind the two preceding CRPs of IAEA<sup>8,9)</sup> clearly pointed to that direction. Going back to *ad hoc* fitting seems to the author like reversion to the ages decades ago.

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# 8. Plan of the PFNS calculation with Semi-Empirical Model and Study of the energy partition

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The programme of this work is to calculate the PFNS of  $^{235}$ U (n, f) in the energy range from thermal to 6 MeV where only one fission channel (n,f) is open by the Point-by-Point method and with a semi-empirical model described below.

The emitted neutron spectrum,  $Sp(A_{L,H}, E_{L,H}^*)$ , for every single fission fragment will be determined. Then, the total FPNS could be deduced by summing up all fragments' spectra with a weight involving the mass yields and average neutron number ( $\overline{v}$ ) emitted from each fragment.  $E_{L,H}^*$  is the excitation energy of the fragment, which can be calculated from the total energy released, total kinetic energy *TKE*, total excitation energy *TXE*, and the energy partition between the light and heavy fragments, etc. The evaporation model would be considered in the spectrum calculation. To improve the result, more physics will be taken into account, such as the scission neutron, the pre-equilibrium emission, and neutron emission during fragment acceleration, etc.

The energy partition is one of the key questions. Our previous work gave the following result by a systematic study for the <sup>235</sup>U(n,f) fission in Fig. 3 [Yongjing Chen *et al*, in print]. Commonly, the fission yield consists of 2 parts, symmetrical and asymmetrical fission. The yield ratio ( $Y_S/Y_a$ ) of the symmetrical to asymmetrical fission were studied [Tinjing Liu *et al*, China physics C, 32(7), 556 (2008)] as shown in fig. 1,



Fig .1. Yield ratio of symmetrical fission to the asymmetrical one, Ys/Ya.

Symmetrical fission could be described with a statistical model, of which the energy partition is proportional to the fragment mass in eq. (1),

$$E_L^*(\text{sym.}) = R_L(\text{sym.}) \cdot TXE = \frac{A_L}{A_C} TXE, \qquad (1)$$

Where,  $E_{\rm L}^*$ ,  $A_{\rm L}$  are resp. the excitation energy and the mass of the light fragment,  $A_{\rm C}$  the mass of the compound nuclei. While the asymmetrical fission is mainly caused by shell effects at N~82 and ~88, which energy partition could be deduced from the  $^{235}$ U(n<sub>th</sub>,f) experimental data (wherein the symmetrical is very small and could be omitted at thermal fission),

$$E_{L}^{*}(\operatorname{asy} m) = R_{L}(\operatorname{asy} m) \cdot TXE$$

$$= \frac{\overline{v}_{\exp}(A_{L}) \langle \eta \rangle_{L}(A_{L}) + \overline{E}_{\exp \psi}(A_{L})}{\overline{v}_{\exp}(A_{L}) \langle \eta \rangle_{L}(A_{L}) + \overline{E}_{\exp \psi}(A_{L}) + \overline{v}_{\exp}(A_{H}) \langle \eta \rangle_{H}(A_{H}) + \overline{E}_{\exp \psi}(A_{H})} \cdot TXE, \quad (2)$$

where,  $\langle \eta \rangle (A_{L,H})$  is the energy the emitted neutron carries, including the kinetic and binding energy. The experimental data involved were available for <sup>235</sup>U fission at thermal energy, which are the  $\gamma$  energy  $\overline{E}_{\exp,\gamma}$  taken from the fragment, the average prompt neutron number  $\overline{\nu}_{\exp}$  and the average energy  $\langle \eta \rangle$  brought by each neutron. The result is shown in Fig. 2.

The overall excitation energy partition could be deduced with eq. (3), and the result is shown in Fig. 3.

$$E_{L}^{*} = \frac{Y_{S}}{Y_{S} + Y_{a}} \cdot R_{L}(\text{sym}) \cdot TXE + \frac{Y_{a}}{Y_{S} + Y_{a}} R_{L}(\text{asym.}) \cdot TXE$$
$$E_{H}^{*} = TXE - E_{L}^{*}$$
(3)





Fig .2. The energy partition (here the ratio is shown of  $E_{\rm L}$ \*/TXE) between the two fragments at thermal incident energy.

