

# ***Compilation and evaluation of fission yield nuclear data***

*Final report of a co-ordinated research project  
1991–1996*



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## FOREWORD

Fission product yields are required at several stages of the nuclear fuel cycle and are therefore included in all large international data files for reactor calculations and related applications. Such files are maintained and disseminated by the Nuclear Data Section of the IAEA as a member of an international data centres network. Users of these data are from the fields of reactor design and operation, waste management and nuclear materials safeguards, all of which are essential parts of the IAEA programme.

In the 1980s, the number of measured fission yields increased so drastically that the manpower available for evaluating them to meet specific user needs was insufficient. To cope with this task, it was concluded in several meetings on fission product nuclear data, some of them convened by the IAEA, that international co-operation was required, and an IAEA co-ordinated research project (CRP) was recommended. This recommendation was endorsed by the International Nuclear Data Committee, an advisory body for the nuclear data programme of the IAEA.

As a consequence, the CRP on the Compilation and Evaluation of Fission Yield Nuclear Data was initiated in 1991, after its scope, objectives and tasks had been defined by a preparatory meeting. The different tasks, such as special evaluations and development of improved methods, were distributed among participants. The results of the research work were discussed and approved by all participants in research co-ordination meetings. For a successful development of theoretical and empirical models, experiments had to be recommended and their results to be awaited, which made necessary an extension of the CRP by two years.

This TECDOC is the result of a joint effort of all participants in this CRP. The individual sections represent CRP tasks and were prepared by the participants responsible for doing the research, some of which comprise significant new scientific developments. The appendices to this book contain voluminous tables and are therefore enclosed as a CD-ROM, which also includes a computer program for calculating fission yields. For the availability of evaluations, data files or tabulations contained or described in this book the reader is referred to the addresses given in Chapter 3 or in the list of CRP participants.

The IAEA wishes to thank all CRP participants who contributed to the success of the CRP and to this publication, namely: H.O. Denschlag (Chapter 2); T.R. England (Section 7.1); A.A. Goverdovski (Section 5.2); M.F. James (Sections 4.4 and 7.2); Liu Tingjin (Sections 5.1, 6.1, 6.3 and 7.4); R.W. Mills (Sections 4.4, 6.2 and 7.2); G. Rudstam (Section 4.3); F. Storrer (Section 3.2); A.C. Wahl (Sections 4.1, 4.2 and 7.4) and Wang Dao (Sections 6.3 and 7.3).

The IAEA officer responsible for the project and this publication was M. Lammer of the Division of Physical and Chemical Sciences.

### *EDITORIAL NOTE*

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## Chapter 1

### INTRODUCTION AND SUMMARY OF THE CO-ORDINATED RESEARCH PROJECT

#### 1.1. REQUIREMENTS FOR FISSION YIELDS

##### 1.1.1. Fission yields in application fields

One can distinguish two main areas of the use of fission yields: in fundamental physics, their significance lies in all aspects of the probability of fragment formation in the fission process, whereas in applied user fields they are needed for calculating the accumulation and inventory of fission products at various stages of the nuclear fuel cycle.

This CRP deals only with evaluations designed, produced and maintained for applied purposes. User needs in all areas of the nuclear fuel cycle have been extensively reviewed in past specialists meetings [1.1–1.4]. Here the most important application fields and their fission yield requirements are briefly summarized.

In *reactor design* and *operation*, fission yields are used in criticality and reactivity calculations, for fuel and reactor core management, for reactor safety (including fission product gas production and release, fuel failure detection and contamination of reactor components) and in determining limits of safe operation in new plants and for nuclear materials transport. *Burnup* determination and *decay heat* calculations are treated separately as special cases. Generally, the required accuracies are met by existing data. It should be noted that for the various reactor types, fission yields should be known as function of incident neutron energy. For contamination and gas production detection, ternary fission yields (tritium, helium) are needed.

For the *reprocessing of spent fuel* and the *management of nuclear waste* (temporary spent fuel storage and final waste depositories) it is important to know the fission products present primarily as a source of radiation (heat production and potential hazard to the environment and personnel). Fission yields enter calculations of fission product inventory and radioactivity (*decay heat*).

The *burnup* is a measure of the total number of fission events that have occurred in nuclear fuel and hence of the consumption of fissionable material. The burnup is predicted by reactor calculations and experimentally determined for actual spent fuel with the purpose of allowing an accurate evaluation of the fuel and reactor performance. For certain methods, fission products are used as burnup monitors and their fission yields are required with high accuracy for the evaluation of measurement results.

Two types of fission product *decay heat* can be distinguished:

- The *residual heat after reactor shutdown* is due to radiation emitted by fission products and actinides present in nuclear fuel in the reactor core, and is most important in the event of a loss-of-coolant accident. Its accurate knowledge, especially in the time from 1 to 1000 seconds after reactor shutdown, is crucial for the dimensioning of the maximum reactor power and the emergency cooling system. The fission product decay heat is obtained by

summation calculations that include all contributing fission products (short lived, with half-lives longer than a few tenth of a second) with their fission yields (and decay data).

- The ***decay heat from spent fuel*** is due to radiation emitted by (well known) long lived fission products (and actinides). Its significance is outlined above (***waste***).

In nuclear materials ***safeguards***, certain methods for ***spent fuel assay*** use fission yields (and other nuclear data) of monitor fission products for the verification (by measurement and/or calculation) of statements by reactor operators. (Similar methods could in principle also be used for the verification of dissolved fuel in reprocessing plants but have not yet been developed and tested).

### 1.1.2. Types of required fission yields

The users of evaluated fission yields can be separated into two distinct groups according to the fission yield type and corresponding evaluation work required:

The ***first group*** needs to calculate ***complete fission product inventories*** as accurately as possible and requires evaluated ***complete yield distributions***. This group includes most reactor and spent fuel handling applications, in particular summation calculations of decay heat at reactor shutdown.

The ***second group*** needs fission yields only for ***specific fission products***, either as ***reference yields*** for measurement standards, or yields for ***monitor*** fission products (burnup determination, safeguards), or because only specific fission products are important (contamination, gas production detection, waste management).

Detailed and exact definitions of fission yields as recommended by the CRP are given in Chapter 2. For the subsequent explanations we shall briefly define the ***independent*** or ***direct*** yield of a fission product as the probability of its formation directly in fission, the ***cumulative*** yield as the probability of its accumulation directly from fission plus via decay of its precursor(s) plus and/or minus via delayed neutron emission (but excluding changes due to neutron reactions in fission products).

The general evaluation method for currently available ***fission yield files*** is to evaluate, for each ***yield set*** (combination of fissionable nucleus and neutron energy), together measured (and corrected) relative and absolute independent and cumulative yields, then combine them with estimated yields (where no measurements are available) and finally adjust the complete yield distributions by applying physical constraints (basically to conform with the conservation of mass and charge in fission). This "global" evaluation method produces accurate and reliable complete yield distributions as required by the ***first user group***.

However, the higher accuracy obtained in the direct evaluation of individual fission yields is lost later on in the evaluation process with the application of the various adjustments. Therefore special careful evaluations of specific fission yields would be of benefit for the ***second user group***. The production of such an evaluation was one of the tasks of this CRP.

## 1.2. FISSION YIELD EVALUATION AND THE NEED FOR A CRP

### 1.2.1. Fission yield evaluation before the CRP and needs for improvements

Up to about 1973, several fission yield evaluations were done on a limited number of yield sets, often for special purposes (see reviews in Refs [1.5–1.7] and evaluations discussed therein). Just before that symposium [1.5–1.7], E.A.C. Crouch, United Kingdom [1.8], and B.F. Rider, United States of America [1.9], started evaluation efforts which later on became the comprehensive UK and US fission yield libraries. In 1976 the Chinese fission yield compilation and evaluation effort by the Chinese group commenced.

In the eighties, after the retirement of the pioneers Crouch and Rider, the co-ordination of the evaluation efforts was taken over by M.F. James (UK) and T.R. England (USA) respectively. They and their co-workers extended the scope of fission yield evaluation as, during that time, the requirements expressed by users of fission yields and the number of fission yield measurements to be evaluated increased drastically. The evaluators were facing requests for the following additional tasks:

- Fission yields were increasingly used in computer codes for reactor calculations using the ENDF format. Suitable **ENDF compatible formats** for independent and cumulative yields and their errors had to be developed and all yield sets transferred into ENDF-formatted files. A future requirement was also the introduction of **correlations** and **covariance matrices** in the evaluated yield files
- The **number of yield sets** requested for inclusion in evaluated data files increased drastically (for example: the evaluated yield sets in the US files increased from 10 in 1972/1974 to 50 in 1987). This meant of course an equally drastic increase in the measured data to be analyzed and evaluated.
- The inclusion of **isomeric yields** in evaluations was required for decay heat calculations.
- Users noted **discrepancies** among evaluations and demanded that they be resolved.
- The evaluation of the **energy dependence** of fission yields was requested to allow more accurate burnup calculations for fast reactor neutron spectra with different mean energies.
- **New techniques** for fission yield **measurements**, such as on-line mass spectrometry, were developed. Evaluators were required to study these methods to allow them a judgement of the type of yield and quality of data (error assignment) obtained in measurements, and how the data fit into the evaluation process (possibly requiring new evaluation methods).
- A consequence of the increasing demands was that also yield sets and charge distributions, for which rather few measurements existed, had to be included in data files. Furthermore, the accuracies of measured independent and isomeric yields and the prediction capabilities of charge distribution and isomeric yield models were insufficient for decay heat calculations. Therefore, **models** and **calculation methods** for **charge** and **mass distributions**, and for **isomeric yield** ratios had to be developed to fill these gaps of unmeasured yields and fulfil the accuracy requirements for decay heat calculations.

- To cope with the evaluation of an *increasing* number of fission yield measurements, the evaluation process and data selection criteria (chi-squared test, down-weighting) had to be *automated* and appropriate *computer programs* had to be developed.

The evaluators worked on improvements, and some support was provided on national levels. In the USA, the fission yield evaluation (see Section 7.1) became part of a nation wide effort (Cross Section Evaluation Working Group, CSEWG) of creating evaluated data files in ENDF format. In the UK the old Crouch evaluation was extended and eventually led to the development of the first UK fission yield file, UKFY1, which was adopted for the first version of the Joint Evaluated File, JEF1, (see Section 7.2). In 1987, the first ENDF-formatted file of evaluated fission yields was created in China and included in the Chinese Nuclear Data Library CENDL-1. The fission yields incorporated in other evaluated files (like the Japanese library JENDL or the French file) were taken from other sources.

At the same time, A.C. Wahl made a thorough evaluation of independent fission yields which he used to obtain best values for the parameters of his models [1.10]. For this purpose he also evaluated cumulative and chain yields for selected fission reactions. His model parameters were used by the other evaluators for the calculation of charge distributions and estimation of unmeasured yields, which was the first, still restricted, form of international co-operation.

In spite of all these efforts, the majority of the necessary improvements were not even attempted: Reliable charge distribution model predictions existed only for the main uranium and plutonium isotopes, and suggested mass distribution models were not applied for fission yield evaluation. Theoretical approaches to predict the energy dependence of yields did not exist and systematics were not studied. Computer programs for the inclusion of correlations and covariances in fission yield evaluations were not developed, and discrepancies among measurements and among evaluations were noted but not analyzed.

### 1.2.2. The need for co-operation

Thanks to the nationally co-ordinated efforts at least continuity in the evaluation work could be ensured and some improvements implemented like the creation of the first ENDF-formatted fission yield files, the development of codes for automated evaluation, the inclusion of model estimates and the application of physical constraints in the evaluation procedures. However, the evaluators complained that this was all they could accomplish with the manpower and funding available, and work on the majority of the improvements listed above could not even be started.

This situation of fission yield evaluation and possible solutions were discussed at several specialists meetings in the eighties [1.3, 1.4, 1.11, 1.12]. As a first step a closer co-operation among evaluating groups was recommended. They should:

- discuss evaluation methods (constraints), procedures and programs, and exchange ideas;
- exchange and compare files of compiled experimental data and references;
- exchange corrections made to experimental data and discrepancies found;
- cross-check evaluated data (file inter-comparisons) and discuss gaps and discrepancies;
- exchange information on experiences made with model estimates for unmeasured yields.

However, to solve the problem of limited manpower and to find ways of working on all the suggested improvements, a wider co-operation would be necessary that involves other

groups. Since national funding was restricted, this could only be done at an international level. The experts recommended [1.11, 1.12] an IAEA Co-ordinated Research Project (CRP) as the best solution to accomplish such an international co-operation.

### **1.2.3. Scope, goals and tasks of the CRP**

In the preparatory meetings [1.11, 1.12] and the CRP proposal, the scope, goals and tasks of the CRP were defined as follows:

*The overall goals of the CRP should be:*

- To produce complete, consistent yield sets for fissioning systems important for applications. The yields are derived from experimental results and reliable model calculations in a work sharing effort between the relevant research groups.
- To establish a network for continued future co-operation and work-sharing in fission yield evaluations and for communication with yield data measurers.

*The main tasks for the CRP, in order to achieve these overall goals, would be:*

- To co-operate in the compilation of yield data into EXFOR, and in the improvement of the EXFOR (and computation) format and coding rules.
- To review available yield data and empirical models, and to recommend measurements needed.
- To further develop models to achieve more reliable fission yield predictions.
- To improve the whole evaluation process from the compilation of experimental data to the final least squares fitting procedure using all suitable physical constraints and correlations.
- To perform the following special tasks in order to improve the deficiencies of existing evaluations:
  - Produce agreed evaluated sets of reference fission yields for fission products used as standards in measurements or monitors in applications.
  - Compare and test existing evaluated fission yield libraries.
  - Continue and extend the study and evaluation of independent yields and empirical models, in particular isotopic and isobaric yields, isomeric yields and the influence of delayed neutron emission.
  - Check and correct results of fission yield measurements.
  - Study discrepancies among experimental data in detail and try to resolve them.
  - Develop methods and computer programs for the introduction of correlations and covariance matrices in the evaluation process.
  - Study the dependence of fission yields on incident neutron energy with the aim to produce energy dependent evaluated yield sets.

*In summary:*

The main objectives of the CRP were to perform the theoretical studies and special evaluation tasks necessary to overcome the identified deficiencies of evaluations, to improve the whole evaluation process, and to establish a network for continued future co-operation in fission yield evaluation.

The tasks listed above have by and large been fulfilled, and the objectives and goals of the CRP have mostly been achieved. The work and achievements are briefly reviewed in Section 1.3, the full details are presented in the rest of this publication.

#### **1.2.4. Current fission yield evaluations**

##### *General improvements adopted for evaluations*

By fulfilling the tasks of this CRP (see Section 1.3), the ground has been prepared for evaluators to adopt the improvements achieved and to incorporate them in their evaluations. Those adopted already in individual evaluations are reported in Chapter 7, others may follow later as manpower permits. The following improvements were adopted in evaluations during the CRP thanks to the co-operation among CRP participants and to the information presented and exchanged during the meetings.

- The exchange of compiled experimental data and references has resulted in a cleanup of the data bases (completeness of input, identification of multiple references to the same work, corrections to original data, clarifications by private communications, etc.).
- Inter-comparisons of evaluated databases have led to the identification of input errors and data discrepancies and have helped to resolve them.
- An agreed upon list of minimum errors for assignment in the evaluation process was created from an analysis and review of measurement methods and inherent experimental errors (Chapter 2).
- Models are now used for estimating unmeasured isomeric yield ratios.
- All evaluations use empirical charge and mass distribution models for estimating unmeasured yields where necessary and appropriate. During the CRP, the model parameters were periodically updated after the presentation of revisions by A.C. Wahl resulting from the progress in his studies of yield systematics that led to model improvements.
- For the first time, estimated errors of model calculated yields were proposed and introduced in evaluations.
- All processes (decay, delayed neutron emission, etc.) to be accounted for in analyses of measurement results and evaluations, as well as conservation laws (e.g. of mass and charge in fission) requiring constraints and adjustments were reviewed and generally accepted for evaluation procedures.

Table 1.1 presents a survey of data sets in current evaluations considered in this CRP. **Thermal (T)** yields include all measurements at 0.025 eV or corrected for that, or in well moderated thermal reactor spectra which should be corrected for a Maxwellian spectrum at 20°C and a 1/E epithermal neutron flux. **Fast (F)** yields include all measurements in fast reactor spectra (mean neutron energies of 150–500 keV) and fission neutron spectra (mean energies around 1 MeV and above). **High (H)** means neutron energies around 14–15 MeV. **S** stands for *spontaneous* fission.

TABLE 1.1. YIELD SETS INCLUDED IN EVALUATED DATA FILES

Fissioning nucleus	ENDF/B-VI				UKFY2/3 (JEF-2)				CENDL		
	S	T	F	H	S	T	F	H	T	F	H
<sup>227</sup> Th		X									
<sup>229</sup> Th		X									
<sup>232</sup> Th			X	X			X	X		X	
<sup>231</sup> Pa			X								
<sup>232</sup> U		X									
<sup>233</sup> U		X	X	X		X	X	X	X		
<sup>234</sup> U			X	X			X				
<sup>235</sup> U		X	X	X		X	X	X	X	X	X
<sup>236</sup> U			X	X			X				
<sup>237</sup> U			X								
<sup>238</sup> U			X	X			X	X		X	X
<sup>237</sup> Np	X	X	X	X		X	X				
<sup>238</sup> Np			X			X	X				
<sup>238</sup> Pu			X			X	X				
<sup>239</sup> Pu		X	X	X		X	X		X	X	
<sup>240</sup> Pu		X	X	X			X				
<sup>241</sup> Pu		X	X			X	X		X		
<sup>242</sup> Pu		X	X	X			X				
<sup>241</sup> Am		X	X	X		X	X				
<sup>242m</sup> Am		X				X	X				
<sup>243</sup> Am			X			X	X				
<sup>242</sup> Cm			X		X						
<sup>243</sup> Cm		X	X			X	X				
<sup>244</sup> Cm	X		X		X	X	X				
<sup>245</sup> Cm		X				X	X				
<sup>246</sup> Cm	X		X								
<sup>248</sup> Cm	X		X								
<sup>249</sup> Cf		X									
<sup>250</sup> Cf	X										
<sup>251</sup> Cf		X									
<sup>252</sup> Cf	X				X						
<sup>253</sup> Es	X										
<sup>254</sup> Es		X									
<sup>254</sup> Fm	X										
<sup>255</sup> Fm		X									
<sup>256</sup> Fm	X										

The **US evaluation** has increased from 10 yield sets in 1972/4 to over 20 sets in 1978, to 50 sets in 1987, and to currently 60 yield sets. The current evaluation is in ENDF-6 format and is part of the **ENDF/B-VI** library. Each yield set consists of cumulative and independent

yields, comprising a total of about 132 000 yield values and their uncertainties. Corrections have been applied, models have been used to estimate unmeasured yields, and adjustments have been applied to all yield sets as described in Section 7.1.

The first **UK evaluation** of complete yield distributions, UKFY1, contained 15 yield sets, and **UKFY2** includes 39 sets. **UKFY3** has just been completed at the end of the CRP, but had still to be tested before release. It has been produced as part of Mills' thesis [1.13], which contains 193 tables of individual evaluated yield sets, but UKFY3 also contains 39 sets of each, cumulative and independent yields. UKFY2 and UKFY3 yields are both in ENDF-6 format and are adopted as the fission yield files for different versions of the **Joint Evaluated File (JEF)**. Models and evaluation procedures as elaborated during the CRP are being used as described in Section 7.2.

The **Chinese** fission yield file is part of the **CENDL library** and still has the status of 1987. However, a new evaluation is in progress and has been completed for several yield sets (see Section 7.3 for details). They will be converted into ENDF-6 formatted files. Furthermore, **reference yield** sets have been evaluated as described in Section 6.3.

Fission yields can be estimated from the empirical model representing systematic trends in yields and incorporated in the CYI computer program described in Section 4.2.3. The estimated yields can be compared to evaluated yields, many of which are also estimated, and used for fission reactions for which there are no evaluations. Estimated uncertainties for the estimated yields are represented by empirical equations. The systematic trends were derived from data for fissioning nuclides with  $Z_F$  in the 90 to 98 range,  $A_F$  in the 230 to 252 range and excitation energies up to about 20 MeV, and yield estimates should be limited to these ranges.

## 1.3. SUMMARY OF WORK, CONCLUSIONS AND RECOMMENDATIONS

### 1.3.1. Work during the CRP

At the beginning of the CRP, during the first Research Co-ordination Meeting (RCM), the work and discussions of participants concentrated on two main topics: the **distribution of tasks** among participants and **communication** with the **measurer** and **user** communities. Also, ways of co-operation and intercommunication among participants were established.

#### *Distribution of tasks*

The tasks included in the CRP proposal (as listed in Section 1.2.3 above) were adopted for the CRP work. All participants co-operated in the literature search for new measurements, the improvement of EXFOR coding rules for fission yield compilation, and the detection and examination of discrepancies among measurements and evaluations. The evaluators continued their work which comprised collection of relevant literature, compilation of experimental data, (development and) testing of models, and improvements of evaluation procedures (descriptions in Chapter 7). Several special tasks were distributed among participants. The work on these special tasks and results achieved are described in Chapters 2 to 6 and are summarized in the subsequent parts of this Section 1.3.



### *Communication with users of fission yield data*

It was noted that users of fission yield data expressed their dissatisfaction with the data available but often did not specify their needs quantitatively. Furthermore, their source of numerical data used was not known or was not a recommended evaluated set of data. The following **recommendations** were issued **to users** at the beginning of the CRP (see [1.14]) so that users could be advised about the best data to use, and that accuracy targets could be obtained for evaluations developed or improved during the CRP:

Users of fission yield data should consult evaluators or data centres (see Section 3.1) about the best yield data sets and the most recent versions of libraries available.

Users, whose requirements for fission yield data have not been met, should specify their needs in terms of yield sets and types, and of data accuracies, backed up by sensitivity studies.

Users should make their requests public at meetings or through national or international request lists. This may help evaluators get more support for their work.

### *Communication with measurers of fission yields*

Whereas users request recommended yield data and accuracies from evaluators, the evaluators request experimental data from measurers to fill gaps in the evaluations and for development and improvement of systematics and empirical models for charge and mass distributions and isomeric yield ratios. The improvement of model parameters and prediction capability of models depends on the experimental data available. Therefore, a **list of measurements required** was issued right at the beginning of the CRP so that measurement results can be used for the CRP work. These request lists were appended to summary reports before [1.12] and after [1.14] the start of the CRP, in a supplement to the World Request List WREND 93/94 [1.15], and in the report series Progress in FPND [1.16]. Appendix B.3 to this publication (on CD) contains the discrepancy file of experimental data at the end of this CRP, from which the measurements still required can be derived.

Another matter of concern is the **publication of experimental details** needed for a judgement of the measurement results, inclusion in evaluations with appropriate weighting and generation of covariances. Corresponding requests to measurers have been issued in general [1.17, 1.18] and with the type of information specified for fission yields [1.12, 1.14]. The latter is summarized again in Section 2.6 of this publication.

### **1.3.2. Fission yield measurements**

The CRP dealt only with those aspects of fission yield measurements which are of relevance for fission yield evaluation: uncertainties, systematic errors, correlations in general, new techniques, and discrepancies among measured data.

Fission yield measurement methods have been analyzed and information for evaluators has been collected on the type of yields obtained, the problems to pay attention to such as sources and samples used and their treatment, limitations of techniques, data analysis and corrections required, etc. Typical and minimum (inherent) uncertainties were derived for use as error limits in evaluations. New measurement techniques were also presented and discussed. Details of this review can be found in Chapter 2.

Discrepancies among measurements have been reviewed continuously during the CRP and presented in tables together with other unmeasured fission yields (see [1.11–1.16] and appendices to this publication). A success of such a study performed by the CRP was that systematic discrepancies between on-line and radiochemical measurements could be resolved: the reasons for the deviations of on-line measurements were accounted and corrected for, and agreement could eventually be reached [1.19].

### 1.3.3. Compilation of fission yields

The EXFOR system (see Section 3.1) was adopted as most suitable for the compilation and storage of experimental fission yield data, and its use by all compilers/evaluators is recommended. Furthermore, it is recommended to treat EXFOR entries as regular publications and cite them accordingly in reference lists.

CRP participants have helped to improve and update the coding rules for fission yields in EXFOR with regard to which important information on details of the experiments (method, samples, measurement conditions, analysis, corrections, uncertainties etc.) should be included and how it should be coded. In a co-ordinated effort, the EXFOR database has been updated and is now almost 100% complete with respect to fission yields.

Chapter 3 gives more details on EXFOR and a survey about information systems (data bases, their access and retrieval systems).

### 1.3.4. Model calculations

The models discussed here are of *empirical* nature. The equations and parameters used are derived from studies of systematic trends in measured yield distributions. They have been developed because predictions using purely theoretical models for the fission process are not sufficiently accurate and reliable for applied purposes. Models are used in evaluations to obtain numerical values where no yields have been measured, or to check and adjust experimental data to the expected distribution of yields. Full information is provided in Chapter 4.

#### 1.3.4.1. Models for mass distribution

Generally, mass distributions are obtained from measurement results, but models can be used to fill gaps that are too large for linear interpolation. The first proposal was to represent mass distributions by summation of five *Gaussian functions*, which has been associated with fission channels by a theoretical model. In the course of the CRP studies, less or more than five Gaussians have been used for different fission reactions and neutron energies. The model developed can be used for fission reactions in nuclides from Th to Es ( $Z = 90–99$ ) with excitation energy of about 20 MeV and below.

Equations for calculating the uncertainty of yields obtained from model estimates have been proposed (Eq. 4.1-1) and applied successfully. This allows, for the first time, the assignment of realistic uncertainties to estimated mass yields in evaluations.

#### 1.3.4.2. Models for nuclear-charge distribution

Two models for the description of the nuclear-charge distribution in fission have been developed by Wahl:

- The  **$Z_p$  model** describes the isobaric charge distribution of yields and uses fractional independent yields as input.
- The  **$A'_p$  model** describes the isotopic charge distribution of yields and uses independent and chain yields as input. Thus it can generate chain yields in good agreement with measurements.

Both distributions have Gaussian shapes with superimposed deviations due to the odd-even effect of neutron and/or proton pairing.

Both models have been further developed and considerably improved in the course of the CRP and allow better estimates of unmeasured yields. The  $A'_p$  model seems to give more general and consistent results than the  $Z_p$  model. However, studies of the features and parameters of the  $Z_p$  model are much more complete, and therefore it continues to be used in fission yield evaluations. Further investigations to improve the  $A'_p$  model are still required.

Also for charge distribution models have, for the first time, equations been proposed (Eqs. 4.2-2 and 4.2-9) that allow the calculation of realistic uncertainties for estimated independent yields which can be used in evaluations.

#### 1.3.4.3. Isomeric fission yields

Many isomers exist among fission products and play an important role mainly for the calculation of the decay heat after reactor shutdown. However, measured yields or yield ratios are fairly complete only for thermal fission of  $^{235}\text{U}$ . Therefore, models are needed to calculate the partitions of independent fission yields of nuclides among their isomeric states. These models use spin distributions of fission fragments and of nuclear levels as fitting parameters.

Isomeric yield ratios predicted with the most widely used but rather simple model by Madland and England [1.20] failed in some cases to even qualitatively reproduce certain systematic trends derived from some measurements and were found to be the cause for observed discrepancies among different decay heat calculations. Therefore, a new model was developed during this CRP (see Section 4.3). Unfortunately the model was tested only for  $^{235}\text{U}$ , for which good results were obtained. It can be concluded that the model can be used with some confidence where many experimental data exist. Whether the derived parameters and their dependencies can be used for other fission reactions remains to be investigated.

#### 1.3.5. Ternary fission yields

The studies were confined to *tritium* and  $^4\text{He}$  which are important for applications. The analysis of experimental data versus incident neutron energy revealed no significant variation between thermal (0.025 eV) and 2 MeV, the region of interest for reactor applications. Therefore, **energy independence** is assumed in evaluations for that range.

Due to the lack of sufficient experimental data, systematics of the variation of ternary yields with different fission reactions are required. Various formulae for dependencies on fission reactions were tested against recommended data. A formula was found to give best fits and was used to calculate missing  $^1\text{H}$ ,  $^2\text{H}$ ,  $^3\text{H}$  and  $^4\text{He}$  yields. Details are given in Section 4.4.

### 1.3.6. Energy dependence of fission yields

For the development of a model for predicting the energy dependence of fission yields, theoretical studies of the expected dependence to obtain a formula for the model, and systematic studies of measurements to obtain the model parameters are required. Results of such studies performed during the CRP are presented in Chapter 5.

The conclusions from these studies are:

- The accuracy of purely theoretical predictions is insufficient for applications. In fact, they are much less accurate and reliable than the evaluated yields for fast reactor spectra.
- The existing measurements of energy dependencies are too scarce for deriving systematics which are complete enough to develop a semi-empirical model for reliable predictions.

Therefore it is recommended that ‘thermal’, ‘fast’ and ‘high’ (= around 14 MeV) yields continue to be evaluated for data files and used in applied calculations. Possibly the dependencies on fast reactor spectra (mean energies) could be studied in more detail. Many more systematic measurements of the energy dependence and for deriving values of the theoretical model components are required for the development of a reliable model for the low energy range.

### 1.3.7. Correlations of experimental yields

Correlations between different experimental data reflect common systematic influences on the measurement results which have then common systematic errors. They are introduced:

- when the same facility, equipment or method is used;
- through common monitors, normalizations, or ratio measurements;
- the use of the same nuclear data or approximations etc. in the processing of measurement results (such correlations between different experiments are generally ignored).

Correlations exist between individual data from the same experiment and between results from different experiments. They are mathematically represented by covariance matrices.

During this CRP, the use of correlations and covariance information has for the first time been introduced in fission yield evaluation. An established computer code has been adapted and tested for fission yield evaluation as described in Section 6.1. It has been applied successfully for the simultaneous evaluation of fission yields for which not only absolute yields but also yield ratios were measured (Sections 6.1 and 6.3).

### 1.3.8. Reference fission yields

Reference fission yields are defined as yields of fission products which are used as yield standards or as monitor products for applied purposes as described in Section 1.1.2. Relevant fission products were identified during the CRP and assembled in lists.

For deriving the best values for such reference fission yields, they are evaluated carefully from (often selected) measurement results only, without application of any models or constraints, but using correlations and covariance information. Such reference fission yields have been evaluated for  $^{235}\text{U}$  at thermal, fission spectrum and around 14 MeV neutron energy and for  $^{238}\text{U}$  at fission spectrum and around 14 MeV neutron energy as a CRP task. A list of monitor fission products and these evaluated reference yields are given in Section 6.3.

### 1.3.9. Evaluation of fission yields

It is ***emphasized*** that *evaluation is necessary as long as measurements are being made*, so that the results can be digested and incorporated in usable computer files; otherwise data obtained at considerable expense will not become available for practical use. It is ***recommended*** that *independent fission yield evaluations at different places should continue* to be funded and performed, as evaluators employ different approaches and methods, and this is the only way to reveal discrepancies and (input) errors. Therefore, a single evaluation was not recommended or produced by CRP participants.

The improvements of existing evaluations during the CRP and their current status are summarized in Section 1.2.4. Detailed descriptions of evaluations are presented in Chapter 7.

***Auxiliary data*** used in fission yield evaluation were under continuous review during the CRP, and several sets of them were evaluated by participants (e.g. [1.21], others are unpublished). They consist of several groups of data as outlined in Section 6.2: the nuclear data used to calculate fission yields from measured raw data, needed by evaluators for a re-analysis of measurements (corrections for obsolete data); parameters for models and final adjustments; data used for production of cumulative yield libraries and for testing of yield evaluations. Evaluators should make sure that they always use the best data available.

Finally, ***discrepancies*** among experimental data and between evaluated yields were also continuously reviewed during the CRP, which helped to reveal cases of erroneous data analysis, use of bad auxiliary data or simple mistakes. Listings of evaluated yield file inter-comparisons, and discrepant experimental data or (ranges of) yields where measurements are lacking are reproduced in the ***Appendices*** on the CD.

### 1.3.10. CRP achievements and open problems

The main ***achievements*** of the CRP are:

- An international co-operation network of fission yield experts has been established and will be maintained for future efforts. This co-operation helped to achieve considerable improvements in all aspects of the evaluation process (e.g., cleanup of data bases, analysis of experimental data, model development, evaluation methods and procedures).

- Empirical models for mass and charge distributions and for ternary fission yields are available that allow the derivation of corresponding fission yields from thermal to 15 MeV neutron energy with sufficient reliability.
- Some computer programs for the introduction of correlations and covariance matrices in the fission yield evaluation are now available.
- Complete yield sets from different evaluations or special evaluated reference yield sets are available for complete fission product inventory calculations or when using monitor fission products.

The main *open problems* are:

- Theoretical models and systematics of the energy dependence of yields do not yet produce fission yield estimates at different energies that are accurate and reliable enough for applied purposes. For improvement more experimental data are required.
- The isomeric yield model in its present form can produce reliable estimates only where sufficient experimental data are available. For an improved, more general model, many more experimental data are necessary to develop the systematics needed for the derivation of appropriate model parameters.

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## Chapter 2

### TECHNIQUES OF FISSION YIELD MEASUREMENTS AND INHERENT ERROR MARGINS

#### 2.1. INTRODUCTION

Sets of evaluated yields are of importance for a fairly large community of people because fission yields have an impact on different fields of interest.

From a theoretical standpoint they are interesting for the understanding of matter, because they allow the description of the phenomena occurring in a nucleus undergoing large collective motion at low excitation energy and, hence, are influenced by nuclear shells that disappear at higher excitation energies.

From a practical standpoint fission yields are of importance for the design of nuclear reactors and for waste management. The design of alternative reactors may be crucial for the future energy supply of mankind. The yields of "delayed neutron precursors" and their neutron emission probability ( $P_n$ -values) determine reactor dynamics and, hence, the safety of a reactor against prompt criticality. The inventory of fission products in a reactor and their decay characteristics determine the possible hazard of contamination in the case of an accident. They also determine the amount of heat produced by a reactor core after shut down and in consequence the amount of emergency cooling that has to be foreseen. The inventory and the neutron absorption cross sections of the fission products finally determine the neutron economy in a reactor and its possibilities in neutron induced reactions (like incineration of actinides and fission products).

In consequence, the measurements of fission yields have started right after the discovery of nuclear fission and are continuing, thus involving a large number of scientists and a large variety of experimental methods. Each of these methods has its merits and disadvantages, that have to be taken into account by the evaluators and — to a certain extent — this has been done at least for the more common fission reactions. However, a critical user should be aware of possible deficiencies of experimental values. On the other hand every experimentalist producing yield values should publish a detailed error analysis and sufficient experimental details to allow the evaluators and users to verify his approach. This chapter is addressing yield measurers, compilers/evaluators and users and has the purpose to point out the mutual requirements. It will concentrate on fission reactions of practical importance as are available in the current compilations (i.e. essentially neutron induced and spontaneous fission). Other fission reactions, like charged particle or photon induced fission will be mentioned only if the results have some bearing on the present topic. The space available here does not allow a complete coverage of the literature, nor does it allow a detailed treatment of the subject. Readers interested in a more detailed presentation of the matter should refer to recent review articles [2.1–2.3] on fission yields and their measurement. The topics of spontaneous fission [2.4] and of charged particle accompanied (ternary) fission [2.5–2.7] have also been reviewed recently.

#### 2.2. YIELD DEFINITIONS

Fission is a complicated process in which at least 500 different nuclides are produced. Most of these products may be formed in different ways — e.g. in a primary event or by the



$\beta^-$  decay of precursors. They may be produced with different kinetic energies and different internal excitation energies leading to different numbers of neutrons emitted from the primary fragments. In addition, binary fission may be accompanied by third light particles (ternary fission) or in rare events by "cluster" emission of somewhat heavier fragments.

In accordance with the complexity of nuclear fission, different types of fission product yields have to be defined:

- \* **Independent fission yield (%)**: number of atoms of a specific nuclide produced directly (not via radioactive decay of precursors) in 100 fission reactions
- \* **Cumulative fission yield (%)**: total number of atoms of a specific nuclide produced (directly *and* via decay of precursors) in 100 fission reactions
- \* **Chain yield (Mass [number] yield) (%)**: Before this CRP, only the term '(total) chain yield' was commonly known and was used to describe the total yield for a specific isobaric mass chain. The exact definition was, however, not quite clear: it was used for the (sum of) cumulative yield(s) of the last (stable or long-lived) chain member(s) as well as for the isobaric sum of independent yields, and also for some yield types in between, e.g. the sum of cumulative yield of a product towards the end of an isobaric chain and the independent yield of its daughter (used for fractional yield determinations — see there). A complication arises due to  $\beta$ -delayed neutron emission: cumulative and — in consequence — chain yields are suffering increases by neutron emission from heavier mass chains and losses to lighter mass chains. In consequence, cumulative yields and chain yields are no more identical to the sum of the independent yields of their precursors. Following a proposal of A.C. Wahl [2.8], two new clear definitions of this type of yields emerged from this CRP:

The **(total) chain yield** is defined as the (sum of) cumulative yield(s) of the last (stable or long-lived) chain member(s). The chain yields apply for fission "products" after the emission of prompt neutrons that takes place in a time of about  $10^{-14}$  s after scission. They are obtained in classical mass spectrometric measurements of long lived and stable end products of mass chains.

The **mass (number) yields** are defined as the sum of all independent yields of a particular mass chain and are this way distinguished from chain yields. It should be made clear that the sum of independent yields is a fictitious number, not realised in most measured yield distributions. Its importance lies, however, in fundamental studies of the fission process and in the development of fission yield systematics. Some of the most modern methods to measure fission yields (e.g. using Lohengrin, to be discussed below) provide sets of truly independent yields which — at summation — will produce "mass number yields" rather than chain yields. In principle, these values would have to be corrected to produce real chain yields. In practice, such correction factors are of importance only for a few mass chains (e.g.  $A = 84, 85, 136$ ).

Some physical methods allow the measurement of yields prior to prompt neutron emission. These yield distributions are designated as "fragment-yields".

- \* **Fractional independent/cumulative yields** represent the independent or cumulative fission yield divided by the chain yield (or mass number yield) (%). A problem similar to the one

discussed above (chain yields) exists: It should be pointed out in every case affected by delayed neutron emission whether the fractional yield refers to chain yield or the sum of independent yields of that chain. Frequently, this is however obvious from the method used.

- \* **Partial yields:** Some methods of measurement provide yields for a specific condition (like a specific kinetic energy or a specific ionic charge state of the fission fragments). Such yields are called partial yields. Only a summation over the whole kinetic energy distribution (or ionic charge distribution) will provide the yields discussed above.

A particular problem concerns "isomeric formation ratios". In some nuclei the  $\gamma$ -ray de-excitation is hindered by a high spin difference between the initial and final level, bringing about a delay in this de-excitation process to be described by an exponential law with half-lives from microseconds to years. In these cases the division of the total independent yield among the isomeric states is of interest and may be described by the "isomeric formation ratio" (independent yield of metastable state(s) divided by the independent yield of the ground state) or by the "fraction of high spin isomer" (independent yield of high-spin isomer divided by the total independent yield of that nuclide). The evaluation of measurements is frequently complicated by the fact that the  $\beta^-$  decay of the precursor may feed both isomers and the decay of the metastable state may occur not only by "isomeric transition" but also by  $\beta^-$  decay. Frequently the corresponding "branching ratios" are not well known.

The following discussion will be dedicated to some general problems of yield measurements like the determination of the absolute fission rate, interference by other nuclear reactions of the fission products and the problems associated to the energy spectrum of the neutrons in nuclear reactors.

## 2.3. GENERAL PROBLEMS IN FISSION YIELD DETERMINATION

### 2.3.1. Absolute fission rate/reference yields

It results from the definitions given above that the fission yields refer to a number of fission reactions. As a consequence, the determination of the absolute fission rate is a prerequisite of a fission yield measurement. The early fission scalars [2.9] — thin layers of the fissile material inside an ionisation chamber counting directly the large fission pulses — are difficult to introduce into a normal irradiation position and may induce local changes of the neutron flux. In consequence, the absolute yields of (only) a few fission products with well known decay properties which are easily accessible have been measured with great care. Once the absolute fission yield of such a reference nuclide is known, the number of atoms of this nuclide isolated from a sample allows the calculation of the absolute number of fission reactions that this sample has undergone and hence the absolute fission yields of the other nuclides isolated from the same sample. The error margins of reference yield values are assessed in Section 6.3 of this volume.

In principle, all mass or chain yields of a particular fission reaction have to add up to 200%. For well studied fission reactions with experimental values for (nearly) all the mass chains this may serve as an independent check. Evaluators have carried out such comparisons and found fair agreement. For the evaluated data files discussed in Chapter 7 a normalisation was carried out forcing the sum of the chain yields to be 200%.

### 2.3.2. Nuclear reactions of fission products

Some fission products have high neutron capture cross sections ( $^{135}\text{Xe}$ ...). In yield measurements this has therefore to be taken into account in a twofold manner: In irradiations with a high neutron flux the yields of  $^{135}\text{Xe}$  and of the subsequent chain members may be found low; yields of  $^{136}\text{Xe}$  may be found high. Corresponding problems with the fission yield standard (reference) nuclide  $^{148}\text{Nd}$  due to neutron capture in  $^{147}\text{Nd}$  were pointed out by Maeck [2.10]. The isotope  $^{134}\text{Cs}$  is generally produced almost exclusively by neutron capture in  $^{133}\text{Cs}$ , because its independent fission yield is small and it is "shielded" from  $\beta^-$  decay by stable  $^{134}\text{Xe}$ . Such reactions have to be taken into account when using fission yield values, e.g. in reactor inventory calculations. In fact, the pertinent codes, like ORIGEN [2.11] and KORIGEN [2.12] take care of this problem to a certain extent.

### 2.3.3. Neutron energy spectra

Neutrons in a reactor are not monoenergetic but show a distribution ranging from about 10 MeV down to thermal energy. Three components of a reactor neutron flux may be distinguished. Each component has a well described shape and only the contribution of each component to the total flux varies.

- (1) The flux of unslowed neutrons ( $\phi(E)$ ) originating from the fission process show a distribution from about  $E = 1\text{--}10$  MeV, which can be described by Watt's equation [2.13]:

$$\phi(E) = 0.484 e^{-E} \sinh \sqrt{2E}$$

- (2) The flux of "epithermal" neutrons ( $\phi(E)$ ) on the way to thermal energy is generally described by a  $1/E$ -law:

$$\phi(E)' = c/E$$

- (3) The flux of thermal neutrons is best described by the well known Maxwellian distribution valid for the velocity or energy distribution of molecules in a gas.

Generally, the contribution of fast ("fission") neutrons (1) is negligible because the cross sections at these energies are small. Epithermal neutrons (2) have, however, to be considered because some fissioning nuclei and some reference nuclei show large resonance integrals [2.14]. This is particularly true for gold which is frequently used as a neutron flux monitor. Due to a high resonance integral, half of the activity of a gold monitor may be due to epithermal neutrons. In consequence, gold should not be used for normalisation of irradiations in different locations of a core or among different reactors. In these cases the normalisation should be carried out using fission product standards such as  $^{99}\text{Mo}$  or  $^{140}\text{Ba}$  as monitors. In publications of experimental yield determinations at least a minimum characterization of the neutron spectrum used should be given (e.g. the cadmium ratio).

### 2.3.4. Self shielding

Self shielding of samples during irradiation is generally considered and samples are made sufficiently thin to avoid this problem. For example, samples of  $^{235}\text{U}$ , which has a

fission cross section ( $\sigma_f$ ) of 582 barn ( $10^{-24}$  cm<sup>2</sup>) must be thinner than 50 mg/cm<sup>2</sup>. In some cases the request may be even more stringent, like <5 mg/cm<sup>2</sup> for <sup>242m</sup>Am with  $\sigma_f = 7000$  barn. For gold (as a monitor), not only the thermal (n, $\gamma$ ) cross section (98.6 barn) has to be taken into account but also the large resonance integral with a sharp resonance of nearly 30000 barn at a neutron energy of 5 eV.

In the following, the classical techniques used for yield measurements will be discussed briefly and specific sources of error will be pointed out. The discussion will be in the order of the different techniques. For each technique the application to the measurement of chain yields will be discussed first and then — in case the technique is suitable for measuring independent yields — the additional problems associated with these measurements will be treated. Very generally, the basic difference between the measurement of chain yields and independent yields is time. In the case of chain yields for most techniques one has to wait until the precursors have decayed to the last radioactive or even stable member(s) of the  $\beta$  decay chain, whereas for independent yields the measurement has to be done sufficiently quickly so that the initial nuclear charge distribution can be observed or at least reconstructed.

## 2.4. SINGLE EXPERIMENTAL TECHNIQUES

### 2.4.1. Radiochemical methods

#### 2.4.1.1. Chain yields

Historically, the first methods for yield determinations were radiochemical. For mass yield determinations the last (or the longest-lived) radioactive member of the  $\beta$  decay chain was isolated radiochemically from the irradiated sample of a fissile nuclide after a waiting time which allowed all precursors to decay into this nuclide.

In order to determine the yield of the chemical isolation (chemical yield), a known amount of the element to be separated was added to the irradiated sample and the fraction collected on the sample for counting was determined. The methods for the chemical yield determination could be any conventional analytical method, neutron activation analysis, X ray fluorescence or radioactive counting — in case a radioactive isotope of the element of interest (not produced in the fission reaction) had been chosen as a monitor. For the determination of the chemical yield, it has to be made certain that the element added behaves like the fission products of the same element. This is not certain if different oxidation states exist, like in the case of iodine where iodide added to the sample may precipitate completely with silver nitrate, but fission iodine present in the form of iodate would remain in solution. In such cases a complete oxidation/reduction cycle would establish "complete chemical exchange". Similar problems exist with elements that tend to form colloids or adsorb on the walls of vessels (Nb, Zr etc.). Such problems have to be taken into account by evaluators and users of fission yields.

After chemical isolation the absolute activity of the fission product has to be counted. In the early days  $\beta$  radiation was used. This non-selective radiation imposes very strict requirements to the chemical separation. Experimental difficulties are also due to many corrections needed to obtain absolute counting efficiencies (absorption and back-scattering problems). The existence of several isotopes and of radioactive daughters requires a decay curve analysis that may introduce large uncertainties, especially when several components of similar half-life have to be analysed. For this reason  $\beta$  counting was abandoned for fission

yield determination in favour of  $\gamma$  ray measurements after the advent of high resolution  $\gamma$  ray spectroscopy. Values derived from  $\beta$  counting should be checked carefully and should be discarded from evaluations in case of doubt.

In the case of  $\gamma$  ray measurements, a frequent problem is that the absolute intensities of the  $\gamma$  rays are not well known — even published values are subject to later correction. Whenever isomers exist in a decay chain, the decay branching of the precursors to the isomeric states as well as that of the metastable state between isomeric transition and  $\beta$  decay influence the evaluation. Updates of nuclear decay properties are evaluated, compiled and published for single mass chains in regular intervals [2.15]. Actually, the updated information has been summarised recently in a comprehensive table of isotopes [2.16]. Therefore it is important that experimenters give the necessary details to allow a recalculation of their decay analysis with improved decay data.

Finally, the absolute count rate has to be related to the absolute fission rate which has to be determined as discussed above using fission counters, standards, monitors etc.

#### *2.4.1.2. Independent yields*

In the measurement of independent yields, the problems are practically identical but generally more stringent. A basic difference is that the irradiation has to be short and the chemical isolation has to be performed sufficiently rapidly so that the precursor in the decay chain has not decayed to a disturbing amount. For this purpose fast chemical separation methods have been developed. They are described in several review articles [2.17–2.19]. The application of such fast chemical separations to fission yield determinations is discussed in [2.20] and [2.2]. It is obvious that conditions, like a complete chemical exchange, are more difficult to establish in a short time. The radiochemical method allows in principle the determination of isomeric yields, provided that the half-lives of both isomers and their precursor are sufficiently long and the decay data (like branching ratios) are known. On the other hand, for the determination of the nuclear charge distribution frequently an absolute measurement can be avoided by measuring a fractional yield. In the case of a decay chain  $A \rightarrow B \rightarrow C$ , a fast separation between A and B and — after decay of A and B to C — a measurement of the distribution of C among the chemical fractions "A" and "B" would provide the fractional cumulative yield of A and the fractional independent yield of B — provided the independent yield of C is negligible. In such measurements, errors due to decay characteristics and calibration problems cancel.

### **2.4.2. Classical mass spectrometry**

#### *2.4.2.1. Chain yields*

In classical mass spectrometry a sample of fission products is introduced into the ion source of the mass separator and heated, and the evaporated elements are ionised and separated according to their mass [2.21, 2.22]. A problem of these — generally very precise — measurements is that yields for each element are relative and need to be normalised to obtain absolute values. Often the normalisation is the major source of uncertainty in absolute yields. Another disadvantage of the method, when measuring stable or long-lived end products of a decay chain, is that large amounts of fissionable material and long exposure

times are required in order to produce amounts of the investigated isotopes sufficient for detection with a conventional detector. There is also the problem that in long times of exposure to high neutron fluxes, the abundance of fission products may be changed due to some interfering reactions as discussed above. Contamination by stable nuclides and their reaction products is another problem. However, generally the accuracy of these measurements is very good and the relative errors do not exceed a few per cent [2.23].

#### *2.4.2.2. Independent yields*

For the determination of independent yields, the transfer of the fission products to the ion source has to be made as quickly as possible. Therefore frequently on-line systems are chosen, where the target is directly connected to the ion source or is even mounted within the ion source. The problems differ from those encountered in chain yield determinations. Whereas the problems due to secondary reactions disappear in the short irradiation times required for independent yield determinations, new serious problems arise because different elements show different volatilisation yields and volatilisation kinetics from the target and are ionised to a different degree in the ionisation source. These problems are further complicated because every isotope of one element has a different kinetics of formation and decay, due to the different half-lives and fractional yields of the chain members. In some cases an attempt has been made to volatilise only specific elements like, e.g., alkali elements from a graphite target [2.24]. In another approach high temperature ion sources ( $T = 2500^{\circ}\text{C}$ ) were developed [2.25, 2.26] with the aim to obtain nearly complete volatilisation within a time short with respect to the half-life of the isotopes measured. In addition, the release and ionisation mechanisms were studied by Rudstam and Kirchner [2.27, 2.28]. The technique has been used by Rudstam et al. [2.29] within the framework of this CRP to measure systematically a large part of the independent yield distribution in the fission of  $^{235}\text{U}$  including the region of symmetry and many isomeric pairs.

### **2.4.3. Direct $\gamma$ ray spectroscopy**

#### *2.4.3.1. Chain yields*

Direct  $\gamma$  ray spectrometry is a suitable method to obtain a wide range of mass yields mainly for exotic targets of limited availability. In this method, a sample of the fissile material is irradiated for varying lengths of time and the decay of the fission products is followed by taking sequential high resolution  $\gamma$  ray spectra. The advantages of the method are the requirement of only small amounts of the fissile nuclide (mg-amounts) and the short time needed to obtain the complete range of the more important yields. The method has the disadvantage of a complicated data analysis relying on the decay characteristics of the nuclides measured that are frequently not well known. The accuracy of the method is limited ( $\pm 10\%$  at best). Low yield regions are not accessible without additional chemical separation. The method has nevertheless been used extensively. A survey on different applications [2.20] (with 40 references) has been given in 1986.

#### *2.4.3.2. Independent yields*

The application of this method to the determination of independent yields is even more difficult, because: (1) the spectrum of fission products — and hence the  $\gamma$  ray spectrum — is

much more complicated; (2) the duration of counting is limited due to the shorter half-lives of the more unstable fission products formed directly. Even though a cycle of irradiation and a subsequent counting program can be repeated many times in this non-destructive method, the short counting times limit the statistical accuracy of the spectra obtained. Nevertheless, Dickens et al. [2.30–2.36] have studied several fissioning systems with irradiation times down to 4 s. Because the evaluation is based on the growth and decay curves of mother/daughter systems the results do generally not depend on  $\gamma$  ray line intensities, that are frequently not well known for these short-lived nuclei. However, the accuracy is limited and data derived from this technique are useful only if data from the more accurate techniques discussed in the following are absent.

#### 2.4.4. Measurements of unstopped fission fragments

##### 2.4.4.1 Chain yields/prompt neutron emission

In low energy fission — when the momentum of the inducing particle may be neglected — the law of momentum conservation allows one to calculate the masses of two complementary fission products when the corresponding kinetic energies or velocities of the fragments are measured. Basically all the combinations possible to determine yields have been used: double energy, double velocity, single energy plus single velocity measurements [2.3, 2.37, 2.38]. In all these cases it is important to have a thin fission source in order to avoid a disturbing energy loss within the source. In all arrangements — except the single armed measurement of energy and velocity of the same fragment — it is also important that the backing of the fission target be thin. Technically, fragment energies may be measured using surface barrier detectors or ionization chambers. Velocities may be determined using a system of a start and stop detector and a flight tube.

A problem arises due to prompt neutron emission which takes place isotropically in the centre of mass system while the fragments are in flight. The effect is twofold:

- (a) The (relative) loss of mass of the fragment, according to  $E = 0.5 mv^2$  leads to a (relative) loss in kinetic energy of

$$\Delta E/E = \Delta m/m = v/A$$

if  $v$  is the number of neutrons emitted. The number  $v$  is distributed around a mean value  $v(A)$  that ranges from 0.5 to 3 as a function of fragment mass. In some cases neutron multiplicities up to 8 are observed. The masses  $A$  range from 70 to 170. In consequence,  $\Delta E$  may reach a few MeV, i.e. several per cent of the fragment kinetic energy.

- (b) The recoil energy ( $E_R$ ) transferred to the fragment by the emission of the neutron (of about 1 MeV in energy) adds vectorially to the energy of the fragment. The absolute amount of this energy is much smaller, namely (for one neutron of 1 MeV)

$$E_R = (1/A) \times 1 \approx 0.01 \text{ MeV}$$

Measurements of fragment **energies** are affected by both effects. **Velocity** measurements are subject only to the recoil effect (b), which induces no systematic error but leads to a widening in velocity and energy distributions. In consequence, velocity experiments are

inherently more accurate. Until recently, however, this advantage was generally compensated in practice by a less good resolution of the velocity measurement.

Appropriate corrections for prompt neutron emission and — in the case of energy measurements — for pulse height defects in the detectors allow the calculation of mass (chain) yield distributions from both types of measurement. The advantages of this approach are:

- (1) A range of chain yields of a total mass region is obtained in a single experiment
- (2) The requirement for fissionable material is very small (often a few  $\mu\text{g}$  are sufficient). This is important for the study of some rare, exotic nuclei.
- (3) The experiment provides simultaneously the information on the kinetic energy of the fission fragments.
- (4) Contrary to the radiochemical measurements this experiment provides the yields of **fragments** (prior to prompt neutron evaporation). The combination of the mass distributions of fragments and products allows the calculation of the number of prompt neutrons evaporated from each mass pair.

A disadvantage of the method is that poor mass resolution leads to high values for low yields and low values for high yields wherever yields vary strongly with mass, e.g. around fine structures and on the flanks of the mass distributions.

A recently developed instrument based on the measurement of the energy and velocity of the fission fragments with a considerably improved resolution is ***Cosi fan tutte*** [2.39, 2.40]. It has a mass resolution sufficient to separate single masses among the light fission products. A similar system under construction in Moscow should also be mentioned here [2.41].

Another interesting variant of this technique is realized by the one-armed instrument ***Hiawatha*** which also measures the velocity and energy of one fragment [2.42]. The velocity is determined by a flight time measurement. The energy is determined by an electrostatic analyser and a surface barrier detector. The analyser has an energy resolution of 0.3% (FWHM), which is better than that achieved by a surface barrier detector. However, because the analyser deflects particles on the basis of the ratio of kinetic energy ( $E$ ) to ionic charge ( $q$ ), and the fragments show a spectrum of charge states, the selection is not unequivocal and the surface barrier detector is required in order to select between different combinations of  $E/q$  by measuring  $E$ .

The third and most elaborate approach using unstopped fission fragments is the mass separator ***Lohengrin*** [2.43, 2.44]. It uses magnetic and electric sector fields, providing a true isolation of fission products as opposed to the instruments discussed above that provide only a mass identification of single fragment pairs. In consequence, fission fragments of one mass can be collected with Lohengrin and may be studied, e.g. by  $\gamma$  ray spectroscopy.

The separation takes place according to the ratio ( $A/q$ ) of mass to ionic charge state. The charge states of the ions range from about 15 to 30 and result from the dynamic equilibrium that arises when fission fragments with their full kinetic energy (of 50 to 120 MeV) are moving through the target material. The resolution of the separator is sufficiently good to allow in general the isolation of single masses in spite of the multiplicity of the  $q$  values.



The electric sector field of Lohengrin separates the fission fragments according to the ratio of kinetic energy to ionic charge state ( $E/q$ ).

In some cases of identical or very similar  $A/q$ -ratios (e.g. 100/25, 96/24, 92/23 etc.) Lohengrin is not able to separate and delivers a multiplet. Such a multiplet can be resolved in a surface barrier detector or an ionization chamber attached to the collector side by measuring the total kinetic energy of the fragments because the different masses of the multiplet transmitted through Lohengrin possess identical velocity and — hence — different kinetic energy.

A general practical difference between the three approaches based on the measurement of unstopped fission fragments should perhaps be stated here explicitly:

- (1) In the classical instruments based on energy/velocity measurements including Cusi fan tutte, all the fission fragments (irrespective of their mass, nuclear or ionic charge state or kinetic energy) falling into the acceptance angle of the detectors are recorded with their pertinent parameters (kinetic energies of coincident complementary fragments, flight times etc.). The events are sorted electronically and added into the appropriate bins of mass, nuclear charge and kinetic energy either on line or subsequently.
- (2) In Hiawatha, the electrostatic analyser selects fragments with a specific ratio of kinetic energy ( $E$ ) to ionic charge state ( $q$ ), discarding all fragments with other ratios of  $E/q$ . Therefore, for the complete study of one fission reaction, the voltage applied to the electrostatic analyser has to be varied systematically and the measurements corresponding to the different settings have to be analysed separately and sorted into the appropriate bins after the end of the series of measurements.
- (3) Lohengrin is even more restrictive than Hiawatha: Only fragments with a certain ratio of  $E/q$  *and*  $A/q$  ( $A$  = mass of fragment) are detected in one run. In consequence, for the measurement of the yield distribution of a fission reaction many single measurements (with different settings of the magnet and electric condensor) have to be performed varying systematically the ionic charges and kinetic energies of the fragments selected.

In all three cases the calculation of yields requires an integration over the distribution in kinetic energies of the fragments. For Hiawatha and Lohengrin in addition an integration over the ionic charge distribution is required. This point will be raised again later during the discussion of independent yield measurements.

In spite of its less effective use of beam-time (due to the requirement of multiple settings) Lohengrin has contributed more complete data sets on mass- charge- and energy distribution than any other method. This may be attributed to its perfect mass resolution, due to the fact that it is not based on momentum conservation and, hence, that it is not influenced by prompt neutron evaporation.

#### *2.4.4.2. Independent yields*

Cusi fan tutte, Hiawatha and Lohengrin can be equipped with a detector identifying the nuclear charge of the fission fragments. The principle of this  $Z$ -identification is as follows: the stopping power of matter for fast heavy ions depends, inter alia, on their atomic number  $Z$ : a higher  $Z$  of the heavy ion causes a stronger interaction with the electronic shells of the atoms

of the stopping material. The attachment of Z-identification to the three instruments, therefore, allows the simultaneous determination of the distribution of mass, nuclear charge and kinetic energy of fission fragments within a time scale of microseconds.

For practical realisation of the Z-identification, different variants have been used that are all based on the same principle: a specific energy loss ( $dE/dx$ ) of the fast fragments when passing through a thin layer of matter (solid or gaseous) in which the fission fragments deposit about half of their kinetic energy. This matter can be an active absorber in the form of a thin (uniform!) surface barrier detector (about 8  $\mu\text{m}$  thick), providing directly the energy deposited by the fragment [2.45]. It can also be a passive absorber like a stack of 160 very thin carbon foils [2.46, 2.47] or a single foil (parylene C) [2.48]. In these cases the energy loss is determined by measuring the remaining energy either by a time of flight method [2.46, 2.47] or by a direct measurement in an ionization chamber [2.49, 2.50] and by subtracting it from the initial kinetic energy as determined by the mass separator Lohengrin. These arrangements have the disadvantage of using only a small part (a few mm) of the total length of the exit slit of Lohengrin (of 70 cm). A more recent development of a "Big Ionization Chamber, BIC" [2.51] allows the use of up to 40 cm of the exit slit and, this way, offers a considerable increase in sensitivity allowing measurements in regions of low yield as will be discussed. The big ionization chamber has a split anode that allows a simultaneous measurement of the energy loss  $\Delta E$  in a first part of the chamber and the remaining energy  $E_R$  in the second part of the chamber. The two values can be summed to give the total energy of the fragment. A last example for the different variants used is the development of an axial or "Bragg" ionization chamber [2.52], as has been attached to Cosi fan tutte. In an axial chamber, the ions travel parallel to the electric field lines between the two plane electrodes. The fission fragments enter at the cathode and come to a stop close to the grid in front of the anode. The arrival of the electrons — and therefore the pulse shape — at the anode as a function of time reflects the energy deposition of the fragment along its path through the ionization chamber and is correlated to the specific energy loss and total kinetic energy of the fragment. It allows one to obtain its mass and nuclear charge  $Z$ . The  $Z$ -resolution of all of these variants is not very different. It depends not only on the properties of the detectors, but also on the nuclear charge and kinetic energy of the fragments. A typical example of a spectrum of different members of a nuclear decay chain is given in Fig. 2.1.

As it appears from Fig. 2.1, the more important elements contributing to the total mass chain can be well separated. A fit of (equidistant) Gaussian normal distributions provides the respective contributions. As obvious from the figure, contributions of fractional yields smaller than about 5% are however difficult to ascertain and should be treated with caution.

Unfortunately, the  $Z$ -resolution of the method at the typical kinetic energies of fission fragments is only good enough to distinguish single isotopes up to about  $Z = 47$ , i.e. for the light mass peak. This problem can, however, be overcome to a certain extent as — except for the rare events of ternary fission — the  $Z$ -distribution of light and heavy mass peak are complementary with respect to  $Z_F$ , the atomic number of the compound nucleus, and, hence, a measurement of the light fission products provides the information on the complementary elements. However for single nuclide yields a correction for prompt neutron emission ( $v(A)$ ) has to be carried out and this correction results in another loss of mass resolution because  $v(A)$  represents a mean of a distribution.

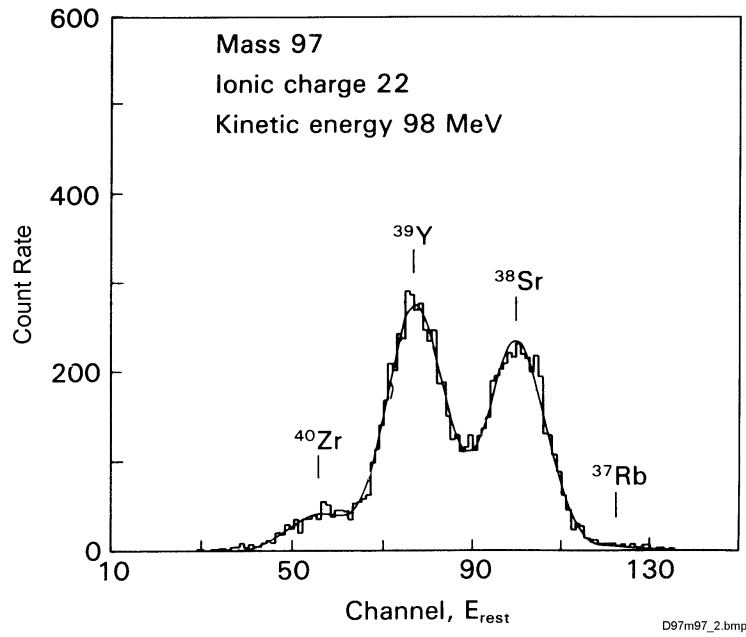


FIG 2.1. Pulse height spectrum of the residual energy of fission products of mass 97 (initial  $E_{kin} = 98$  MeV, Ionic charge  $q = 22$ ) after passing through an absorber foil of parylene C. The experimental data are shown in histogram form. The smooth curve is a computer fit of four normal "Gaussian" curves. The areas under these bell shaped curves represent the relative independent yields of the isotopes indicated (after Quade et al. [2.49]).

The time of analysis is given essentially by the flight time through the separator and is in the order of microseconds. Hence, it is short relative to the time of  $\beta$  decay ( $>100$  ms). Therefore, these methods provide automatically an undisturbed primary distribution. Strictly speaking the summation of the independent yields of a decay chain in these measurements does not provide chain yields but the "sum of independent yields" which may be converted to true chain yields after correction for delayed neutron emission (see above: "yield definitions": chain yields).

As discussed already briefly in the context of mass yields for Lohengrin and Hiawatha, measurements as shown in Fig. 2.1 have to be repeated for various (about 5) different values of the kinetic energy of the fragments in order to determine the energy distribution of the yields. In practice the energy distribution can generally be described fairly well by a Gaussian normal distribution with possibly a low energy tail due to inhomogeneities of the targets.

In addition, in the case of Lohengrin the measurements as shown in Fig. 2.1 have to be performed for various ionic charge states of the fragments. Again, normally the ionic charge distribution can be described by a Gaussian normal distribution. An example is shown in Fig. 2.2 for the mass chain with  $A = 84$  and for the main isobars of that chain. In some cases, however, deviations from the normal behaviour are observed. Such a case is shown in Fig. 2.3 (chain with  $A = 86$ ). In this case the element with  $Z = 35$  ( $^{86}\text{Br}$ ) shows an increased ionic charge.

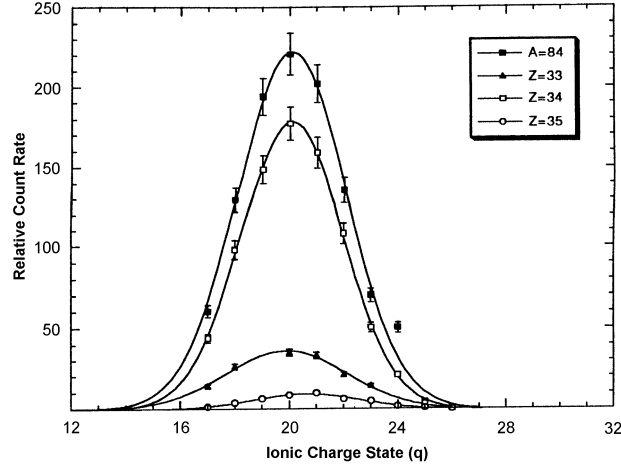
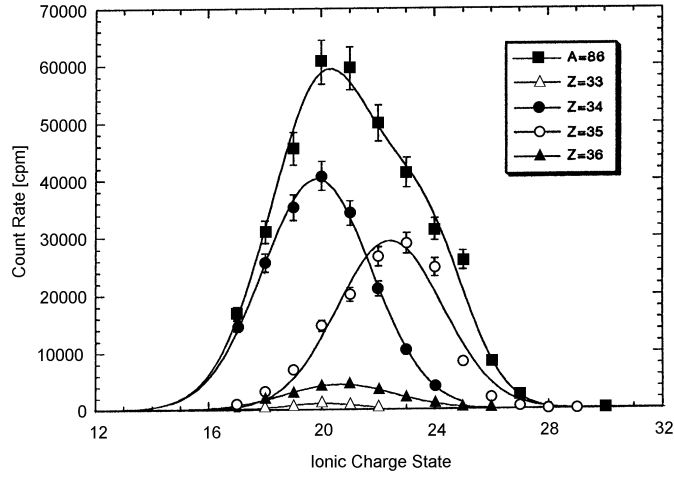


FIG. 2.2: Relative count rates of  $^{84}\text{Br}$ ,  $^{84}\text{Se}$ , and  $^{84}\text{As}$  and of all fragments of mass 84 (curves in ascending order) for various ionic charge states of the fragments during separation in Lohengrin from [2.53].



D97m86\_2.bmp

FIG. 2.3: Relative count rates of  $^{86}\text{Kr}$ ,  $^{86}\text{Br}$ ,  $^{86}\text{Se}$ , and  $^{86}\text{As}$  and of all fragments of mass 86 (curves in ascending order) for various ionic charge states of the fragments during separation in Lohengrin from [2.53].

This is due to Auger cascades taking place in the  $^{86}\text{Br}$  ions after they have left the target and while they are travelling in the vacuum of the separator. Depending on the contribution of the Auger cascades, the increase in ionic charge may affect only part of the nuclide concerned producing 2 overlapping normal distributions. Fortunately, this complication is characteristic only for specific fission products and affects different fission reactions in the same way.

In any case, for the calculation of fission yields the relative count rates have to be integrated over both the distributions of kinetic energies and ionic charge states. The resulting count rates of different nuclides reflect relative yields and can be converted to absolute yields, when the absolute yield of at least one nuclide and its relative count rate are known.

The fact that the sum of all count rates has to correspond to 100% yield offers another possibility for normalisation, when complete sets of yields of, e.g. the light fission products are determined.

The method has been used to measure (nearly) complete sets of the (light mass) fission products in the neutron induced fission of the following nuclides:  $^{233}\text{U}$  [2.54],  $^{235}\text{U}$  [2.55],  $^{239}\text{Pu}$  [2.56], and  $^{249}\text{Cf}$  [2.57] using Lohengrin,  $^{229}\text{Th}$  [2.58] and  $^{241}\text{Pu}$  [2.59] using Cosi fan tutte, as well as  $^{233}\text{U}$  [2.60] and  $^{235}\text{U}$  [2.61, 2.62] using Hiawatha.

A limitation of the method is the fact that small *fractional* yields ( $\text{YFI} < 5\%$ ) cannot be obtained with good accuracy, due to the limited resolution in  $Z$ , as may be seen already by inspection of Fig. 2.1. This problem has some bearing especially on the calculation of delayed neutron yields. On the other hand, there is no problem to measure yields in regions of even very small *mass* yields as has been demonstrated for the thermal neutron fission of  $^{235}\text{U}$  [2.63],  $^{239}\text{Pu}$  [2.53], and  $^{249}\text{Cf}$  [2.64].

Another limitation of the method of specific energy loss is that naturally it cannot differentiate between isomeric states of a nuclide (as they do not differ in  $Z$ ). In the case of Lohengrin, isomeric ratios can be measured by intercepting the separated fragments and submitting them to a  $\gamma$ -spectroscopic measurement [2.65]. Otherwise for the measurement of isomeric pairs the traditional methods based on radiochemistry and on mass separation with a high temperature ion source discussed above remain of primary importance.

## 2.5. OUTLOOK: NEW TRENDS AND DEVELOPMENTS

### 2.5.1. Multiparameter measurements

Two recent trends in measurements of yields (formation cross sections) originate from the goal of fundamental studies of the fission process but may turn out useful for acquiring data for practical purposes dealt with in the present CRP.

The first one is the tendency to characterize as completely as possible every single fission event and to store the multiparameter information. Such a complete characterization would involve the masses of the complementary fragments, their nuclear charges, any ternary charged particles, the number of neutrons and the number of  $\gamma$  rays emitted in the fission event, the angular distribution of ternary particles, prompt neutrons and  $\gamma$  rays and the corresponding energies. A first step in this direction goes back to C. Butz-Jørgensen and H.-H. Knitter [2.66]. The fission fragment detectors of a twin ionisation chamber were supplemented by a scintillation neutron detector. Recently a number of variants of this approach have been or are being developed: For neutron and  $\gamma$  ray detection, Gd-loaded scintillation tanks [2.67–2.70], and a sphere of 162 NaI-detectors (Heidelberg-Darmstadt Crystal-Ball-Spectrometer) [2.71] were used. If all the neutrons emitted in every fission reaction could be detected, this would allow the production of complete yield data sets for every fission reaction studied. Unfortunately, so far the efficiency for neutron detection in these systems is not 100% yet and some neutrons may be registered in more than one segment of the detector ("cross talk").

### 2.5.2. Fission in inverse kinematics

Another new approach is to study "fission in inverse kinematics" [2.72–2.74]. In classical fission studies a target of fissile material is exposed to neutrons or accelerated light particles (protons, photons etc.). For fission in inverse kinematics the fissile nuclides are projected at high (relativistic) energies on a still-standing target of varying composition. A transfer of excitation energy occurs to the fissile ions when they pass through the target and they subsequently undergo fission — in flight. An important difference to classical fission is that the fission fragments carry a kinetic energy which results from the vectorial addition of the fission energy (60 to 100 MeV) and the momentum of the incoming fissile nucleus (e.g. 100 GeV). In consequence, the kinetic energy of the fission fragments is much higher and the fission fragments form a beam of a small divergence and therefore may be analysed in a fragment separator and by time of flight instrumentation quite effectively. In addition, at these high kinetic energies the resolution of the detectors for Z-identification by specific energy loss discussed above for Lohengrin, Hiawatha and Cosi Fan Tutte is considerably better than at the energy of about 1 MeV per nucleon as provided by the fission process alone.

Another advantage of "fission in inverse kinematics" is that the fission of almost any nuclide (whether stable or unstable) in the region above lead can be studied. For this purpose a primary relativistic beam of, e.g.  $^{238}\text{U}$  can be interacted with a first target (e.g. of copper, 1 g/cm<sup>2</sup> thick). This results in the ablation of one to a few neutrons and protons from the projectiles and in consequence to a spectrum of reaction products with lower nuclear charge and neutron number than the projectile. Any one of these products can be isolated by in-flight mass separation and submitted to fission in a second target as described above. The fission properties of any of these nuclides — even inaccessible up to now due to a very short half life or other reasons — may be investigated. This would allow systematic studies not possible otherwise. Such studies would certainly be very interesting in the context of the new projects of the accelerator driven nuclear reactors and of the incineration of actinides in nuclear reactors and, actually, a patent has been sought [2.75].

It should, however, be pointed out that the technique of "fission in inverse kinematics" has the problem that the excitation of the fissioning nuclei is due to two different processes (electromagnetic excitation of the giant resonance, "EM", and peripheral nuclear collisions, "NC") and is not well defined. Whereas for the EM-mode an excitation energy of 12 MeV (with a FWHM of about 10 MeV) is estimated, the NC-mode produces somewhat higher excitation energies. Therefore for both modes second- and higher-chance fission cannot be excluded. In principle a distinction between the two modes is possible because the EM-mode is favoured in heavy target materials like lead whereas light target materials favour the NC-mode of excitation. Nevertheless an uncertainty concerning the excitation energy and the composition of the compound nucleus remains.

## 2.6. REQUIREMENTS FOR PUBLICATIONS

At the end of this chapter a word should be said to experimentalists publishing data in general — but in particular publishing data that ought to be entered in data compilations.

Besides the obvious requirement of a careful and self-critical error analysis which is a prerequisite of any scientific work, it is important, as has been said in the introduction (Section 2.1) already, to give sufficient details in order to allow evaluators and users to make

their own judgement of the error limits. In addition, whenever nuclear decay data, such as half-lives, line intensities, branching ratios are used in a measurement analysis, they should be given with the necessary additional information in order to allow an evaluator to do a complete recalculation in the event that some of the decay data would change due to later more accurate measurements.

Unfortunately, it is frequently not possible to include all the necessary details in a publication. In many cases the necessary information may be put into a doctoral thesis, a report or even an electronic data file that ought to be cited in the paper with a source from which it would be available on request.

The general problem was already recognized by the IAEA in 1965 and an attempt has been made to solve the problem at least in part by the creation of the EXFOR system. EXFOR is designed for storage, retrieval and exchange of nuclear reaction data in a standardised EXchange FORmat, performed by four co-operating data centres (see Chapter 3 for further information). Even though EXFOR was created for nuclear reactions less complicated than fission and may not fulfil all the wishes concerning fission yield data, many publications on fission yield measurements — especially from the USA — are available in EXFOR with more experimental details than given in the corresponding papers. Information may be obtained from the IAEA. For the creation of these files, compilers of the 4 data centres contact the authors of pertinent publications to ask for the necessary information.

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## Chapter 3

### COMPUTERIZED INFORMATION SYSTEMS

#### 3.1. INTERNATIONAL INFORMATION SYSTEMS

##### 3.1.1. The Nuclear Data Centres Network

The Nuclear Data Centres Network, a worldwide co-operation of nuclear data centres under the auspices of the International Atomic Energy Agency (IAEA), has been established to co-ordinate the collection, compilation and dissemination of nuclear data on an international scale. The scope of the Data Centres Network includes nearly all nuclear data required for energy and non-energy nuclear applications, as well as data for basic sciences. The data centres provide the essential link between the producers and users of nuclear data. The Nuclear Data Centres Network has been established to organize this important activity on an international scale.

The Network is co-ordinated through regular meetings organized by the IAEA Nuclear Data Section (NDS), and through direct communication between the centres. The rules and procedures for the compilation and exchange of data files, in particular for CINDA and EXFOR (described below), are, among other things, determined during Network meetings.

The Network consists of four ‘core’ nuclear data centres, which are identical to the Neutron Data Centres described below, and a group of regional, national and/or specialised data centres. This second group compiles data from a restricted geographical region and/or for special data types (nuclear structure data, charged particle or photon induced reactions, etc.). All data are exchanged within the Network, but disseminated worldwide only by the ‘core’ Nuclear Data Centres.

##### *3.1.1.1. The four neutron data centres*

In 1964, the four neutron data centres formed a network, originally to co-ordinate the worldwide compilation of bibliographic information into CINDA. In the later 1960s they created and defined the EXFOR format and system for the compilation of experimental data. The actual compilation started in 1970, until 1976 only for neutron reaction and spontaneous fission data.

The four data centres not only compile and exchange data in the CINDA and EXFOR system (described in this Section 3.1) but also maintain and exchange evaluated data files for nuclear reaction, structure and decay data which are disseminated worldwide (see Section 3.2). The evaluated data files include general purpose files as well as specialized data files (e.g. for fission products, activation, thermonuclear fusion, dosimetry, etc.). The Data Centres compile data originating and disseminate data to customers from a defined geographical area:

National Nuclear Data Center: United States of America and Canada;  
Nuclear Energy Agency Data Bank: OECD countries in Western Europe and Japan;  
Russian Nuclear Data Centre: former USSR countries in Europe and Asia;  
IAEA Nuclear Data Section: all remaining countries in Eastern Europe, South and Central America, Asia, Africa and Australia.

The reader may contact the responsible (geographical area) data centre for further information or retrieve data directly via internet, using one of the addresses given below:

Centre name	<b><i>National Nuclear Data Center</i></b>	<b><i>Nuclear Energy Agency Data Bank</i></b>
Contact person	C.L. Dunford	C. Nordborg
Address	Bldg. 197D Brookhaven National Laboratory PO.Box 5000 Upton NY 11973-5000	Le Seine Saint-Germain 12, boulevard des Iles F-92130 Issy-les Moulineaux France
Telephone	+1 516-344-2902	+33 (1) 45 24 10 90
Fax	+1 516-344-2806	+33 (1) 45 24 11 10
e-mail	nndc@bnl.gov	nea@nea.fr
World Wide Web	<a href="http://www.nndc.bnl.gov/">http://www.nndc.bnl.gov/</a>	<a href="http://www.nea.fr/">http://www.nea.fr/</a>
FTP file transfer	bnlnd2.dne.bnl.gov username: bnlndc (no password required)	ftp.nea.fr username: open password: neadb
Centre name	<b><i>Nuclear Data Section</i></b>	<b><i>Russian Nuclear Data Centre</i></b>
Contact person	D.W. Muir	V.N. Manokhin
Address	IAEA Wagramerstrasse 5 P.O. Box 100 A-1400 Wien Austria	Leipunsky Institute of Physics and Power Engineering Centr Jadernykh Dannykh Ploschad Bondarenko 249 020 Obninsk Kaluga Region Russia
Telephone	+43 (1) 2600-21709	+7 08439-9-8982
Fax	+43 (1) 26007	+7 095-883-3112 +7 095-230-2326
e-mail	services@iaea.org	manokhin@ippe.obninsk.ru
World Wide Web	<a href="http://www.nds.iaea.org">http://www.nds.iaea.org</a>	<a href="http://rndc.ippe.obninsk.ru/">http://rndc.ippe.obninsk.ru/</a>
FTP file transfer	iaeand.iaea.org username: ndsopen	acjd.ippe.rssi.ru username: cjd

### 3.1.1.2. EXFOR

EXFOR is the **EX**change **FOR**mat used by the data centres for the exchange of the compiled experimental data. Initially designed for neutron reaction data, the format is flexible enough to be adjusted and extended to the needs for the compilation of a great variety of data types, presently neutron, charged particle and photon induced reactions as well as spontaneous fission data. Changes to the format are discussed and agreed between the data centres. EXFOR is also the name of the database itself that contains the experimental data compiled by the reaction data centres.

Each entry consists of a “bib-section” containing bibliographic information, where under (retrievable) keywords authors, institutes, references, as well as information on the

experiment can be entered, partly also in coded (retrievable) form. In this section, also numerical values used for corrections are entered, which includes decay data or auxiliary data used in data analysis. A “common-section” contains numerical data common to all reactions compiled in a particular entry. The actual numerical data are entered in tabular form under coded reactions, in one or more so-called subentries. More details can be obtained in brochures from the data centres or directly from the respective Web pages.

The flexibility of the format as well as the possibility for the storage of all experimental details relevant for the evaluators make **EXFOR the ideal medium** for the worldwide compilation of all experimental data. Furthermore, in a joint effort of the four neutron data centres have all the fission yield data from the Meek&Rider file (see Chapter 1 and [1.7]) that were still missing in EXFOR, been converted into the EXFOR format and added to the EXFOR database, which makes the latter almost 100% complete with regard to fission yields. Therefore is a **recommendation from this CRP** to all evaluators of fission yields to use EXFOR as the format and database for compiled fission yield data.

### 3.1.1.3. CINDA

CINDA, the **C**omputer **I**ndex of **N**eutron **D**ata, contains bibliographic references to measurements, calculations, reviews and evaluations of neutron reaction and spontaneous fission data. An extension of the scope to include also charged particle and photon induced reaction data is envisaged. CINDA is also the index for EXFOR entries and the evaluated data libraries available from the data centres. The information contained in the CINDA file is available as printed books with an annual update issue, and by direct retrievals via internet, where the customer is guided with instructions.

A CINDA entry consist of coded information and free text “comments”, and appears as one line in printed form. An entry is prepared for each target-reaction (data type) combination contained in a publication. Codes are used for the target, data type (reaction), institute, neutron energy and reference. The comments part contains the name of the first author and brief descriptions of the information contained in the publication. For fission yields, there is only one code foreseen in CINDA (plus another one for ‘fragment spectra’); hence the comments should include further specification of the type of yield and how data are presented. The CINDA database is almost complete with respect to fission yields.

### 3.1.2. INIS

INIS, the **I**nternational **N**uclear **I**nformation **S**ystem, is a co-operative, decentralised information system which contains bibliographical information on the peaceful uses of nuclear science and technology, as well as on economic, environmental and health aspects of nuclear and other energy sources. The information is provided by INIS Liaison Officers in several international organisations in addition to the IAEA Member States, who have also the right to disseminate that information within their restricted areas (as laid down in agreements).

An entry into the INIS database consists of several pieces of information: title of publication, author(s) and reference citation, an abstract, if available, and sets of descriptors, plus some information on origin and availability of the published information. Searches of the database can be made either by bibliographic information (author, reference) or by keywords (to be found in title, abstract or descriptors).

The collected information is published by the IAEA on a regular basis in the *INIS Atomindex*, which is available as hard copy, microfiche, magnetic tapes and cartridges, and CD-ROM. Updated versions of the whole database are also available. The INIS services are not cost-free. Further information can be obtained from:

***IAEA INIS Section***

P.O. Box 100

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Fax: +43 (1) 26007 22882

e-mail: [INIS.CentralServicesUnit@iaea.org](mailto:INIS.CentralServicesUnit@iaea.org)

Web home page: <http://www.iaea.org/inis/inis.htm>

subscription to INIS Database: <http://www.iaea.org/inis/inisdbm.htm>

### 3.2. OTHER SYSTEMS AND RETRIEVAL PROGRAMS

#### 3.2.1. Use of computerized information systems

This is a brief survey of some of the new on-line services (mainly in Europe and in the USA), user friendly PC programs and CD ROMs, for searching, displaying and/or retrieving:

- the ***evaluated nuclear data files*** currently available now, e.g. dealing with fission yield, nuclear structure, mass, radioactive decay and cross-section data.
- the ***scientific works recently published in the literature*** and collected in various databases, e.g. such as the NSR (Nuclear Science References, a computer file indexed references maintained by NNDC, Brookhaven National Laboratory, USA) or INIS (International Nuclear Information System operated by the International Atomic Energy Agency, Vienna, Austria) systems, or the references system of the Nubase evaluation (Atomic Mass Data Center, Orsay, France), with some regular updates accessible through internet.

Each of these tools may be used as a kind of electronic (and up-to-date) nuclear data Handbook by both the ***reactor physicist*** to have a look at the available evaluated data (e.g. ENSDF, AME, NUBASE, ENDF/B, JEF, ...), and by the ***nuclear data evaluator*** as a complementary source of information for comparison purposes or to check if new data have recently been released and have to be taken into account in the evaluation process. Such online services or PC programs may also be very useful for any other ***research*** or ***educational*** purposes.

We do not intend to present here the numerous existing special purpose files, or the various decay data files, which are restricted to some special applications such as radionuclide metrology (e.g. the LARA reference file for gamma and alpha spectroscopy, developed in France at CEA/DAMRI/LPRI, see papers within [3.1–3.4]), medical applications, dosimetry, detector calibration, and so on.

#### 3.2.2. Software recently developed to display nuclear data

We would like to mention here that some of the recently developed software is now available on CD-ROM or on floppy disk:

### 3.2.2.1. In the frame of the US Nuclear Data Network

As a result of the cooperation between different US National Laboratories such as the Los Alamos National Laboratory (LANL-T2), Oak Ridge National Laboratory (ORNL), Brookhaven National Laboratory (BNL), Triangle Universities Nuclear Laboratory (TUNL), Lawrence Berkeley National Laboratory (LBL), and the San José State University, the following PC programs and data services are available through internet or on CD-ROM:

- The CD-ROM version 1.0 of the 8th edition of the Table of Isotopes (TOI), associated with the Adobe Acrobat viewer to display the hypertext data, has been released in March 1996 by Richard B. Firestone (LBL), S.Y. Frank Chu, CD-ROM Editor, Virginia S. Shirley, Editor, John Wiley & Sons, Inc. (ISBN 0-471-14918-7 Volume set, ISBN 0-471-16405-5 CD-ROM). *Updated in 1998*: ISBN 0-471-24699-9 Volume set, ISBN 0-471-29090-4.  
Five folders are available on this CD-ROM corresponding to the:
  - \* Table of Isotopes
  - \* Table of superdeformed nuclear bands and fission isomers
  - \* Tables of Atoms, Atomic nuclei and subatomic particle.
  - \* Description of Nuclear Structure and Decay Data Bases
  - \* ENSDF Manual
- In the framework of the NSR/ENSDF, related Nuclear Structure Data & References, and the associated PC tools, a very useful CD-ROM has recently been released, entitled: Nuclear Data and References, PC Applications for Nuclear Science, PCNudat and Papyrus™ NSR by P. Ekström, R. Kinsey and E. Browne.

For further information:

- a) contact persons in the USA:  
Edgardo Browne (email: [ebrowne@lbl.gov](mailto:ebrowne@lbl.gov))

In particular for PCNudat:  
Robert Kinsey (email: [kinsey@bnl.gov](mailto:kinsey@bnl.gov))

- b) the US Nuclear Data Network (USNDN) Home Page via Internet:  
**<http://www.nndc.bnl.gov/usndp>**

- The EXFOR CD-ROM produced at the IAEA (more information see the Web site: **<http://www-nds.iaea.or.at/>**)
- The MacNuclide project (see C. A. Stone in [3.2]) at the San Jose State University

### 3.2.2.2. At the University of Lund (Sweden)

One should also mention the Lund Nuclear Data WWW Service at:  
**<http://nucleardata.nuclear.lu.se/nucleardata/>**

Among the available data and services, there is the Isotope Explorer 2.0 program which is a Windows application to interactively access and display nuclear data and to search for



literature references. Isotope Explorer can retrieve data via the Internet or it can use data stored locally. It was developed by:

S Y F Chu<sup>a</sup>, H Nordberg<sup>ab</sup>, R B Firestone<sup>a</sup>, and L P Ekström<sup>ab</sup>

<sup>a</sup> Isotopes Project, LBNL, Berkeley

<sup>b</sup> Department of Physics, Lund University

For further information on Isotope Explorer: <http://ie.lbl.gov/isoexpl/isoexpl.htm>

For further information on the Lund Nuclear Data Center and Services:

Contact person: Peter Ekström

e-mail: [peter.ekstrom@nuclear.lu.se](mailto:peter.ekstrom@nuclear.lu.se)

Telephone: (+46) 46-22 27647

Mobile phone: (+46) 073-995 7984

Fax: (+46) 46-22 24709

Address: Department of Physics, Lund University, Box 118, Office: B201  
SE-221 00 Lund, Sweden

Visiting address: Professorsgatan 1, Internal post: Hämtställe 14

### 3.2.2.3. *At the Atomic Mass Data Center, Paris-Orsay*

One can easily look at the information available on the Web site of the Atomic Mass Data Center via internet at the following address: <http://csnwww.in2p3.fr/amdc>

This centre is devoted to nuclear and mass spectroscopic data, and provides the users also with some news (in the AMDC news letter which is available on the server) about the ongoing experimental, theoretical and evaluation work on the atomic masses, and with some feedback about important conferences in the field (such as ICRM'95, the 9th Symposium on capture gamma ray spectroscopy, ...).

Of particular interest is the 1995 update of the Atomic Mass Evaluation by G. Audi and A.H. Wapstra (AME'95, [3.5]). One should also mention the NUBASE data evaluation by G. Audi et al. (NUBASE'97, [3.6]) displayed by the NUCLEUS PC program or by the 'jvNubase JAVA applet' on the Web. This database contains the main nuclear and decay properties of the known nuclides in their ground and isomeric states as derived from ENSDF, AME, and a critical compilation of recent literature (see references to the NUBASE table in [3.6]).

For further information please contact Georges Audi:

Address: Atomic Mass Data Center, C.S.N.S.M. (IN2P3-CNRS)  
Batiment 108, 91405 Orsay Campus, France

Telephone: (+33 1).6915.5223

Fax: (+33 1).6915.52.68

e-mail: [audi@csnsm.in2p3.fr](mailto:audi@csnsm.in2p3.fr)

### 3.2.2.4. *At the OECD/NEA Data Bank (Paris)*

The evaluated nuclear data available through on-line services are often stored in the common and international Evaluated Nuclear Data File (ENDF) format. This standardized format is very suitable for large data sets and computational applications, but quite complex and difficult to read for casual users. Thus the NEA Data Bank, in co-operation with the

CSNSM-Orsay (France), and the University of Birmingham (UK), has developed and released the very convenient JEF-PC program. JEF-PC uses the jointly developed NUCLEUS driver program, also used within the NUBASE library, to give a “Chart Of The Nuclides” interface for displaying data from a number of evaluated data libraries, including the Joint Evaluated File (JEF) version 2.2, ENDF/B-VI.4, JENDL-3.2, etc., supplied on CD-ROM as part of the package. Three internal modules are devoted to radioactive decay, fission product yield and cross-section data (see [3.7]). The ability to plot and compare evaluated and experimental cross-section data is also included, with experimental data on a second accompanying CD-ROM. Version 2.0 of JEF-PC was released towards the end of 1997 and is available from NEA at a cost of 950 FF (US\$ 157).

For more information, please contact the NEA Nuclear Data Service Section:

	M. Kellett	or	A. Nouri (Nuclear Data Services)
E-mail:	kellett@nea.fr		nouri@nea.fr
Tel:	+33 (1)45.24.10.85		+33 (1)45.24.10.84
Fax:	+33 (1)45.24.11.10		+33 (1)45.24.11.10

OECD Nuclear Energy Agency, Le Seine-Saint Germain, 12, boulevard des Iles  
F-92130 Issy-les-Moulineaux, France  
NEA Data Bank web site: <http://www.nea.fr/html/databank/>

More information on JEF-PC are available on the web at:  
<http://www.nea.fr/html/dbdata/>

### **3.2.3. The three main nuclear data centres in Europe and in the USA providing databases and on-line services**

In [3.2–3.4, 3.8, 3.9] one can find a general description of the main international Data Bases and Services available at:

#### *3.2.3.1. The National Nuclear Data Center (NNDC), Brookhaven National Laboratory (BNL), USA*

- Anonymous FTP server.  
Address: ftp.nndc.bnl.gov.  
User name: anonymous  
Password: your e-mail address.
- Terminal Access (using Telnet).  
Address: telnet.nndc.bnl.gov (IP address: 130.199.112.132)  
User name: NNDC (no password)  
At the prompt for assigned authorization code, enter the code. The new users may use the user name GUEST for a time-limited trial, and can apply for registration using an electronic form which appears on exit from the system.
- A home page through the World Wide Web: <http://www.nndc.bnl.gov/>

Nuclear Data and Programs available:

NSR, ENSDF, NUDAT, MIRD, PHYSCO, CINDA, CSISRS/EXFOR, ENDF Files.

For further information: e-mail: [services@bnlnd2.dne.bnl.gov](mailto:services@bnlnd2.dne.bnl.gov)

NNDC Web effort co-ordinator: Thomas W. Burrows

NNDC Online service co-ordinator: Victoria McLane

Address: Online Data Service.

National Nuclear Data Center.

Brookhaven National Laboratory,

Upton, NY 11973, USA.

Tel: 516-344-2901

Fax: 516-344-2806

e-mail: [NNDC@BNL.GOV](mailto:NNDC@BNL.GOV).

#### *3.2.3.2. The IAEA Nuclear Data Section (NDS), Vienna*

- Services are available through the Web at: <http://www-nds.iaea.or.at/>
- Telnet access (NDIS - Nuclear Data Information System):  
Address: [iaeand.iaea.or.at](telnet://iaeand.iaea.or.at)  
User name: [iaeands](#) (no password).  
At the prompt for assigned authorization code, enter the code. The new users may use the user-name GUEST for a time-limited trial, and can apply for registration using an electronic form which appears on exit from the system

Nuclear data and programs available: Basically the same as available at the US NNDC, see above.