Fig. 3. The excitation energy partition (here the ratio is shown of  $E_{\rm L}$ \*/TXE) between the two fragments at 3, 5, 14 and 20 MeV.

#### 9. Prompt Fission Neutron Spectra of actinides

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The T-2 Nuclear Physics Group at Los Alamos National Laboratory is involved in various modeling and evaluation work related to prompt fission neutron spectra (PFNS). Three main thrusts can be identified:

- Monte Carlo Hauser-Feshbach calculations of PFNS
- Uncertainty Quantification on Evaluated PFNS
- Benchmarking Evaluations & Uncertainties

A Monte Carlo implementation of the Hauser-Feshbach equations has been coded, allowing the computation of the evaporation of excited primary fission fragments step-by-step, by following the probabilities of neutron and  $\gamma$ -ray emissions at each stage of the compound nucleus evaporation. Such calculations start by sampling the initial fission fragment distribution in mass, charge and kinetic energy. Assuming a particular distribution of the total excitation energy between the two complementary fragments, detailed characteristics of prompt neutrons can be inferred. At this stage, the competition between neutrons and  $\gamma$  rays is neglected, and  $\gamma$  rays are only emitted when the residual excitation energy becomes too low for further neutron emission. Exclusive data such as neutron multiplicity distribution, P(v), average neutron multiplicity as a function of the mass of the fragment,  $\langle v \rangle$ (A), exclusive spectrum for a given neutron multiplicity,  $\prod(E)_{|v|}$ , etc can be computed.

Examples of results have been shown, in particular for the neutron-induced fission of <sup>235</sup>U, in the incident-neutron energy range below the threshold for second-chance fission. Experimental fission fragment yields Y(A,TKE) measured by F.-J. Hambsch were used as input in these calculations. The assumption used to share the total excitation energy between the two fragments was shown to have a strong influence on  $\langle v \rangle$ (A), as well as on the total calculated spectrum. While only one parameter was used in the present calculations to distribute this energy, such a parameter should ultimately be predicted from theoretical considerations, and depend on the particular nuclear configurations present near the scission point. A summary of results for other isotopes, <sup>252</sup>Cf (sf),  $n_{th}$ +<sup>239</sup>Pu,  $n_{th}$ +<sup>235</sup>U and <sup>236</sup>Pu (sf), was also discussed. Such results should be viewed as preliminary as the optimization of various model input parameters remains to be done.

The second part of the talk was devoted to the quantification of uncertainties associated with evaluated PFNS. In particular, the PFNS for the reaction  $n+^{239}$ Pu for 0.5 MeV incident neutrons, evaluated for the ENDF/B-VII.0 library, was studied, and a covariance matrix was calculated. The Uncertainty Quantification (UQ) methodology used follows closely the one already successfully applied in the case of neutron-induced reaction cross-sections for many isotopes, and whose covariance matrices will appear in the ENDF/B-VII.1 library- those matrices are already available in the ENDF/A library and can be downloaded on the BNL National Nuclear Data Center web site. The Los Alamos (LA) or Madland-Nix model was used for the ENDF/B-VII.0 evaluated PFNS, and this UQ work follows the evaluation procedure as closely as possible. New LA model calculations were performed, using the same model input parameters as *prior* parameters. A selection of experimental data sets for low-energy neutron-induced fission on <sup>239</sup>Pu was then collected, and simple estimates were made regarding statistical and systematic uncertainties associated with these experiments. A Bayesian statistical approach was then used to combine experimental data and model parameter sensitivity calculations, and obtain *posterior* parameters and PFNS.

A final covariance matrix was produced for the  $n(0.5 \text{ MeV})+^{239}\text{Pu}$  PFNS. It was compiled in ENDF format (MF35, MT18), and processed in 33 energy-groups using the NJOY-99.296 data processing code. Transport calculations using the deterministic PARTISN code were performed in the case of a 1D model of the Jezebel critical assembly. Uncertainties were propagated from the PFNS covariance matrix, resulting in a ~0.2% uncertainty on the calculated multiplication factor  $k_{eff}$ .