- FTP server:  
Address: [iaeand.iaea.or.at](ftp://iaeand.iaea.or.at)  
User names: ANONYMOUS for FTP file transfer  
FENDL2 for FENDL-2.0 files  
RIPL for RIPL files

For further information:

Nuclear Data Section

Address: International Atomic Energy Agency,

P.O. Box 100,

A-1400 Vienna, Austria.

Tel: (43-1) 2600-21710

Fax: (43-1) 26007

e-mail: [online@iaeand.iaea.or.at](mailto:online@iaeand.iaea.or.at)

#### *3.2.3.3. The Nuclear Energy Agency (NEA) Data Bank*

The NEA Online Services are open to registered scientific users in the seventeen countries participating in the NEA Data Bank. New users are asked to register via the online form at the web address [www.nea.fr](http://www.nea.fr). The site offers access to a wide range of databases, in

particular NSDD (ENSDF, NSR, NUDAT) as well as experimental data (EXFOR), bibliographic reference to neutron induced reactions (CINDA), evaluated data files (including JEF, ENDF/B, JENDL) and the NEA Thermochemical Data Base (TDB). All data libraries have online search and download facilities, with new services coming soon to include online plotting of experimental and evaluated data.

The Online services can be accessed through the Web at the following address:

***<http://www.nea.fr/>***

For further information please contact:

Pierre Nagel - Network & Online-services

e-mail: [nagel@nea.fr](mailto:nagel@nea.fr)

Telephone: +33 (1) 45.24.10.82

### REFERENCES TO CHAPTER 3

- [3.1] Proc. International Committee for Radionuclide Metrology, Paris, 1995; Nucl. Instr. Meth. Phys. Res. A **369** (1996) 333.
- [3.2] INTERNATIONAL ATOMIC ENERGY AGENCY, Co-ordination of the International Network of Nuclear Structure and Decay Data Evaluators (Summary, IAEA Advisory Group mtg, Livermore, 1994), Rep. INDC(NDS)-307, IAEA, Vienna (1994).
- [3.3] LEMMEL, H.D. (Ed.), Development of an International Nuclear Decay Data and Cross-Section Database (Summary IAEA Specialists Mtg Vienna, 1994), Rep. INDC(NDS)-328, IAEA, Vienna (1994).
- [3.4] LEMMEL, H.D. (Ed.), Development of an International Nuclear Decay Data and Cross-Section Database, (Proc. IAEA Specialists Mtg Vienna, 1994; Rep. INDC(NDS)-329, IAEA, Vienna (1994).
- [3.5] AUDI, G., WAPSTRA, A.H., The 1995 atomic mass evaluation. Nucl. Phys. A **595** (1995) 409.
- [3.6] AUDI, G., BERSILLON, O., BLACHOT, J., WAPSTRA, A.H. The Nubase evaluation of nuclear and decay properties. Nucl. Phys. A **624** (1997) 1.
- [3.7] KONIECZNY, M., et al., "JEF-PC Version 2.0: A PC program for viewing evaluated and experimental data", Nuclear Data for Science and Technology, (Proc. Int. Conf. Trieste, 1997) (REFFO, G., VENTURA, A., GRANDI, C., Eds).
- [3.8] IAEA Nuclear Data Newsletter **20** (1994).
- [3.9] Fifth edition of the Nuclear Wallet Cards (TULI, J.K., Ed.), NNDC, BNL, USNDN, 1995.

## Chapter 4

### MODEL CALCULATIONS

#### 4.1. MODELS FOR MASS DISTRIBUTIONS AND WAHL'S MODEL

Mass distributions can be represented approximately by summations of Gaussian functions [4.1.1–4.1.4]. Considerable success has been achieved by use of five Gaussian curves, but even fewer can be used, under some circumstances, as discussed below. The  $A'_p$  model [4.1.3] (see Section 4.2.2) makes use of many Gaussian functions, one for each fission product element.

The experimental chain-yield data,  $Y(A)$ , of products from fission of nuclei with atomic numbers  $Z_F = 90\text{--}99$  and excitation energies  $E^* \leq \sim 20$  MeV were fitted by the sum of 2 to 5 Gaussian functions using the method of least squares. Then the parameters determined for the Gaussian functions were fitted by mathematical functions of the atomic numbers ( $Z_F$ ), the mass numbers ( $A_F$ ), and the excitation energies ( $E^*$ ) of fissioning nuclei again using the method of least squares. Reciprocal variance weighting was used in both types of calculations. The functions derived, given below, allow calculation of mass distributions for fissioning nuclei in the ranges given above and with an estimated accuracy that will be discussed.

The sources of the mass-yield data were evaluated data files from UKFY2 [4.1.4] (UK2 in figures) and ENDF/B-VI [4.1.5] (EB6 in figures), 4  $Y(A)$  data sets for fission induced by monoenergetic neutrons (0.1–10 MeV) [4.1.6–4.1.9], 4  $Y(A)$  data sets for light-wing masses from the LOHENGRIN fission-product separator [4.1.10–4.1.14] (LO in figures), and data from LOHENGRIN for AM242T ( $^{241}\text{Am}(2n_{th},f)$ ) [4.1.15]. Since ENDF/B-VI data files differentiate between experimental and estimated  $Y(A)$  by the magnitude of assigned errors, only ENDF/B-VI  $Y(A)$  values with assigned errors  $<12\%$  of the value were used as data. Gaussian  $Y(A)$  parameters for fast-neutron fission were not used in the second stage of the analysis because fission is induced by neutrons with a large range of energies. However, the mass distributions for fast-neutron fission reactions were calculated by a summation method from the model parameters derived, as will be discussed.

It was found that the central Gaussian curve was not needed to represent chain yields from spontaneous fission reactions (S.F.). This finding is consistent with experimental results showing that the valley yields from spontaneous fission are more than an order-of-magnitude less than those from thermal-neutron induced fission reactions [4.1.16, 4.1.17].

It was also found that for fission induced by 14 MeV neutrons (high-energy fission) and for thermal-neutron induced fission of the heavier actinides ( $Z_F > 94$ ), one curve per peak represented the chain-yield data reasonably well. For the heavier actinides, the representation is improved by modifying the peak Gaussian functions to include an exponential drop in  $Y(A)$  for  $A_H < 130$  and in the complementary range for the light peak. The modified peak functions are renormalized to achieve a 200% sum for all yields (see Eq. 14.1-14e). The sharp drop in heavy chain yields occurs in about the same place (below  $A_H = 130$ ) for all fission reactions investigated and may be associated with the influence of the 50-proton shell on yields.

Uncertainties in model calculated yields were estimated from the following empirical equation proposed earlier for the percent uncertainty (PER) [4.2.6].

$$\text{PER} = (25) \exp\{-0.25[\ln Y(A)]\} \quad (4.1-1)$$

Estimated range of uncertainty:  $Y(A)/(1 + \text{PER}/100)$  to  $Y(A)(1 + \text{PER}/100)$

#### Examples

Y(A), %	PER	1 + PER/100
10.	14.	1.14
1.	25.	1.25
0.1	44.	1.44
$10^{-3}$	141.	2.41
$10^{-6}$	791.	8.91
$10^{-10}$	7910.	80.10

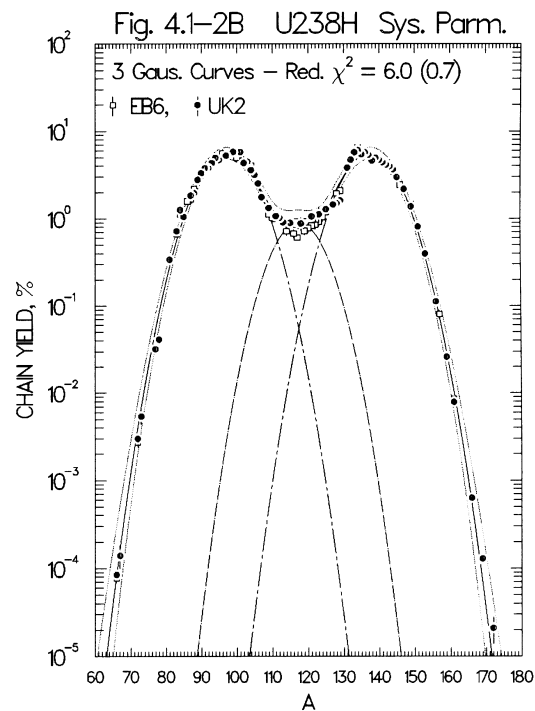
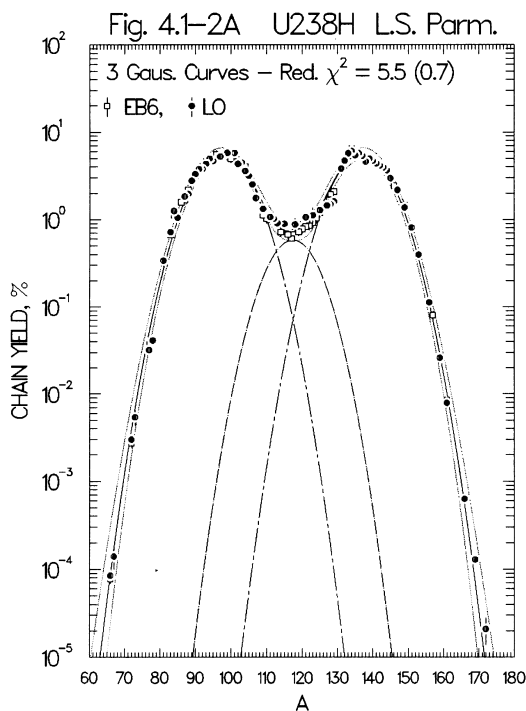
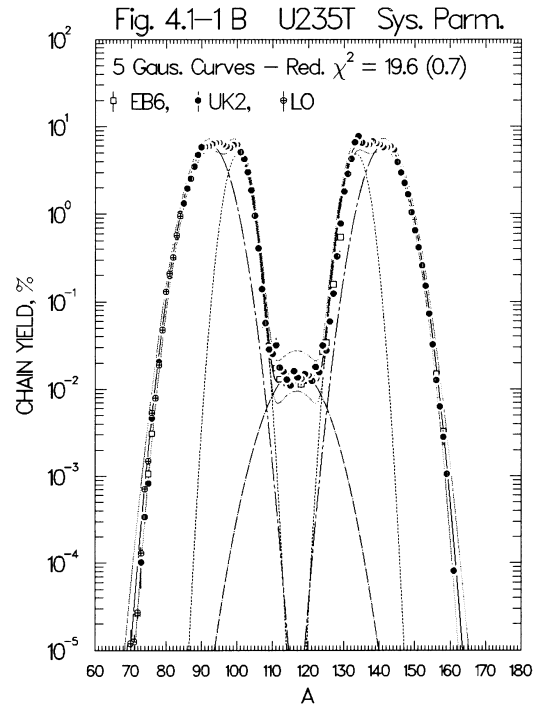
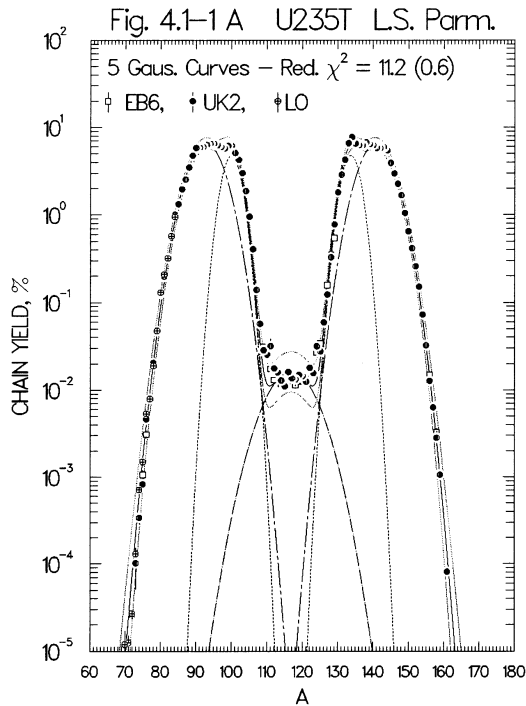
As shown in the accompanying figures, most experimental chain yields fall within the estimated range of uncertainties (dotted lines), which suggests that most estimated chain yields, calculated from the equations below, should be reliable to within the estimated uncertainties.

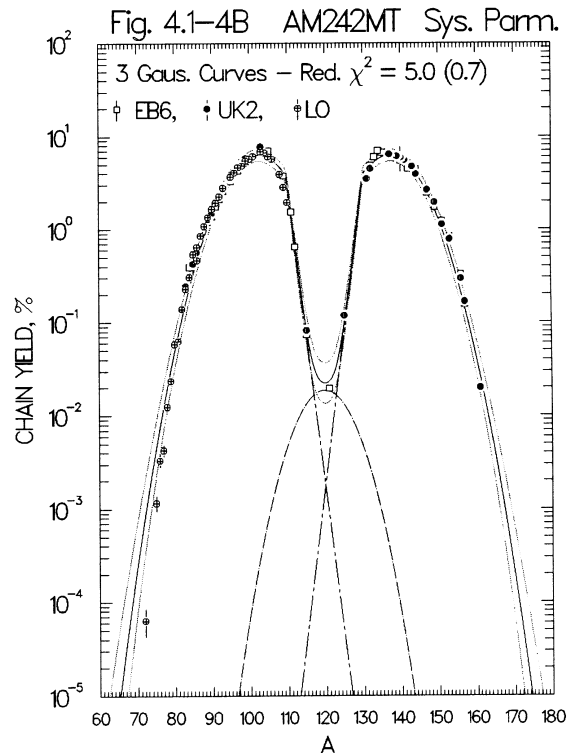
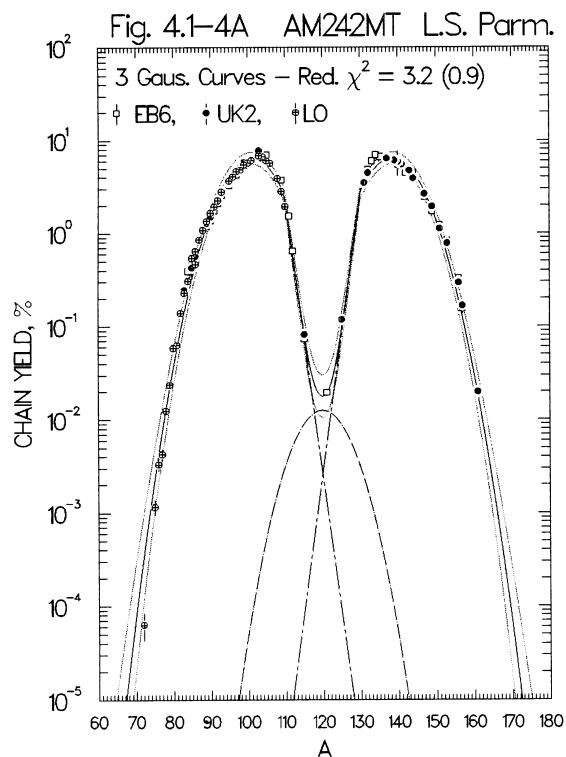
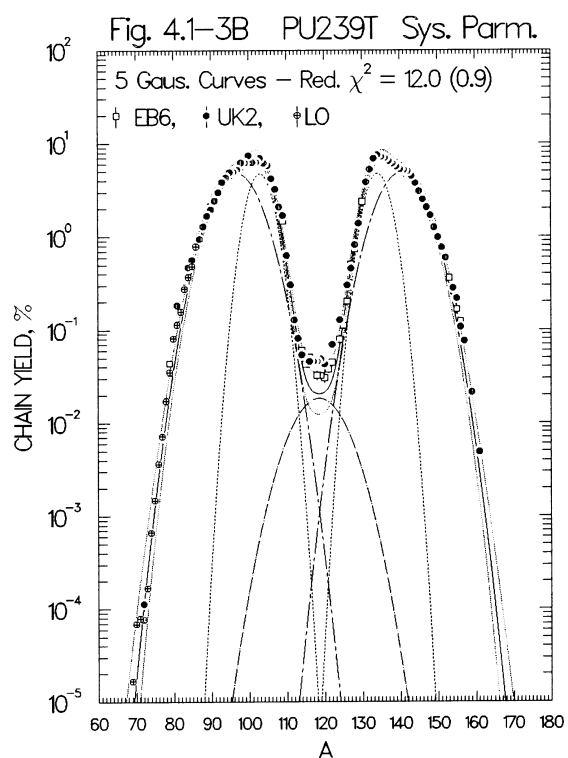
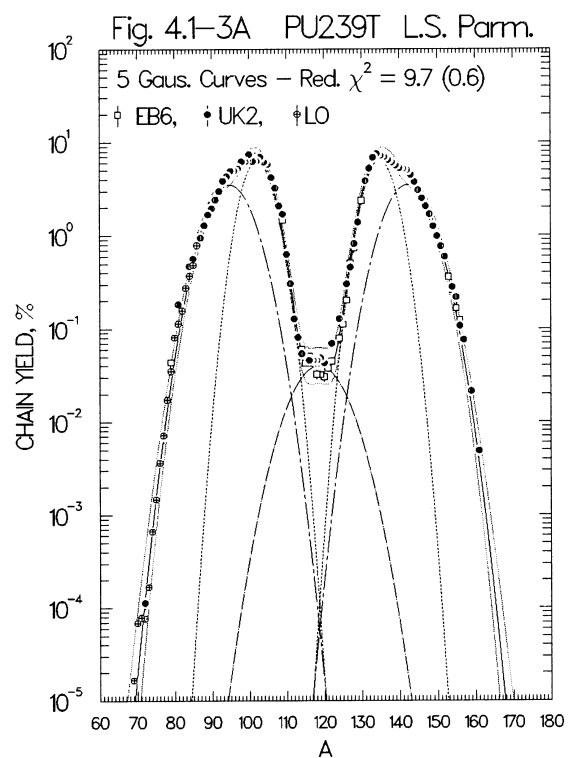
The accompanying figures compare for a number of fission reactions the calculated curves (solid lines) and range of uncertainties (dotted lines) with experimental data (points). Contributions of individual Gaussian functions are also shown (dashed lines, etc.). Curves are shown both for the results of least squares calculations (labelled A and L.S. Parm.) and for results of calculations using systematic trends expressed in the equations below (labelled B and Sys. Parm.). As can be seen, the measured chain yields show fine structure that cannot be reproduced by sums of smooth Gaussian functions. Also, a number of the higher yields have been measured very accurately, to ~1%. These facts contribute to the reduced chi-square values being considerable greater than one. If the estimated model uncertainties, discussed above, are included as contributing errors in reduced chi-square calculations, the results are mostly less than one and are shown in parentheses in the figures.

Figures 4.1-2A,B, 4.1-4A,B, 4.1-5A,B, and 4.1-6A,B illustrate that complementary single Gaussian curves for each peak represent the experimental data reasonably well for high excitation energies and for  $Z_F > 94$ . The Gaussian functions for the large  $Z_F$  are modified to include an exponential decrease toward symmetry, as discussed above. The differences between the experimental data and the wings of the model curves for fission of californium isotopes, Figs 4.1-5A,B and 4.1-6A,B, suggest that addition of a small Gaussian contribution to the wings could improve representation of experimental data by the model. This suggested model modification is illustrated in Figs 4.1-3C, 4.1-4C, 4.1-5C, and 4.1-6C.

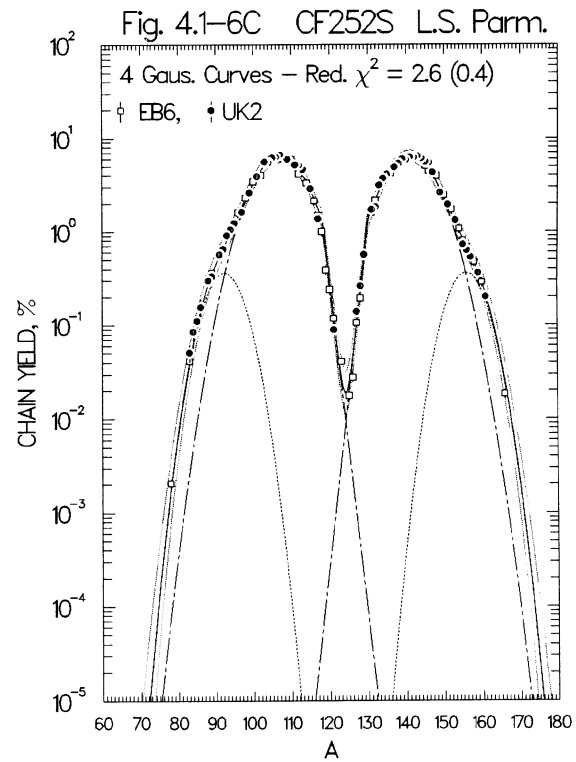
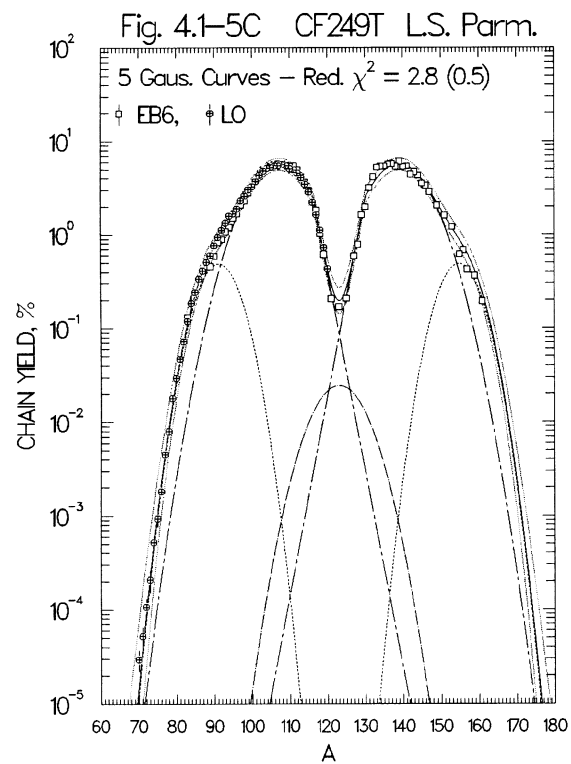
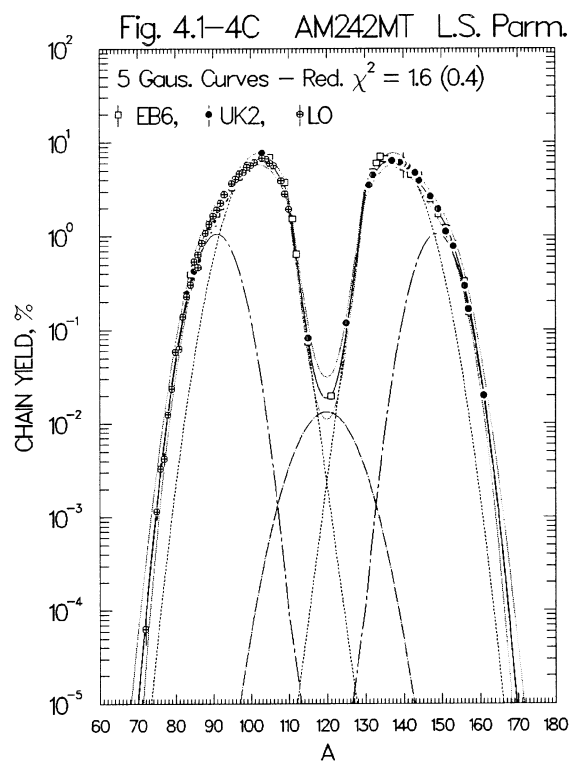
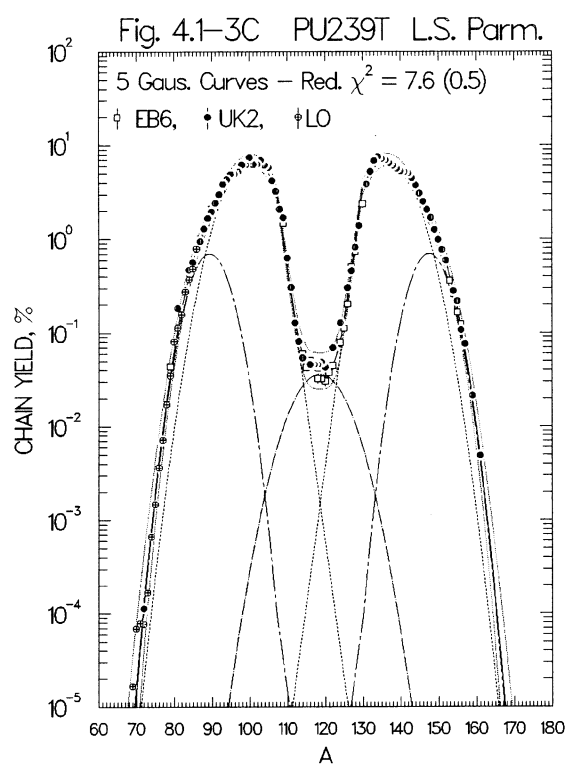
The mass distributions for the peaks from fission of thorium isotopes are quite narrow, and the central peaks are quite well resolved, as shown in Figs 4.1-7A,B and 4.1-8A,B. The average width parameter for the central curve,  $\sigma_3$ , for several light fissioning nuclei, including the thorium nuclei, is  $6.0 \pm 0.5$ . The value for TH232F determined by least squares is considerably larger ( $9.5 \pm 1.5$ ) than the average — compare Figs 4.1-8A and 4.1-8B.

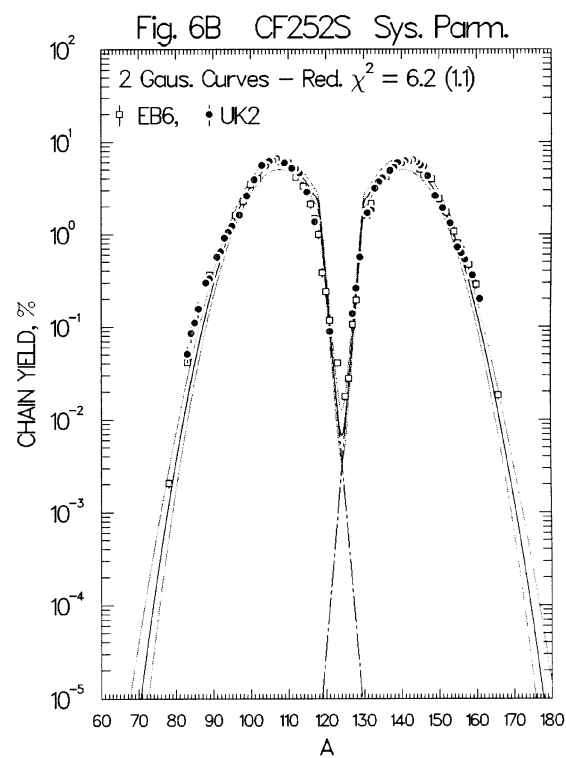
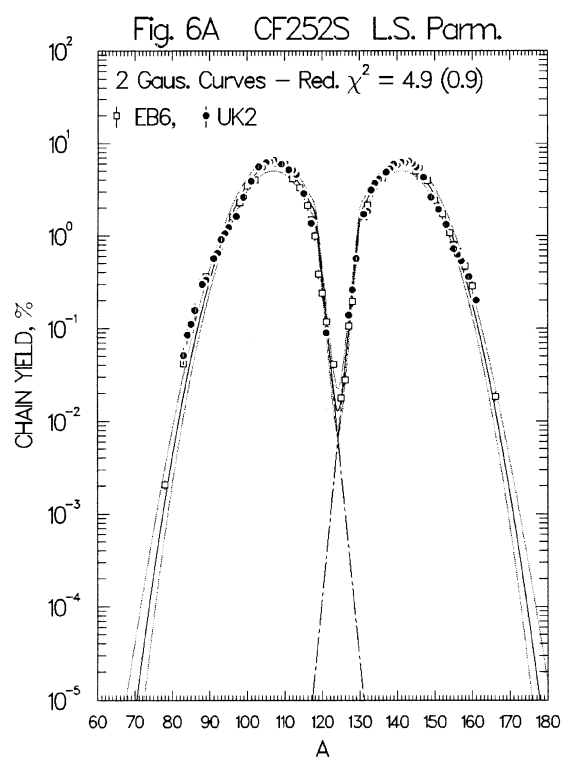
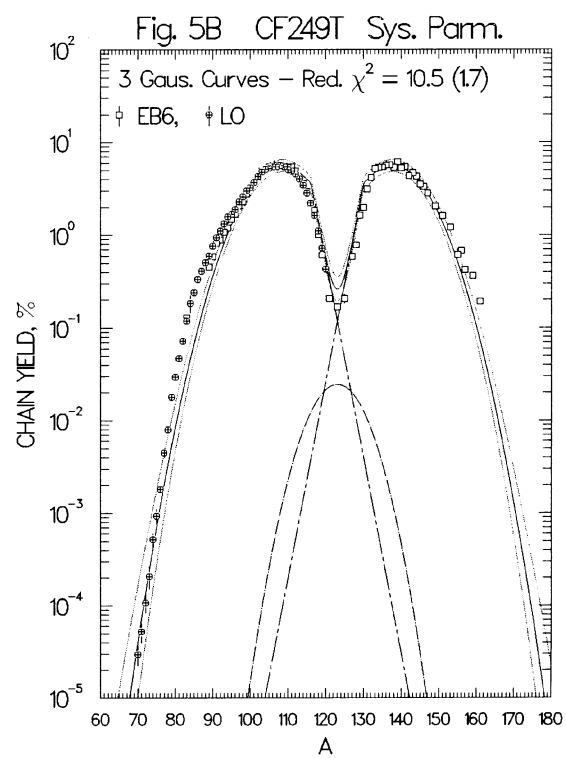
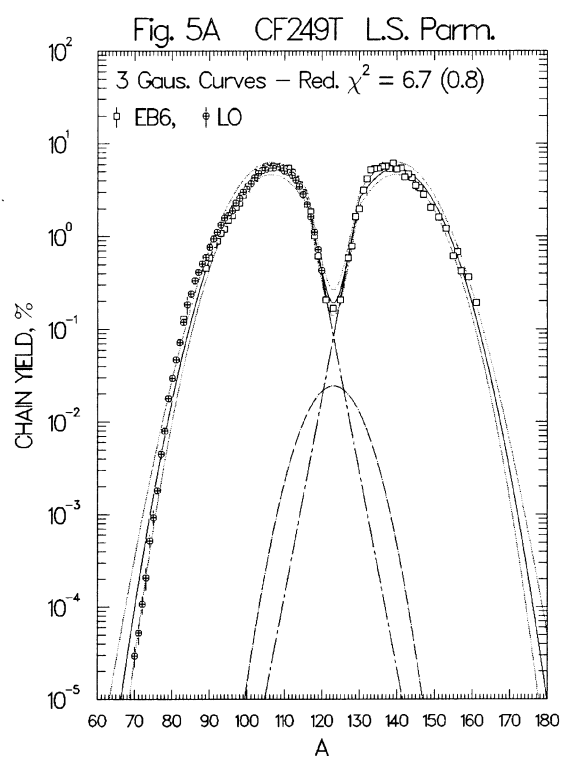
The intensity of the central Gaussian peak,  $Y_3$ , increases exponentially with the kinetic energy of the incident neutron (See Eq. 4.1-11). For fast-neutron-induced fission the incident neutrons have a wide range of energies,  $E_n$  or  $EN$ , so a summation calculation was carried out in which  $Y_3(E)$  for each energy increment was weighted for the relative cross section,  $\sigma_E$ , and the relative yields of neutrons,  $R_E$ , at the average neutron energy,  $E$ , for each energy increment (Eq. 4.1-15). The average number of neutrons emitted,  $\bar{\nu}$ , and their average kinetic energy,  $\bar{E}_n$ , for fast-fission reactions was determined similarly (Eqs. 4.1-16, 4.1-16a).

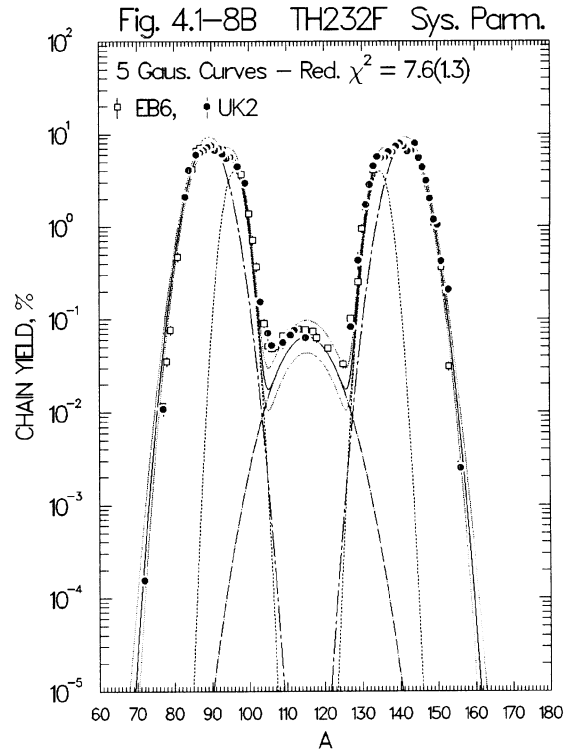
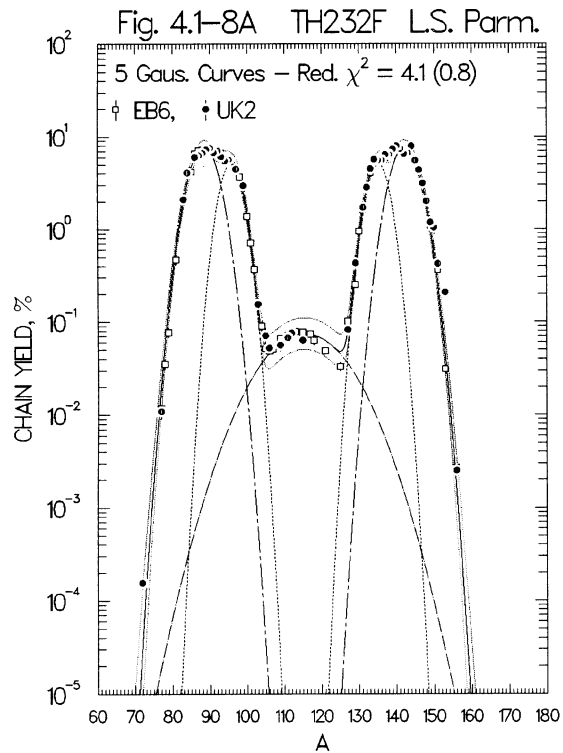
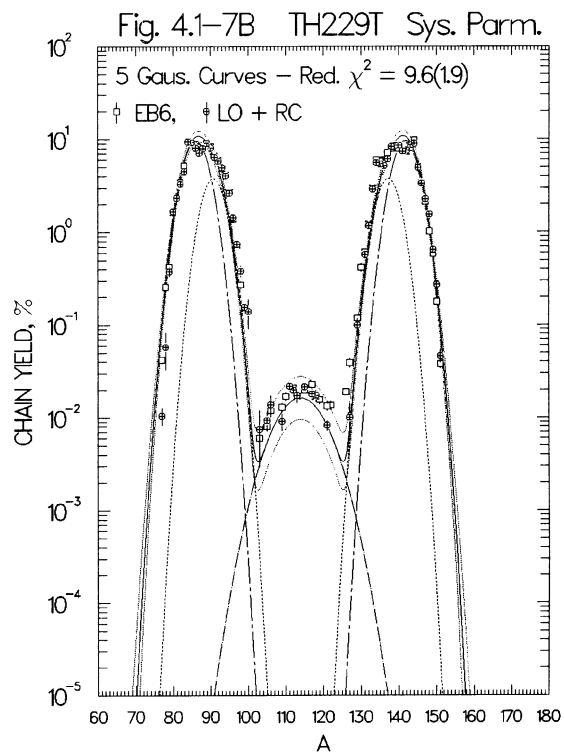
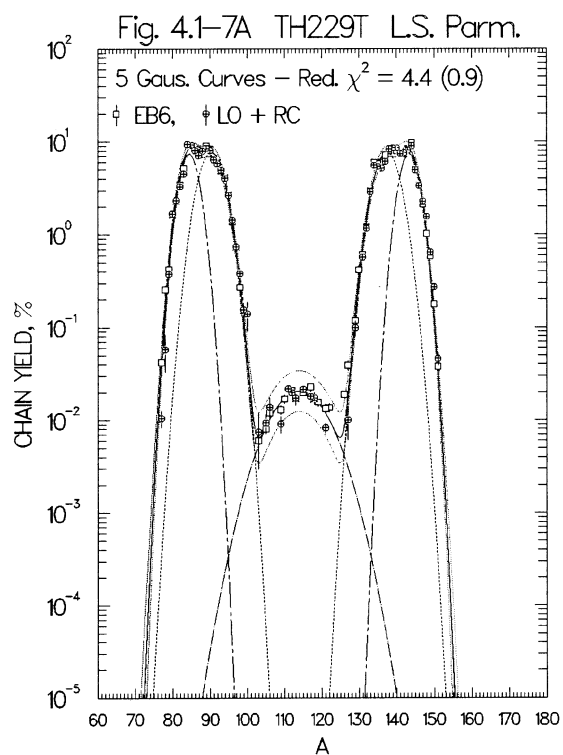


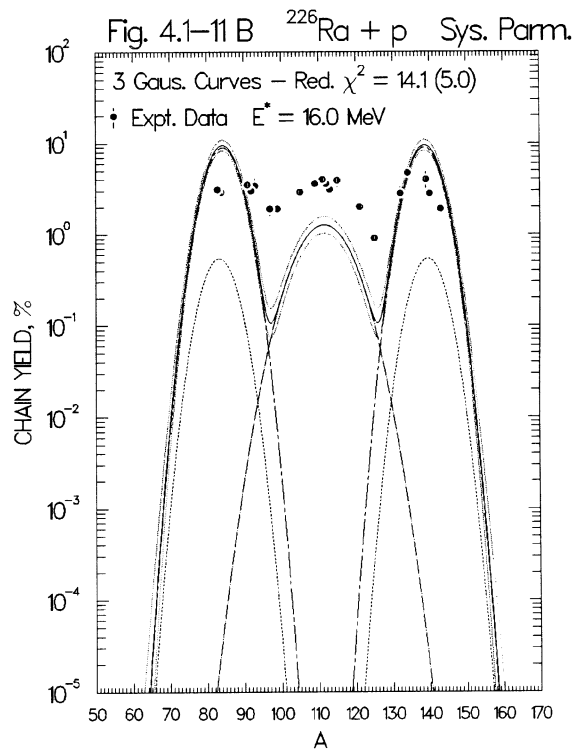
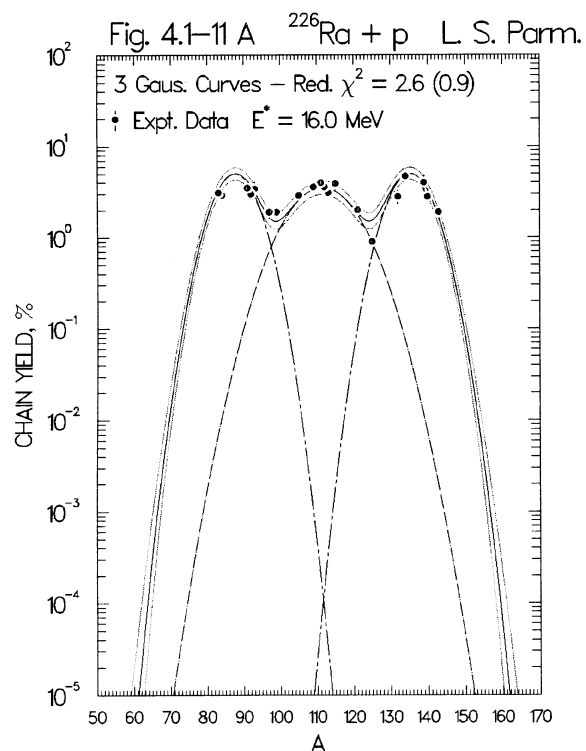
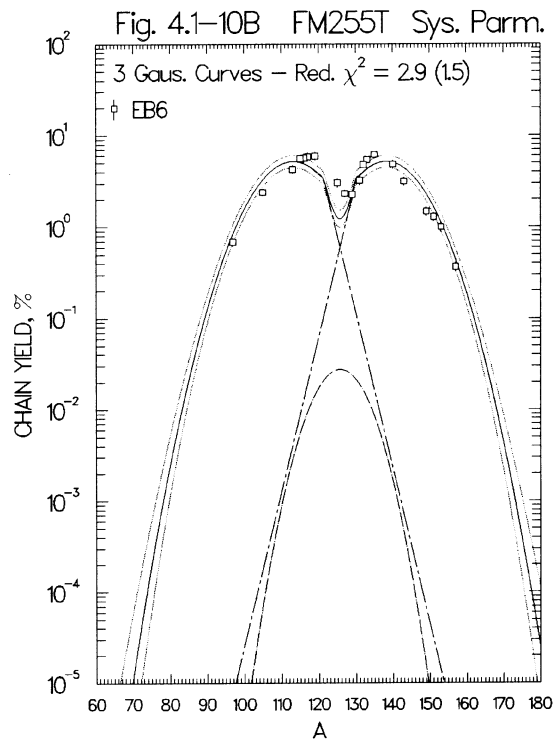
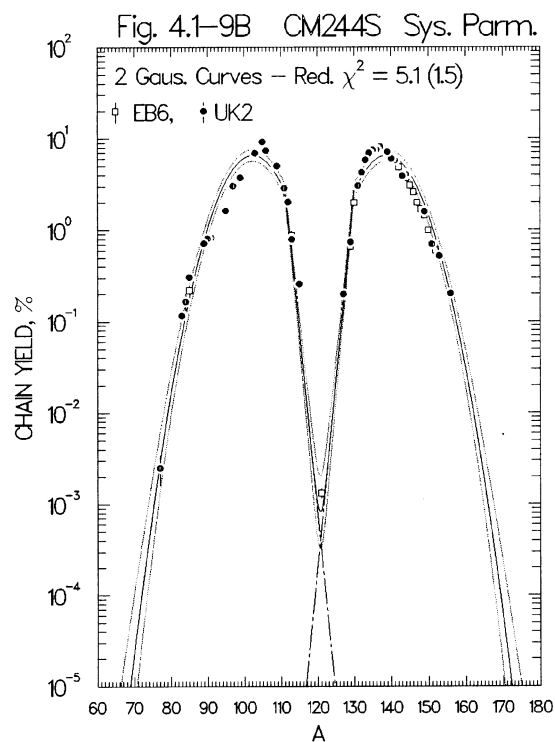












The relative neutron yield,  $R_E$ , was determined from Eq. 4.1-15c [4.1.18], the constant  $C = 1.29$  for U235T being multiplied by 3/4 giving  $C = 0.9675$  to allow for degradation of fission-neutron spectra in reactors.

The relative fission cross section,  $\sigma_E$ , which approximated experimental curves [4.1.19], was taken to be 1.00 over much of the energy range; 0.5 to 6.0 MeV if the threshold were less than 0.0 ( $TH < 0.0$ );  $TH + 1.0$  to the neutron binding energy (BN) if  $TH > 0.0$ . Below 0.5 MeV and if  $TH < 0.0$  MeV,  $\sigma_E$  increases with decreasing energy as  $1/\sqrt{E}$  (Eq. 4.1-17). Above threshold, if  $TH > 0.0$ ,  $\sigma_E$  increases linearly from 0.0 at  $TH$  to 1.0 at  $TH + 1.0$ . To approximate the increase in  $\sigma_E$  due to second-chance fission above 6.0 MeV, if  $TH < 0.0$ , or above BN, if  $TH > 0.0$ ,  $\sigma_E$  rises linearly over an 1.0 MeV interval to the value of STEP (Eq. 4.1-17d).  $TH$  was calculated from Eq. 4.1-20; the constants in Howerton's [4.2.8] original equation were recalculated by least squares using published threshold and barrier values [4.2.8, 4.1.21, 4.1.22]. ( $TH = BARRIER - BN - 0.4$ ; the neutron binding,  $BN = 8.071 + MEX(A - 1) - MEX(A)$ , 8.071,  $MEX(A - 1)$ , and  $MEX(A)$  being mass excesses of the neutron, the target, and the fissioning nuclide, respectively [4.1.20].)

Errors introduced by the approximations made in expressions for  $R_E$  and  $\sigma_E$  are minimized in the treatment because the sum of their products is used for normalization (Eqs. 4.1-15 and 4.1-16).

$Y(A)$  data sets can be created in which the arithmetic average of evaluated experimental values and recent experimental values replace model estimated values. These files can include listings of differences between evaluated experimental values greater than the error in the average and listings of differences between average evaluated experimental values and model estimated values greater than estimated uncertainties. These listings can help to resolve discrepancies in evaluated experimental values and indicate where the model needs improvement. Examples of the listings are given in Appendix B.1.

Calculations were made using the systematic model parameters derived from the equations below (also see Section 4.2.3), and the results were compared in plots to 28 sets of experimental chain yields available from UKFY2 [4.1.4], 60 sets from ENDF/B-VI [4.1.5], and 33 sets for monoenergetic-neutron induced fission [4.1.6–4.1.9]. The calculated curves represent the experimental data reasonably well; as shown in the figures with B in the label. For many fission reactions the data were not used in deriving the model parameters, two examples are shown in Figs 4.1–9B and 4.1–10B. Note that FM255T is just above the recommended range of  $Z_F$ . A test of the model below the recommended range of  $Z_F$  shows in Fig. 4.1–11B that the model underestimates the intensity of the central peak and overestimates the intensity of the other peaks. However, as shown in Fig. 4.1–11A, the experimental data can be represented reasonably well by a least squares model calculation in which Gaussian parameters are free to vary. Thus some modification of the equations below will be required for extension of systematic trends beyond the regions presently considered.

### Equations for Estimation of Chain Yields, $Y(A)$

$A_F$ ,  $Z_F$ ,  $EN$ , are needed;  $\bar{v}$  can be given; if  $\bar{v} = 0.0$ ,  $\bar{v}$  is calculated from Eq. 4.1-19. Program keys:  $EN < 0$  for S.F.,  $\bar{v} < 0$  for fast-neutron fission. Parameter subscripts 1 and 2 refer to light peak components, 4 and 5 to heavy peak components, and 3 to the central peak.

$$Y(A) = \sum_{i=1}^{i=5} \frac{Y_i(NF_i)}{\sigma_i \sqrt{2\pi}} \exp[-(A - \bar{A} + \Delta_i)^2 / 2\sigma_i^2] \quad (4.1-2)$$

$$\bar{A} = (A_F - \bar{\nu})/2 \quad NF_i \text{ are normalization factors so that } \Sigma Y(A) = 200\% \quad (4.1-2a)$$

( $\bar{\nu}$  value given or calculated from Eq. 4.1-19)

$$\sigma_1 = \sigma_5 = 3.303 + 0.235(Z_F - 92) + 0.140(A_F - 236) + 0.154E^* \quad (4.1-3)$$

$$E^* = \text{excitation energy of fissioning nucleus} = EN + BN \quad (= 0 \text{ for S.F.}) \quad (4.1-3a)$$

(BN = binding energy of incident neutron calculated from mass excesses [4.1.20])

$$\sigma_2 = \sigma_4 = 2.268 + E^* \{0.064 + 0.08(Z_F - 92) - 0.03(A_F - 236)\} \quad (4.1-4)$$

$$\sigma_2 = 6.0 \quad (4.1-5)$$

$$-\Delta_1 = \Delta_5 = 26.204 - 0.570(Z_F - 92) - 0.373(A_F - 236) - 0.262E^* \quad (4.1-6)$$

$$-\Delta_2 = \Delta_4 = PD - (A_F - \bar{\nu})/2.0 \quad PD = 132.211 + 0.18348(A_F - 230) \quad (4.1-7)$$

$$\text{if } Z_F \leq 91, \quad PD = 137.6 - 0.87903(A_F - 230) \quad (4.1-7a)$$

$$\text{if S.F. and } A_F < 240, \quad PD = 137.0 \quad (\text{for U238S}) \quad (4.1-7b)$$

$$\Delta_3 = 0.0 \quad (\text{model definition}) \quad (4.1-8)$$

$$Y_1 = Y_5 = 100 - Y_2 - Y_3/2 \quad (\text{normalization}) \quad (4.1-9)$$

$$Y_2 = Y_4 = 46.21 + 2.837(Z_F - 92) - 2.036E^* \quad \text{if } EN > 12, Y_2 = 0.0 \quad (4.1-10)$$

if  $Y_2 > 35.0$ ,  $Y_2 = 35.0$

$$Y_3 = P_1 \exp[P(E^* - BP)] \quad BP = \text{Break Point (start of 2<sup>nd</sup> chance fission)} \quad (4.1-11)$$

$$BP = 2(BN) - TH1 + 0.5 \quad (0.5 \text{ MeV} \approx \text{emitted neutron K.E.}) \quad (4.1-11a)$$

$$TH1 = \text{Threshold for } (A_F - 1) \quad (4.1-11b)$$

$$P_2 = 0.492 - 0.0544(Z_F - 92) \quad (P \text{ for } E^* < BP) \quad (4.1-11c)$$

$$P_3 = 0.162 - 0.0684(Z_F - 92) + 0.0268(A_F - 236) \quad (P \text{ for } E^* > BP) \quad (4.1-11d)$$

$$P_1 = 0.447 \exp[P_2(BP - 7.79)] \quad (4.1-11e)$$

$$P = P_2 \quad \text{if } EN < BP, \quad P = P_3 \quad \text{if } EN \geq BP \quad (4.1-11f)$$

$$Y_3 = 0 \text{ for S.F.} \quad (4.1-11g)$$

***for  $Z_F > 94$ :***

$$Y_2 = Y_4 = 0$$

$$\sigma_1 = \sigma_5 = 5.71 + 0.096(A_F - 236) + 0.129E^* \quad (4.1-12)$$

$$-\Delta_1 = \Delta_5 = 26.07 - 0.055(Z_F - 92) E^* - 0.373(A_F - 236) \quad (4.1-13)$$

$$\text{for } A > AC, \quad AC = A_F - 230 - \bar{\nu} \quad (4.1-14)$$

$$Y(A)_1 = Y(A=130) \exp[-PE(A-AC)] \quad (4.1-14a)$$

for  $A < 130$ :

$$Y(A)_5 = Y(A=130) \exp[-PE(130-A)] \quad (4.1-14b)$$

$$Y(A)_1 = 0 \quad \text{if } A \geq 155, \quad Y(A)_5 = 0 \quad \text{if } A < (A_F - 155 - \bar{\nu}) \quad (4.1-14c)$$

$$PE = 1.116 - 0.169(Z_F - 92) + 0.062(A_F - 236) - 0.018E^* - 0.0039(A_F - 236)E^* \quad (4.1-14d)$$

$$Y_1 = Y_5 = 100 - Y_3/2 \quad (\text{renormalization}) \quad (4.1-14e)$$

*for fast neutron fission:*

$$Y_3 = \frac{\int_0^\infty (R_{EN})(\sigma_{EN})(Y_{EN})dEN}{\int_0^\infty (R_{EN})(\sigma_{EN})dEN} \approx \frac{\sum_0^{11\text{Mev}} (R_E)(\sigma_E)(Y_E)(0.01)}{\sum_0^{11\text{Mev}} (R_E)(\sigma_E)(0.01)} \quad (4.1-15)$$

$$E = EN + 0.005 \quad (4.1-15a)$$

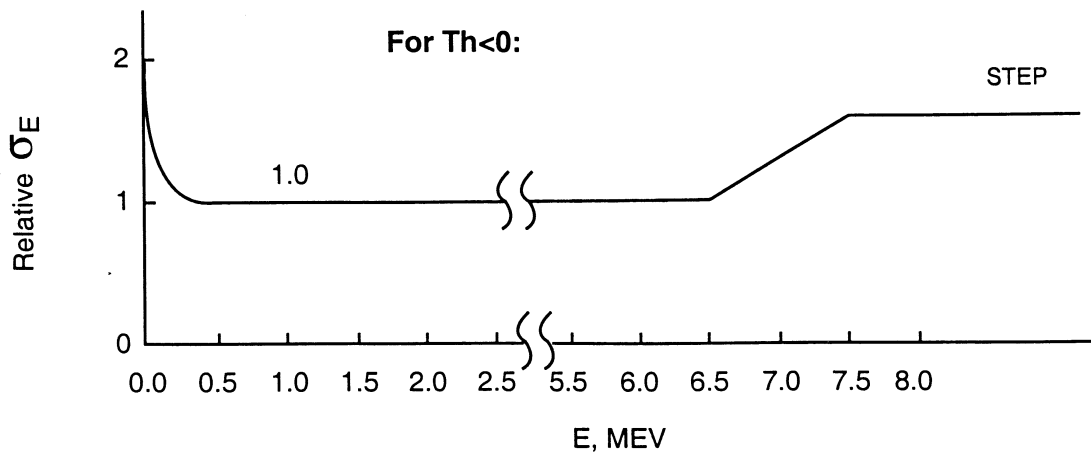
$$Y_E = Y_3 \quad \text{from Eqs. (4.1-11, 11a-f)} \quad (4.1-15b)$$

$$R_E = \sqrt{E} e^{-E/C}, \quad C = 1.29 \text{ U235T (undegraded)}, \quad (4.1-15c)$$

$$C = 0.9675 \text{ U235T (degraded } \{3/4\})$$

$$\bar{\nu} = \frac{\int_0^\infty (R_{EN})(\sigma_{EN})(\nu_{EN})dEN}{\int_0^\infty (R_{EN})(\sigma_{EN})dEN} \approx \frac{\sum_0^{11\text{Mev}} (R_E)(\sigma_E)(\nu_E)(0.01)}{\sum_0^{11\text{Mev}} (R_E)(\sigma_E)(0.01)}, \quad \nu_E = \bar{\nu}_E \quad \text{from Eq. 4.1-19} \quad (4.1-16)$$

$$\bar{E}_n = \frac{\int_0^\infty (R_{EN})(\sigma_{EN})(EN)dEN}{\int_0^\infty (R_{EN})(\sigma_{EN})dEN} \approx \frac{\sum_0^{11\text{Mev}} (R_E)(\sigma_E)(E_n)(0.01)}{\sum_0^{11\text{Mev}} (R_E)(\sigma_E)(0.01)}, \quad (4.1-16a)$$

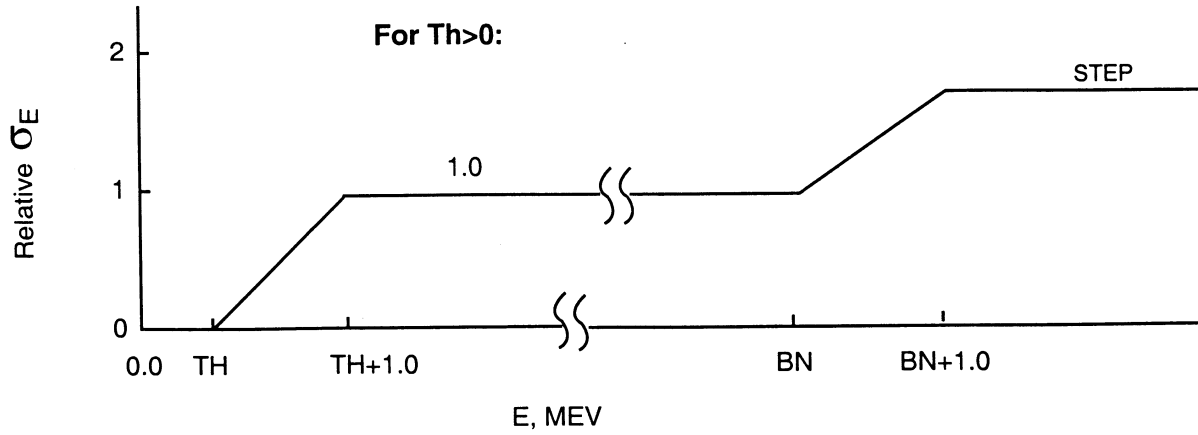


$$\sigma_E = 1.0 + 0.13/\sqrt{E} - 0.13/\sqrt{0.5} \quad (E < 0.5) \quad (4.1-17)$$

$$\sigma_E = 1.0 \quad (E = 0.5-0.6) \quad (4.1-17a)$$

$$\sigma_E = 1.0 + (\text{STEP} - 1.0)(E-6.0) \quad (E = 6.0-7.0) \quad (4.1-17b)$$

$$\sigma_E = \text{STEP} \quad (E > 7.0, \text{ limits: } 1.0-2.5) \quad (4.1-17d)$$



$$\sigma_E = (E-TH) \quad (E = TH \text{ to } TH + 1.0) \quad (4.1-18)$$

$$\sigma_E = 1.0 \quad (E = TH + 1.0 \text{ to } BN) \quad (4.1-18a)$$

$$\sigma_E = 1.0 + (\text{STEP}-1.0)(E-BN) \quad (E = BN \text{ to } BN + 1.0) \quad (4.1-18b)$$

$$\sigma_E = \text{STEP} \quad (E > BN+1.0, \text{ limits: } 1.0 - 2.5, \text{ see Eq. 4.1-17d for STEP}) \quad (4.1-18d)$$

$$\begin{aligned} \bar{\nu}_E = & 2.286 + 0.147(Z_F - 92) + 0.054(A_F - 236) + 0.040[2 - (-1)^{N_F} - (-1)^{Z_F}] + \\ & + [0.145 - 0.0043(A_F - 236)](E - TH) \end{aligned} \quad (4.1-19)$$

$$TH = 11.47 - 0.166 Z_F^2 / A_F + 0.093[2 - (-1)^{N_F} - (-1)^{Z_F}] - BN \quad (4.1-20)$$

## Summary and Conclusions

An empirical multi-Gaussian model for calculating chain yields,  $Y(A)$ , has been developed for fission reactions with nuclear charge,  $Z_F$ , from 90 to 99 and with excitation energy,  $E^*$ ,  $\leq \sim 20$  MeV. Uncertainties in the calculated  $Y(A)$  can be estimated by use of an empirical equation (Eq. 4.1-1). Averages of experimental data can replace calculated values, and, after normalization, the new  $Y(A)$  data set can be treated by the modified Terrell method [4.1.3] to derive  $\bar{\nu}_A$  values. See, also, Section 4.2.3.



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## 4.2. WAHL'S MODELS FOR NUCLEAR-CHARGE DISTRIBUTION

Nuclear-charge distribution describes the dispersion of yields with mass and atomic numbers,  $A$  and  $Z$ , of the  $\sim 1000$  primary fission products from each of many fission reactions. The yields are for products after prompt neutron emission and before beta decay. Since only a small fraction of the yields have been measured, models are needed for estimation of unmeasured yields. Theoretical models are not sufficiently advanced to give reliable yield estimates, so two empirical models have been proposed that include mathematical functions derived from available experimental data [4.1.3].

The two models, called the  $Z_p$  and the  $A'_p$  models, give different perspectives of nuclear-charge distribution from fission, and comparison of results of calculations with the two models has been helpful in improving both models [4.1.3]. The  $Z_p$  model treats dispersion of fractional independent yields, FI, with  $Z$  for each  $A$  of primary fission products [4.1.3, 4.2.1–4.2.6]. The  $A'_p$  model treats the dispersion of independent yields, IN, with  $A'$  for each  $Z$  of primary fission products,  $A'$  being the average mass number of the precursor primary fragments that give products with  $A$  by prompt neutron emission ( $A' = A + \bar{\nu}_A$ ) [4.1.3, 4.2.3–4.2.6]. Use of  $A'$  in both models results in approximate complementarity for light and heavy fission products ( $A'_L + A'_H = A_F$ ), which allows treatment of yield data for light and heavy products together to derive model parameters applicable to both [4.1.3]. Gaussian dispersion, modulated by even-odd proton and neutron effects, is assumed for both models.

The  $\bar{\nu}_A$  values are calculated by a modification of Terrell's  $Y(A)$  summation method [4.2.7] as described in reference [4.1.3]. The  $\bar{\nu}_A$  values, weighted by  $Y(A)$ , are normalized to sum to  $\bar{\nu}$ . Experimental  $\bar{\nu}$  values are used for normalization, if available, or else they are calculated from a modification of Howerton's equation [4.2.8], Eq. 4.1-19. The  $Y(A)$  values used in the  $\bar{\nu}_A$  summation and normalization calculations are the averages of evaluated experimental values [4.1.4–4.1.5] and recent experimental values [4.1.10–4.1.15] supplemented by values calculated from the multi-Gaussian model, described in Section 4.1, if experimental values were not available.

$Z_p$  and  $A'_p$  model calculations for U233T, U235T, PU239T, and CF252S [4.1.3] were extended in 1989 and 1992 to include TH229T, TH232F, NP238T, U238F, U238H, PU241T, and CF249T, and some systematic trends in model parameter values were deduced [4.2.5–4.2.6]. The equations used for least-squares calculations with the  $Z_p$  model [4.1.3] were modified to accommodate the additional systematic trends shown by the expanded data set [4.2.6]. The equations used for least-squares calculations with the  $A'_p$  model were those published in 1988 [4.1.3].

Experimental independent yields, supplemented by yields calculated from the  $Z_p$  or  $A'_p$  models for a fission reaction of interest, can be divided among isomeric states using either the Madland-England treatment [4.2.9] or Rudstam's treatment [4.2.10]. The spins of the states, needed for both treatments, are available from files furnished by England [4.2.11] and by Blachot [4.2.12]. Also, isomeric cumulative yields may be calculated by summation of independent yields using either England's [4.2.11] or Blachot's [4.2.12] sets of values for spins and for decay branching fractions to isomeric states.

### 4.2.1. The $Z_p$ Model

The  $Z_p$  model [4.1.3] has been used for charge distribution calculations in the two major fission yield evaluations, ENDF/B-VI [4.1.5] and UKFY2,3 [4.1.4, 4.2.13]. In the versions of

the model used [4.1.3, 4.2.5],  $Z_p$ -model parameters either are constant or are linear functions of  $A'$  in each of the regions considered. For most  $A'$ , all except those near symmetry, the parameters are constant, except  $\Delta Z$ , which has a small negative slope. ( $\Delta Z$  is the displacement of  $Z_p$  from unchanged charge distribution,  $\Delta Z = \{Z_p - A'[Z_F / A_F]\}_H = \{A'[Z_F / A_F] - Z_p\}_L$  [4.1.3].) Near symmetry the width parameter  $\sigma_Z$  changes abruptly twice to a lower value, the even-odd proton and neutron factors  $F_Z$  and  $F_N$  become 1.0, and the  $\Delta Z$  function undergoes a zig-zag transition from positive values for light fission products to negative values for heavy products (see central regions of plots in Fig. 4.2–1).

Recently, the  $Z_p$  model has been modified to include parameter slopes vs.  $A'$  and slope changes in the wing regions. Existing evaluated data sets [4.1.3, 4.2.5, 4.2.6] were used, supplemented by yield data from the fission-product separator LOHENGRIN for  $^{241}\text{Am}(2n_{th},f)$  (AM242T) from Mutterer and Siegler [4.1.15] and data for the light wings of U235T, PU239T, AM242T, and CF249T from Denschlag et al. [4.1.10–4.1.14]. All parameters were assumed to be linear functions of  $A'$  in each of several regions of  $A'$ : near symmetry, the two peaks, the two wings, and the two far wings; see Figs 4.2–1A to D where region boundaries are indicated by short vertical dotted lines and labeled B1–B6 and Ba, Bb<sup>1</sup>.

Parameters for the far wing regions,  $A' < B1$  and  $A' > B6$ , where few data exist, were assumed to be constant. For the  $\Delta Z$  and  $\sigma_Z$  parameters, the symmetric region is subdivided into three regions in which  $\Delta Z$  sharply decreases, increases, and decreases again with increasing  $A'$ , as illustrated in Fig. 4.2–1B. The  $\sigma_Z$  parameter decreases abruptly in two regions, B3–Ba and Bb–B4 in Fig. 4.2–1A, where  $Z_p$  is close to  $Z = 50$  and its complement,  $Z = 42$  for fission of uranium isotopes. The value of  $\sigma_Z$  at symmetry (Ba–Bb) is assumed to be constant with the value of the peak  $\sigma_Z$  slopes extrapolated to Ba or Bb. The wing effects, showing a sharp decrease for  $\sigma_Z$  and sharp increases for  $F_Z$  and  $F_N$  [4.1.10–4.1.14], occur in regions B1–B2 and B5–B6 close to  $Z = 28$  and its complement,  $Z = 64$  for fission of uranium isotopes. Equations defining the boundaries and parameters in each region are given below.

Values of  $Z_p$ -model parameters were determined for each of 12 fission reactions (TH229T, TH232F, U233T, U235T, U238F, U238H, NP238T, PU239T, PU241T, AM242T, CF249T, and CF252S)<sup>2</sup> by the method of least squares, weighting data equally<sup>3</sup>. However, only for U235T could all model parameters be determined by this method, although for several other fission reactions all but a few parameters could be determined. For these parameters and for a number of parameters for fission reactions for which there are few data, Eq. 4.2-1 [4.2.5, 4.2.6] was used to estimate  $Z_p$ -model parameter values, PM, that could not be determined by the least method of squares.  $Z_F$ ,  $A_F$ , and  $E^*$  are, respectively, the atomic number, mass number, and excitation energy above the ground state of a fissioning nuclide. The constants are the corresponding values for U235T<sup>4</sup>.

<sup>1</sup> Data for  $A = 68$  and  $69$  from PU239T [4.1.13] are not consistent with linear functions and were not used in the calculations reported.

<sup>2</sup> The corresponding symbols used in figures are: T9T, T2F, U3T, U5T, U8F, U8H, N8T, P9T, P1T, A2T, C9T, and C2S.

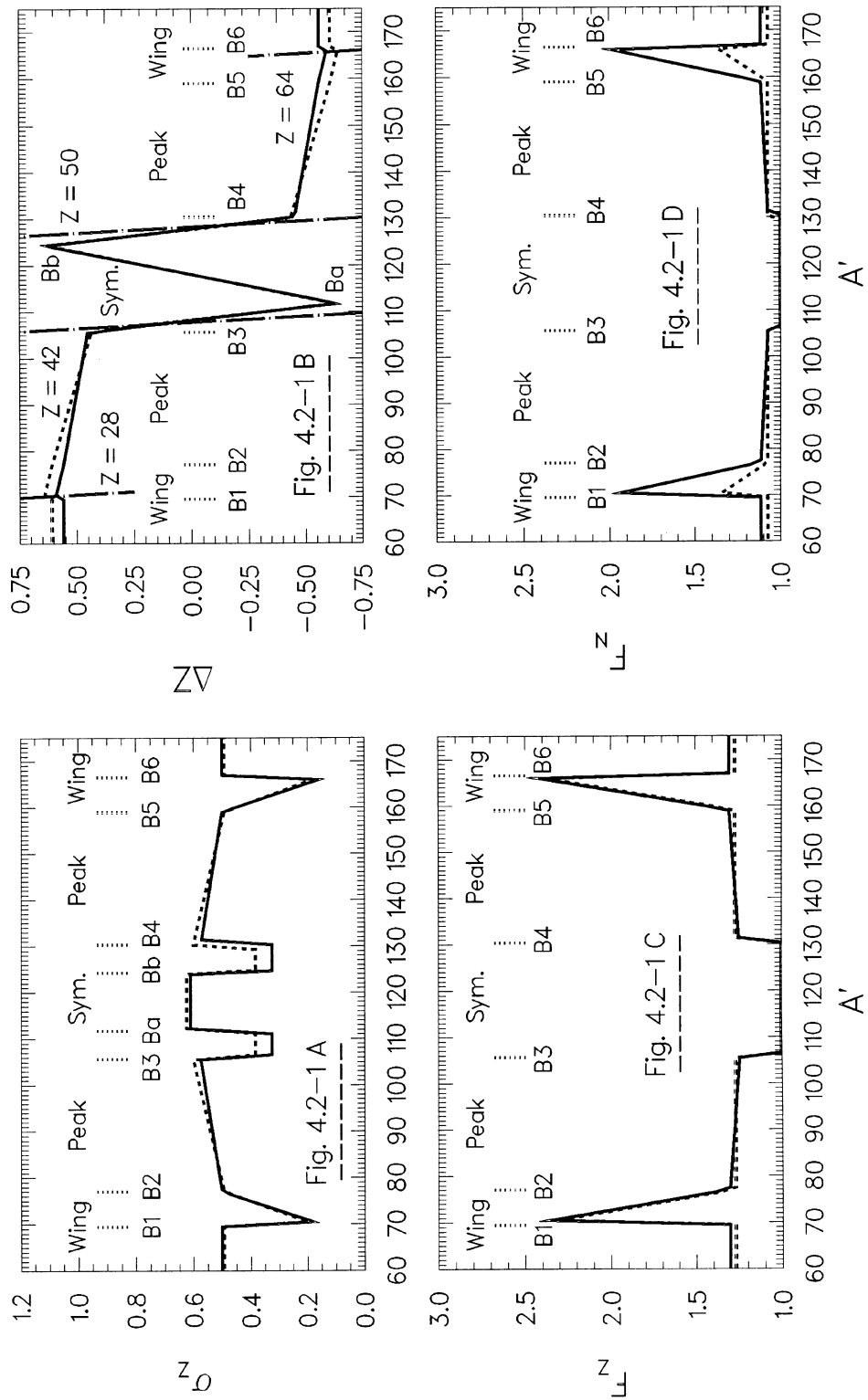
<sup>3</sup> Assignment of equal weights to all FI values allows use of all data without undue weight for small values with very small errors, as occurs with  $1/\text{variance}$  weighting.

<sup>4</sup> It was found that use of excitation energy,  $E^*$ , in the fourth term of Eq. 4.2-1 gave better representation of parameter values than did the term with  $\bar{v}$  used earlier [4.2.5–4.2.6].

## $Z_p$ Functions for U235T

Solid lines calculated by L.S., red.  $\chi^2 = 3.6(0.7)$

Dashed lines from systematics, red.  $\chi^2 = 6.6(0.6)$



$$PM = P(1) + P(2)(Z_F - 92) + P(3)(A_F - 236) + P(4)(E^* - 6.551) \quad (4.2-1)$$

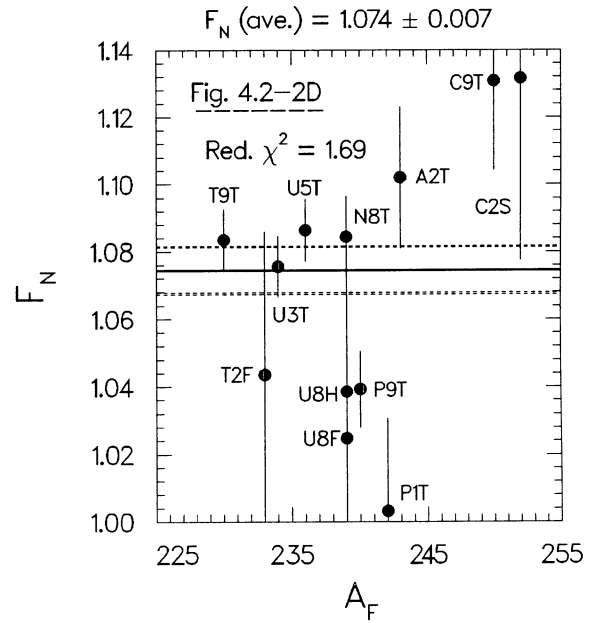
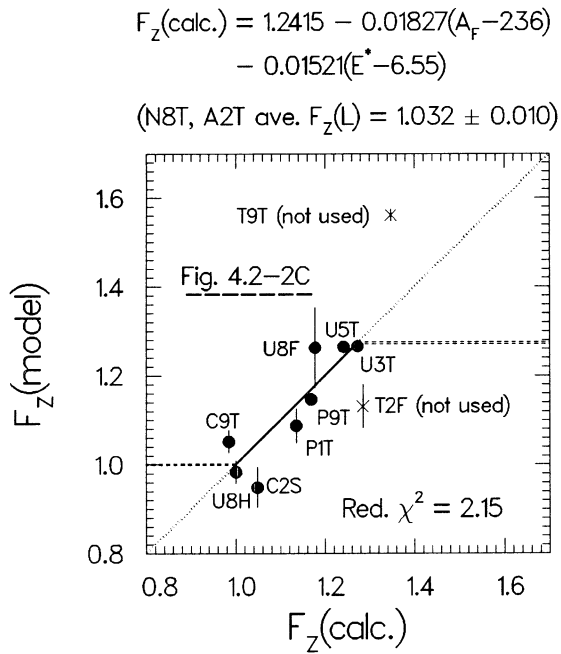
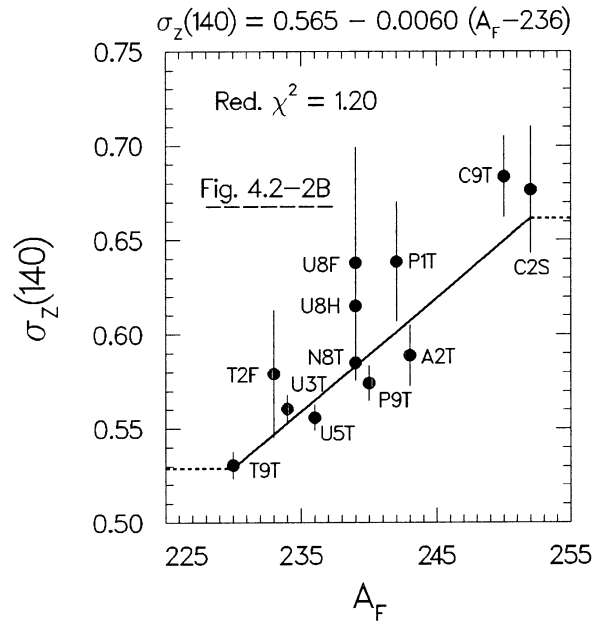
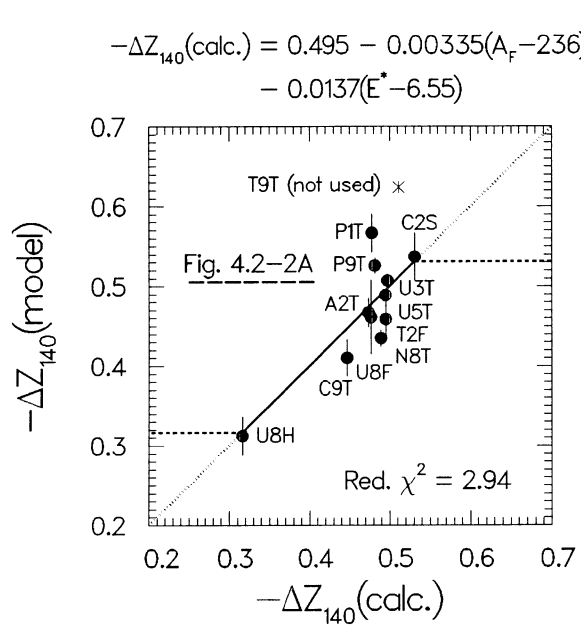
Values of  $P(i)$  were determined by the method of least squares, weighting by  $1/\text{variance}$ , from the PM values that could be determined by least squares for individual fission reactions. A  $P(i)$  value with an error as large or larger than the value was set equal to zero and not allowed to vary in subsequent calculations. For a number of parameters only  $P(1)$  could be determined to give the weighted average value.

The parameter values used are shown as points and the functions derived as lines in Figs 4.2-2 to 4.2-5. Figs 4.2-2A,C are correlation plots because the  $\Delta Z$  and  $F_Z$  parameters were found to be functions of both  $A_F$  and  $E^*$ . The largest differences between parameter values determined by least squares and the values from functions representing systematic trends were for TH229T, the lightest nuclide investigated. Since the differences may be due to changing systematic trends for light fissioning nuclides, TH229T parameter values with large deviations from values determined from systematic trends for other fissioning nuclides were not used in calculations with Eq. 4.2-1 and are shown as X in Figs 4.2-2 and 4.2-3. Also, the TH232F value for  $F_Z$  was not used and is shown by X in the Fig. 4.2-2C, and the value for the  $\Delta Z$  slope,  $\partial\Delta Z/\partial A'$ , was not used for U238H, the only highly excited nuclide investigated, and is shown by X in the Fig. 4.2-3A.

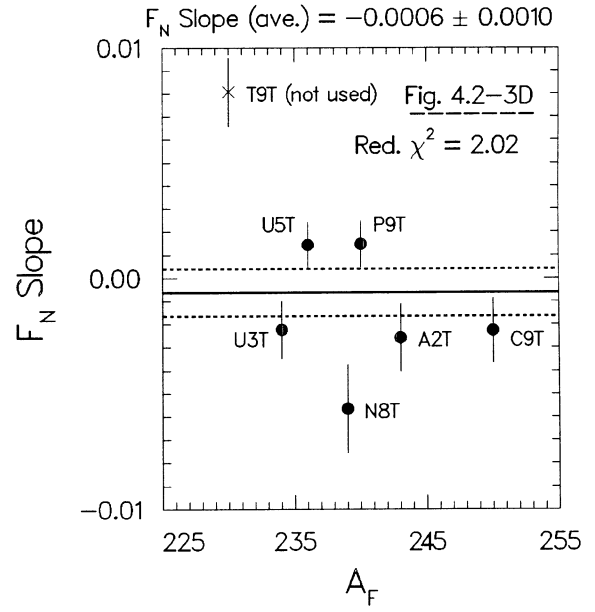
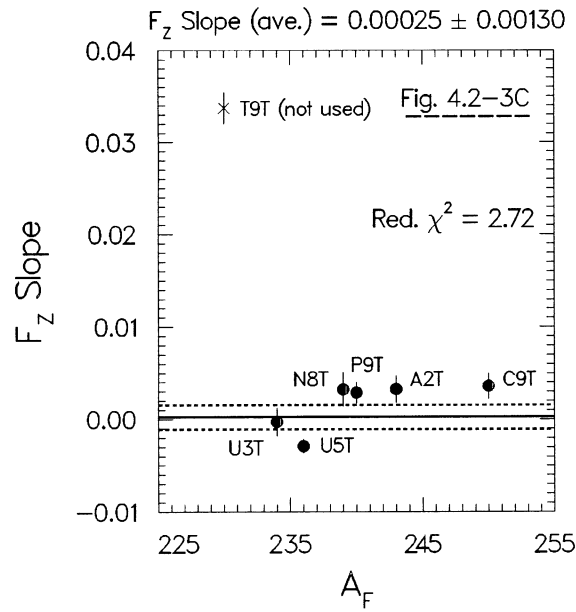
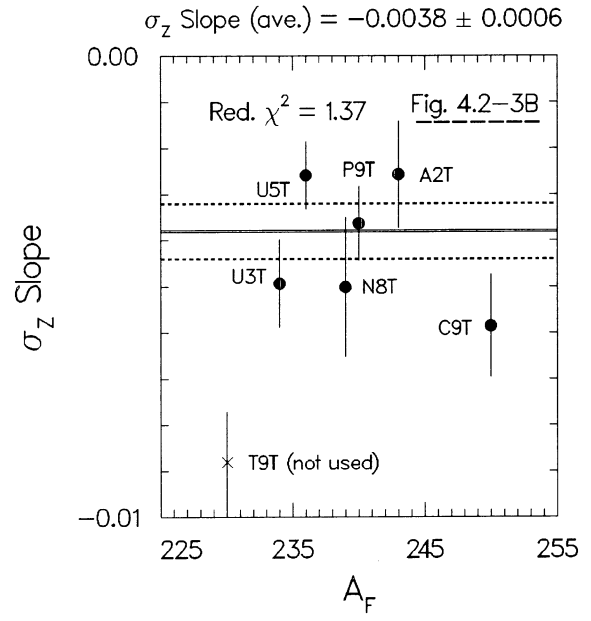
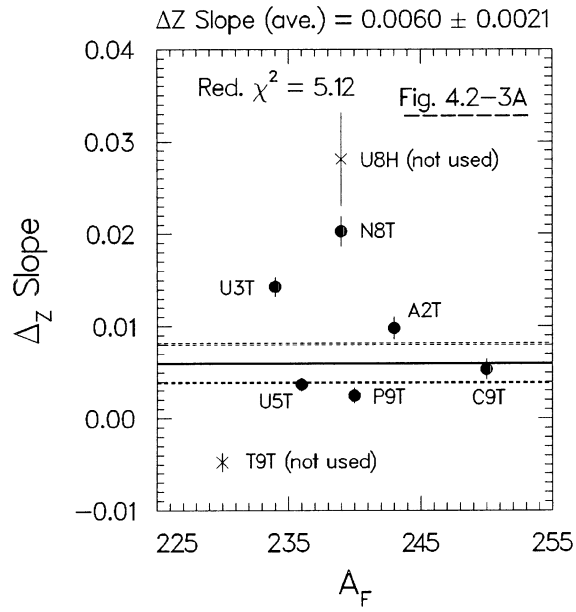
Allowing the even-odd proton factor  $F_Z$  to vary for the  $^{237}\text{Np}(2n_{th},f)$  (NP238T) and  $^{241}\text{Am}(2n_{th},f)$  (AM242T) reactions, for which only data for light products exist, values were found to be  $1.028 \pm 0.012$  and  $1.044 \pm 0.020$ , respectively, the average being  $1.032 \pm 0.010$ . In order to conserve charge for odd- $Z$  fissioning nuclides,  $F_Z$  for heavy products was set equal to  $1/F_Z$ .

$P(i)$  values determined for the parameter functions are listed in Table 4.2.1. The calculated uncertainty in the last several significant figures of  $P(i)$  values is shown in parentheses in the table. Limits are set for parameter values calculated from Eq. 4.2-1 to avoid unreasonably small or large values for fission reactions outside of the range studied. The limits, listed in Table 4.2.1, are the values of the Eq. 4.2-1 functions for the lowest and highest  $A_F$  or  $Z_F$  for which parameter values were available. No limits are necessary for average values (only  $P(1)$  determined). The parameter values below  $A' = 70$  and above  $A' = A_F - 70$  are assumed to be constant at values of the peak parameter functions where the wing functions start at B2 and B5 (see Eq. 4.2-4b). Extrapolation of the large slope parameters derived from data for the wings (regions B1-B2 and B5-B6) would give unreasonably large or small parameter values for very low and very high  $A'$  (below B1 and above B6 - the far wings). This can be seen in Figs 4.2-1A to D, which show plots of two sets of parameter functions for U235T: (1) those determined by least squares (solid lines) and (2) those derived from systematic trends using Eq. 4.2-1 and values from Table 4.2.1 (dashed lines). The two sets of functions show the same trends and are similar in magnitude except the systematic  $F_N$  function considerably underestimates the wing values for U235T.

# PEAK PARAMETERS FOR A'(140)



# SLOPES OF PEAK PARAMETERS



# WING PARAMETER SLOPES

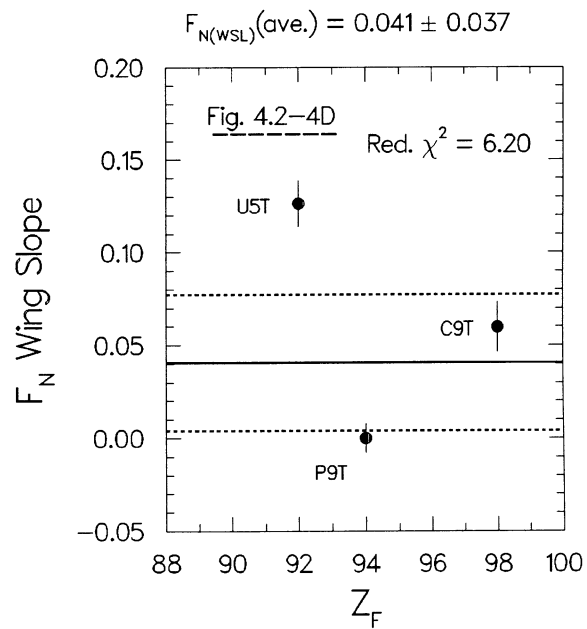
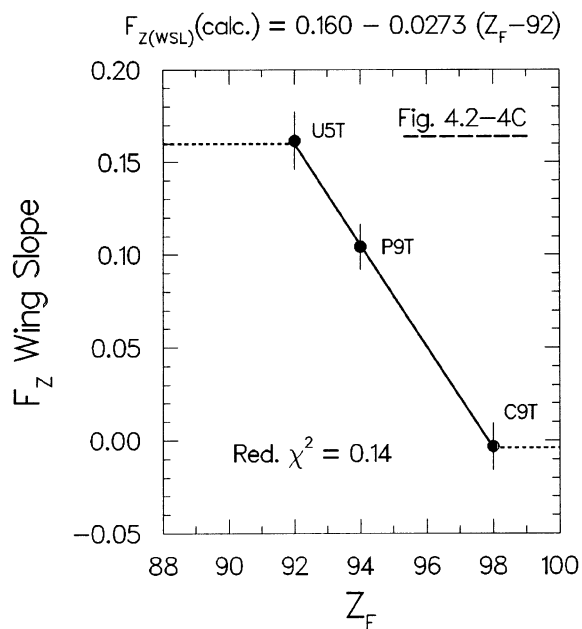
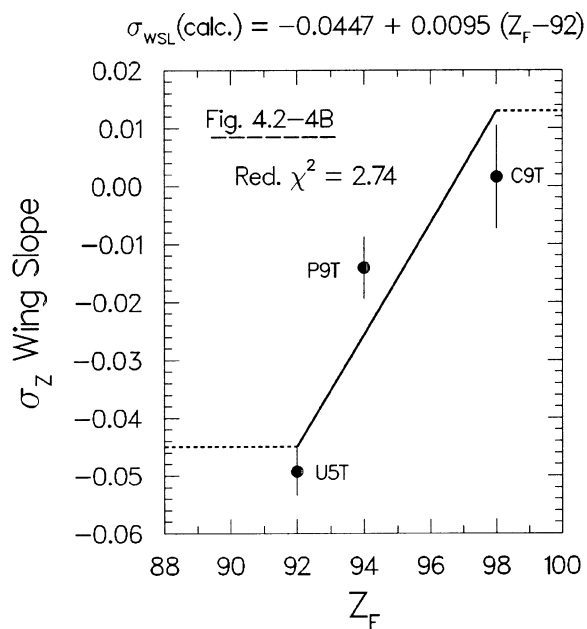
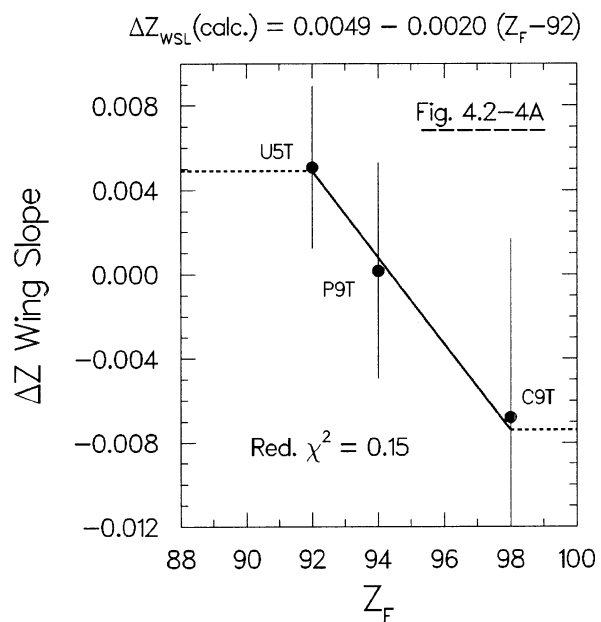




TABLE 4.2.1.  $Z_p$  - MODEL PARAMETERS FOR EQUATION (4.2-1)

$$PM = P(1) + P(2)(Z_F - 92) + P(3)(A_F - 236) + P(4)(E^* - 6.551) \quad (4.2-1)$$

Parameter	P(1)	P(2)	P(3)	P(4)	Red. $\chi^2$	No. <sup>#</sup>	Limits
<b>Parameters for regions near peaks (B2-3, B4-5)</b>							
$\sigma_Z(140)$	0.565(4)	0.0	0.0060(9)	0.0	1.19	12	0.529-0.662
$\Delta Z(140)$	-0.495(13)	0.0	0.0034(29)	0.0137(54)	2.94	11	-0.531-0.316
$F_Z^*$	1.242(15)	0.0	-0.0183(30)	-0.0152(43)	2.15	8	1.000-1.274
$F_N^*$	1.074(7)	0.0	0.0	0.0	1.69	12	—
$\partial\sigma_Z/\partial A'$	-0.0038(6)	0.0	0.0	0.0	1.37	6	—
$\partial\Delta Z/\partial A'$	-0.0060(21)	0.0	0.0	0.0	5.12	6	—
<b>Parameters for regions near symmetry (B3-4)</b>							
SL50	0.189(22)	0.0	-0.0063(28)	0.0	3.36	4	0.100-0.201
$\sigma_{50}$	0.343(20)	0.061(9)	0.0	0.0	1.95	4	0.343-0.710
$\Delta Z_{\max}^{**}$	0.632(32)	0.0	0.0	0.0	—	1	—
$A'_{\max}$	124.35(23)	0.29(15)	0.0	0.0	1.26	3	124.35-126.09
<b>Parameters for wing regions (B1-2, B5-6)</b>							
$\sigma_Z WS$	-0.0447(102)	0.0095(44)	0.0	0.0	2.74	3	-0.045-0.013
$\Delta Z WS$	0.0049(35)	-0.0020(15)	0.0	0.0	0.15	3	-0.0074-0.0049
$F_Z WS$	0.160(12)	-0.0273(32)	0.0	0.0	0.14	3	-0.004-0.160
$F_N WS$	0.041(37)	0.0	0.0	0.0	6.20	3	—

<sup>#</sup> No. = Number of fission-reaction parameter values used in the least-squares calculation.

\* The weighted averages for the  $F_Z$  and  $F_N$  slopes from 6 values for each are: 0.00025(130) and -0.0006(10), with reduced  $\chi^2$  of 2.72 and 2.02, respectively. For odd-Z fissioning nuclides the  $F_Z$  for light products is taken to be 1.032(10), the average of 2 values, and the reciprocal of this value is used for heavy products. (See discussion.)

\*\* The value of  $\Delta Z_{\max}$  could be determined only for U235T.

As a check on the validity and possible usefulness of the  $Z_p$ -model parameters derived from Eq. 4.2-1, calculations of FI values were made for each of the 12 fission reactions investigated using systematic  $Z_p$ -model parameter values calculated from Eq. 4.2-1 with parameter values from Table 4.2.1. The reduced  $\chi^2$  values determined were within a factor of about 2 of those determined by the method of least squares with variation of as many parameters as possible.

Application of Eq.4.2-2, proposed in 1992 [4.2.6], for estimation of uncertainties in FI values calculated from  $Z_p$  model parameters was investigated using parameter values both determined by least squares calculations and estimated from systematic trends (Eq. 4.2-1 and Table 4.2.1).

$$\% \text{ Error} = 6 + 6 \exp[(Z-Z_p)/\sigma_Z] \quad (4.2-2)$$

Reduced  $\chi^2$  values calculated by taking the error to be the square root of the sum of the experimental variance and the model variance estimated from Eq. 4.2-2 were mostly  $< 1.0$ .

### ***Equations for the $Z_p$ Model***

The equations for the  $Z_p$  model given below involve the error function of  $x$ ,  $\text{erf}(x)$ . Complementarity of light and heavy fission products is approximated by  $A' = A + \bar{v}_A$ <sup>5</sup> to allow the same or complementary functions to be used for both light and heavy products.

$$\text{FI}(A,Z) = [0.5][F(A)][N(A)][\text{erf}(V) - \text{erf}(W)] \quad (4.2-3)$$

$$V = \frac{Z(A) - Z_p(A) + 0.5}{\sigma_Z(A')\sqrt{2}} \quad (4.2-3a)$$

$$W = \frac{Z(A) - Z_p(A) - 0.5}{\sigma_Z(A')\sqrt{2}} \quad (4.2-3b)$$

$$Z_p(A_H) = A'_H[Z_F/A_F] + \Delta Z(A'_H) \quad (4.2-3c)$$

$$Z_p(A_L) = A'_L[Z_F/A_F] - \Delta Z(A'_{Hc}), (A'_{Hc} = A_F - A'_L) \quad (4.2-3d)$$

	for Z	for N	
$F(A) = [F_Z(A')][F_N(A')]$	even	even	
$F(A) = [F_Z(A')]/[F_N(A')]$	even	odd	(4.2-3e)
$F(A) = [F_N(A')]/[F_Z(A')]$	odd	even	
$F(A) = 1/[F_Z(A')][F_N(A')]$	odd	odd	

The normalization factor,  $N(A)$ , applied to achieve  $\Sigma(\text{FI}) = 1.00$  for each  $A$ , is required because the even-odd factors,  $F(A)$ , destroy the inherent normalization properties of Gaussian distributions. Values of  $N(A)$  seldom deviated by more than 10% from unity.

The functions used for  $\Delta Z(A')$ ,  $\sigma_Z(A')$ ,  $F_Z(A')$ , and  $F_N(A')$  depend on  $A'$  and the region in which  $A'$  falls. The region boundaries, shown as short dotted lines and labelled B1–6 and Ba,b in Figs 4.2–1A to D, are defined below. The parameters in the functions for each region may be calculated by the method of least squares, or they may be estimated from derived systematic trends by using Eq. 4.2-1 with parameter values from Table 4.2.1.

---

<sup>5</sup> Values of  $\bar{v}_A$  were calculated using the modified [4.1.3] Terrell method [4.2.7] from average experimental chain yields supplemented by model calculated values if there were no experimental values (see Section 4.1).

$$B1 = 70 \quad (4.2-4a)$$

$$B2 = 77 + 0.036(A_F - 236) \quad (4.2-4b)$$

$$B3 = A_F - B4 \quad (4.2-4c)$$

$$B4 = \frac{\Delta Z_{\max} - \Delta Z(140) + A'_{\max} [SL50] + \partial \Delta Z / \partial A' [140]}{SL50 + \partial \Delta Z / \partial A} \quad (4.2-4d)$$

$$B5 = A_F - B2 \quad (4.2-4e)$$

$$B6 = A_F - B1 \quad (4.2-4f)$$

$$Ba = A_F - A'_{\max} \quad (4.2-4g)$$

$$Bb = A'_{\max} \quad (4.2-4h)$$

( $\Delta Z_{\max}$ ,  $\Delta Z(140)$ ,  $A'_{\max}$ ,  $SL50$ , and  $\partial \Delta Z / \partial A'$  are model parameters determined by least squares or from Eq. 4.2-1 with values from Table 4.2.1.)