This work has been submitted for publication in Nuclear Science and Engineering, and can be found in the LANL technical report LA-UR-10-00646 (2010).

A similar study was performed in the case of  $n+^{240}$ Pu PFNS. In this case however, no experimental spectrum data was found, and model parameters were constrained by data on the average neutron multiplicity  $\langle v \rangle$  only. A similar ENDF-formatted PFNS covariance matrix was produced.

Much remains to be done to test both the evaluated PFNS and the PFNS covariance matrices. A first example of such benchmarking calculations was shown with the use of the PARTISN transport code to simulate the multiplication factor  $k_{eff}$  and its uncertainty for the Jezebel <sup>239</sup>Pu critical assembly. Similar calculations will be performed for other isotopes and other assemblies.

# 10. Prompt fission neutron spectrum calculations in the frame of Point by Point and extended Los Alamos models. Application for <sup>233</sup>U(n,f) and <sup>239</sup>Pu(nf)

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In the last 10 years prompt neutron emission models based on Los Alamos (LA) model features [1] were developed and successfully used in prompt neutron data evaluations. Refinements were brought to the initial LA model by taking into account a more realistic form of the fragment residual nuclear temperature distribution and the anisotropy effect (details in Ref. [2]). In the case when only one compound nucleus (CN) is involved (spontaneous fission and neutron induced fission in the energy range of the first chance) the model can be used in three ways:

a) by taking only one fragmentation (the so-called "most probable fragmentation" approach) with average values of model parameters ( $\langle Er \rangle$ ,  $\langle TKE \rangle$ ,  $\langle Sn \rangle$ ,  $\langle C \rangle = A_{CN}/\langle a \rangle$ ,  $\langle E\gamma \rangle$ );

b) by using the multi-modal fission concept: the total prompt fission neutron spectrum (PFNS) and multiplicity (PFNM) being calculated as superposition of the spectrum and multiplicity of each mode weighted with the modal branching ratios. Average model parameters are determined for each mode;

c) the Point by Point (PbP) model (see [3] and references therein) that takes into account the entire fission fragment (FF) range covered by the Y(A,TKE) distribution. Total PFNS and PFNM are calculated as superposition of spectrum and multiplicity of each pair weighted with the charge and mass distributions of the FF. The PbP treatment is the most accurate because it takes into account the full range of possible fragmentations while the other two approaches consider only one or few fragmentations (subsets) and average model parameters. When more fission chances are involved, only the "most probable fragmentation" approach is used because it is impossible to distinguish Y(A,TKE) of each chance and more, over the secondary CN are formed at many excitations energy leading to a very large amount of calculations. The "most probable fragmentation" approach was extended to take into account the contribution in multiplicity and spectrum of the secondary nucleus chains and ways formed by charged particle emission at high incident energies (see [4]). Also the spectrum of neutrons emitted prior to scission is obtained from (n,xn) spectra provided by nuclear reaction codes (GNASH-FKK, Talys, Empire) from which the contribution of neutrons leading to excitation energies of the residual nucleus less than the fission-barrier height were substracted [5]. The average values of model parameters (and their dependences on the excitation energy) are usually determined from the PbP treatment. Systematic behaviours of these parameters were developed for Th, Pa, U, Np, Pu and Am isotopes (details in Ref. [6]). The PbP model provides all quantities referring to each FF (not depending on FF distributions Y(A,Z,TKE)) that can be compared with experimental data. Such quantities are: the multi-parametric matrix v(A, TKE), the multiplicity of the FF pair  $v_{pair}(A)$ , the sawtooth v(A),  $\varepsilon(A)$ ,  $E\gamma(A)$  and so on. Also the PbP model provides all total average quantities (obviously depending on FF distributions), such as PFNM, PFNS,  $< v_p >$ (TKE) and so on. The PbP model is able to provide the prompt neutron distribution P(v) in very good agreement with all existing experimental data [7].