*Peak regions (B2–B3, B4–B5)*

$$\Delta Z(A'_H) = \Delta Z(140) + \partial \Delta Z / \partial A' [A'_H - 140] \quad (4.2-5a)$$

$$\sigma_Z(A'_H) = \sigma_Z(140) + \partial \sigma_Z / \partial A' [A'_H - 140] \quad (4.2-5b)$$

$$\sigma_Z(A'_L) = \sigma_Z(A'_{Hc}), \quad (A'_{Hc} = A_F - A'_L) \quad (4.2-5c)$$

$$F_Z(A') = F_Z(140) \quad (4.2-5d)$$

$$F_N(A') = F_N(140) \quad (4.2-5e)$$

*Near symmetry region (B3–B4)*

$$F(A) = 1.00 \quad (4.2-6a)$$

$$\mathbf{B3-Ba:} \quad \Delta Z(A') = \Delta Z(B3) - SL50[A' - B3] \quad (4.2-6b)$$

$$\sigma_Z(A') = \sigma_{50} \quad (4.2-6c)$$

$$\mathbf{Ba-Bb:} \quad \Delta Z(A') = \Delta Z(Ba) + \{A' - Ba\} \{[\Delta Z(Bb) - \Delta Z(Ba)] / [Ba - Bb]\} \quad (4.2-6d)$$

$$\sigma_Z(A') = \sigma_Z(140) - \partial \sigma_Z / \partial A' [140 - Bb] \quad (4.2-6e)$$

$$\mathbf{Bb-B4:} \quad \Delta Z(A') = \Delta Z(B4) + SL50[B4 - A'] \quad (4.2-6f)$$

$$\sigma_Z(A') = \sigma_{50} \quad (4.2-6g)$$

*Wing regions (B1–B2, B5–B6)*

$$\Delta Z(A'_L) = \Delta Z(B2) + \Delta ZWS[B2 - A'_L] \quad (4.2-7a)$$

$$\Delta Z(A'_H) = \Delta Z(B5) - \Delta ZWS[A'_H - B5] \quad (4.2-7b)$$

$$\sigma_Z(A'_L) = \sigma_Z(A'_{Hc}), \quad (A'_{Hc} = A_F - A'_L) \quad (4.2-7c)$$

$$\sigma_Z(A'_H) = \sigma_Z(B5) + \sigma_ZWS[B2 - A'_L] \quad (4.2-7d)$$

$$F_Z(A'_L) = F_Z(140) + F_ZWS[B2 - A'_L] \quad (4.2-7e)$$

$$F_Z(A'_H) = F_Z(140) + F_Z WS[A'_H - B5] \quad (4.2-7f)$$

$$F_N(A'_L) = F_N(140) + F_N WS[B2 - A'_L] \quad (4.2-7g)$$

$$F_N(A'_H) = F_N(140) + F_N WS[A'_H - B5] \quad 4.2-7h)$$

*Far wing regions (<B1, >B6)*

$$\Delta Z(A'_L) = \Delta Z(B2) \quad (4.2-8a)$$

$$\Delta Z(A'_H) = \Delta Z(B5) \quad (4.2-8b)$$

$$\sigma_Z(A') = \sigma_Z(B5) \quad (4.2-8c)$$

$$F_Z(A') = F_Z(140) \quad (4.2-8d)$$

$$F_N(A') = F_N(140) \quad (4.2-8e)$$

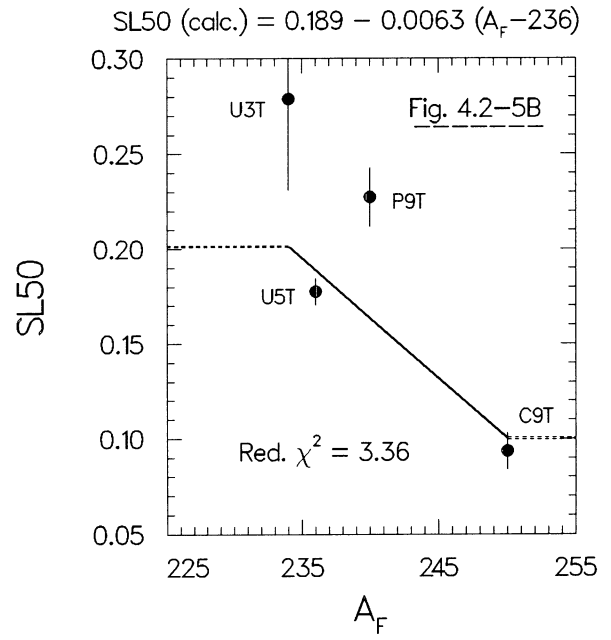
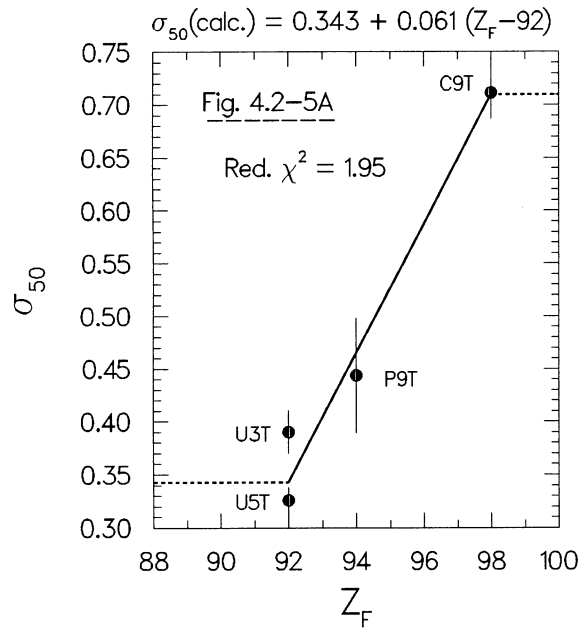
#### 4.2.2. The A'<sub>p</sub> model

Calculations have been started to determine if data that have become available since 1988 [4.1.10–4.1.15, 4.2.14] require model modifications. Also, an investigation has been started of a simplified A'<sub>p</sub> model that involves only five Gaussian functions to represent element yields, instead of the many needed for association of one model parameter with each element yield [4.1.3, 4.2.3–4.2.6]. Investigation of the effects of new data [4.1.10–4.1.15] for the wings of mass-yield curves on the A'<sub>p</sub> model and calculations with the simplified model are not sufficiently advanced to report. Results of investigations of yields near symmetry are discussed below.

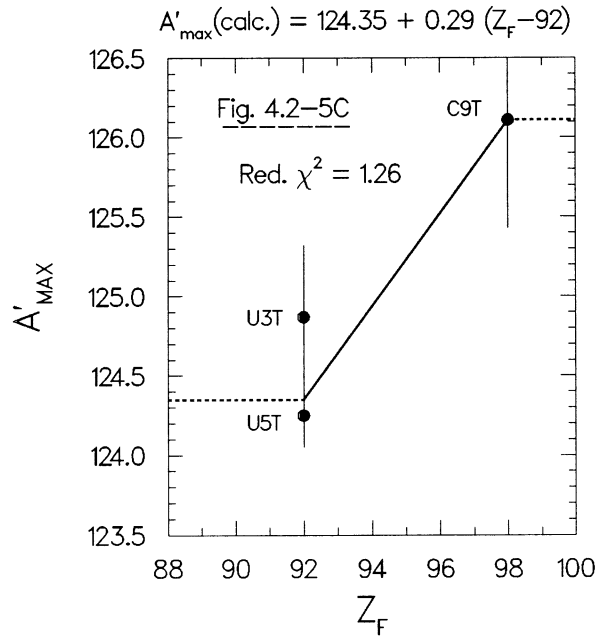
Results of calculations by the method of least squares with currently available data for U235T fission products near symmetry ( $Z = 42$ – $50$ ) are shown in Figs 4.2–6A to D. The data include independent yield values for individual elements, or for complementary element pairs when data were available, from radiochemical (RC) measurements [4.1.3] and from on-line isotope-separator measurements with OSIRIS [4.2.14] and SOLIS [4.2.15]. Filled symbols represent  $^{43}\text{Tc}$  yields; open symbols represent yields for heavy fission products. For  $^{47}\text{Ag}$  and the  $^{50}\text{Sn}$ – $^{42}\text{Mo}$  pair, the determined Gaussian width parameters,  $\sigma = \sigma_{A'}$ , are close to the average global parameter of 1.50 [4.1.3]. However, for  $^{48}\text{Cd}$  and the  $^{49}\text{In}$ – $^{43}\text{Tc}$  pair the values of  $\sigma$  are considerably larger, suggesting that isotopes of these elements could be formed by more than one process.

The data for  $^{48}\text{Cd}$  and the  $^{43}\text{Tc}$ – $^{49}\text{In}$  pair can be represented better by two Gaussian functions, called symmetric (S) and asymmetric (A), each with a  $\sigma$  value of 1.50 and peak separation of about 4 A', as shown in Figs 4.2–7B and C. The greatest deviation of the curves from data points are for the  $^{43}\text{Tc}$ – $^{49}\text{In}$  pair; also, for this pair there are discrepancies between some data values, so a reevaluation of the data is needed. For the  $^{47}\text{Ag}$  and the  $^{42}\text{Mo}$ – $^{50}\text{Sn}$  pair, the second curves contribute < 1% to the total yields, so have little effect on the curves for the sums. However, the small independent yields of  $^{121}\text{Sn}$  and  $^{123}\text{Sn}$ , shown to the lower left of the curve in Fig. 4.2–6D, are represented well by the dotted symmetric curve in Fig. 4.2–7D. The dashed asymmetric curve in Fig. 4.2–7A for  $^{47}\text{Ag}$  is estimated from the semi-global model discussed below. The parameter values with errors shown in Figs 4.2–6 and 4.2–7 are from least-squares calculations; those without errors are from the semi-global model.

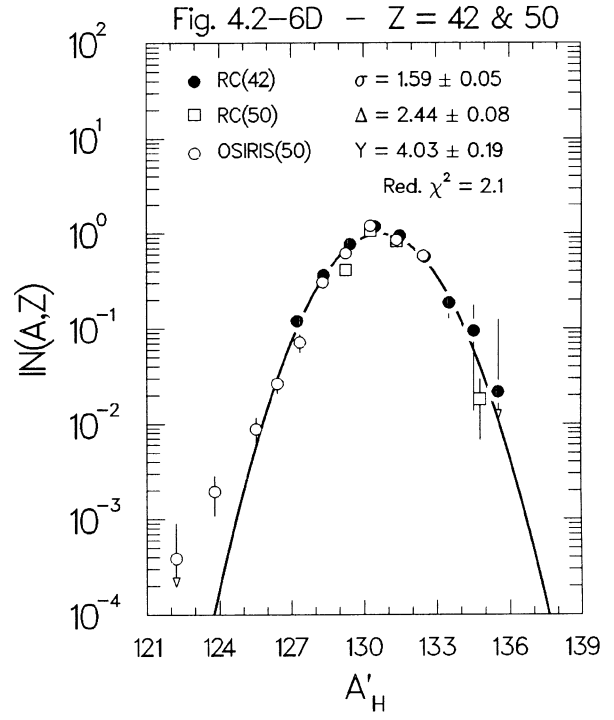
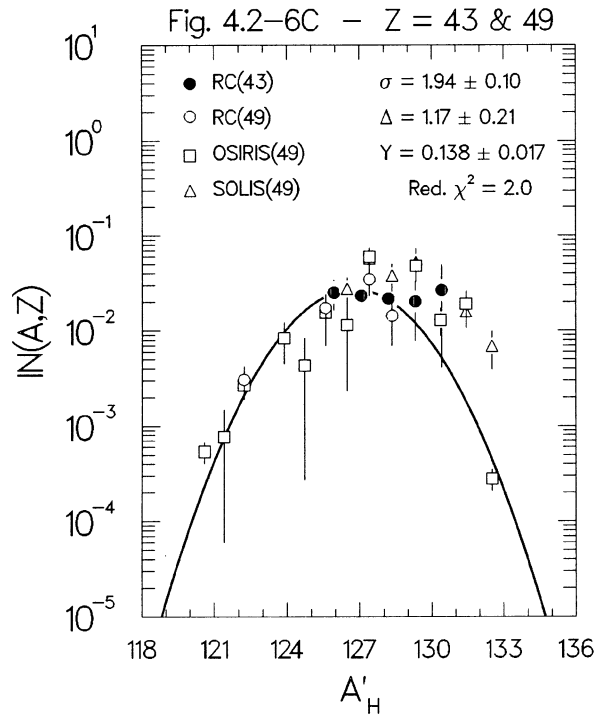
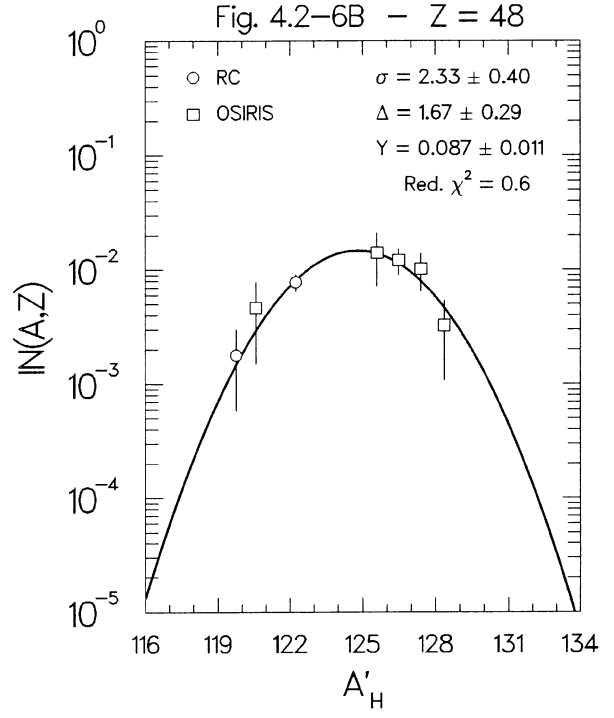
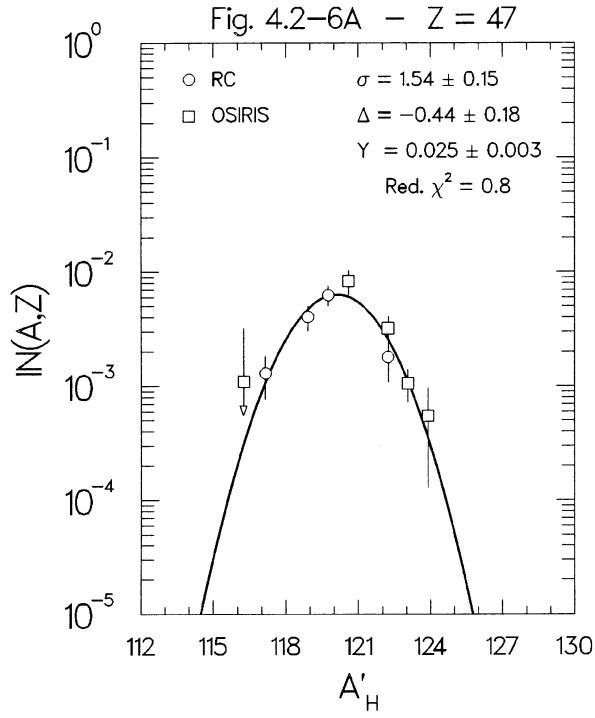
# PARAMETERS NEAR SYMMETRY



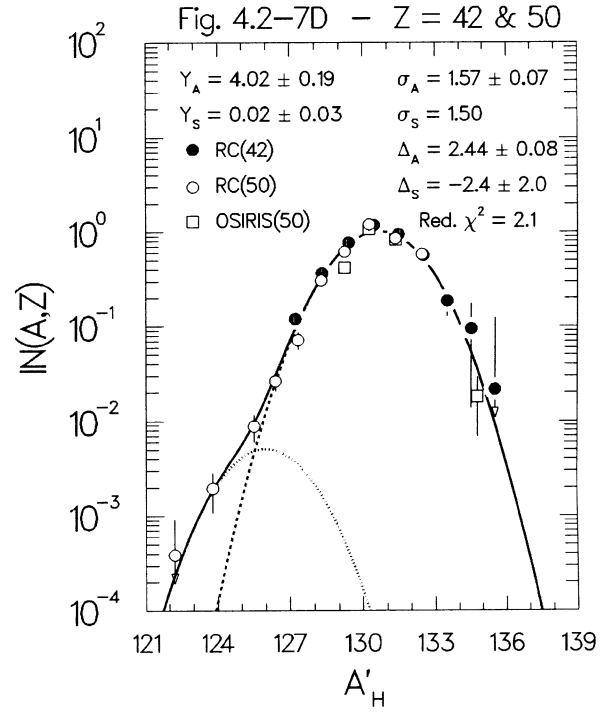
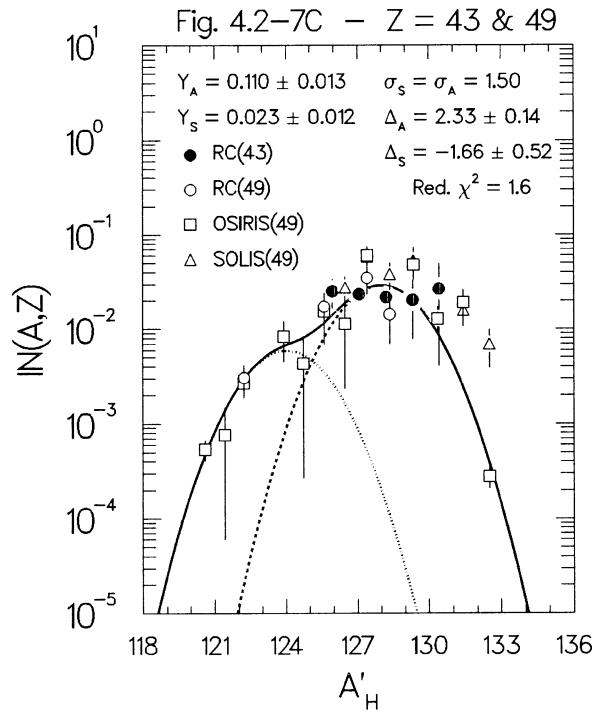
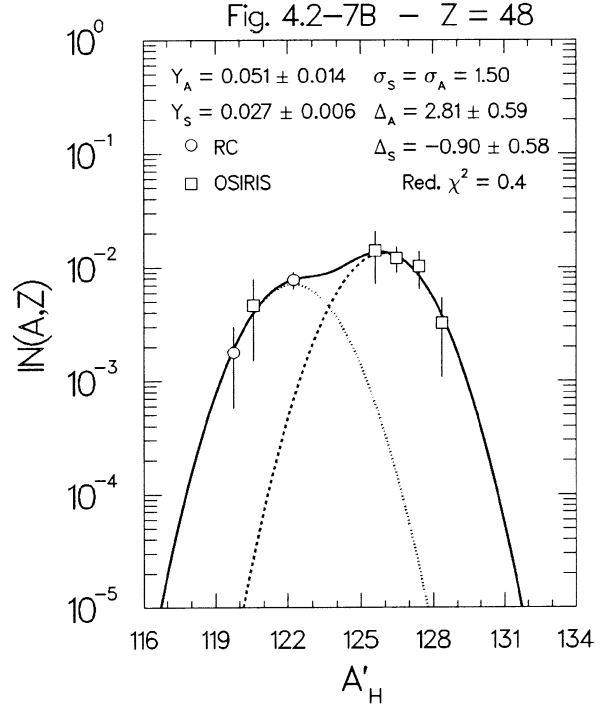
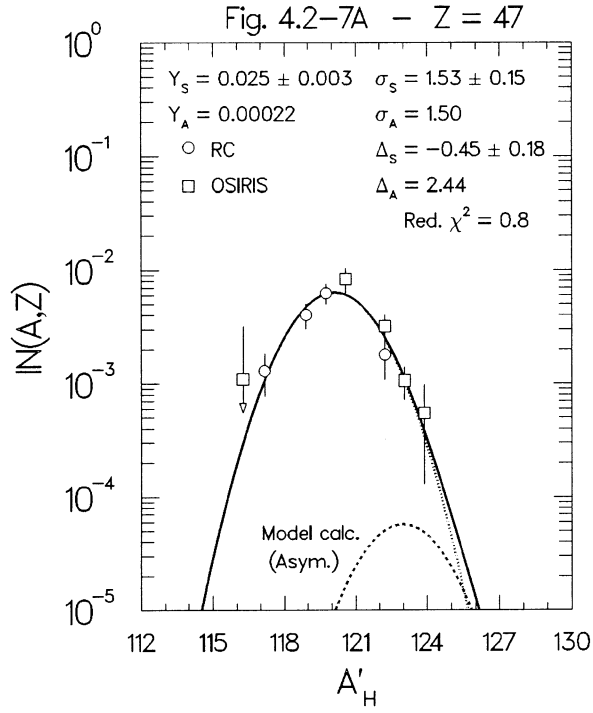
$$\Delta Z_{\text{max}}(\text{U5T}) = 0.632 \pm 0.032$$



# U235T – $A'_p$ Model Near Symmetry



# U235T – 2-Mode $A'_p$ Model Near Symmetry



The transition from asymmetric to symmetric charge distribution occurs just below  $_{50}\text{Sn}$  and just above the complementary element,  $_{42}\text{Mo}$  for fission of uranium nuclides. Both processes contribute to the yields of  $_{48}\text{Cd}$  and the  $_{43}\text{Tc}$ – $_{49}\text{In}$  pair from  $\text{U235T}$ . The transition can be represented by a semi-global 2-mode model for the range  $Z = 42$ – $50$ . It was assumed in calculations that the even-odd factors for the asymmetric mode had near average global values,  $F_Z = 1.25$  and  $F_N = 1.07$  [4.1.3, 4.2.5, 4.2.6], and were unity for the symmetric mode, as would be appropriate for fission by a liquid-drop mechanism. Least squares calculations allowing  $F_Z$  and  $F_N$  to vary gave values consistent with the assumptions within rather large calculated errors due to the limited data available.

The results of calculations are summarized in Figs 4.2–8A and B. The  $\Delta A'$  functions, shown in Fig. 4.2–8B, are assumed to be 0.0 at  $Z = 46$  (symmetry), as was the 1988  $\Delta A'$  function [4.1.3]. The width,  $\sigma_{YS}$ , of the central Gaussian function representing the symmetric mode could not be determined by least-squares calculations and was estimated to be about  $2.4 Z$  from the average width,  $\sigma_{YAS}$ , of  $6.0 A$  for the central  $Y(A)$  Gaussian function discussed in Section 4.1 ( $6.0 [Z_F / A_F] \approx 2.4$ ). These calculations should be extended, if possible, to other fissioning systems.

The 2-mode concept was proposed in 1951 [4.2.16]. The bases of this and later 2-mode proposals has been the pattern of chain yields near symmetry, suggesting a small central peak. The current proposal is based on patterns both of chain yields and of independent yields near symmetry and includes the suggestion that the symmetric mode may be associated with a liquid-drop mechanism. The symmetric mode is not observed for spontaneous fission reactions since there is no evidence for a central chain-yield peak (see Section 4.1), but the central peak becomes important in higher energy fission and increases with increasing excitation energy.

The  $A'_p$  model could be improved near symmetry by incorporating the 2-mode model. As shown in Fig. 4.2–7C, the symmetric mode contributes very little to peak element yields ( $Z \geq 50$  and light complements) so parameters for the peaks would not be affected appreciably by the change, at least for low-energy fission.

The following equation, analogous to Eq. 4.2-2, was proposed in 1992 [4.2.6] for estimation of uncertainties in independent yield values calculated from the  $A'_p$  model.

$$\% \text{ Error} = 6 + 6 \exp\{(A' - A'_p)/\sigma_{A'}\} \quad (4.2-9)$$

Preliminary studies using the above equation gave reasonable uncertainties [4.2.6], but more detailed investigations, as was done for the  $Z_p$  model, have not yet been carried out.



## U235T – 2–Mode A' Model Near Symmetry

Fig. 4.2–8A – Yield Functions

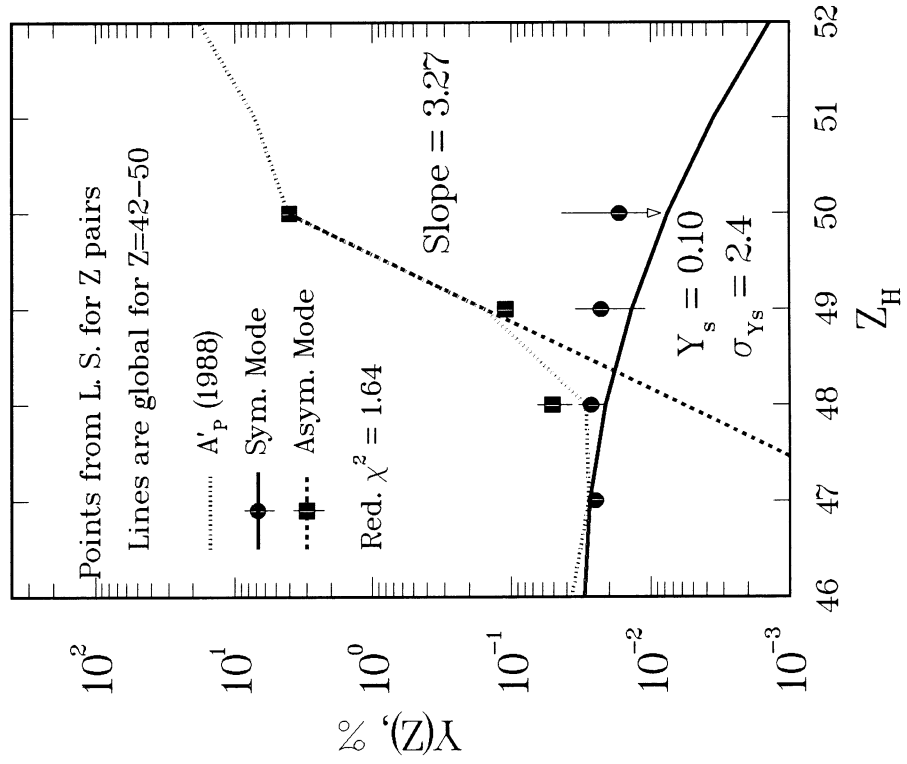
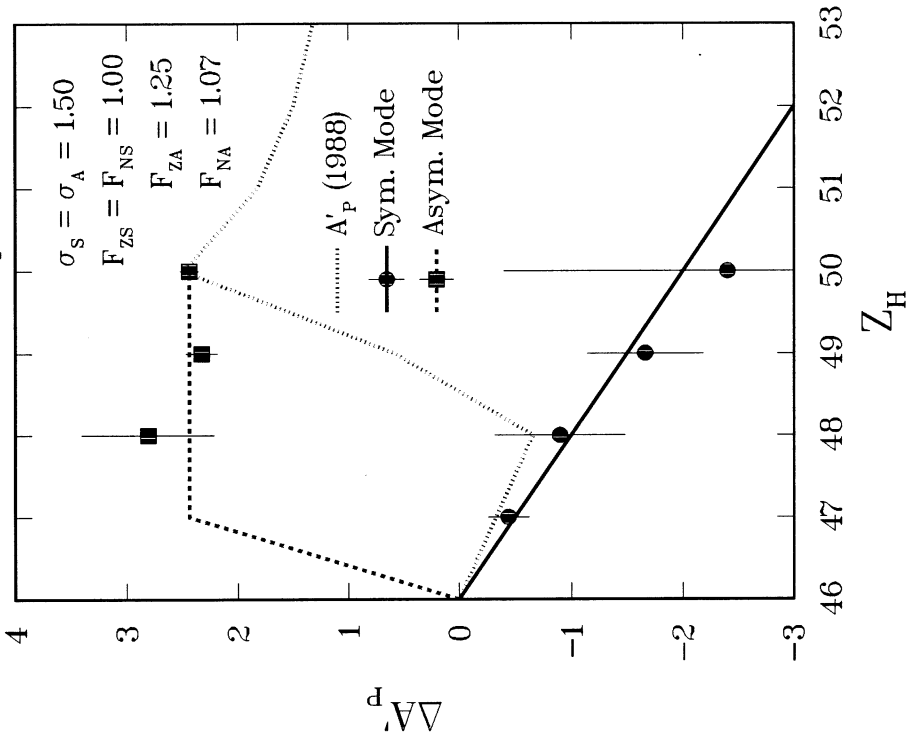


Fig. 4.2–8B –  $\Delta A'_P$  Functions



### 4.2.3. Yield calculations

A computer program has been developed to calculate from equations representing systematic trends, presented in Sections 4.1 and 4.2.1, values for the following quantities:  $\bar{\nu}$ ,  $\bar{\nu}_A$ ,  $Y(A)$ , and  $FI(Z,A)$  with only values of  $A_F$ ,  $Z_F$ , and  $E_n$  needed as input. Estimated uncertainties in  $Y(A)$  and  $FI(Z,A)$  are also calculated from Eqs. 4.1-1 and 4.2-2. The program includes the option to use experimental  $\bar{\nu}$ ,  $Y(A)$ , and  $FI(Z,A)$  values and uncertainties in place of calculated values and to use model parameter values derived by least squares from data for the fission reaction of interest in place of those derived from systematic trends. Independent, cumulative, and fractional cumulative yields and the uncertainties in each of them are then calculated for each fission product. Examples of tables of yields that can be produced are given in Appendix A.1. Treatment of isomeric-state yields is included in the current version (CYI) of the computer program.

### 4.2.4. Summary and conclusions

The  $Z_p$  model for nuclear charge distribution has been modified to represent changing slopes of model functions near symmetry and on the mass-yield wings. Model parameters determined for 12 fission reactions, in the ranges  $Z_F = 92-98$  and  $E^* \leq \sim 20$  MeV, show systematic trends with  $Z_F$ ,  $A_F$ , and  $E^*$ , and these trends are represented by Eq. 4.2-1 with parameters listed in Table 4.2.1. Uncertainties in calculated fractional independent yields,  $FI(Z,A)$ , can be estimated from empirical Eq. 4.2-2.

The systematic trends in fission-product yields that have been described allow reasonable estimates to be made of unmeasured yields and increase the understanding of fission processes. Most estimates of yields in the range investigated, 92 to 98 in  $Z_F$  and  $\leq \sim 20$  MeV in  $E^*$ , are believed reliable to within the estimated uncertainties given by Eqs. 4.2-2 and 4.2-9.

The 50-proton shell has a pronounced effect on fission yields as shown by the large increase in independent yields of fission-product pairs from U235T with  $Z = 50, 42$  compared to the yields of products with  $Z = 49, 43$  and  $48$  and  $47$  (see  $A'_p$ -model plots in Fig. 4.2-7A to 4.2-7D — note changes in the ordinate scales). The effect is represented by the  $Z_p$  model as very large  $FI$  for  $_{50}\text{Sn}$  and  $_{42}\text{Mo}$  fission products over several  $A'$  causing the  $\Delta Z$  function to remain close to  $Z = 50$  and  $42$  and  $\sigma_Z$  to be small over the same  $A'$  ranges [4.1.3]. The 50-proton and 82-neutron shells may be the principal causes of asymmetric mass and charge division for most fissioning actinide nuclides. The nearly constant position of the light side of the heavy mass peak [4.2.17] can be associated with the large increase in  $_{50}\text{Sn}$  product yields compared to  $_{49}\text{In}$  products. However, the 2-mode treatment, discussed above, indicates that the transition from the symmetric (liquid drop) to the asymmetric mode occurs over several  $Z$ .

The wing effects, a rapid decrease in  $\sigma_Z$  and rapid increases in  $F_Z$  and  $F_N$  with decreasing  $A'_L$  [4.1.10–4.1.14], may be due to the influence of the 28-proton shell on yields. The experimental  $FI$  for  $_{28}\text{Ni}$  fission products near  $A = 70$  are quite large [4.1.10–4.1.14] and influence values determined for  $\sigma_Z$  and  $F_Z$ . Also, the  $Z = 28$  shell seems to influence chain yields. Although the average  $A$  of the light mass-yield peak shifts linearly to higher  $A$  with increasing  $A$  of fissioning nuclides [4.2.18], the lower portion of the light side of the light peak shifts much less [4.1.10–4.1.14] causing peaks to broaden.

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### 4.3. ISOMERIC YIELDS IN FISSION AND RUDSTAM'S MODEL

#### 4.3.1. Background

At present several data files (JEF-2 in Europe, ENDF/B-VII in the USA and JENDL-3 in Japan) containing fission yields are being assembled. In these files the independent yields of isomeric states are needed, but the experimental basis for these yields is meager except for thermal fission of  $^{235}\text{U}$ . Therefore, it is necessary to rely on model estimates. One widely used model is due to Madland and England [4.3.1]. The present work is an attempt to further develop this model in order to get a more reliable way to estimate unmeasured isomeric yields.

#### 4.3.2. Model for the partition of the nuclear yield on isomeric states

##### 4.3.2.1. Main model

Madland and England assume the spin distribution of the fragments after the evaporation of neutrons to be given by the formula [4.3.1]:

$$P(J) = \text{const} \times (2J + 1) e^{-[(J+1/2)/J_{rms}]^2}, \quad (1)$$

with  $J_{rms}$  characterizing the shape of the spin distribution. They then assume that fragments with  $J$  nearer to the spin of a particular isomeric state will feed that state. This assumption is not retained here. Instead, the probability that the spin will decrease by one unit is taken to be proportional to the density of nuclear states of spin  $J - 1$ , and the probability that the spin will increase is then proportional to the density of states of spin  $J + 1$  (only  $E1$  transitions are considered). The spin distribution of the nuclear states is again of the form (1) but now with another parameter  $J_{nuc}$ .

The ratio between the number of nuclear states of spin  $(J - 1)$  and those of spin  $(J + 1)$  is denoted by  $Z(J)$ . It is found to be equal to

$$Z(J) = \frac{2J-1}{2J+3} e^{(4J+2)/J_{nuc}^2}. \quad (2)$$

With this notation the relative probability to decrease the spin by one unit will be  $Z/(1 + Z)$ , and the probability to increase it by one unit will be  $1/(1 + Z)$  if the energy effects are disregarded. That simplification will lead however to erroneous results if the isomeric state is situated highly above the ground state. Such a case has been found at Studsvik [4.3.2]: the  $^{131}\text{In}$  isomer of spin  $21/2^+$  is situated about 4 MeV above the ground state. If the fragment should end up with an excitation energy, after neutron evaporation, of less than 4 MeV that isomer obviously cannot be formed. This seriously decreases the fractional yield of the isomer in question. Experimentally it was found to be only 0.40% [4.3.3].

In the following it is assumed that all fragment excitation energies are equally probable. This means that the ground state will always be fed if the energy is below the position of the isomer at energy  $E_x$ , something which happens with a frequency of  $E_x/E_{max}$ .  $E_{max}$  is here the maximum excitation energy after the neutron evaporation. It can be approximated by the neutron separation energy +50 keV.

If the spin of the low-spin isomer is  $J_l$ , and  $J_l = J - 1$ , this isomer can be reached by the emission of 1, 3, 5, 7, ... gamma-rays (only gamma emission leading to a spin change is considered). The probability for this is proportional to

$$\frac{Z(J)}{1+Z(J)}(1 + S2 + S4 + \dots). \quad (3)$$

The term S2 has the form  $\frac{Z}{(1+Z)^2}$ ; S4 has the form  $S2^2$ , etc. These terms decrease rapidly. Energy restrictions also limit the number of terms. If  $J_l = J - 2$ , the probability to reach the low-spin isomer is proportional to

$$\frac{Z(J)}{1+Z(J)} \times \frac{Z(J-1)}{1+Z(J-1)}(1 + S2 + S4 + \dots), \quad (4)$$

etc. Similar relations are valid for the high-spin isomer but with  $Z/(1+Z)$  exchanged for  $1/(1+Z)$ .

Neglecting the small terms S2, S4,... we can calculate the fractional independent isomeric yields (*fiiy*) using the following assumptions:

If the low-spin isomer is the ground state all  $P(J)$  values for  $J$  smaller than or equal to  $J_l$  are assumed to feed the low-spin isomer. In the spin range  $J_l < J < J_h$  ( $J_h$  being the spin of the high-spin isomer), the probabilities are calculated using the arguments above. Since either of the isomers must be reached the sum of the probabilities to reach the low-spin isomer and the high-spin isomer is normalized to unity. For  $J \geq J_h$  the high-spin isomer is assumed to be fed unless the energy is smaller than  $E_x$  in which case the low-spin isomer is fed. The final formula will then be

$$\begin{aligned} fiiy(low) &= \sum_{J \leq J_l} P(J) + \frac{E_x}{E_{max}} \sum_{J \geq J_l+1} P(J) + \\ &\quad \frac{E_{max} - E_x}{E_{max}} \sum_{k=J_l+1}^{J_h-1} P(k)N(k) \prod_{m=J_l+1}^k \frac{Z(m)}{1+Z(m)}. \\ fiiy(high) &= \frac{E_{max} - E_x}{E_{max}} \left[ \sum_{J \geq J_h} P(J) + \sum_{k=J_l+1}^{J_h-1} P(k)N(k) \prod_{m=k}^{J_h-1} \frac{1}{1+Z(m)} \right]. \end{aligned} \quad (5)$$

$$N(k) = 1 / \left( \prod_{m=J_l+1}^k \frac{Z(m)}{1+Z(m)} + \prod_{m=k}^{J_h-1} \frac{1}{1+Z(m)} \right).$$

If the low-spin state is the isomeric state the following formulae are used:

$$\begin{aligned} fiiy(low) &= \frac{E_{max} - E_x}{E_{max}} \left[ \sum_{J \leq J_l} P(J) + \sum_{k=J_l+1}^{J_h-1} P(k)N(k) \prod_{m=J_l+1}^k \frac{Z(m)}{1+Z(m)} \right]. \\ fiiy(high) &= \sum_{J \geq J_h} P(J) + \frac{E_x}{E_{max}} \sum_{J \leq J_h-1} P(J) + \\ &\quad \frac{E_{max} - E_x}{E_{max}} \sum_{k=J_l+1}^{J_h-1} P(k)N(k) \prod_{m=k}^{J_h-1} \frac{1}{1+Z(m)} \end{aligned} \quad (6)$$

The factor  $N(k)$  is the same as above.

The formulae above contain only two parameters  $J_{rms}$  and  $J_{nuc}$ . These parameters have to be determined by a comparison with experimental results. It is possible to choose the parameters in such a way that the experimental yield split is reproduced for each isomeric pair. This is not an interesting solution, however, because it cannot be used to estimate the independent yield split for isomeric pairs where no experimental data exist. One should rather search for correlations between groups of nuclides. Thus, it is tempting to keep the parameter  $J_{rms}$  constant for a given fissile system and to try to find  $J_{nuc}$ -values representative for groups of nuclides, for instance one value for odd-mass nuclides and another for even-mass nuclides. If this can be done, estimates for unknown cases would be easy to carry out.

The experimental data at hand are mainly from thermal neutron induced fission of  $^{235}\text{U}$  and  $^{233}\text{U}$  and for fast fission of  $^{238}\text{U}$ . The same formulae should apply to other systems and with the same values of  $J_{nuc}$ . We do not know, however, how  $J_{rms}$  will depend on the fissile system. In order to establish that, more experimental results are needed.

The formulae can also be used for cases with three isomeric states by combining first those with lower spins and then those with higher spins. This determines yield values for the low-spin and high-spin isomers. That of the intermediate spin isomer is then obtained from the requirement that the sum of fractional yields should add up to unity.

#### 4.3.2.2. Alternative treatment of the energy effect

The model outlined in the preceding section takes the energy effect into account in a very crude way, simply by reducing the excitation energy range available to the high-lying isomer by the fraction  $\frac{E_{max}-E_x}{E_{max}}$ . It is possible to go one step further in this direction by taking into account that the excitation energy will be reduced by the energy  $\Delta E$  for each emitted gamma-ray, where  $\Delta E$  is some average gamma-ray energy. Then the gamma-ray emission will sooner or later lead to a remaining excitation energy which prohibits reaching the high-lying state. If the ground state has the lower spin, following formulae apply in this model:

$$f_{iiy}(low) = \sum_{J \leq J_l} P(J) + \frac{E_x}{E_{max}} \sum_{J \geq J_h} P(J) + \sum_{k=J_l+1}^{J_h-1} P(k) \left[ \frac{(J_h - k)\Delta E + E_x}{E_{max}} + N(k) \frac{E_{max} - (J_h - k)\Delta E - E_x}{E_{max}} \prod_{m=J_l+1}^k \frac{Z(m)}{1 + Z(m)} \right]; \quad (7)$$

$$f_{iiy}(high) = \frac{E_{max} - E_x}{E_{max}} \left[ \sum_{J \geq J_h} P(J) + \sum_{k=J_l+1}^{J_h-1} P(k) N(k) \frac{E_{max} - (J_h - k)\Delta E - E_x}{E_{max}} \times \prod_{m=k}^{J_h-1} \frac{1}{1 + Z(m)} \right].$$

If the low-spin state is the metastable state, the following formulae are to be used:

$$\begin{aligned}
fiiy(low) &= \frac{E_{max} - E_x}{E_{max}} \sum_{J \leq J_l} P(J) + \\
&\quad \sum_{k=J_l+1}^{J_h-1} P(k) N(k) \frac{E_{max} - (k - J_l) \Delta E - E_x}{E_{max}} \prod_{m=J_l+1}^k \frac{Z(m)}{1 + Z(m)}; \\
fiiy(high) &= \sum_{J \geq J_h} P(J) + \frac{E_x}{E_{max}} \sum_{J \leq J_h} P(J) + \\
&\quad \sum_{k=J_l+1}^{J_h-1} P(k) \left[ \frac{(k - J_l) \Delta E + E_x}{E_{max}} + \right. \\
&\quad \left. N(k) \frac{E_{max} - (k - J_l) \Delta E - E_x}{E_{max}} \prod_{m=k}^{J_h-1} \frac{1}{1 + Z(m)} \right].
\end{aligned} \tag{8}$$

The factor  $N(k)$  is the same as above.

#### 4.3.3. Experimental data basis

The experimental data basis for various fissile nuclides is tabulated at the end of the article. The nuclides for which information is available have been grouped according to their spins. In certain cases the *fiiy*-values are very uncertain, and only approximate values can be given. Only nuclides with well determined fractional independent isomeric yields are used for evaluating the parameters  $J_{rms}$  and  $J_{nuc}$ . This is done only for thermal neutron fission of  $^{235}\text{U}$  where many data have been collected.

Yields from thermal neutron fission of  $^{233}\text{U}$  and  $^{235}\text{U}$  can be compared in the tables as well as yields of thermal neutron fission of  $^{235}\text{U}$  and fast neutron fission of  $^{238}\text{U}$ . The tables show that the product for  $^{235}\text{U}$  can be grouped into two groups: odd-mass nuclides and even-mass nuclides. The former have larger yields of the high-spin isomer than the latter. If the  $J_{rms}$ -values are the same, this might depend on different  $J_{nuc}$ -values for the two groups. For  $^{233}\text{U}$  the situation seems to be different. The *fiiy*-values of the high-spin isomers are generally lower. There the  $J_{nuc}$ -values must be the same, and the reason could be the different  $J_{rms}$  for the two fissile systems. For  $^{238}\text{U}$  the experimental data are more scarce, but a similarity of the yield ratios with those of  $^{235}\text{U}$  are evident.

#### 4.3.4. Choice of $J_{rms}$ and $J_{nuc}$

As noted above the odd-mass nuclides and even-mass nuclides behave differently for thermal fission of  $^{235}\text{U}$ . In the former group the high-spin isomer is more favoured than in the latter group. Therefore, the nuclides were grouped into odd-mass and even-mass nuclides, and these groups were treated separately. When trying to find the best values of  $J_{rms}$  and  $J_{nuc}$ , only those nuclides with the best data in Tables 1–13 were used. For odd-mass nuclides  $J_{rms}$  was varied between 6.00 and 8.50, in each case combined with  $J_{nuc}$  from 4.00 to 6.50. For even-mass nuclides the corresponding ranges were 5.00 to 7.50 and 1.00 to 3.50, respectively. The cases with the smallest sum of chi-squared are tabulated in Table 17. Table 17 shows the best combination of  $J_{rms}$  and  $J_{nuc}$  to be 6.50 and 6.00 for odd-mass nuclides and 6.00 and 1.00–2.00 for even-mass nuclides. It is interesting to note that the optimal  $J_{rms}$ -value is very similar for the

two groups of nuclides, whereas the  $J_{nuc}$ -value is very different. It is therefore tempting to try to use an average of the  $J_{rms}$ -values whilst keeping the optimal  $J_{nuc}$ -values. This has been done using for  $J_{rms}$  the average of 6.25 and for  $J_{nuc}$  the value 6.00 for odd-mass nuclides and 2.00 for even-mass ones. All the isomeric pairs from Tables 1–13 are included. Different measurements of the same isomeric pair are shown in Table 18. In some cases the values are very approximate with no known error limit. Then an error of  $\pm 0.50$  is adopted in the figure to illustrate the uncertainty.

In general the agreement of experimental values and model calculations is quite satisfactory, but there are cases where the discrepancy is large. To this group belong  $^{82}\text{As}$ ,  $^{99}\text{Nb}$ ,  $^{146}\text{La}$ , and  $^{148}\text{Pr}$  where the experimental values are lower than the calculated ones and  $^{90}\text{Rb}$  (one of the determinations),  $^{119}\text{Cd}$ ,  $^{128}\text{Sb}$ , and  $^{148}\text{Pm}$  where they are higher. The reason for the deviations can be unusual  $J_{nuc}$ -values or simply erroneous spin- or yield-values. Nevertheless, it seems possible to use the model outlined in the present work to estimate isomeric yields for unmeasured nuclides.

In this connection it may be interesting to note that [4.3.1] use the  $J_{rms}$ -value 7.5 for thermal neutron-induced fission of  $^{235}\text{U}$  which is somewhat higher than the value 6.25 used here. Their model has been applied to the selected nuclides used in the present work and with  $J_{rms} = 7.5$ . For odd-mass nuclides the model works well except that yields of  $^{133}\text{I}$  cannot be reproduced at all, which is mainly responsible for the high value of  $\chi^2$  of 281. For the even-mass nuclides  $\chi^2 = 995$ , mainly caused by large contributions from  $^{128}\text{Sn}$ ,  $^{130}\text{Sn}$ ,  $^{132}\text{Sb}$ , and  $^{134}\text{Sb}$ , whose low-spin yields are greatly overestimated. It must be remembered, though, that Madland and England only use one parameter in their formula compared to two in the present work.

Note also the formula for the spin distribution  $P(J) = \text{const} \times (2J+1)e^{-(J+1/2)^2/2\sigma^2}$ , with  $\sigma = (0.98 \pm 0.23)$ , derived by von Egidy, Schmidt and Rehkami [4.3.4]. This would correspond to  $J_{nuc} = 5.1_{2.1}^{3.2}$  for  $A = 90$  and  $5.7_{2.4}^{3.7}$  for  $A = 130$  in good agreement to our value 6.0 for odd-mass nuclides and somewhat higher than the value 2.00 used for even-mass nuclides.

Results for nuclides with two isomeric states in addition to the ground state are shown in Table 19. For the even-mass indium isotopes the high-spin isomer is underestimated and the low-spin isomer overestimated by the model whereas the yield of the intermediate-spin isomer is well reproduced. For  $^{131}\text{In}$  the model predicts a low *fiy*-value for the high-spin isomer but not as low as found experimentally. Also the yields of the other two isomers are badly reproduced.

#### 4.3.5. Choice of $J_{rms}$ and $J_{nuc}$ for the alternative model

The best values of  $J_{rms}$  are 7.00 for odd-mass nuclides and 6.00 for even-mass ones. We choose the average value 6.50 which should be combined with the  $J_{nuc}$ -value 6.50 for the former group of nuclides and 1.00–2.00 (2.00 is chosen) for the latter. The sum of  $\chi$ -squares is hardly improved as compared to the first model, and it is therefore questionable whether it is worth while to use this second model containing one additional parameter.



### 4.3.6. Summary

The aim of the present work was to find a method to describe the partition of independent nuclidic yields on isomeric states, with the practical application to estimate isomeric yields in mind. A semi-empirical formula containing two free parameters has been developed, one ( $J_{rms}$ ) describing the angular momenta of the fission fragments after neutron evaporation, and the other one ( $J_{nuc}$ ) for the spin distribution of the nuclear levels. For the latter parameter different values have to be used for odd-mass nuclides and even-mass nuclides. With this formula the fractional independent isomeric yields are quite well reproduced for the bulk of isomeric pairs. The formula can therefore be used with some confidence in spite of the fact that there are a few cases for which the agreement between experimental data and model predictions is unsatisfactory.

All results presented here are for thermal neutron-induced fission of  $^{235}\text{U}$ . It should be possible to use the model also for other fission systems and with the same  $J_{nuc}$ -values. Whether the same  $J_{rms}$ -value can be used remains to be checked. So far, there are not sufficient isomeric yields known to enable extensive tests except possibly for thermal neutron fission of  $^{233}\text{U}$  and fast neutron fission of  $^{238}\text{U}$ .

There are now extensive experimental data available for a more ambitious approach, perhaps along the lines of Ford, Wolfsberg, and Erdal [4.3.5]. Then, the experimental yield data combined with level density data for the various nuclides would be used to get a deeper insight into the fission process itself, for instance, evaluating the angular momentum distribution of those fragments which lead to a particular isomeric pair.

### 4.3.7. Tables

#### 4.3.7.1. Odd-mass nuclides

TABLE 1. SPINS  $7/2 - 1/2$

Spin	$^{79}\text{Ge(a)}$	$^{79}\text{Ge(b)}$	Experimental $fiiy$ , %			
			$^{113}\text{Ag(c)}$	$^{115}\text{Ag(c)}$	$^{117}\text{Ag(a)}$	$^{117}\text{Ag(c)}$
$\frac{7}{2}$	$44 \pm 5$	$100 \pm 11$	$\sim 100$	$\sim 100$	$38 \pm 6$	$\sim 100$
$\frac{1}{2}$	$56 \pm 5$	$0 \pm 11$	$\sim 0$	$\sim 0$	$62 \pm 6$	$\sim 0$

- (a) [4.3.6].
- (b) [4.3.3] with isotopic yield from [4.3.7] used to estimate the ground state yield
- (c) Independent isomeric yields from [4.3.3]. The ground-state yields were not measured. They are assumed to be small.

TABLE 2. SPINS  $9/2 - 1/2$

Spin	$^{81}\text{Ge(a)}$	$^{81}\text{Ge(b)}$	$^{81}\text{Ge(c)}$	Experimental $fiiy$ , %				
				$^{83}\text{Se(b)}$	$^{99}\text{Nb(d)}$	$^{123}\text{In(a)}$	$^{123}\text{In(b)}$	$^{125}\text{In(a)}$
$\frac{9}{2}$	$28 \pm 6$	$70 \pm 6$	$54 \pm 8$	$89 \pm 7$	$6 \pm 14$	$17 \pm 4$	$94 \pm 11$	$14 \pm 3$
$\frac{1}{2}$	$72 \pm 6$	$30 \pm 6$	$46 \pm 8$	$11 \pm 7$	$94 \pm 14$	$83 \pm 4$	$6 \pm 11$	$86 \pm 3$

TABLE 2. (cont.)

Spin	Experimental $fiiy$ , %						
	$^{125}\text{In(e)}$	$^{127}\text{In(a)}$	$^{127}\text{In(b)}$	$^{127}\text{In(c)}$	$^{129}\text{In(a)}$	$^{129}\text{In(b)}$	$^{129}\text{In(c)}$
$\frac{9}{2}$	$88 \pm 8$	$18 \pm 1$	$87 \pm 6$	$67 \pm 9$	$41 \pm 7$	$76 \pm 7$	$60 \pm 8$
$\frac{1}{2}$	$12 \pm 8$	$82 \pm 1$	$13 \pm 6$	$33 \pm 9$	$59 \pm 7$	$24 \pm 7$	$40 \pm 8$

- (a) [4.3.6].
- (b) [4.3.3].
- (c) [4.3.8].
- (d) [4.3.9].
- (e) [4.3.3] with independent isomeric yield obtained by subtracting the parent contribution from the cumulative isomeric yield with branching from [4.3.10].

TABLE 3. SPINS 11/2 – 1/2

Spin	Experimental $fiiy$ , %		
	$^{119}Cd(a)$	$^{121}Cd(b)$	$^{121}Cd(c)$
$\frac{11}{2}$	$39 \pm 20$	$.72 \pm .79$	$89 \pm 11$
$\frac{1}{2}$	$61 \pm 20$	$99.3 \pm .8$	$11 \pm 11$

- (a) [4.3.3] with independent isomeric yield obtained by subtracting the parent contribution from the cumulative isomeric yield assuming 22% branching.  
(b) [4.3.6].  
(c) [4.3.3].

Table 4. Spins 11/2 – 3/2

Spin	Experimental $fiiy$ , %								
	$^{123}Cd(a)$	$^{123}Cd(b)$	$^{123}Cd(c)$	$^{125}Cd(a)$	$^{125}Cd(b)$	$^{125}Cd(c)$	$^{123}In(b)$	$^{127}Sn(a)$	$^{127}Sn(e)$
$\frac{11}{2}$	$77 \pm 18$	$68 \pm 2$	$70 \pm 12$	$65 \pm 10$	$66 \pm 8$	$91 \pm 9$	$100 \pm 28$	$13 \pm 5$	$90 \pm 11$
$\frac{3}{2}$	$23 \pm 18$	$32 \pm 2$	$30 \pm 12$	$35 \pm 10$	$34 \pm 8$	$9 \pm 9$	$0 \pm 28$	$87 \pm 5$	$10 \pm 11$

TABLE 4. (cont.)

Spin	Experimental $fiiy$ , %									
	$^{129}Sn(a)$	$^{129}Sn(b)$	$^{129}Sn(c)$	$^{133}Te(a)$	$^{133}Te(b)$	$^{133}Te(f)$	$^{133}Te(g)$	$^{133}Te(c)$	$^{133}Xe(h)$	$^{135}Xe(h)$
$\frac{11}{2}$	$24 \pm 6$	$43 \pm 6$	$57 \pm 6$	$72 \pm 5$	$78 \pm 4$	$57 \pm 4$	$61 \pm 8$	$87 \pm 5$	$75 \pm 4$	$65 \pm 4$
$\frac{3}{2}$	$76 \pm 6$	$57 \pm 6$	$43 \pm 6$	$28 \pm 5$	$22 \pm 4$	$43 \pm 4$	$39 \pm 8$	$13 \pm 5$	$25 \pm 4$	$35 \pm 4$

- (a) [4.3.6].  
(b) [4.3.3].  
(c) [4.3.8].  
(d) [4.3.3] with independent yields obtained by subtracting parent contributions from cumulative yields using branchings from [4.3.11]. The cumulative yield of the ground state is taken from [4.3.12].  
(e) [4.3.3] with independent yields obtained by subtracting parent contributions from cumulative yields using branchings from [4.3.13].  
(f) [4.3.14].  
(g) [4.3.15].  
(h) [4.3.5].

TABLE 5. SPINS 19/2 – 7/2

Spin	Exp. $fiiy$ , % $^{133}I(a)$
$\frac{19}{2}$	$7 \pm 2$
$\frac{7}{2}$	$93 \pm 2$

- (a) [4.3.3].  
(b) [4.3.6].

#### 4.3.7.2. Even-mass nuclides

TABLE 6. SPINS 4 – 1

Spin	Experimental $fiiy$ , %				
	$^{90}Rb(a)$	$^{90}Rb(b)$	$^{90}Rb(c)$	$^{90}Rb(d)$	$^{148}Pr(e)$
4	$77 \pm 12$	$58 \pm 5$	$90 \pm 1$	$38 \pm 5$	$12 \pm 3$
1	$23 \pm 12$	$42 \pm 5$	$10 \pm 1$	$62 \pm 5$	$88 \pm 3$

- (a) [4.3.6].  
(b) [4.3.3].  
(c) [4.3.16]. The difference between (b) and (c) is large.  
(d) [4.3.8].  
(e) [4.3.17].

TABLE 7. SPINS 5 – 2

Spin	Experimental $fiiy$ , %					
	$^{82}As(a)$	$^{82}As(b)$	$^{82}As(c)$	$^{84}Br(a)$	$^{84}Br(d)$	$^{84}Br(e)$
5	$38 \pm 7$	$17 \pm 7$	$8 \pm 4$	$11 \pm 2$	$30 \pm 4$	$38 \pm 4$
2	$62 \pm 7$	$83 \pm 7$	$92 \pm 4$	$89 \pm 2$	$70 \pm 4$	$62 \pm 4$

- (a) [4.3.6].  
(b) [4.3.3].  
(c) [4.3.8].  
(d) [4.3.3] with isotopic yield  $0.057 \pm 0.007$  % (average of values in [4.3.7], [4.3.18], and [4.3.19]).  
(e) [4.3.20].

TABLE 8. SPINS 6 – 1

Spin	Experimental $fiiy$ , %	
	$^{116}Ag(a)$	$^{148}Pm(b)$
6	$\sim 100$	$71 \pm 6$
1	$\sim 0$	$29 \pm 6$

- (a) [4.3.3]. No value is available for the ground-state yield. It is probably small.  
(b) [4.3.21].

TABLE 9. SPINS 6 – 2

Spin	Experimental $fiiy$ , %		
	$^{136}I(a)$	$^{136}I(b)$	$^{146}La(c)$
6	$76 \pm 3$	$79 \pm 14$	5.5
2	$24 \pm 3$	$21 \pm 14$	94.5

- (a) [4.3.6].  
(b) [4.3.3].  
(c) [4.3.20].

TABLE 10. SPINS 6 – 3

Spin	Experimental $fiiy$ , %							
	$^{118}Ag(a)$	$^{118}Ag(b)$	$^{120}Ag(a)$	$^{120}Ag(b)$	$^{120}Ag(d)$	$^{138}Cs(a)$	$^{138}Cs(c)$	$^{138}Cs(d)$
6	$68 \pm 4$	$44 \pm 10$	$65 \pm 19$	$85 \pm 15$	$86 \pm 4$	$48 \pm 8$	$58 \pm 8$	$19 \pm 3$
3	$32 \pm 4$	$56 \pm 10$	$35 \pm 19$	$15 \pm 15$	$14 \pm 4$	$52 \pm 8$	$42 \pm 8$	$81 \pm 3$

- (a) [4.3.6].  
(b) [4.3.3].  
(c) Isomeric yield of ground-state obtained by subtracting isomeric yield ([4.3.3]) from isotopic yield ([4.3.22]).  
(d) [4.3.8].

TABLE 11. SPINS 7 – 0

Spin	Experimental $fiiy$ , %					
	$^{126}Sn(a)$	$^{130}Sn(b)$	$^{130}Sn(c)$	$^{130}Sn(d)$	$^{134}Sb(b)$	$^{134}Sb(e)$
7	$11 \pm 7$	$19 \pm 6$	$13 \pm 2$	$14 \pm 2$	$71 \pm 16$	$19 \pm 5$
0	$89 \pm 7$	$81 \pm 6$	$87 \pm 2$	$86 \pm 2$	$29 \pm 16$	$81 \pm 5$

- (a) [4.3.3]. The yield of the ground state is obtained by subtracting the contribution from the low-spin parent. That of the isomeric state is then obtained by subtracting the ground-state independent yield from the independent isotopic yield from [4.3.23].  
(b) [4.3.6].  
(c) [4.3.3].  
(d) [4.3.8].  
(e) [4.3.3] (high-spin yield) and [4.3.24] (low-spin yield).

TABLE 12. SPINS 8 – 3

Spin	Experimental $fiiy$ , %						
	$^{124}In(a)$	$^{124}In(b)$	$^{126}In(a)$	$^{126}In(c)$	$^{128}In(a)$	$^{128}In(b)$	$^{128}In(d)$
8	$96 \pm 4$	$86 \pm 80$	$27 \pm 7$	$42 \pm 17$	$25 \pm 7$	$30 \pm 7$	$36 \pm 7$
0	$4 \pm 4$	$14 \pm 80$	$73 \pm 7$	$58 \pm 17$	$75 \pm 7$	$70 \pm 7$	$64 \pm 7$

- (a) [4.3.6].  
(b) [4.3.3].  
(c) [4.3.3]. Low-spin yield obtained by subtracting the contribution from the parent from the cumulative ground-state yield.  
(d) [4.3.8].

TABLE 13. SPINS 8 – 4

Spin	Experimental $fiiy$ , %								
	$^{128}Sb(a)$	$^{130}Sb(b)$	$^{130}Sb(a)$	$^{132}Sb(b)$	$^{132}Sb(c)$	$^{132}Sb(d)$	$^{134}I(b)$	$^{134}I(e)$	$^{134}I(f)$
8	$61 \pm 6$	$80 \pm 34$	$45 \pm 11$	$54 \pm 8$	$20 \pm 2$	$19 \pm 3$	$21 \pm 6$	$20 \pm 2$	$10 \pm 3$
4	$39 \pm 6$	$20 \pm 34$	$55 \pm 11$	$46 \pm 8$	$80 \pm 2$	$81 \pm 3$	$79 \pm 6$	$80 \pm 2$	$90 \pm 3$

- (a) [4.3.3]. Independent isomeric yield obtained by subtracting the parent contribution.  
(b) [4.3.6].  
(c) [4.3.3].  
(d) [4.3.14].  
(e) Independent yield of ground-state taken from [4.3.3] and that of the ground state from [4.3.25].  
(f) [4.3.8].

#### 4.3.7.3. Nuclides with two isomeric states in addition to the ground states

TABLE 14. SPINS 8 – 5 –1

Spin	Experimental $fiiy$ , %		
	$^{120}In(a)$	$^{122}In(b)$	$^{122}In(a)$
8	$40 \pm 15$	$35 \pm 16$	$48 \pm 20$
5	$31 \pm 13$	$14 \pm 3$	$24 \pm 10$
1	$29 \pm 24$	$51 \pm 7$	$28 \pm 29$

- (a) [4.3.3].  
(b) [4.3.6].

TABLE 15. SPINS 10 – 5 –2

Spin	Experimental $fiiy$ , %		
	$^{130}In(a)$	$^{130}In(b)$	$^{130}In(c)$
10	$18 \pm 6$	$24 \pm 4$	$25 \pm 5$
5	$43 \pm 42$	$44 \pm 5$	$41 \pm 7$
2	$39 \pm 10$	$32 \pm 4$	$34 \pm 7$

- (a) [4.3.6].  
(b) [4.3.3].  
(c) [4.3.8].

TABLE 16. SPINS  $21/2 - 9/2 - 1/2$ 

Spin	Experimental $fiiy$ , %	
	$^{131}In(a)$	$^{131}In(b)$
$\frac{21}{2}$	$0.40 \pm 0.17$	$3.6 \pm 1.3$
$\frac{9}{2}$	$16 \pm 7$	$21 \pm 6$
$\frac{1}{2}$	$83 \pm 7$	$75 \pm 7$

- (a) [4.3.3].  
(b) [4.3.8].

#### 4.3.7.4. Other Tables

TABLE 17.  $\chi^2$  VERSUS  $J_{rms}$  AND  $J_{nuc}$

$J_{rms}$	$J_{nuc}$	$\chi^2$ Odd-mass nuclides	$J_{nuc}$	Even-mass nuclides
5.50			1.00	114
5.50			1.50	114
5.50			2.00	114
6.00	5.50	90	1.00	93
6.00	6.00	81	1.50	93
6.00	6.50	76	2.00	94
6.50	5.50	66	1.00	116
6.50	6.00	66	1.50	116
6.50	6.50	70	2.00	118
7.00	5.50	69		
7.00	6.00	78		
7.00	6.50	88		

TABLE 18. EXPERIMENTAL AND CALCULATED FRACTIONAL INDEPENDENT YIELD OF THE HIGH-SPIN ISOMER OF NUCLIDES FORMED IN THERMAL-NEUTRON INDUCED FISSION OF  $^{235}\text{U}$

Nr	Nuc- lide	$f_{iiy}(high)$ experimental	calculated	Nr	Nuc- lide	$f_{iiy}(high)$ experimental	calculated
1	$^{79}\text{Ge}$	$1.00 \pm 0.11$	0.829	25	$^{127}\text{In}$	$0.87 \pm 0.06$	0.814
2	$^{81}\text{Ge}$	$0.70 \pm 0.06$	0.835	26	$^{127}\text{Sn}$	$0.90 \pm 0.11$	0.626
3	$^{82}\text{As}$	$0.17 \pm 0.07$	0.597	27	$^{128}\text{In}$	$0.30 \pm 0.07$	0.190
4	$^{83}\text{Se}$	$0.89 \pm 0.07$	0.816	28	$^{128}\text{Sn}$	$0.11 \pm 0.07$	0.209
5	$^{84}\text{Br}$	$0.30 \pm 0.04$	0.504	29	$^{128}\text{Sb}$	$0.61 \pm 0.06$	0.192
6	$^{84}\text{Br}$	$0.38 \pm 0.04$	0.504	30	$^{129}\text{In}$	$0.76 \pm 0.07$	0.815
7	$^{90}\text{Rb}$	$0.58 \pm 0.05$	0.659	31	$^{129}\text{Sn}$	$0.43 \pm 0.06$	0.621
8	$^{90}\text{Rb}$	$0.90 \pm 0.01$	0.659	32	$^{130}\text{Sn}$	$0.13 \pm 0.02$	0.212
9	$^{99}\text{Nb}$	$0.06 \pm 0.14$	0.819	33	$^{130}\text{Sb}$	$0.45 \pm 0.11$	0.190
10	$^{113}\text{Ag}$	$\sim 1.00$	0.847	34	$^{132}\text{Sb}$	$0.20 \pm 0.02$	0.190
11	$^{115}\text{Ag}$	$\sim 1.00$	0.846	35	$^{132}\text{Sb}$	$0.19 \pm 0.03$	0.190
12	$^{116}\text{Ag}$	$\sim 1.00$	0.391	36	$^{133}\text{Te}$	$0.78 \pm 0.04$	0.590
13	$^{117}\text{Ag}$	$\sim 1.00$	0.848	37	$^{133}\text{Te}$	$0.57 \pm 0.04$	0.590
14	$^{118}\text{Ag}$	$0.44 \pm 0.10$	0.388	38	$^{133}\text{Te}$	$0.61 \pm 0.08$	0.590
15	$^{119}\text{Cd}$	$0.39 \pm 0.20$	0.724	39	$^{133}\text{I}$	$0.07 \pm 0.02$	0.140
16	$^{120}\text{Ag}$	$0.85 \pm 0.15$	0.382	40	$^{133}\text{Xe}$	$0.75 \pm 0.04$	0.603
17	$^{121}\text{Cd}$	$0.89 \pm 0.11$	0.730	41	$^{134}\text{Sb}$	$0.19 \pm 0.05$	0.279
18	$^{123}\text{Cd}$	$0.68 \pm 0.02$	0.614	42	$^{134}\text{I}$	$0.20 \pm 0.02$	0.183
19	$^{123}\text{In}$	$0.94 \pm 0.11$	0.817	43	$^{135}\text{Xe}$	$0.65 \pm 0.04$	0.575
20	$^{123}\text{Sn}$	$1.00 \pm 0.28$	0.627	44	$^{136}\text{I}$	$0.79 \pm 0.14$	0.391
21	$^{124}\text{In}$	$0.86 \pm 0.80$	0.215	45	$^{138}\text{Cs}$	$0.58 \pm 0.08$	0.392
22	$^{125}\text{Cd}$	$0.66 \pm 0.08$	0.614	46	$^{146}\text{La}$	$0.06 \pm 0.03$	0.391
23	$^{125}\text{In}$	$0.88 \pm 0.08$	0.813	47	$^{148}\text{Pr}$	$0.12 \pm 0.03$	0.660
24	$^{126}\text{In}$	$0.42 \pm 0.17$	0.215	48	$^{148}\text{Pm}$	$0.71 \pm 0.06$	0.388

TABLE 19. FRACTIONAL INDEPENDENT ISOMERIC YIELDS FOR NUCLIDES WITH TWO ISOMERS IN ADDITION TO THE GROUND STATE (thermal fission of  $^{235}\text{U}$ )

Nuclide	experimental	$f_{iiy}(low)$ calculated	experimental	$f_{iiy}(intermediate)$ calculated	experimental	$f_{iiy}(high)$ calculated
$^{120}\text{In}$	$0.29 \pm 0.24$	0.483	$0.31 \pm 0.13$	0.327	$0.40 \pm 0.15$	0.190
$^{122}\text{In}$	$0.28 \pm 0.29$	0.483	$0.24 \pm 0.10$	0.327	$0.48 \pm 0.20$	0.190
$^{130}\text{In}$	$0.32 \pm 0.04$	0.485	$0.43 \pm 0.05$	0.440	$0.24 \pm 0.04$	0.075
$^{131}\text{In}$	$0.83 \pm 0.07$	0.197	$0.16 \pm 0.07$	0.761	$0.0040 \pm 0.0017$	0.042

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## 4.4. STUDIES OF NEUTRON INDUCED TERNARY FISSION PRODUCT YIELDS

### 4.4.1. Introduction

During fission there is a probability of producing a light charged particle, for example an alpha particle or a triton, as well as the two main fragments and neutrons. This emission occurs in less than one in a hundred fissions. The process is sometimes called ternary fission as a third fragment is produced.

Since the discovery, in 1946, of the emission of light charged particles during fission this process has been extensively studied and reviewed [4.4.1, 4.4.2]. These reviews have probed the fission process and extended the understanding of fission. The experimental evidence suggests that the light charged particles are formed between the two main fragments very close in time to the scission of the compound nucleus and before the fragments have been separated significantly by Coulombic repulsion. Thus, the alpha particles are repelled by the fragments away from the axis upon which the fission products are being accelerated.

Most experimental studies of ternary fission are concerned with determining properties of the compound nucleus at scission by studying the light charged particle. These measurements, however, cannot usually be used to determine yields. On the other hand the yields of these ternary particles are important in reactors, giving rise mainly to hydrogen and helium isotopes. The tritium,  $^3\text{H}$ , produced from fission is a  $\beta^-$  emitter with a half-life of  $12.33 \pm 0.02$  years [4.4.3] and which can be easily absorbed into living tissue. The tritium fission yield is thus important for calculations concerned with handling and reprocessing of irradiated fuels, and for modeling of accident scenarios. These requirements have led to many measurements of such yields, primarily of  $^4\text{He}$  and  $^3\text{H}$ . The  $^4\text{He}$  is not, of itself, significant radiologically but is important in terms of materials properties of nuclear fuels and is used as a standard relative to which the tritium yield can be measured.

The yield measurements can be of two types: radiochemical measurements of a sample after irradiation or measurements of the number and parameters (energy, nuclear charge or mass) of the long-ranged charged particles from fission. It should be noted that the second type of measurement can be achieved by several techniques. One of these is to measure the particle kinetic energy since this is related to the fragments mass and nuclear charge. The kinetic energy being imparted by Coulombic repulsion from the fissioning compound nucleus. Unfortunately, the energy distributions of the different particles overlap considerably. Thus the alpha peak which is considerably larger has to be modeled and then subtracted to estimate the other particle yields. However, work by D'Hondt et al. [4.4.4] and Cañtucoli et al. [4.4.5] showed that the low energy tail of the alpha particle spectrum deviated significantly from the Gaussian distribution which had been assumed frequently in the past. This means that measurements of low probability yields by this method have an implicit systematic error; this affects many reported measurements of tritium yields.

An improved technique uses  $\Delta E$ , E detectors, one detector being very thin and absorbing only part of the particle's energy and the other absorbing the remaining energy. From the energy loss in the thin detector and the total energy the charge of the particle can be determined. This technique usually plots the energy loss against total energy; regions on this plot can then be defined where the particle's charge and mass are uniquely defined. Thus the

yield for a given mass and charge can be determined uniquely. This technique can accurately measure yields of tritium and other nuclides, usually relative to the alpha yield.

#### 4.4.2. Evaluations

An evaluation consists of collection and analysis of experimental measurements to produce a recommended set of values from both the analysis and, where no measurements exist, by models. Evaluations of ternary yields have been produced in the work of Madland and Stewart [4.4.6] and more recently in UK evaluations [4.4.7–4.4.10]. All uncertainties quoted below and error bars shown in figures are one standard deviation.

The results for alpha yields are shown in Table 4.4.1 for the four main thermal neutron reactor fissile fuel nuclides. The alpha yields are measured by identification of particles resulting directly from fission.

TABLE 4.4.1. THE THERMAL NEUTRON FISSION YIELDS OF HELIUM-4 FROM RECENT EVALUATIONS

Nuclide	UKFY3 [4.4.10] (1993)	UKFY2 [4.4.7] (1990)	UKFY1 (1986)	ENDF-247 [4.4.6] (1977)
<sup>233</sup> U	0.2065 ± 0.0085 {9}	0.2397 ± 0.0034 {7}	0.2072	0.2268 ± 0.0088
<sup>235</sup> U	0.1699 ± 0.0061 {12}	0.1882 ± 0.0030 {13}	0.1680	0.1950 ± 0.0179
<sup>239</sup> Pu	0.2080 ± 0.0069 {8}	0.2232 ± 0.0025 {8}	0.2148	0.2326* ± 0.0107
<sup>241</sup> Pu	0.2015 ± 0.0202 {2}	0.1938 ± 0.0213 {2}	0.1860	2.273* ± 0.1454

The number of experimental measurements analysed in these works are given in { }.

\* Estimate of total light charged particle emission, not just alpha particle yield.

TABLE 4.4.2. THE THERMAL NEUTRON FISSION YIELDS OF TRITIUM FROM RECENT EVALUATIONS

Nuclide	UKFY3 [4.4.10] (1993)	UKFY2 [4.4.7] (1990)	UKFY1 (1986)	ENDF-247 [4.4.6] (1977)
<sup>233</sup> U	0.009691 ± 0.002354 {4}	0.01006 ± 0.00069 {3}	0.01030	0.01043 ± 0.00209
<sup>235</sup> U	0.009314 ± 0.000354 {15}	0.01004 ± 0.00020 {14}	0.01084	0.01219 ± 0.00305
<sup>239</sup> Pu	0.01442 ± 0.00076 {5}	0.01471 ± 0.00032 {5}	0.01479	no value reported
<sup>241</sup> Pu	0.01410 ± 0.00141 {1}	0.01410 ± 0.00061 {1}	0.01410	no value reported

The number of experimental measurements analysed in these works are given in { }.

The tritium fission yields for the same set of fissioning systems as in Table 4.4.1 are shown in Table 4.4.2. It should be noted that tritium yields can be measured radiochemically from the tritium gas produced in an irradiated sample or by the identification of particles resulting directly from fission.

In order to estimate yields for fissioning systems where experimental results are absent it is necessary to understand how yields vary between different fissioning systems and incident neutron energies.



#### 4.4.3. Effects of incident neutron energy on LCP yields

The results of analysis of experimental yield data from the UKFY3 data-set for those three systems with the most information on the variation of yield with neutron energy are shown in Tables 4.4.3 and 4.4.4 for alpha and tritium yields respectively.

TABLE 4.4.3.  $^4\text{He}$  FISSION PRODUCT YIELDS FROM UKFY3

Nuclide	Thermal Spectra	Fast Spectra (mean energy $\sim 0.6$ MeV)	High (14 MeV)
$^{233}\text{U}$	$0.2065 \pm 0.0085$ {9}	$0.2003 \pm 0.0216$ {1}	$0.1980 \pm 0.0210$ {1}
$^{235}\text{U}$	$0.1699 \pm 0.0061$ {12}	$0.1980 \pm 0.0170$ {4}	$0.1667 \pm 0.0088$ {3}
$^{239}\text{Pu}$	$0.2080 \pm 0.0069$ {8}	$0.2029 \pm 0.0177$ {2}	no measured data

The number of experimental measurements analysed in these works are given in { }.

TABLE 4.4.4.  $^3\text{H}$  FISSION PRODUCT YIELDS FROM UKFY3

Nuclide	Thermal Spectra	Fast Spectra (mean energy $\sim 0.6$ MeV)	High (14 MeV)
$^{233}\text{U}$	$0.009691 \pm 0.001386$ {4}	no measured data	$0.02480 \pm 0.00560$ {1}
$^{235}\text{U}$	$0.009314 \pm 0.000354$ {15}	$0.01352 \pm 0.00158$ {4}	$0.01742 \pm 0.00361$ {1}
$^{239}\text{Pu}$	$0.01442 \pm 0.00076$ {5}	$0.01413 \pm 0.00225$ {1}	no measured data

The number of experimental measurements analysed in these works are given in { }.

The results for alpha yields suggest that there is only significant variation, relative to the experimental uncertainties, between the yields for  $^{235}\text{U}$  at thermal and fast energies. The other alpha yields appear energy independent. The  $^{235}\text{U}$  alpha yield data for fast neutron induced fission contain some discrepant data, and until these discrepancies are resolved by new measurements, it is not possible to justify an energy dependence of this alpha yield.

Similarly for tritium, only  $^{235}\text{U}$  shows a trend with energy. However, the fast and 14 MeV yield values derive from single measurements with significant uncertainties and thus it is difficult to justify an energy dependence without further measurements.

It is thus useful to consider experiments which measure the yields of ternary particles at mono-energetic neutron energies relative to the yield at thermal energies. These results were extracted from the UKFY3 experimental yield database. Figure 4.4.1 shows measurements of the alpha yield for  $^{235}\text{U}$  and  $^{239}\text{Pu}$  against energy relative to the respective thermal yield. Figure 4.4.2 shows the measurements of the tritium yields for these fission systems again relative to the thermal yield.

Examining Figure 4.4.1 for  $^{235}\text{U}$  between 0 and 10.5 MeV shows there is no significant differences from the thermal value. Only two of these measurement show significant variation from the thermal value, but these measurements claim much smaller experimental uncertainties than similar measurements. Without further lower uncertainty measurements it is difficult to exclude an assumption that the data does not vary with energy. It should also be noted that between 4 and 10 MeV there is no experimental data. Thus more measurements are needed to demonstrate the yield variation, if any, in this region.

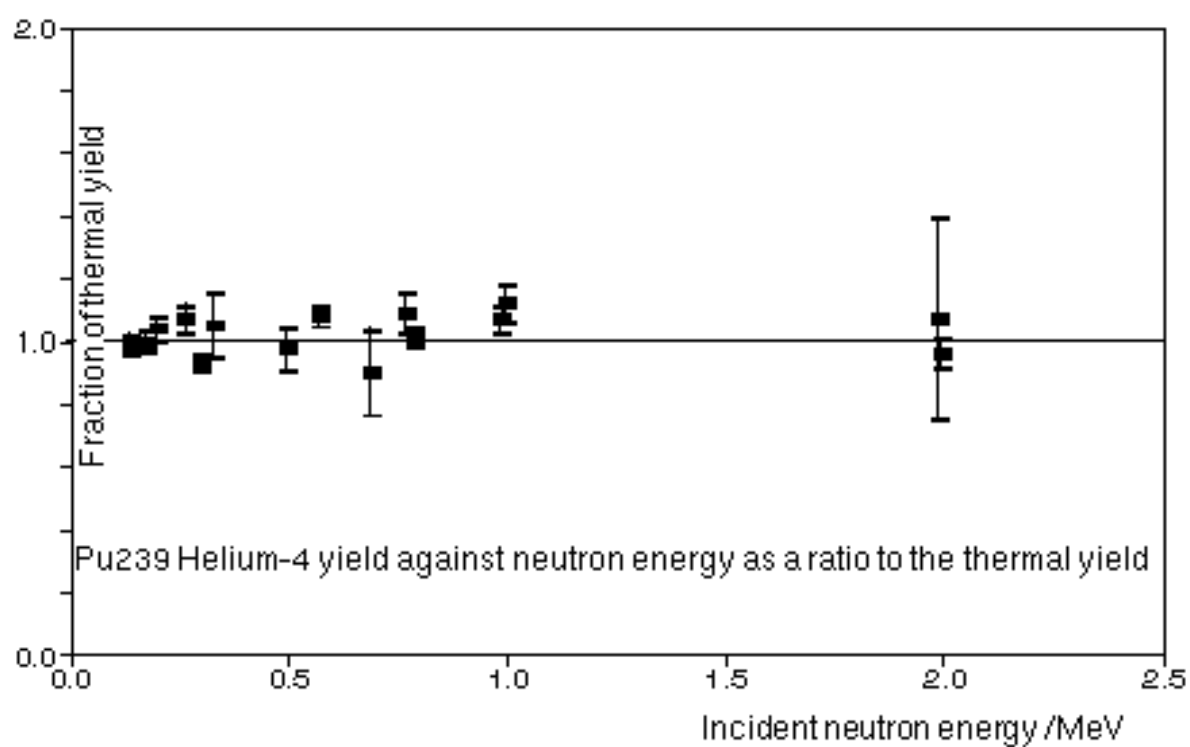
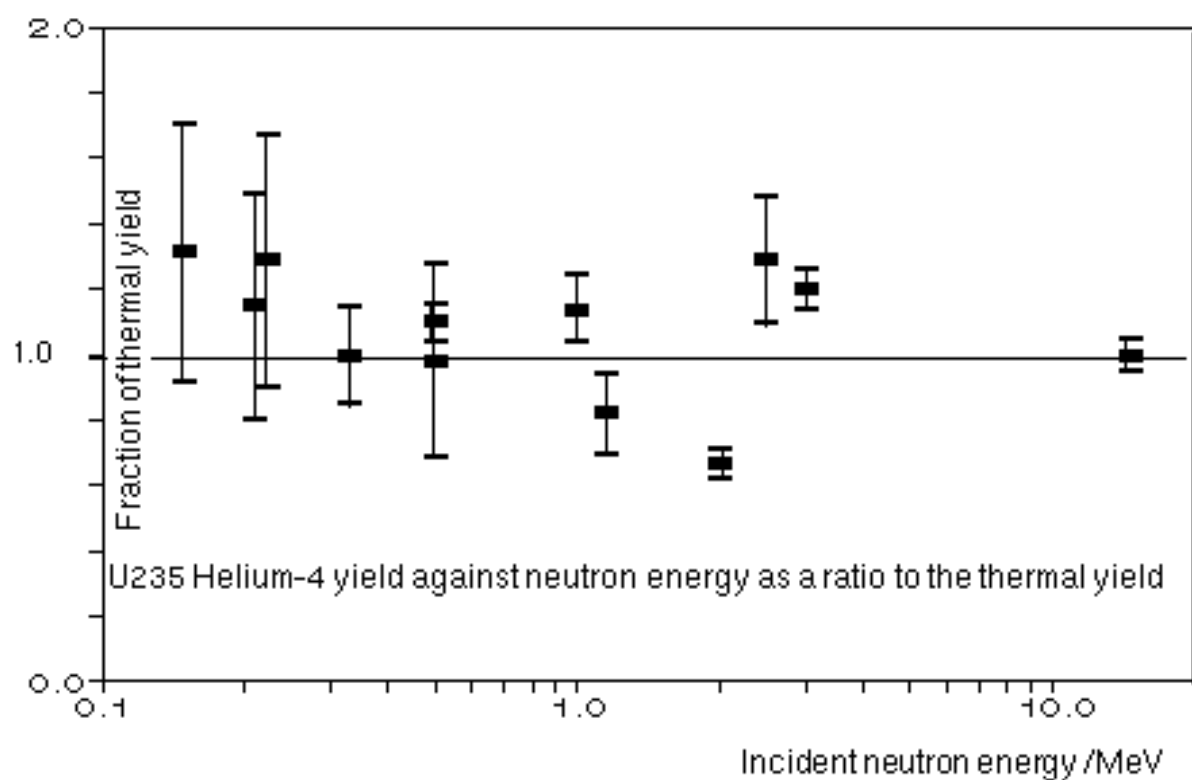


FIG. 4.4.1. Alpha yield as a function of energy for  $^{235}\text{U}$  and  $^{239}\text{Pu}$ .

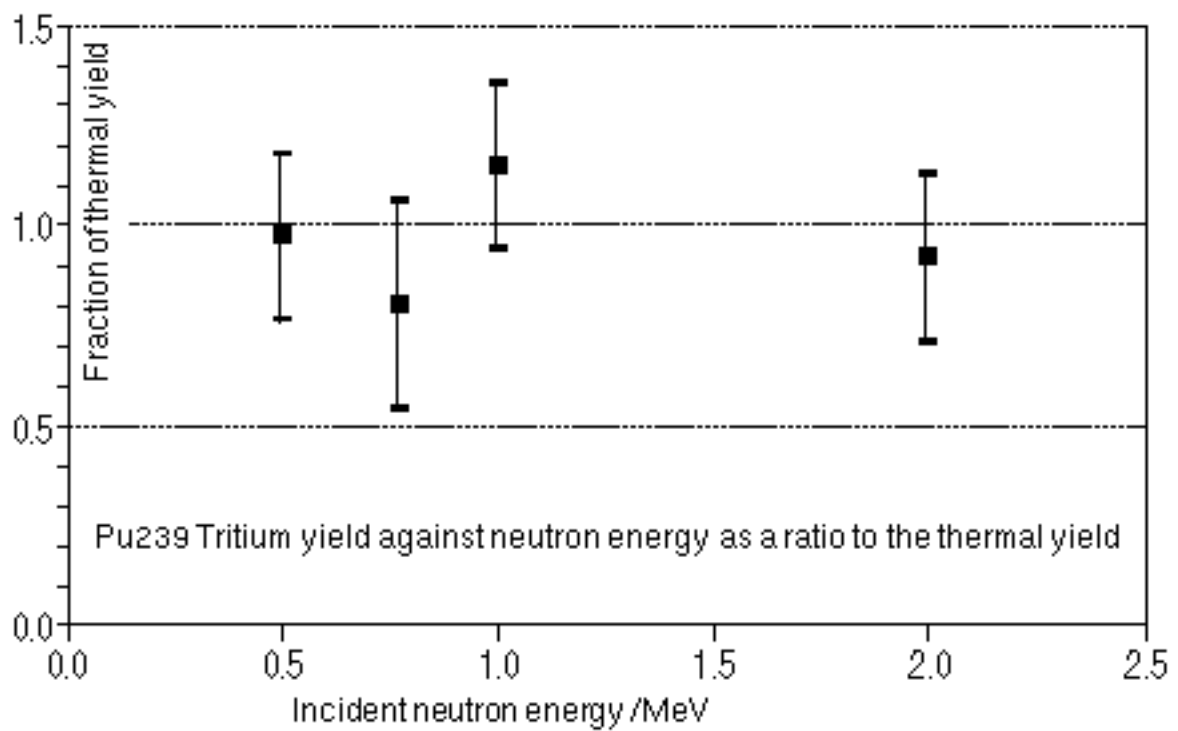
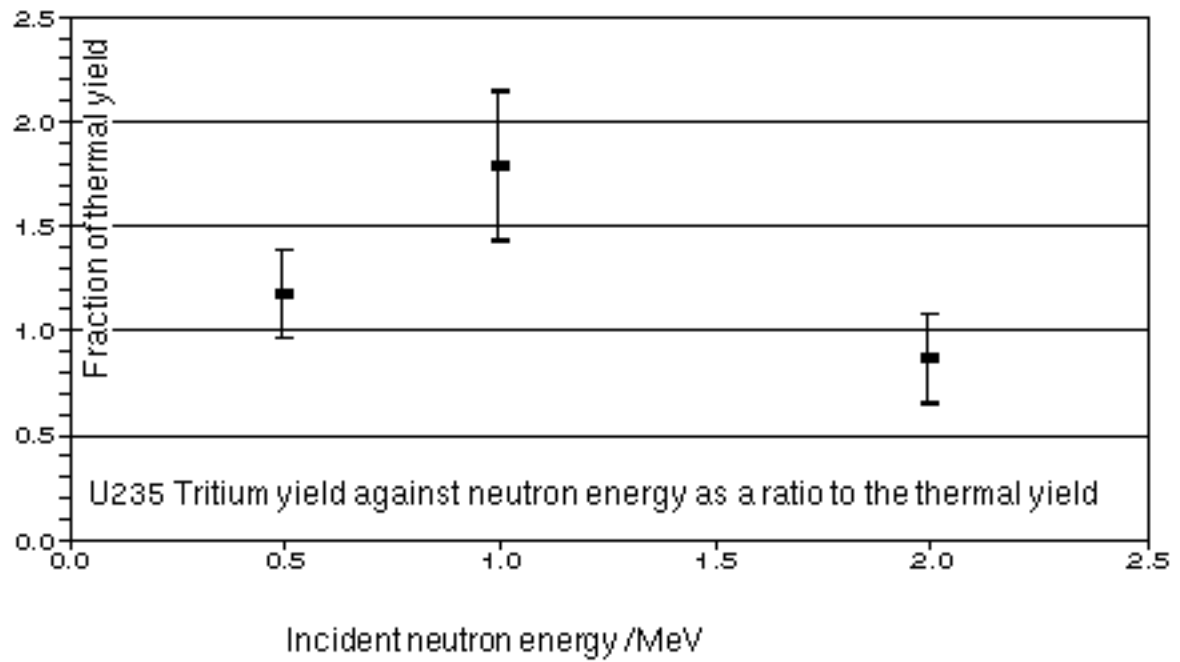


FIG. 4.4.2. Tritium yields as a function of energy for  $^{235}\text{U}$  and  $^{239}\text{Pu}$ .

Similarly the  $^{239}\text{Pu}$  data, although confined to neutron energies between thermal and 2 MeV, show excellent agreement with the thermal yield value.

From the results in Figure 4.4.1 it can be concluded that there is little evidence that the alpha yield varies significantly with energy between thermal and 10.5 MeV.

Figure 4.4.2 shows a similar comparison for tritium yields against incident neutron energy. The only results available measured by the  $\Delta E, E$  method are those of Ouasti [4.4.11]. These results show no significant variation up to 2 MeV, which is the most important region for yields in fission reactors. Thus it can be assumed that the tritium yields are energy independent in thermal and fast reactors, although this may not be so at higher energies.

#### 4.4.4. Variation of LCP yields for different fissioning systems

There have been several attempts to model the light charged particle yields using empirical methods. These techniques are based upon the experimental data and functions of parameters of the compound, or target, nucleus. In the past, several models have been suggested for the alpha yields, primarily through fitting the measured yields to functions of the compound nucleus mass  $A$  and charge  $Z$ .

Nobles [4.4.12] suggested the total ternary yield (alphas, tritons etc.) per 1000 fissions varied within a range of approximately  $\pm 30\%$  as:

$$Y = 0.561 \frac{Z^2}{A} - 18.229 \quad (1)$$

The  $\frac{Z^2}{A}$  term is the fissility parameter related to the “surface tension” term in liquid drop model [4.4.1].

Halpern [4.4.13] on the other hand tried to model the total ternary yield using an expression of the form:

$$Y = m(A + \beta Z) \quad (2)$$

He found that  $b = -4$  and  $m = 0.125$  gave the best fit to his data-set.

For UKFY2 [4.4.7], fits to both  $A - 4Z$  and  $Z^2/A$  were tried. It was found that the alpha yields fitted, to  $\pm 20\%$ , using the expression  $Y(^4\text{He}) = 0.0647 \frac{Z^2}{A} - 2.1292$ . The tritium yields were shown to fit, to  $\pm 25\%$ , as a constant ratio to the alpha yield; the best fit to this form was  $Y(^3\text{H}) = 0.06554 Y(^4\text{He})$ .

The following work is based upon new results from the analysis of the UKFY3 database [4.4.10]. The LCP yield results for hydrogen isotopes and  $^4\text{He}$  are summarized in Table 4.4.5.

From UKFY3 recommended data it can be seen that many alpha and tritium yields have been measured. However very few of these fissioning systems have more than two independent measurements. Also, not all systems of significance have measurements of both alpha and tritium yields. Thus it is necessary to fit the yields to models and then extrapolate and interpolate on the basis of the modeling to estimate the yields for the other fissioning systems within UKFY3.

TABLE 4.4.5. RECOMMENDED LIGHT CHARGED PARTICLE YIELDS FOR HYDROGEN ISOTOPES AND <sup>4</sup>HE RESULTING FROM ANALYSIS OF THE UKFY3 MEASUREMENT DATABASE.

Neutron energy	System	Mass	Yield per 100 fissions	Standard deviation (%)	Number of data points
Thermal	U233	1	6.542E-03	40.2	1
Thermal	U235	1	1.711E-03	10.8	3
Thermal	Pu239	1	4.080E-03	10.0	1
Fast	U235	1	1.174E-02	51.1	1
High	U233	1	9.018E-03	41.4	1
High	U235	1	6.335E-03	40.4	1
High	U238	1	2.001E-03	100.5	1
High	Np237	1	1.902E-02	41.4	1
Spontaneous	Cf252	1	6.086E-03	23.4	2
Spontaneous	Cf250	1	9.000E-03	25.0	1
Spontaneous	Fm256	1	7.000E-03	30.0	1
Spontaneous	Cm244	1	1.221E-02	41.0	1
Thermal	U233	2	8.466E-04	15.6	1
Thermal	U235	2	8.400E-04	17.9	2
Thermal	Pu239	2	1.347E-03	14.2	2
Spontaneous	Cf252	2	1.500E-03	20.0	1
Thermal	U233	3	9.691E-03	14.3	4
Thermal	U235	3	9.314E-03	3.8	15
Thermal	Pu239	3	1.442E-02	5.3	5
Thermal	Pu241	3	1.410E-02	10.0	1
Fast	U235	3	1.352E-02	11.7	4
Fast	Pu239	3	1.413E-02	15.9	1
High	U233	3	2.480E-02	22.6	1
High	U235	3	1.742E-02	20.7	1
High	U238	3	6.499E-03	22.1	1
High	Np237	3	3.329E-02	22.6	1
Spontaneous	Cf252	3	2.244E-02	3.9	8
Spontaneous	Cf250	3	2.700E-02	20.0	1
Spontaneous	Fm256	3	3.900E-02	15.0	1
Spontaneous	Cm244	3	2.197E-02	22.0	1
Thermal	U233	4	2.065E-01	4.1	9
Thermal	U235	4	1.699E-01	3.6	12
Thermal	Pu239	4	2.080E-01	3.3	8
Thermal	Pu241	4	2.015E-01	10.0	2
Fast	U233	4	2.003E-01	10.8	1
Fast	U235	4	1.980E-01	8.6	4
Fast	Pu239	4	2.029E-01	8.7	2
High	Th232	4	7.181E-02	36.4	2
High	U233	4	1.957E-01	10.6	1
High	U235	4	1.667E-01	5.3	3
High	U238	4	8.226E-02	9.5	4
High	Np237	4	2.010E-01	10.6	1
Spontaneous	Pu240	4	3.190E-01	10.0	1
Spontaneous	Pu242	4	2.740E-01	10.0	1
Spontaneous	Cf252	4	3.102E-01	6.3	4
Spontaneous	Cf250	4	3.980E-01	10.0	1
Spontaneous	Fm256	4	4.742E-01	8.3	2
Spontaneous	Fm257	4	3.994E-01	7.1	2
Spontaneous	Cm244	4	2.849E-01	9.1	3
Spontaneous	Cm242	4	3.601E-01	12.1	2

For UKFY3 many variations of functions based upon Z and A were fitted ( $mZ^2/A + c$ ,  $mZ + c$ ,  $mA + c$ ,  $m(3Z - A) + c$ ,  $m(4Z - A) + c$ ,  $m(5Z - A) + c$ ,  $m(5Z - 2A) + c$ ,  $m(6Z - A) + c$ ,  $m(7Z - A) + c$ ,  $m(7Z - 2A) + c$ ,  $mA + uZ + c$  and  $mA + uZ$ ). Also the effect of fitting to the target nucleus mass and charge, rather than the compound nucleus was tried. The best fit was found in all cases to be the function  $Y = mA + uZ + c$ . This was the case where there were sufficient data to fit this function, which there were for masses one to four (hydrogen, deuterium, tritium and helium-4). As the energy dependence had been shown to be minimal, all the data were merged; however a separate attempt at fitting the energies groups individually did not give any better fit. The results for masses one to four are summarized in Table 4.4.6.