In the present work the PbP model and the most probable fragmentation approach (with average model parameters issues from PbP treatment and from [6]) were used to calculate all prompt neutron emission data of <sup>239</sup>Pu(n,f) and <sup>233</sup>U(n,f), with focus on PFNS. The influences in the spectrum shape of different optical model parameterizations (used to calculate the CN cross-sections of the inverse process) and of different methods of TXE partition between the two FF forming a pair were investigated. In the region of low emitted neutron energies the spectrum experimental data description is considerably improved by taking into account the possible scission neutron contribution (with an amount of  $\langle v_{SCN} \rangle / \langle v_p \rangle = 1.1\%$ , according to [8]). Two examples of PFNS calculations are given in the following figures.

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Fig.1:  ${}^{233}U(n_{th},f)$  spectrum calculation in comparison with experimental data taken from EXFOR.



Fig.2: <sup>239</sup>Pu(n<sub>th</sub>,f) spectrum calculation in comparison with experimental data taken from EXFOR

# 11. Measurement of the total prompt neutron spectrum of <sup>235</sup>U(nth, f) relative to <sup>252</sup>Cf(sf)

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#### Introduction

A series of experiments has been performed at the PNPI WWR-M research reactor in Gatchina, Russia, to measure prompt neutron angular and energy distributions from thermal neutron-induced fission of <sup>235</sup>U in correlation with the fission fragments [1].

The obtained angular and energy distributions have been analyzed with the assumption of neutron emission from accelerated fission fragments. The performed analysis demonstrates that all obtained experimental results can be described within 5% accuracy using this assumption. It is to be noted that this conclusion, in a systematic sense, is dependent on the choice of the total prompt neutron spectrum used for the neutron detector efficiency correction [2]. To exclude this uncertainty, the additional measurements of prompt neutron angular and energy distribution have been performed recently for  $^{252}Cf(sf)$  and  $^{235}U(n_{th}, f)$  with the same experimental conditions and set-up.

#### 1. Experimental procedure and results

Four measurement cycles have been carried out at the PNPI WWR-M research reactor to measure the total prompt fission neutron spectrum (PFNS) of <sup>235</sup>U(n<sub>th</sub>, f) relative to <sup>252</sup>Cf(sf). During data processing, the following corrections were taken into account: for detector efficiency, for neutron detector background, for angular and energy resolution, for the fragment detector efficiency and for complementary fragment contribution. So the PFNS of <sup>235</sup>U,  $N_U(E_n)$ , was obtained using measured partial spectra,  $N^{exp}(E_n, \theta)$ , by following equation:

$$N_{U}(E_{n}) = F(E_{n}) \cdot N_{Cf}^{Std}(E_{n}) \cdot \frac{N_{U}^{\exp}(E_{n})}{N_{Cf}^{\exp}(E_{n})} = f_{res}^{A}(E_{n}) \cdot f_{res}^{E}(E_{n}) \cdot I(E_{n}) \cdot N_{Cf}^{Std}(E_{n}) \cdot \frac{N_{U}^{\exp}(E_{n})}{N_{Cf}^{\exp}(E_{n})}$$
(1)

where  $f_{res}^{A}(E_n)$ ,  $f_{res}^{E}(E_n)$  are the coefficients taking into account the finite angle and energy resolution;  $I(E_n)$  is due to summing over angle (because there is experimental histogram instead of continuous distribution) and  $N_{Cf}^{Std}(E_n)$  is the reference standard spectrum of <sup>252</sup>Cf(sf) [3]. The obtained PFNS of <sup>235</sup>U after efficiency correction and additional corrections are presented in Figs. 1, 2.





**Fig. 1.** The total PFNS of  $^{235}$ U as a ratio to a Maxwellian (T = 1.314) only background and efficiency corrections were applied (log scale).

**Fig. 2.** The values of corrections taken into account during data processing.

#### 2. Degree of reliability – experimental error determination

For 11 fixed angles between the neutron and light fragment direction (from  $0^0$  to  $180^0$  in  $18^0$  interval) the prompt neutron energy spectra were obtained independently for two neutron detectors as weighted averages of 4 measurement cycles which were analyzed separately.