TABLE 4.4.6. FIT OF LIGHT CHARGED PARTICLE YIELDS TO  $MA+UZ+C$

Mass	No. of yields	m	u	c	A and Z	% ssd <sup>a</sup>	maxfd <sup>b</sup>	$\chi^2$ <sup>c</sup>	max diff <sup>d</sup>
1	12	-7.342E-04	2.655E-03	-6.923E-02	Target	18.1	1.204	17.84	2.04
1	12	-1.256E-03	3.627E-03	-3.851E-02	Compound	13.2	0.849	11.51	1.68
2	4	-3.337E-05	2.249E-04	-1.198E-02	Target	5.5	0.141	1.470	1.00
2	4	-3.047E-05	2.102E-04	-1.138E-02	Compound	5.5	0.143	1.480	1.00
3	14	-8.018E-04	4.347E-03	-0.2012	Target	7.9	0.633	31.36	2.80
3	14	-1.209E-03	5.168E-03	-0.1837	Compound	7.1	0.552	28.84	2.63
4	20	-2.077E-02	7.972E-02	-2.273	Target	3.1	0.292	58.44	-4.02
4	20	-1.877E-02	7.525E-02	-2.394	Compound	4.5	-0.54	115.4	-5.73

Notes: a.  $\%ssd = 100 \sqrt{\frac{1}{n(n-1)} \sum_i \frac{(y_i - y_{calc})^2}{y_{calc}^2}}$

c.  $\chi^2 = \sum_i \left( \frac{y_i - y_{calc}}{\sigma_i} \right)^2$

b.  $maxfd = \frac{y_i - y_{calc}}{y_i}$

d.  $max\ diff = \frac{y_i - y_{calc}}{\sigma_i}$

Table 4.4.6 shows the  $^4\text{He}$  yields are best fitted by using the target nucleus parameters for which the calculated yields have an estimated percentage uncertainty (%ssd) of 3.1% of the calculated value. As the  $\chi^2$  is above 1, this should be multiplied by the square root of  $\chi^2$  to give the best estimate of the uncertainty of the calculated value, in this case 5.8%. The fit is shown in Figure 4.4.3 along with a plot of the experimental yields divided by the model calculation.

The fit shown in Figure 4.4.3 can be used to predict all the unmeasured alpha fission yields required for the UKFY3 evaluation. However, similar fits for masses 1, 2 and 3, when extrapolated, predict some unphysical negative yields for fissioning systems that are required for UKFY3. Thus, as an alternative, the ratios of the these yields to the alpha yield were investigated. Interestingly, for each light charged particle mass, the ratios of each yield to the alpha fission yield appear similar, apparently independent of the fissioning system. Thus a fit to a constant ratio was attempted, which is effectively a weighted mean of the ratios. The weighted mean of the ratios and the internal and external standard deviations are shown in Table 4.4.7 for each mass from one to ten. The higher of the internal and external standard deviations was accepted as the recommended error and is quoted in the table as a percentage. The maximum difference between the experimental and calculated yields, expressed as a multiple of the experimental standard deviation, is also shown.

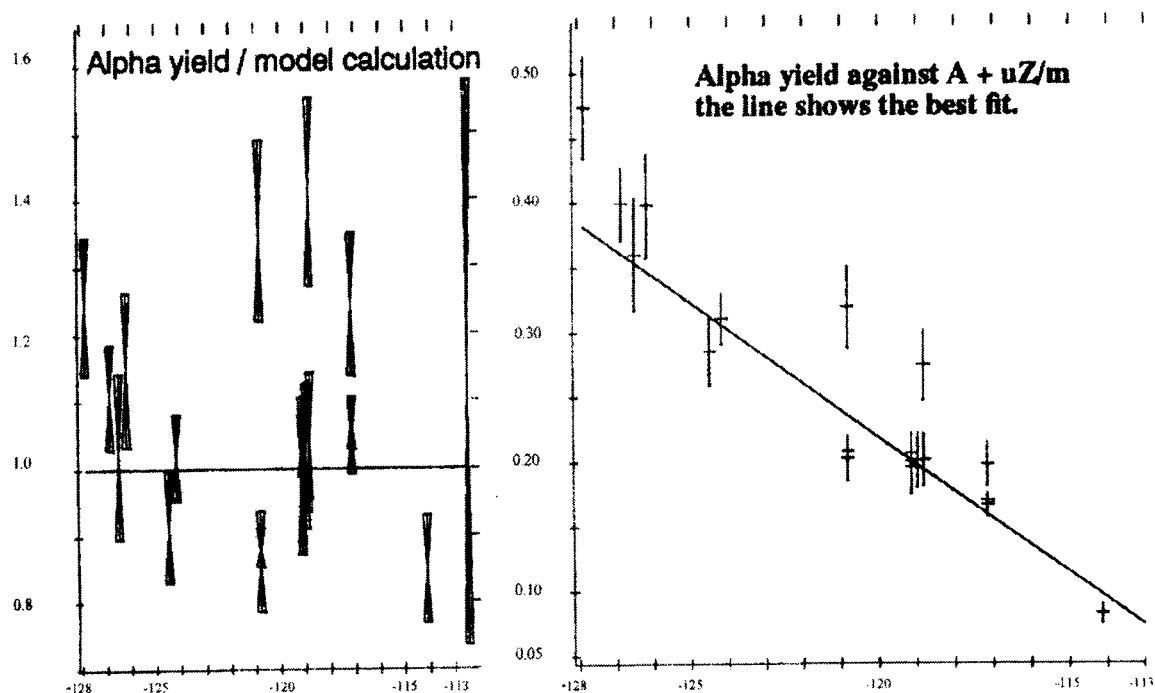


FIG. 4.4.3. Fit of alpha yields to  $mA + uZ + c$ .

TABLE 4.4.7. WEIGHTED MEANS OF YIELD RATIO TO  $^4\text{He}$  YIELDS

Mass	Weighted mean	Internal standard deviation	External standard deviation	Recommended % error of mean	Max. diff. /expt. error
1	1.58184E-02	1.98492E-03	2.53297E-03	16.0	-2.018
2	4.83424E-03	6.92095E-04	4.39668E-04	14.3	0.896
3	6.89694E-02	5.49694E-03	5.20911E-03	8.0	1.934
4	1.00000				
6	1.39646E-02	2.03853E-03	2.06317E-03	14.8	-1.200
7	4.33226E-04	8.18369E-05	8.13437E-05	18.9	1.260
8	2.61057E-04	4.01731E-05	6.46611E-05	24.8	1.664
9	3.08327E-04	5.30162E-05	7.18898E-05	23.3	1.547
10	3.78596E-03	6.32407E-04	5.76288E-04	16.7	0.884

The experimental yield data for masses 1, 2 and 3 are shown in Figures 4.4.4, 4.4.5 and 4.4.6. The weighted mean of the ratio with  $\pm$  one standard deviation uncertainties are shown by three lines across the plots.

These figures and Table 4.4.7 show that a reasonable assumption is that the hydrogen yield is a constant fraction of the alpha yield, and independent of mass and charge of the fissioned nuclide.

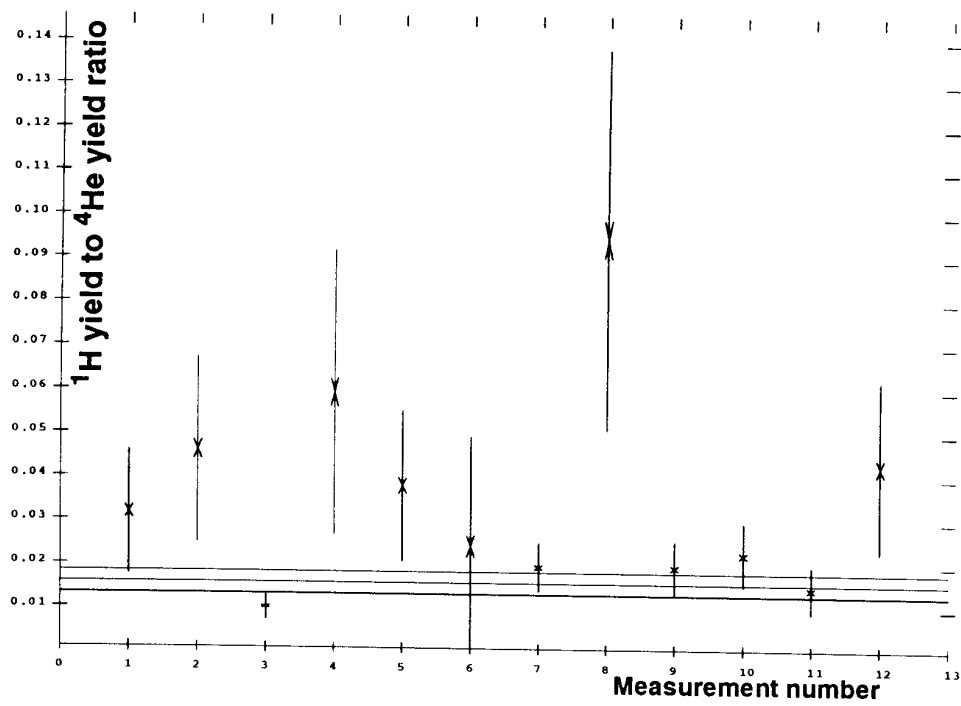


FIG. 4.4.4. Ratio of  $^1\text{H}$  yield to  $^4\text{He}$  yield.

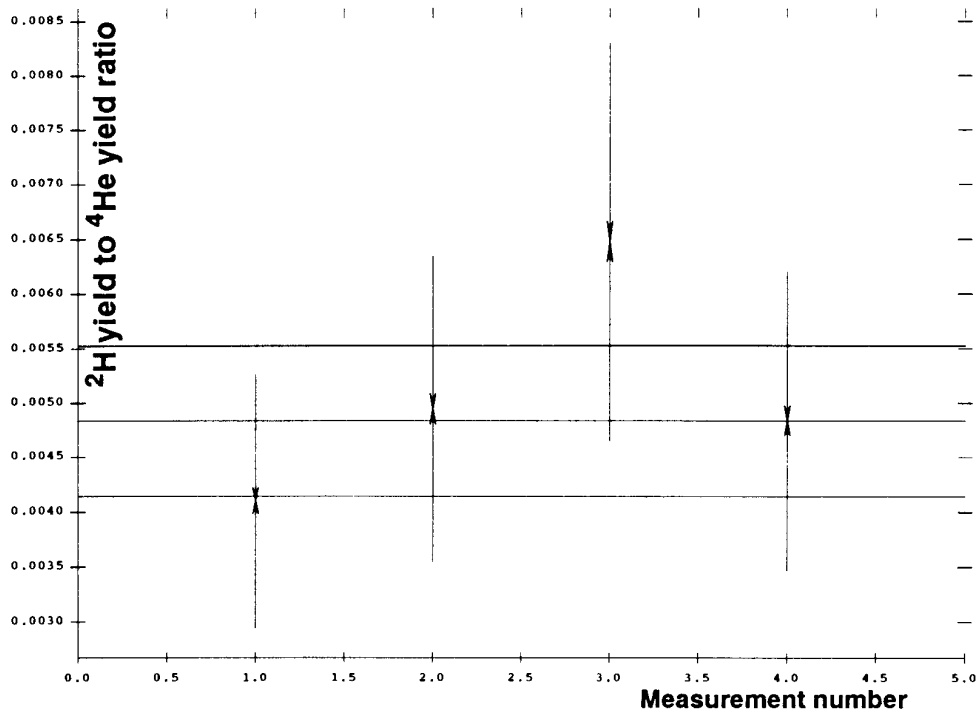


FIG. 4.4.5. Ratio of  $^2\text{H}$  yield to  $^4\text{He}$  yield.



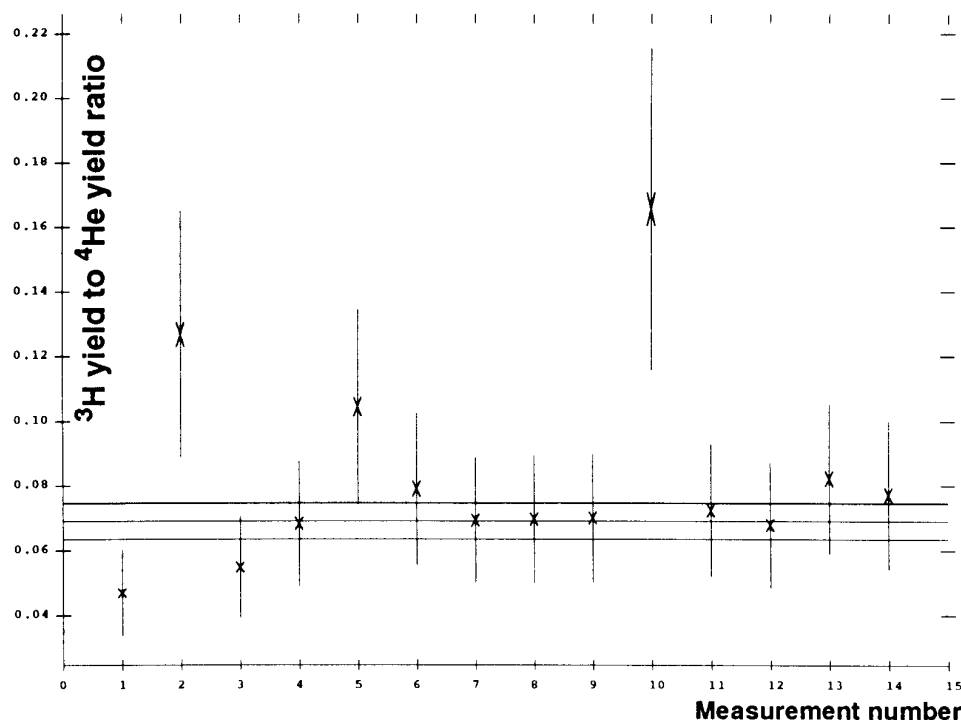


FIG. 4.4.6. Ratio of  $^3\text{H}$  yield to  $^4\text{He}$  yield.

#### 4.4.5. Conclusion

For the UKFY3 evaluation the primary source of recommended data was from the analysis of experimental results. Missing  $^4\text{He}$  yields were calculated from  $Y = mA + uZ + c$ , where  $m = -2.077 \times 10^{-2}$ ,  $u = 7.972 \times 10^{-2}$  and  $c = -2.273$  with a 5.8% standard deviation. Missing  $^1\text{H}$ ,  $^2\text{H}$  and  $^3\text{H}$  yields were then calculated using the  $^4\text{He}$  yields together with the ratios given in Table 4.4.7 and with the fractional standard deviations calculated by quadrature.

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## Chapter 5

### ENERGY DEPENDENCE OF FISSION YIELDS

#### 5.1. MEASUREMENTS OF THE ENERGY DEPENDENCE

The dependence of fission yields on (neutron) energy is a quite complex problem, due to the complicated fission mechanism. At present there are still many open questions and further studies in experiment and theory are needed. But some observed correlations based on existing experimental data can be summarized.

##### 5.1.1. Thermal energy point and eV region

R.W. Mills [5.1.1] gave a summary review on the experimentally measured data in this energy region. The Los Alamos Radiochemistry Group [5.1.2] measured R-values for  $^{235}\text{U}$  epithermal to thermal neutron fission relative to  $^{99}\text{Mo}$  for  $^{115}\text{Cd}$ ,  $^{112}\text{Pd}$  etc. The results are listed in Table 5.1.1. It can be seen that the R-values are smaller than 1 for all product nuclides in the valley, which means that the fission yield in the epithermal energy range is lower than that at thermal energy. This is abnormal in the sense that, in general, the fission yield for fission products in the valley increases with increasing excitation energy.

TABLE 5.1.1. R-VALUES OF VARIOUS FISSION PRODUCTS

Fission product	R (Cd ratio = 8)	R (Cd ratio = 30)
$^{97}\text{Zr}$	1.00	0.98
$^{109}\text{Pd}$	0.98	1.00
$^{111}\text{Ag}$	0.89	0.92
$^{112}\text{Pd}$	0.85	0.86
$^{115}\text{Cd}$	0.84	0.83
$^{136}\text{Cs}$	0.98	0.96

R. Nasuhoglu et al. [5.1.3] measured R-values of  $^{111}\text{Ag}$ ,  $^{115}\text{Cd}$  and  $^{127}\text{Sb}$  relative to  $^{89}\text{Sr}$  from  $^{235}\text{U}$  fission with monoenergetic (1–10 eV) neutrons relative to thermal neutrons with a crystal spectrometer. The data are listed in Table 5.1.2.

TABLE 5.1.2. R-VALUES OF VARIOUS FISSION PRODUCTS

Fission product	1.1 eV	3.1 eV	9.5 eV
$^{111}\text{Ag}$	$1.11 \pm 0.20$	$1.06 \pm 0.20$	$1.00 \pm 0.20$
$^{115}\text{Cd}$	$1.18 \pm 0.20$	$0.73 \pm 0.20$	$0.91 \pm 0.20$
$^{127}\text{Sb}$	$1.10 \pm 0.20$		

Although the uncertainty of the data is fairly large, the trends of yield changes can still be seen. The R-value for  $^{111}\text{Ag}$  is larger than 1, but is decreasing with increasing neutron energy. For  $^{115}\text{Cd}$ , it is larger than 1 at 1.1 eV, and then becomes less than 1, which agrees with the results of [5.1.2].

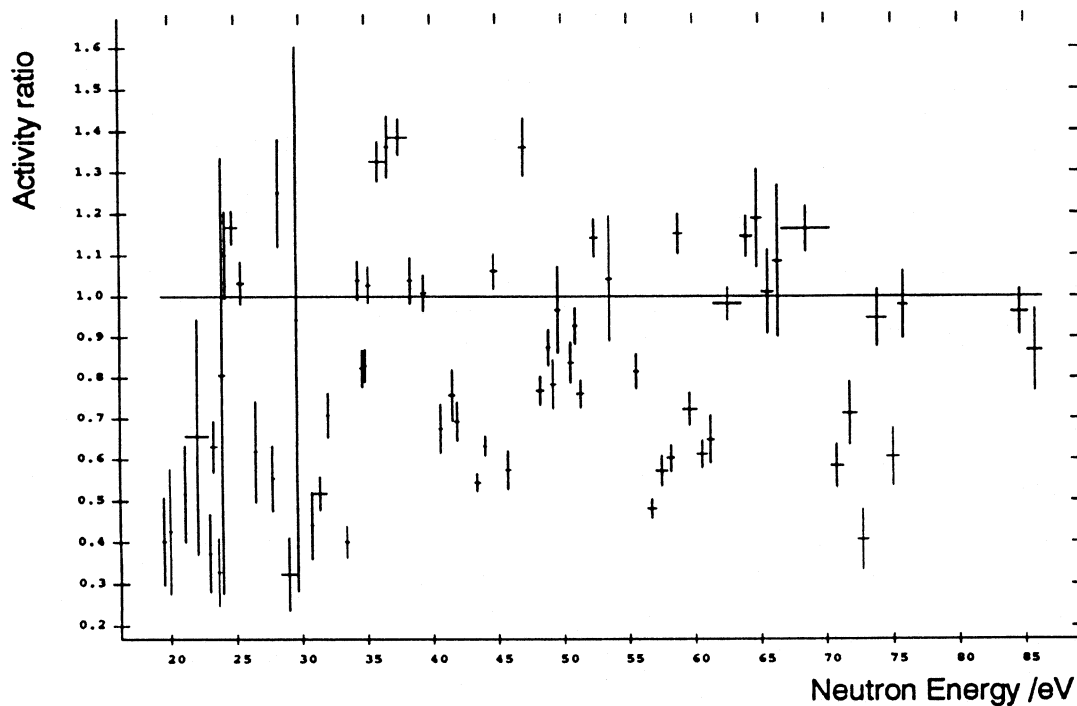


FIG. 5.1.1.  $^{115}\text{Cd}$  R-values (relative to  $^{99}\text{Mo}$  and thermal energy) for  $^{235}\text{U}$ .

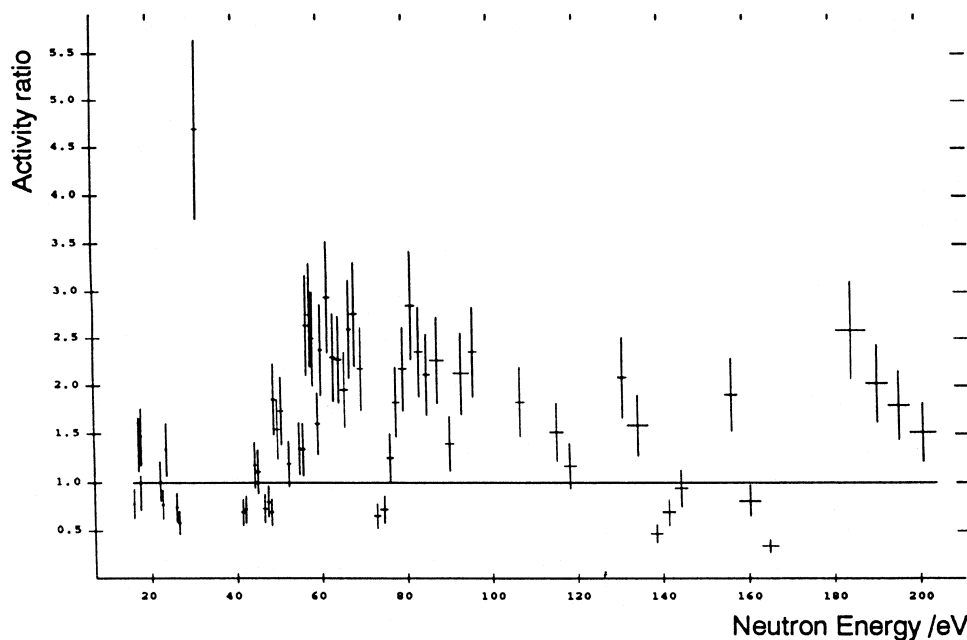


FIG. 5.1.2.  $^{115}\text{Cd}$  R-values (relative to  $^{99}\text{Mo}$  and thermal energy) for  $^{239}\text{Pu}$ .

G.A. Cowan et al. measured R-values of monoenergetic to thermal neutron fission relative to  $^{99}\text{Mo}$  for  $^{235}\text{U}$  ( $E_n = 19.3\text{--}86.3$  eV) [5.1.4–5.1.6] and for  $^{239}\text{Pu}$  ( $E_n = 15.8\text{--}204$  eV) [5.1.7], using the time-of-flight method (flight distance 200 m) and a white neutron source from an underground nuclear explosion. Some results are shown in Figs 5.1.1–5.1.2 (taken from [5.1.1]).

It can be seen that the variation of fission yields at different energy points relative to the thermal yield is significant (assuming that the yield of  $^{99}\text{Mo}$ , a fission product in the light mass peak, is constant), exhibiting a resonance like structure around the thermal yield value.

F.J. Hamsch et al. [5.1.8] measured the fission fragment yield versus mass and total kinetic energy versus incident neutron energy in the energy range 0.006–130 eV at the Geel Electron Linear Accelerator (GELINA) using a Frisch-gridded ionization chamber. The results are shown in Figs 5.1.3 and 5.1.4. It can be seen from Fig. 5.1.3 that the mass distributions for neutron energies between the resonances are almost the same as for thermal neutrons. The asymmetric peaks of mass distributions change their shape with the resonances or resonance groups. The yield differences,  $\text{YIELD}(E_n) - \text{YIELD}(\text{Thermal})$ , are first negative and then positive for the light mass peak, and opposite, namely first positive and then negative, for the heavy mass peak. Figure 5.1.4 demonstrates the drops in symmetric fission yields for all resonances.

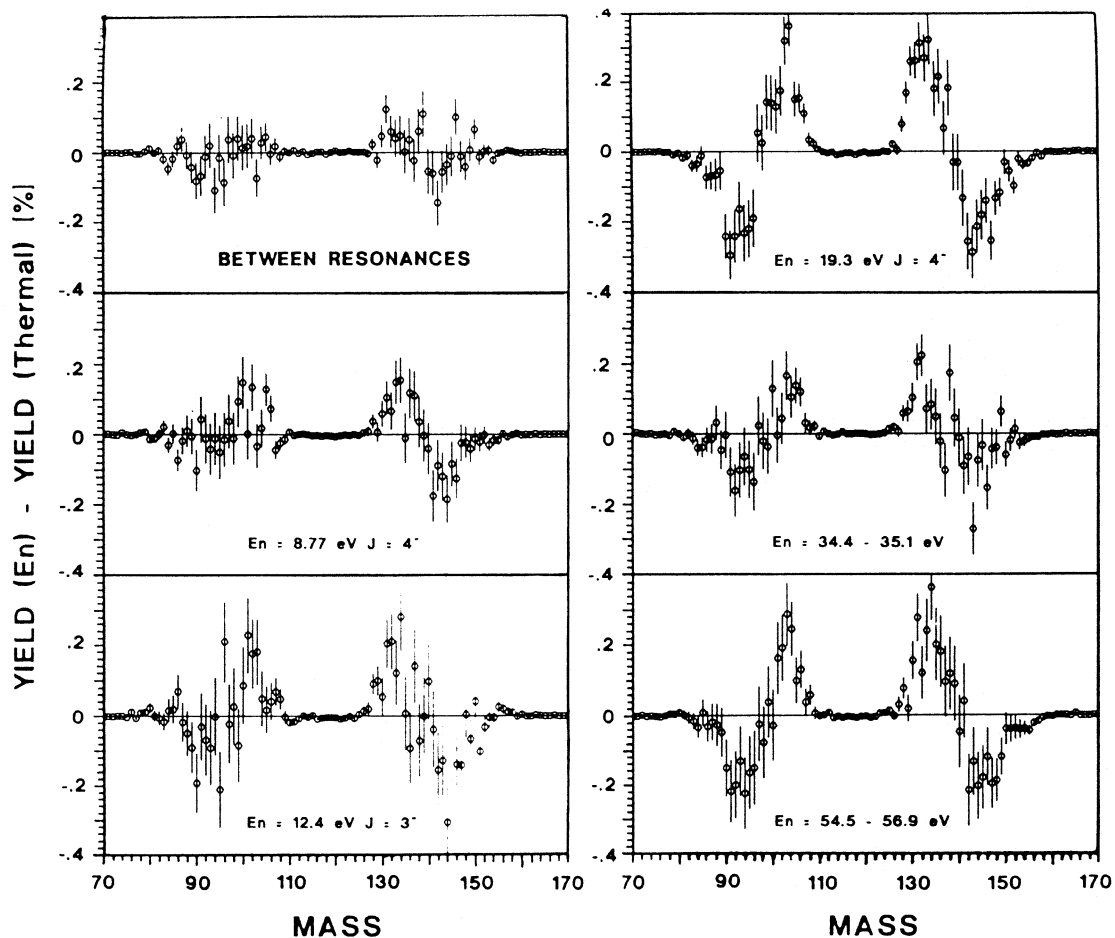


FIG. 5.1.3. Absolute fission fragment yield differences with respect to thermal values.

In summary, there are many experimental data for fission yield ratios or R-values relative to thermal in the energy range up to 100 eV. All of them show a similar behaviour, namely that the fluctuation of the ratios or R-values show a resonance like structure, especially for fission products in the valley (symmetric fission), where the ratios or R-values are generally smaller than 1. But so far it could not be ascertained whether these fluctuations can be explained by a certain resonance character, as the  $^{235}\text{U}$  yield data could not be correlated with the resonance spins. A possible explanation is multiple chance fission for each spin, and each channel has a characteristic yield distribution.

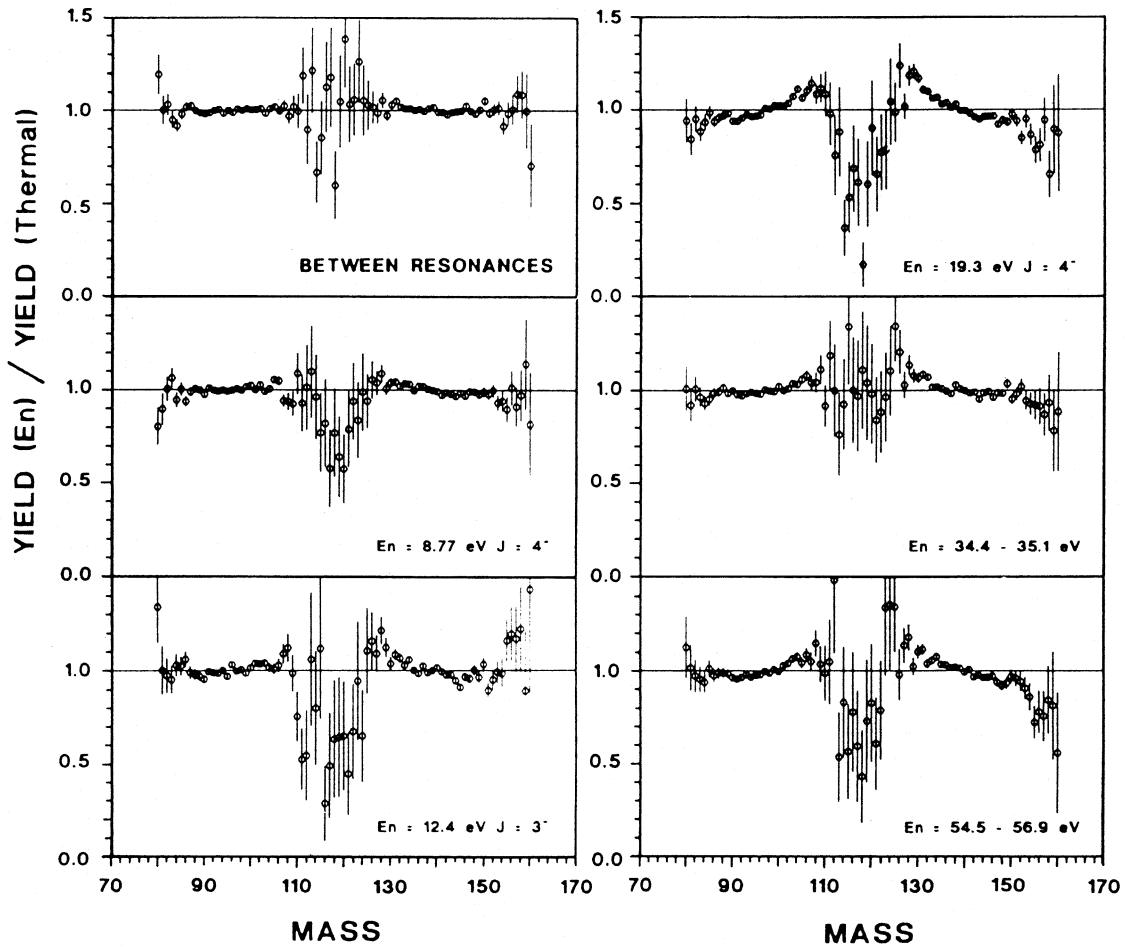


FIG. 5.1.4. Fission fragment yield ratios with respect to thermal values.

### 5.1.2. MeV region

A lot of fission yield measurements exist in this energy region for  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  and others. There are two kinds of measurements, one with monoenergetic neutron sources like Van de Graaff, Cockcroft-Walton or Tandem accelerators, the other employing neutron spectra like thermal or fast reactors, fission neutrons or accelerators with white neutron sources. For the latter, the measured yields are spectrum averaged and could not be used for studying its energy dependence exactly, although some trends of the dependence could be observed. So, here only some typical, important measurement results using monoenergetic neutrons were considered, especially for  $^{235}\text{U}$ , to show the trend in the energy dependence.

G.P. Ford et al. (Los Alamos Scientific Lab.) [5.1.9] measured the mass yield from  $^{235}\text{U}$  fission induced by 4–18 MeV neutrons and  $^{232}\text{Th}$  fission induced by 19–29 MeV  $\alpha$  particles, which both produce the same compound nucleus  $^{238}\text{U}$ . The measurements were performed at a variable energy cyclotron for  $\alpha$  particles, and Van de Graaff, and Cockcroft-Walton accelerators for neutrons using D(d,n), T(d,n) reaction as neutron sources, combined with a radiochemical method. The results show that there is evidence of a slight dip in the symmetric fission yield at 20 MeV excitation energy ( $E_n \approx 13.5$  MeV,  $E_\alpha = 25$  MeV), the onset of (x,2nf) fission. Symmetric yields from fission induced by neutrons and  $\alpha$  particles are both about the

same at excitation energies below 15.7 MeV, but are generally 9% greater for  $\alpha$  fission than for neutron fission at an excitation energy of 20 MeV.

L.E. Glendenin et al. (Argonne National Lab.) [5.1.10] measured mass yields from  $^{235}\text{U}$  neutron fission at 9 energy points from 0.17 to 8.1 MeV. The 37 product nuclides range from  $^{84}\text{Br}$  to  $^{151}\text{Pm}$ , including products from both asymmetric (peaks) and symmetric (valley) fission. The measurements employed direct  $\gamma$  spectrometry and radiochemical separation of the fission products followed by  $\beta$  counting and/or  $\gamma$  spectrometry. The measured data show a sensitive increase of fission yields in the near symmetric mass region with increasing neutron energy, but only small changes for yields from asymmetric fission, the peak-to-valley ratio decreasing from 590 to 13. The curves of valley yields as a function of  $E_n$  display a flat step in the region of second-chance fission (above  $\sim 6$  MeV). The authors also made a comparison between the yields from  $^{235}\text{U}$  and  $^{238}\text{U}$  fission, using their earlier data. In Fig. 5.1.5, the yields of  $^{115}\text{Cd}$ , a typical symmetric fission product, and  $^{140}\text{Ba}$ , a typical asymmetric fission product, are plotted as a function of incident neutron energy  $E_n$  and excitation energy  $E_x = E_n + B_n$  ( $B_n$  is the neutron binding energy). It can be seen that the yields versus excitation energy for the two fissioning systems are the same for peak fission products and nearly the same for valley fission products, at least in the region where first-chance fission only occurs.

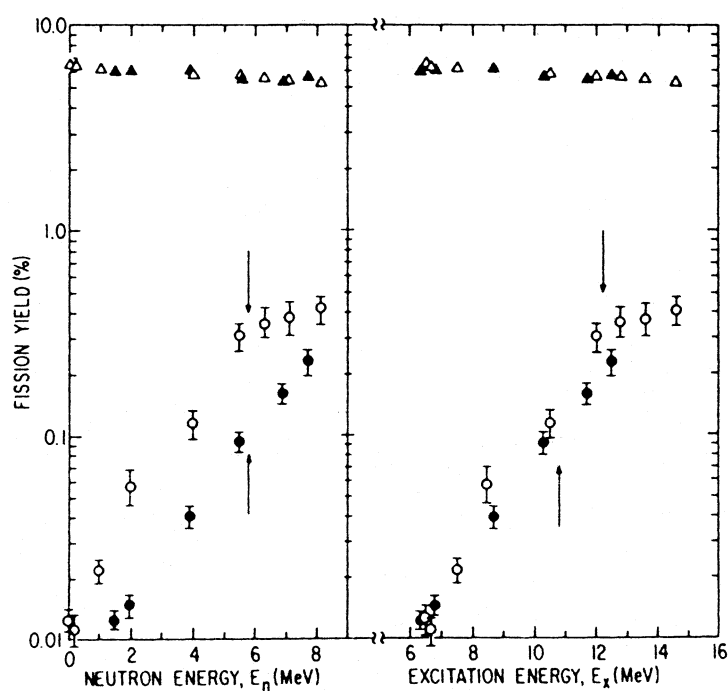


FIG. 5.1.5. The yield of  $^{140}\text{Ba}$  and  $^{115}\text{Cd}$  as a function of incident neutron energy and excitation energy of compound nucleus.

$\Delta$   $^{140}\text{Ba}$ ,  $^{235}\text{U}$  fission       $\circ$   $^{115}\text{Cd}$ ,  $^{235}\text{U}$  fission  
 $\blacktriangle$   $^{140}\text{Ba}$ ,  $^{238}\text{U}$  fission       $\bullet$   $^{115}\text{Cd}$ ,  $^{238}\text{U}$  fission

T.C. Chapman et al. (Lawrence Livermore Lab.) [5.1.11] measured the yields from 6–9 MeV neutron induced fission of  $^{235}\text{U}$  and  $^{238}\text{U}$ , using direct  $\gamma$  spectrometry as well as radiochemical separation followed by  $\gamma$  (Ge(Li)) or  $\beta^-$  (gas-flow proportional counter) counting for 28 fission products from  $^{84}\text{Br}$  to  $^{156}\text{Eu}$ . The measurements were performed at an

insulated core transformer (ICT) accelerator and Cyclograaff accelerator using the T(d,n) reaction for 14.8 MeV and the D(d,n) reaction for 6.0–9.1 MeV neutrons. The most interesting feature of these results is the observed dependence of the fission yield on the neutron energy between 6 and 9 MeV, where the steady increase in yield is interrupted by a dip for symmetric fission, e.g.  $^{111}\text{Ag}$  and  $^{112}\text{Pd}$ . Both  $^{235}\text{U}$  and  $^{238}\text{U}$  fission yields show the same behaviour.

There are other fission yield measurements with monoenergetic neutrons [5.1.12–5.1.20], but they are only at one energy point or only for one product [5.1.19], most of them around 14.5 MeV. A more important measurement was reported by Li Ze et al [5.1.20] in 1995 for the asymmetric fission products  $^{95}\text{Zr}$ ,  $^{140}\text{Ba}$  and  $^{147}\text{Nd}$  from  $^{235}\text{U}$  fission by 8.0 MeV neutrons, where the data are scarce. The results show that the yields deviate from the exponential dependence on neutron energy.

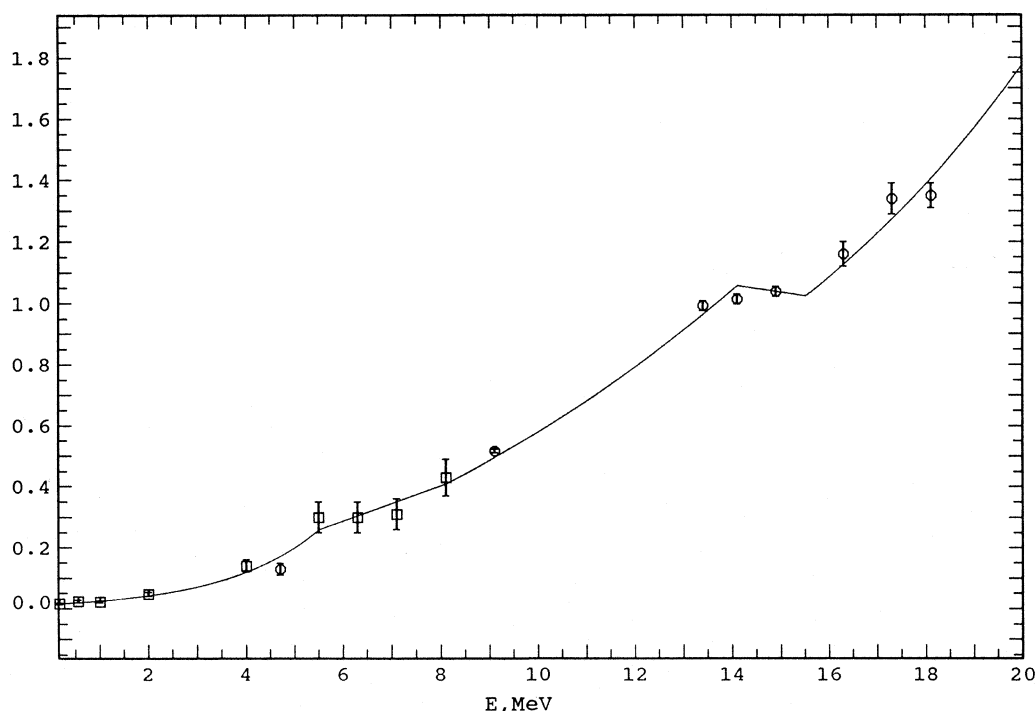


FIG. 5.1.6. Monoenergetic fission yields for  $^{111}\text{Ag}$  from  $^{235}\text{U}$  fission.

— Systematics,  $\square$  L.E. Glendenin (ANL),  $\circ$  G.P. Ford.

Comparing these monoenergetic fission yields for symmetric fission products, e.g.  $^{111}\text{Ag}$ ,  $^{112}\text{Pd}$ ,  $^{115}\text{Cd}$ ,  $^{127}\text{Sb}$  etc., it was found that the data measured by Chapman [5.1.11] are systematically lower than Glendenin's [5.1.10] data at low energies and Ford's [5.1.9] data at high energies. Therefore these data were not used in the following plots and analysis. The data measured by Ford and Glendenin are, generally, in good agreement. Therefore it can be concluded that their observed trends are reliable.

For symmetric fission, the yield increases with incident neutron energy in two steps: first exponentially up to about 5.5 MeV, then a first step-like flattening of the increase up to about 8.0 MeV, then another exponential increase up to  $\sim 14.0$  MeV, followed by a second shoulder roughly from 14.0 MeV to 15.5 MeV, finally again an exponential increase up to 20 MeV. The two shoulders correspond to the onsets of (n,n'f) and (n,2n'f) (second and third



chance fission) respectively, which reduce the excitation energy of the corresponding compound nucleus. These features are characteristic for fission products with  $A = 109\text{--}125$ , and some typical examples are shown in Figs 5.1.6 and 5.1.7.

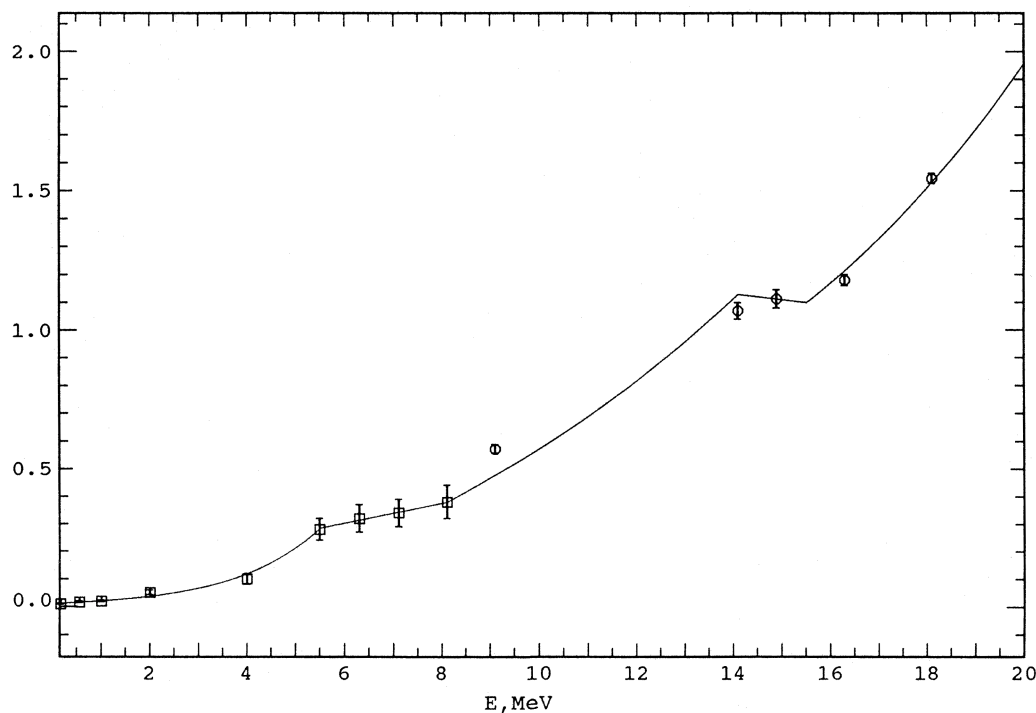


FIG. 5.1.7. Monoenergetic fission yields for  $^{115}\text{Cd}$  from  $^{235}\text{U}$  fission.  
 — Systematics,  $\blacksquare$  L.E. Glendenin (ANL),  $\circ$  G.P. Ford.

For asymmetric fission, the fission product yield decreases slowly and roughly linearly (not exponentially) with increasing neutron energy. Some typical examples are shown in Fig. 5.1.8 for the light mass peak and Fig. 5.1.9 for the heavy mass peak. However, a careful study of the dependence revealed that the linear dependence on energy is true for some products like  $^{95}\text{Zr}$  and  $^{140}\text{Ba}$ , but is not true for others like  $^{99}\text{Mo}$  and  $^{143}\text{Ce}$ . The energy dependence of the yield of  $^{99}\text{Mo}$  (Fig. 5.1.10) from the light mass peak exhibits a concave at  $E_n \approx 500$  keV, whereas that of  $^{143}\text{Ce}$  (Fig. 5.1.11) from the heavy mass peak shows first an increase, then a decrease, starting at about 2 MeV, followed by a concave around 4 MeV, then another increase up to 6 MeV, and finally a linear decrease. This feature is shown more clearly for  $^{147}\text{Nd}$  (Fig. 5.1.12), which is already on the right wing of the heavy mass peak.

For the product nuclides at the wings of the peaks of the mass distribution, the yield dependence on energy is quite complicated. The curve shape is quite sensitive to the mass number and could change significantly with only a small change in mass number. Examples are given in Figs 5.1.13–5.1.14. Up to now there are not enough measured data to reveal the structure and to find the parameters for systematics. However, some characteristic features are noticeable, for example the behaviour of the  $^{147}\text{Nd}$  yield as discussed above is similar for products with mass numbers from 143 to 149 on the right end/wing of the heavy mass peak.