The errors of the  $N^{\exp}(E_n, \theta)$  spectra are the RMS deviation from weighted means. These errors include the possible instability of the electronics (uncertainties of neutron threshold determination etc.) as well as the statistical and energy determination uncertainties.

The total prompt neutron spectrum uncertainties were defined as the deviation of the perturbed total spectra from the total spectrum obtained by summation of the measured angle-energy distribution over all angles. By varying the counts in each energy point within the obtained errors, twenty perturbed spectra were obtained for each of the two neutron detectors. Herewith, it was assumed that any correlation between the energy points for a fixed angle relative to the fission fragment direction was absent. The ratio between the relative errors is presented in Fig. 3.

The result is a weighted average of two total prompt neutron spectra obtained by individual detectors. The obtained PFNS after all corrections is presented in Fig. 4 in comparison with literature data which were normalized to the recommended value of the total average neutron multiplicity,  $v_{tot} = 2.42$ .





**Fig. 3.** The ratio between the relative errors summed up into total uncertainties.

Fig. 4. The PFNS of  $^{235}$ U as ratio to Maxwellian (log scale).

#### 3. Results and discussion

The comparison of the existing experimental data demonstrates the agreement between them within their errors in the energy range from ~1 MeV to 10 MeV. There is some discrepancy in the low energy region. To verify the assumption about neutron emission from accelerated fragments the PFNS was calculated using neutron spectra for small angles relative to the fission fragment direction obtained in our earlier experiment. The calculated (method 2) and experimentally obtained (method 1) spectra are shown in Figs. 5, 6. It is seen that the average of the two PFNS obtained from different experiments and methods is in good agreement with ENDF/B-VII.



Fig. 5. The PFNS of  $^{235}$ U as ratio to ENDF/B-VII evaluation (log scale).



Fig. 6. The PFNS of  $^{235}$ U as ratio to ENDF/B-VII evaluation (linear scale).

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#### 12. Measurements of prompt fission neutron spectra at Bruyères le Châtel

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There is a long tradition in prompt fission neutron spectra measurements (PFNS) at the CEA in Bruyères le Châtel. In the eighties, André Bertin et al., measured PFNS in neutron-induced fission of <sup>238</sup>U and <sup>235</sup>U at 0.6 and 7 MeV using the fission chamber technique [1]. More recently, our laboratory proposed and realised experiments at the Los Alamos Neutron Science Center with the Figaro setup in collaboration with R.C. Haight. These experiments are based on the double time-of-flight method. They allowed us to obtain the mean energy of the prompt fission neutrons and the associated mean multiplicity as a function of incident neutron energy from 1 to 200 MeV for <sup>238</sup>U and <sup>235</sup>U [2-4]. It was the first unambiguous experimental data to confirm the dip predicted by the models in the mean energy of the prompt fission. In 2007, we have performed experiments at Figaro on <sup>239</sup>Pu which provided unprecedented measurements of PFNS for this isotope [5]. These results are currently being finalized and will be published soon. Another experiment has also been performed on <sup>232</sup>Th and analysis is ongoing.

Due to the renewed interest in the problematic of PFNS new measurements have been started at Bruyères le Châtel using monoenergetic neutrons produced at the 4 MV Van de Graaff accelerator. The goal of these experiments is to obtain precision measurements in the 200 keV-5 MeV portion of the PFNS at a few incident neutron energies for <sup>238</sup>U, <sup>235</sup>U, <sup>239</sup>Pu, <sup>237</sup>Np and <sup>232</sup>Th. In these experiments, the fission neutrons are detected thanks to a paraterphenyl crystal detector which is known to be more efficient at low energy than stilbene or liquid organic scintillators [6]. The targets are fission chambers containing hundreds of milligrams of actinides. The energy of the prompt fission neutrons is obtained by the time-of-flight method over a flight path of about 1 m. The neutron detector is shielded from the neutrons and gamma-rays coming from the neutron production target and room return. The detection efficiency is determined with respect to Mannhart reference data using a <sup>252</sup>Cf fission chamber. Typical incident neutron energies being investigated are 500 keV, 2MeV, 5 MeV, 6.5 MeV, 15 MeV.

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