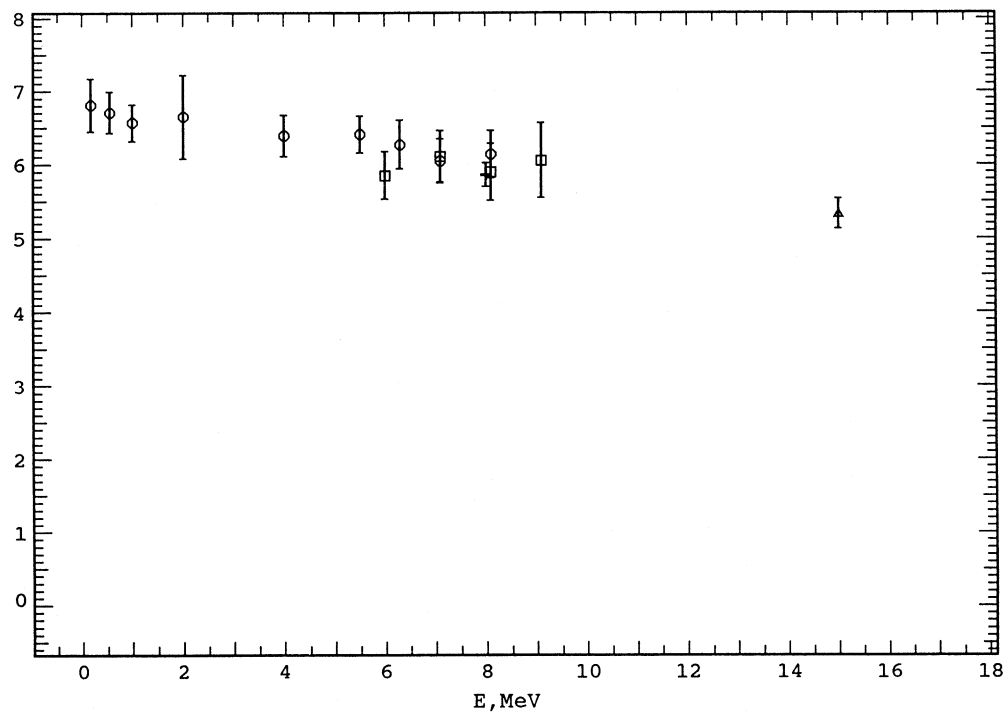


FIG. 5.1.8. Monoenergetic fission yields for  $^{95}\text{Zr}$  from  $^{235}\text{U}$  fission.

■ T.C. Chapman (LR), ○ L.E. Glendenin (ANL), △ J. Laurec (FRBRC),  
+ LI Ze (CAEP).

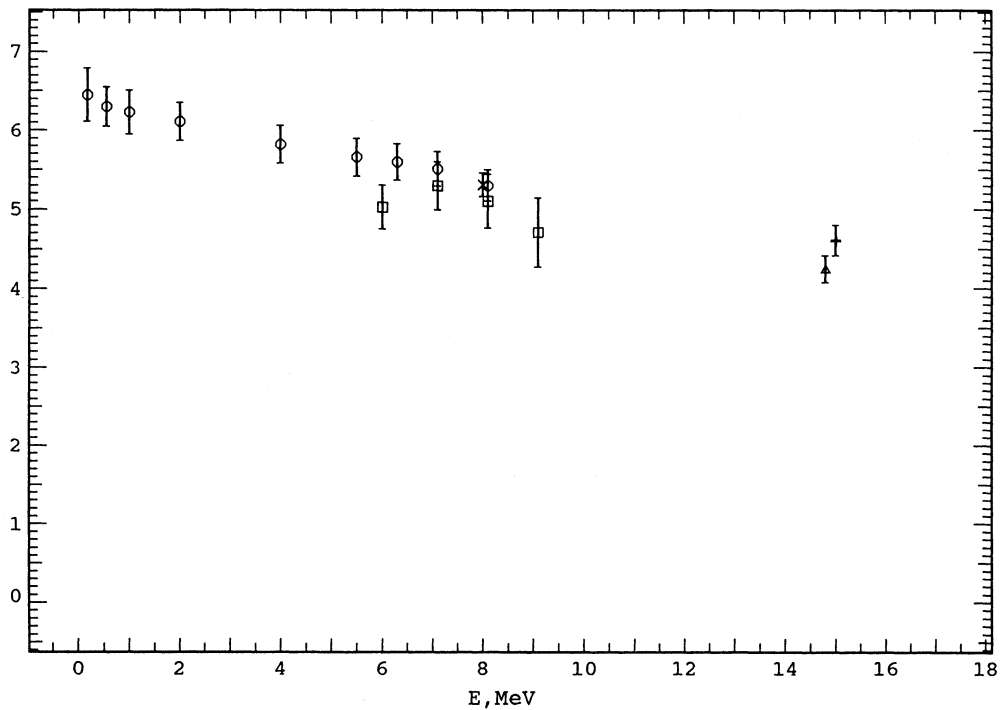


FIG. 5.1.9. Monoenergetic fission yields for  $^{140}\text{Ba}$  from  $^{235}\text{U}$  fission.

■ T.C. Chapman (LR), ○ L.E. Glendenin (ANL), △ D.R. Nethaway (IRL),  
+ J. Laurec (FRBRC); × LI Ze (CAEP).

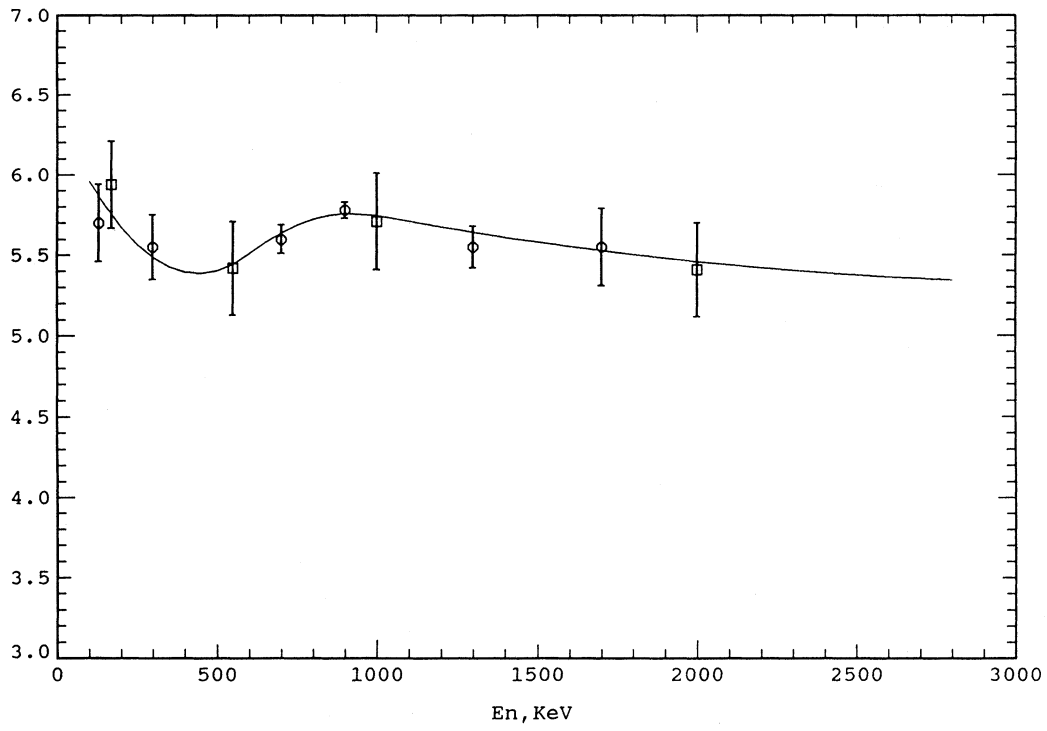


FIG. 5.1.10. Monoenergetic cumulative fission yields for  $^{99}\text{Mo}$  from  $^{235}\text{U}$  fission.  
— spline fit,  $\square$  12729002,  $\circ$  20769002.

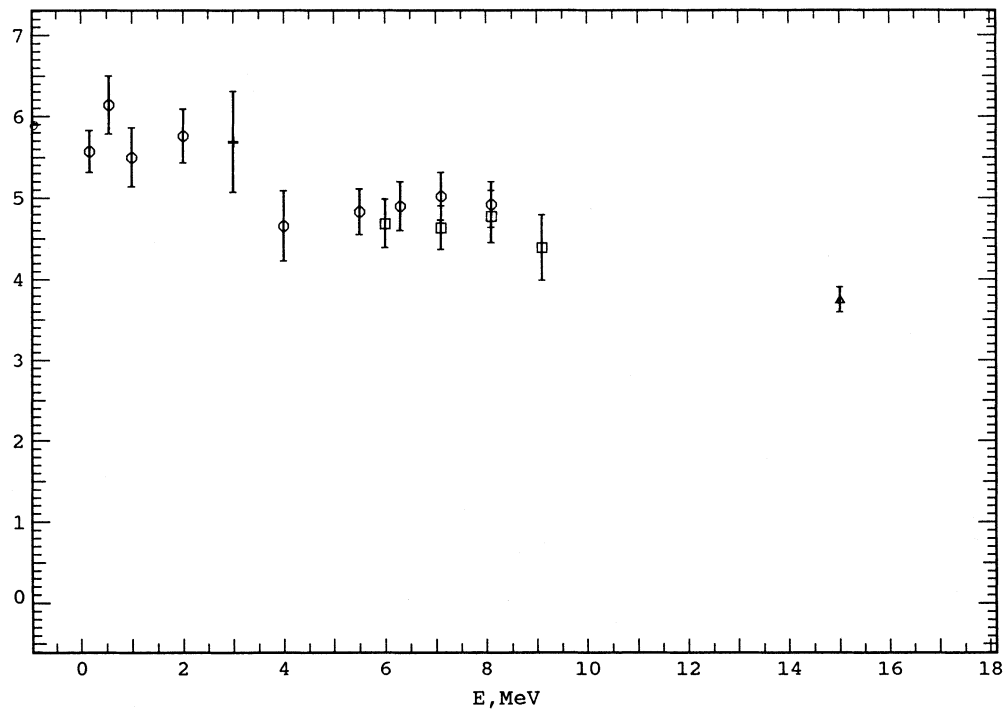


FIG. 5.1.11. Monoenergetic fission yields for  $^{143}\text{Ce}$  from  $^{235}\text{U}$  fission.  
 $\square$  T.C.Chapman (LRL),  $\circ$  L.E.Glendenin (ANL),  $\Delta$  J.Laurec (FRBRC),  
+ A.N.Gudkov (MIF).

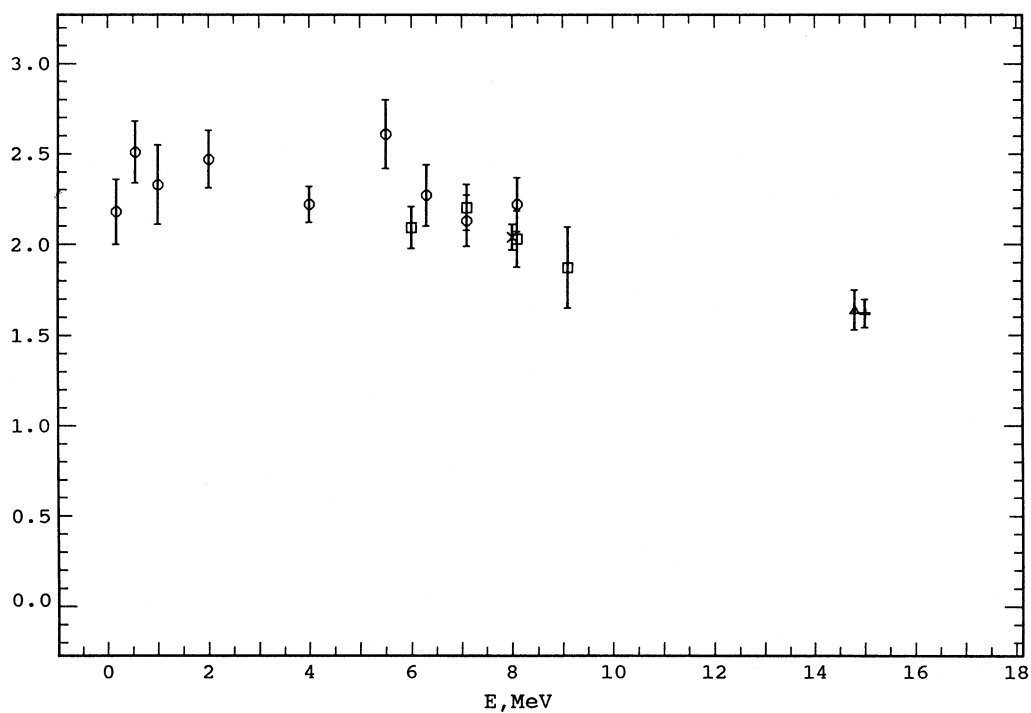


FIG. 5.1.12. Monoenergetic fission yields for  $^{147}\text{Nd}$  from  $^{235}\text{U}$  fission.

□ T.C. Chapman (LRL), ○ L.E. Glendenin (ANL), Δ D.R. Nethaway (LRL), + J. Laurec (FRBRC), × LI Ze (CAEP).

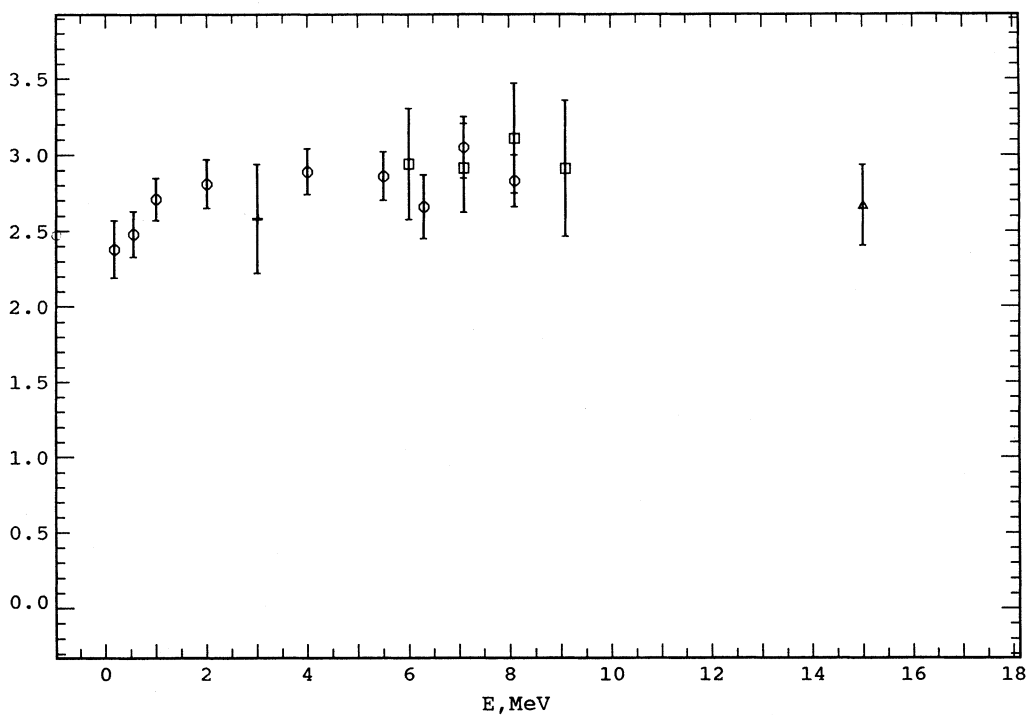


FIG. 5.1.13. Monoenergetic fission yields for  $^{87}\text{Kr}$  from  $^{235}\text{U}$  fission.

□ T.C. Chapman (LRL), ○ L.E. Glendenin (ANL), Δ N.E. Ballou (BNW), + A.N. Gudkov (MIF).

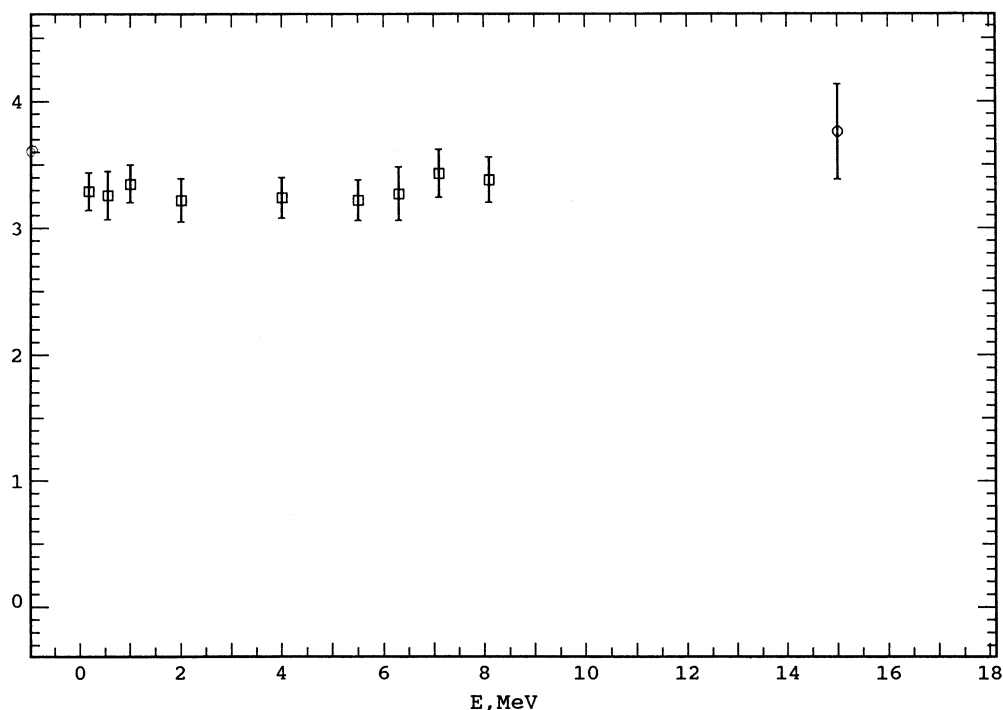


FIG. 5.1.14. Monoenergetic fission yields for  $^{88}\text{Kr}$  from  $^{235}\text{U}$  fission.

■ L.E. Glendenin (ANL), ○ N.E. Ballou (BNW).

### 5.1.3. Resonance (keV) region

Cunninghame et al. (Harwell) [5.1.21] measured the ratios of asymmetric to symmetric fission in p-wave neutron fission of  $^{235}\text{U}$  using radiochemical separation methods followed by  $\beta^-$  or  $\gamma$  counting. The monoenergetic neutrons were produced in a Van de Graaff accelerator by the  $\text{T}(\text{p},\text{n})$  reaction for  $E_n = 65$  keV and  $\text{Li}(\text{p},\text{n})$  reaction for  $E_n = 125\text{--}1000$  keV. 14 MeV neutrons were produced by the  $\text{T}(\text{d},\text{n})$  reaction using a Cockcroft-Walton generator. The results are listed in Table 5.1.3. It can be seen that the ratios of symmetric to asymmetric fission are abnormally larger at 65 and 125 keV than at thermal energy. This means that at around 100 keV, symmetric fission is abnormally smaller or asymmetric fission is abnormally larger than at thermal energy.

TABLE 5.1.3. RATIO OF ASYMMETRIC TO SYMMETRIC FISSION

Energy (keV)	thermal	65	125	200	300	540	1000	14000
$^{99}\text{Mo} / ^{113}\text{Ag}$	551	956	803	458	458	392	404	441
$^{97}\text{Zr} / ^{99}\text{Mo}$	0.974	1.08						
$^{97}\text{Zr} / ^{115}\text{Cd}$	625	809						

Cunninghame [5.1.22] also measured absolute yields of  $^{235}\text{U}$  fission induced by monoenergetic neutrons from 130 to 1700 keV with the same method as above. As shown in Fig. 5.1.10, the data for  $^{99}\text{Mo}$  are in agreement with Glendenin [5.1.10], and show a dip at  $E_n$

$\approx 500$  keV, which certainly deviates from the straight line. Similar features are also observed for other nuclides, for example  $^{147}\text{Nd}$  where the yields at neutron energies below 200 keV are lower, or  $^{140}\text{Ba}$ , where the yield of  $5.79 \pm 0.13$  at 300 keV is obviously lower than  $6.1 \pm 2.2$  at 130 keV and  $6.08 \pm 2.13$  at 700 keV. The differences exceed the experimental errors.

In summary, it can be concluded that the variation of fission yields in the keV region shows abnormal deviation from the linear dependence on energy, or even from a smooth change, but it resembles more the resonance behaviour of neutron cross sections. This is indeed caused by a reaction resonance mechanism, which is, however, very complicated for fission yields, and could not be described clearly by theory.

The discussions and conclusions in this paper are mainly for  $^{235}\text{U}$  neutron fission. However, the measurements of Ford et al. [5.1.9] and Glendenin et al. [5.1.10] show, that the fission yield behaves similarly for different incident particles and targets as long as they form the same compound nucleus and the latter has the same excitation energy. For example, the yield distributions of  $n+^{235}\text{U}$  fission at  $E_n = 10$  MeV and  $\alpha + ^{232}\text{Th}$  fission at  $E_\alpha = 21.5$  MeV are about the same; both have the compound nucleus  $^{236}\text{U}$  with an excitation energy of 16.5 MeV.

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## 5.2. THEORETICAL DESCRIPTION OF THE ENERGY DEPENDENCE

Traditionally, all fission product yield data are divided into three large groups of different incident neutron energies: thermal, fast and so-called high-energy (around 14–15 MeV). The differences in the main parameters of charge and mass distributions in these three groups are so high that the problem of the energy dependence has to be solved on a basis of physical assumptions. Real success in this approach is strongly influenced by the correct knowledge of the mechanism of nuclear fission and the statistical properties of the corresponding neutron-induced reaction. The potential energy surface landscape of the fissile nucleus plays very essential role in the formation of primary fragments which are transforming into fission products just after emission of prompt and delayed neutrons. Therefore it is necessary to perform as completely as possible the analysis of all of data about mass, kinetic energy and charge distributions of primary fragments in the fission of different fissioning nuclides and for a wide range of excitation energies to develop an adequate theoretical model for the precise prediction of product yields. One of the first characteristics which can be investigated is the fragment mass spectrum reflecting the potential barrier shape. This kind of work has been done at the Institute of Physics and Power Engineering, within the frame of this CRP from 1988 to 1996.

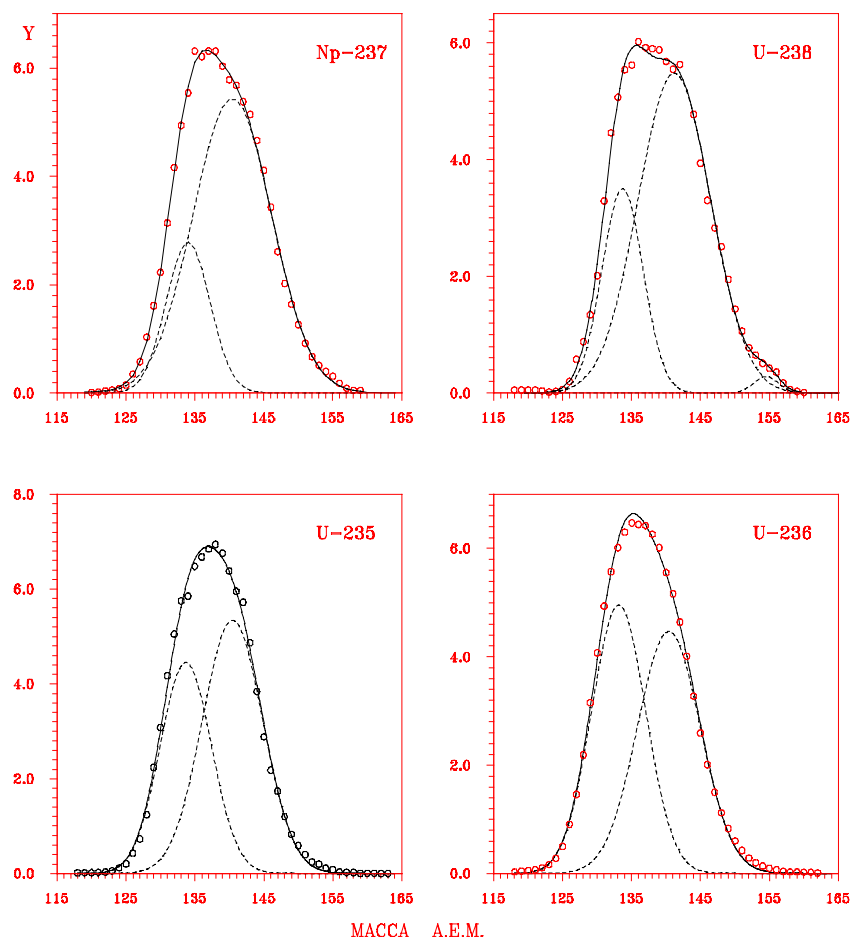


FIG. 5.2.1. Some fission fragment mass-spectra and their fits by Gaussian curves.

The main task of the experimental activity was to demonstrate the significance of the energy dependence of fission yields and to investigate the possibility of a prediction procedure. Systematic studies of fission fragment mass distributions were based on the idea by Berger [5.2.1] - Brosa [5.2.2] - Pashkevich [5.2.3] that the fission barrier has a multi-valley structure. Each valley corresponds to a separate path from the ground state of the fissioning nucleus to the scission point. Differences in the mass-asymmetry parameters give so-called standard mass components (see Fig. 5.2.1). One of them is the component Standard-I (S-I), determined by the closed double shells  $N = 82$ ,  $P = 50$ . The corresponding mass spectrum is centred around mass 134 of the heavy fragment. Another component is called Standard-II (S-II), which is centred around mass 140 for practically all fissile systems available for low excitation energies. The mass-symmetric component is named Superlong (SL). The population probability for all components will definitely fluctuate if each mass component (or mass channel) has a separate fission barrier. Of course this effect is most pronounced around the threshold area or in the resolved resonance region. In experiments at IPPE only threshold isotopes were investigated. The experimental setup and instrumentation were described in detail in [5.2.4] together with the measurement results. Here we shall only show the most important experimental data demonstrating a typical situation.

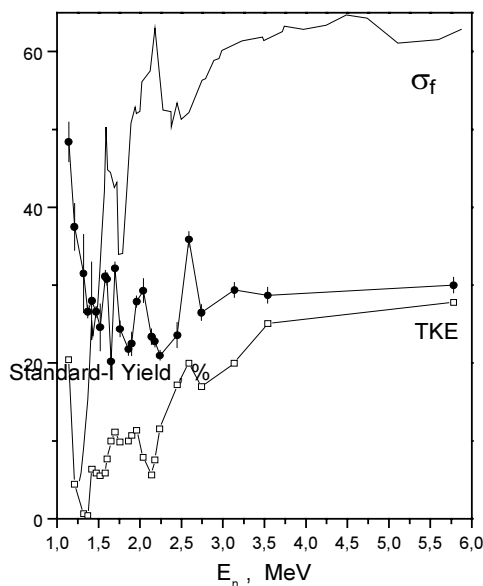


FIG. 5.2.2. Fluctuations of S-I component yield and total kinetic energy of  $^{232}\text{Th}$  fission fragments versus incident neutron energy.

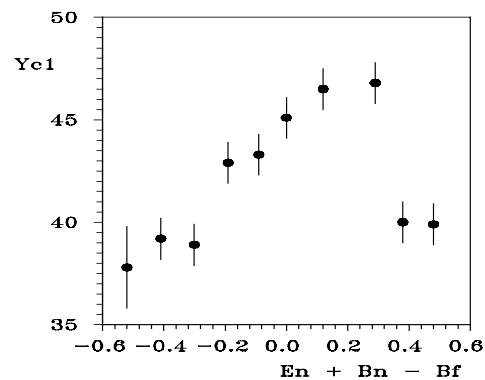


FIG. 5.2.3. S-I yield for fission fragment of  $^{237}\text{Np}$  as a function of excitation energy at the saddle point (relative to barrier top  $B_f$ ).

Figure 5.2.2 shows the energy dependence of the S-I component for the  $^{232}\text{Th}(n,f)$  reaction together with the total fission cross-section. The work was done with semiconductor detectors for forward emission angles. One can see very strong and pronounced fluctuations of S-I. The effect ranges from 50% to 20%. This means essentially a change of the total mass distribution of thorium fission fragments. The theoretical analysis showed that the fission barriers in both standard mass channels are different and a mass component around fragment mass  $m = 140$  amu (second standard mode) can be only realised via a rotational band with  $K = 1/2$  but Standard-I ( $m \cong 134$  amu) via both  $K = 1/2$  and  $K = 3/2$ . This conclusion opens a definite way for a fission yield evaluation where measurements are not possible. In this



approach first of all the fission barrier has to be calculated carefully. Our thorium data confirmed the data by Hambsch [5.2.5] for uranium-235 fission by resonance neutrons. The effects are very similar both qualitatively and quantitatively. Unfortunately, up to now nobody was able to calculate the barriers for different mass channels with satisfactory accuracy. To achieve this, detailed systematics of experimental data on S-I and S-II would be helpful.

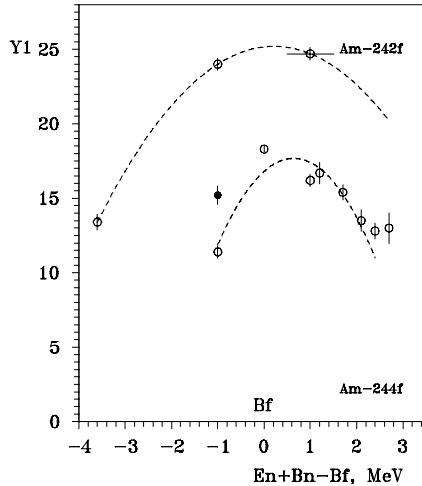


FIG. 5.2.4. Same as in fig.3 but for  $^{242}\text{Am}$  and  $^{244}\text{Am}$  fissile systems. All data were obtained with semiconductor detectors.

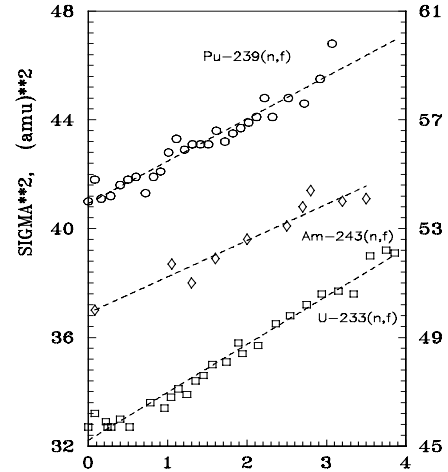


FIG. 5.2.5. Fission fragments mass-dispersions for  $^{233}\text{U}$ ,  $\square$   $^{239}\text{Pu}$  (O),  $^{243}\text{Am}$  ( $\diamond$ ) plus neutron fissile systems.

Another example of the energy dependence can be presented using the data for double-odd nuclei with fission threshold like  $^{238}\text{Np}$  and  $^{244}\text{Am}$ . The neptunium data are presented in Fig. 5.2.3 analogous to those of thorium. The independent variable is the excitation energy of the nucleus relative to the common observable fission barrier top. One can see that coming from a subbarrier region, the contribution of the S-I channel increases first and then decreases again. Practically the same is seen in Fig. 5.2.4 for americium fission [5.2.4]. In the last two cases sharp fluctuations of the S-I component are missing compared to  $^{232}\text{Th}$  fission or resonance neutron induced fission of  $^{235}\text{U}$ . However, the nature of the apparent correlation between S-I and the fission threshold is the same — differences in fission barriers correspond to different mass-asymmetric fission valleys. That means that the barrier parameters have to be calculated or evaluated from systematics before prediction or evaluation of fission yield data.

Fragment mass distributions will be completely determined by two sets of parameters describing both total and partial population probabilities of S-I and S-II channels and their internal properties. The suggested Gaussian shape of the mass components means that the dispersion should be known for all mass channels as a function of excitation energy and nuclear composition of fissioning system. Some obvious systematics of  $\sigma_m^2$  for selected nuclides as a function of neutron energy is shown in Fig. 5.2.5. In all cases,  $\sigma_m^2(E_n)$  can be correctly approximated by a linear function in a relatively wide range of excitation energies. Slopes of  $\sigma_m^2(E_n)$  decrease rapidly with the fissility parameter from 4 (amu) $^2$ /MeV for  $^{232}\text{Th}$  to 1.5 (amu) $^2$ /MeV for californium. Anyway, sufficient experimental data are available at present to predict  $\sigma_m^2$  values for any nucleus.

Assuming that in the process of mass equilibrium the corresponding mode is commonly described by a harmonic oscillator having a phonon energy  $\hbar\omega$  one can write following equation for asymmetric mass distribution variance  $\sigma_m^2$ :

$$\sigma_m^2 = (\hbar\omega/2 k_m) \coth(\hbar\omega/2\theta)$$

Where  $\omega = k_m/B_m$  ( $k_m$  = stiffness coefficient of the mass equilibrium mode,  $B_m$  = effective inertia) and  $\theta$  = nuclear temperature. The last quantity determines the incident neutron energy dependence of the fission fragment mass curve width. One example of the calculations of  $\sigma_m^2$  for  $^{233}\text{U}$  is presented in Fig. 5.2.6 with different assumptions about the dissipation energy in fission (see figure caption). Details of the calculations and a review of experimental results can be found in [5.2.6]. Of course, analogous calculations should be done for separate mass components. This kind of work has really to be done.

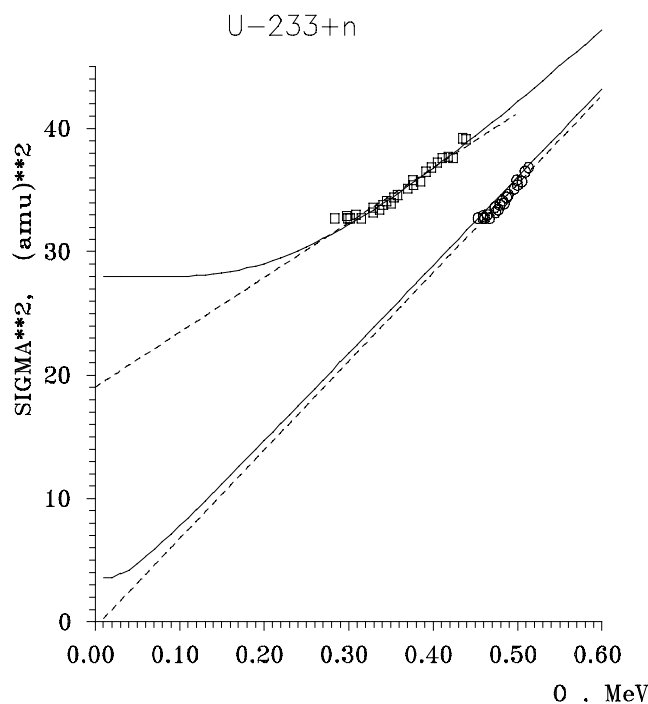


FIG. 5.2.6. Temperature  $\theta$  dependence of mass dispersions for  $^{233}\text{U}$  with two assumptions: top - dissipation energy  $E_{\text{DISS}} = 0$ ; bottom - dissipation energy  $E_{\text{DISS}} = 5 \text{ MeV}$ . dashed curve - high temperature approximation.

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## Chapter 6

### EVALUATION OF EXPERIMENTAL FISSION YIELDS

#### 6.1. CORRELATIONS OF EXPERIMENTAL YIELDS

For the last two decades, with the development of the application of computer techniques in nuclear reactor physics, correlations of data and the introduction of covariance matrices has become more and more important for the analysis of the results from various reactor benchmark measurements and sensitivity studies. During this period there has also been substantial growth in the development and application of covariance methods in other areas of nuclear data research.

As is well known, the data, especially experimental data must be given with their errors or uncertainties to describe their accuracy. But often enough only an unspecified “error” or a total error is given without any specification of its “systematic” or “statistical” nature, or is no information on correlations given. From the application point of view, sometimes “systematic” errors are more important, because, as opposed to statistical ones they are not random and influence the result only in one direction. As far as evaluators and experimenters are concerned, complete data information is given only when both types of errors and their covariance are given.

The covariance matrix  $V_{xy}$  is defined as

$$\text{Cov}(x,y) \equiv \langle (x - \langle x \rangle)(y - \langle y \rangle) \rangle = \langle xy \rangle - \langle x \rangle \langle y \rangle \quad (1)$$

where  $x,y$  are continuous random variables, and  $\langle x \rangle, \langle y \rangle$  are their mean values. The correlation coefficient is defined as

$$\rho(x,y) \equiv \frac{\text{Cov}(x,y)}{\sqrt{\text{Var}(x) \text{Var}(y)}}, \quad (2)$$

where

$$\text{Var}(x) \equiv \langle (x - \langle x \rangle)^2 \rangle = \langle x^2 \rangle - \langle x \rangle^2 \quad (3)$$

is the variance and, obviously,  $-1 \leq \rho \leq +1$ .

A covariance matrix must be symmetric, positive definite, of full rank, and have positive eigenvalues. In physics, this means that the information content is consistent and not redundant.

##### 6.1.1. Error analysis and correlation of experimental fission yield data

Correlations are induced by systematic errors in experimental measurements that are not independent. For constructing the covariance matrix, not only total, but also the systematic errors have to be determined, although this may sometimes be difficult.

For illustration we give some typical examples of systematic and statistical errors and how to determine them in fission yield measurements employing different methods.

The first one is for the  $\gamma$  spectrometric method, which is widely used, taken from Li's paper [6.1.1]. The measurements were performed with a Ge(Li) detector for direct  $\gamma$  counting of fission products and an ionization chamber for the determination of the fission rate. Corrections for neutron transmission rate and background,  $\gamma$  ray self-absorption and sample geometry were made. The sources of error are listed in Table 6.1.1.

TABLE 6.1.1. UNCERTAINTY SOURCES OF ABSOLUTE FISSION YIELD MEASUREMENT WITH THE  $\gamma$  spectroMETRIC METHOD

<i>error sources</i>	<i>values (%)</i>	<i>remarks</i>
a. statistical (random) $\Delta y_{\text{sta}}$	0.3-2.1	depends on measuring conditions <sup>a</sup>
b. fission rate	1.5	
c. detector efficiency calibration	2.0	depends on $\gamma$ ray energy
d. geometry correction	0.3	
e. $\gamma$ self-absorption correction	0.3-0.4	
f. neutron transmission correction	0.4	(for thermal neutrons)
g. neutron background correction	0.6-1.0	(for high energy neutrons)
total systematic error	2.6-2.7	$\Delta y_{\text{sys}} = \sqrt{\sum_i \Delta y_{\text{sys}_i}^2}$
total error	2.6-3.4	$\Delta y_{\text{tot}} = \sqrt{\Delta y_{\text{sys}}^2 + \Delta y_{\text{sta}}^2}$

<sup>a</sup> counting statistics, peak-to-background ratio, measuring time, fission products investigated, etc.

The next example for the radiochemical method, is taken from [6.1.2] (fission yield measurement group of the CIAE). The irradiated samples were first radiochemically separated, followed by  $\gamma$  or  $\beta$  counting of different fission products. The number of fissions was measured with a back to back double ionization chamber. The uncertainty sources are listed in Table 6.1.2.

TABLE 6.1.2. UNCERTAINTY SOURCES OF ABSOLUTE FISSION YIELD MEASUREMENT WITH THE RADIOCHEMICAL METHOD

<i>error sources</i>	<i>values (%)</i>	<i>remarks</i>
a. statistical (random) $\Delta y_{\text{sta}}$	0.3-2	depends on measuring conditions <sup>a</sup>
b. sample quantification	0.1	
c. fission rate	1.5	
d. detector efficiency	1.5-3.3	depend on energy
e. chemical yield		negligible
total systematic error	2.1-3.6	$\Delta y_{\text{sys}} = \sqrt{\sum_i \Delta y_{\text{sys}_i}^2}$
total error	2.7-7.0	$\Delta y_{\text{tot}} = \Delta y_{\text{sys}} + 2\Delta y_{\text{sta}}$

<sup>a</sup> counting statistics, peak-to-background ratio, measuring time, fission products investigated, etc.

The total and systematic errors need to be analyzed and determined carefully according to the individual experimental conditions. This can and should be done by the measurers as they know the experimental conditions in detail. Evaluators, if not provided with that information, can make some general assumptions on the errors from a survey of measurement methods and the inherent error limits associated with them (see Chapter 2). For example, in the fission yield evaluation for ENDF/B-6 [6.1.3], the following error limits were taken: not less than 0.5% for mass spectrometric measurements, 10% for radiochemical measurements made after 1955 and 20% before 1955. In the file, the errors of chain yields in the peak regions are about 1% for the well measured fissionable nuclides, and about 14% for the fissionable nuclides with less measurements. The chain yields in the wings and the valley are less well known, and the errors range from about 8% to 30%. The errors of the most accurate yields of  $^{235}\text{U}$  thermal fission products are around 0.5%, because they are averages of large numbers of measurements rather than due to abnormally small errors assigned to individual measurements. The average error of individual (more accurate) measurements is about 3%.

In the evaluation of CENDL-FY [6.1.4], the error limits are taken as:

- 20% for Geiger-counter era measurements before 1955;
- 10% for NaI(Tl)-detector era measurements between 1955 and 1965;
- 5% for direct  $\gamma$  ray spectrometry in the Ge(Li)-detector era, radiochemical, and other special measurements made since 1965;
- 2% for mass spectrometric measurements;
- 30% for estimated values.

Systematic errors in fission yield measurements include errors from sample quantification, determination of the number of fissions (fission rate measurement or neutron flux measurement plus fission cross section used), detector efficiency calibration, and various corrections, such as decay (data used),  $\gamma$  ray self-absorption, sample geometry, neutron transition etc. With present equipment, the total systematic errors of more accurate absolute cumulative fission yield measurements with  $\gamma$  spectrometry is generally about 2–3% (but not smaller than 2%).

Special attention should be paid to two features of systematic errors, which are distinct from those of statistical errors:

Firstly they cannot be reduced by increasing the measurement time under the same experimental condition. This means that, for example, if the systematic error of an experiment in one laboratory is 2%, the total measurement error cannot be smaller than 2%, no matter how many times the measurements are repeated, and how many individual results are averaged. If measurements are made in different laboratories, but some experimental conditions (e.g. detector or sample) are the same, or the same (nuclear) data (for decay, activation, or fission) are used for the analysis of results, the corresponding systematic error can also not be reduced. This has to be taken into account in the data processing and evaluation, as otherwise the errors of the (recommended) average values would be smaller than the inherent systematic errors. For example when a fission yield is determined in several experiments by the measurement of a  $\gamma$  line with its emission probability known to 2%, then the (evaluated) average of these measurements cannot have a total error less than 2%.

Secondly, systematic errors common to fission yield data (e.g. at different energy points or for different fission products) measured at the same or even different laboratories will result

in correlations among the measured data. The data are not correlated only if the systematic errors in measurements are independent. Correlations are introduced through use of the same experimental conditions, samples, detectors, methods, standards and/or nuclear data in corrections and analysis of results. In the example from above, the measurements using the same  $\gamma$  line data would be correlated, even if performed at different laboratories.

### 6.1.2. Construction of the covariance matrix for experimental fission yield data

The covariance matrix of experimental fission yield data can be constructed using so-called parameter analysis method [6.1.5, 6.1.6].

Suppose the fission yield  $Y$  can be expressed as a function

$$Y = f(x_1, x_2, \dots, x_N), \quad (4)$$

where  $x_1, x_2, \dots, x_N$  are the parameters, which can be directly measured, then

$$\text{Cov}(Y_i, Y_j) = \sum_k \sum_{k'} \frac{\partial f}{\partial x_k} \bigg|_i \frac{\partial f}{\partial x_{k'}} \bigg|_j \rho_{ij}^{kk'} \sigma_{ki} \sigma_{k'j} = \sum_k \frac{\partial f}{\partial x_k} \bigg|_i \frac{\partial f}{\partial x_k} \bigg|_j \rho_{ij}^k \sigma_{ki} \sigma_{kj} \quad (5)$$

$$(\text{Suppose } \rho_{ij}^{kk'} = 0 \text{ for } k \neq k'),$$

where  $i, j$  express the different measurement points (e.g. energies or fission products),  
 $k$  expresses the different parameters,

$\frac{\partial f}{\partial x_k} \bigg|_i$  is the (partial) derivative of function  $f$  with respect to parameter  $x_k$ , at point  $i$ ,  
 $\sigma_{ki}$  is the standard error of the  $k$ -th parameter  $x_k$  at  $i$ ,  
 $\rho_{ij}^{kk'}$  is the correlation coefficient between parameter  $x_k$  at point  $i$  and  $x_{k'}$  at point  $j$ ,  
 $\rho_{ij}^k$  is the correlation coefficient of the parameter  $x_k$  at points  $i$  and  $j$ .

It can be seen from the equations that the covariance matrix can be constructed for the fission yield data, if the function (4) as well as the errors and correlation coefficients of the respective directly measured parameters in (5) are known.

The derivatives  $\frac{\partial f}{\partial x_k} \bigg|_i$ , which form the so called sensitive matrix, can be calculated analytically or numerically from the function (4), and the standard errors of the directly measured parameters can be determined according to the experimental conditions, as given above for the general case. The correlation coefficients  $\rho_{ij}^{kk'}$  can be determined as discussed above. For the general case, they are listed in Table 6.1.3.

TABLE 6.1.3. CORRELATION COEFFICIENTS OF DIRECTLY MEASURABLE PARAMETERS IN FISSION YIELD MEASUREMENTS

Parameters	$\rho_{ij}^k$	Remark
counting statistics	0	random (uncorrelated)
standard yield	1.0	same standard used
fission rate	1.0	same run
sample quantification	1.0	same sample used
chemical yield	1.0	same run
detector efficiency	1.0/0.5	same/different energy
$\gamma$ self absorption correction	1.0/0.5	same/different $\gamma$ -energy
sample geometry correction	1.0/0.5	same/different $\gamma$ -energy
neutron flux correction	1.0	same run
neutron background correction	1.0	same run

### 6.1.3. Processing of correlated fission yield data

Traditional data processing, that employs weighted averages, curve fitting, error propagation etc., is based on the assumption that the data considered are independent. This method cannot, however, be applied to correlated data, which are not independent

#### 6.1.3.1. Average of correlated data

When evaluating a set of measured fission yield data  $x_1, x_2, \dots, x_N$  which are correlated, we can express the data set as vector  $\mathbf{Y}$ , and its covariance matrix  $\mathbf{V}_y$  can be constructed using the method discussed above. In this case, the mean  $\bar{y}$  and its error  $\Delta\bar{y}$  are:

$$\left. \begin{aligned} \bar{y} &= (\mathbf{e}^* \mathbf{V}_y^{-1} \mathbf{e})^{-1} (\mathbf{e}^* \mathbf{V}_y^{-1} \mathbf{Y}) \\ \Delta\bar{y} &= \left[ (\mathbf{e}^* \mathbf{V}_y \mathbf{e})^{-1} \right]^{\frac{1}{2}} \end{aligned} \right\} \quad (6)$$

where  $\mathbf{e}$  is a unit vector:  $\mathbf{e}^* = (1, 1, \dots, 1)$  and  $\mathbf{e} \times \mathbf{e}^* = 1$ .

If the correlation coefficients are all zero, i.e. the data are independent, the equations become

$$\left. \begin{aligned} \bar{y} &= \sum_i \mathbf{V}_{ii}^{-1} y_i / \sum_i \mathbf{V}_{ii}^{-1} \\ \Delta y &= 1 / \sum_i \mathbf{V}_{ii}^{-1} \end{aligned} \right\} \quad (7)$$

these are just the usual weighted average calculation formulae. In the two dimensional case, the formulae become:

$$\left. \begin{aligned} y &= (\Delta y_2^2 - \rho_{12} \Delta y_1 \Delta y_2) y_1 + (\Delta y_1^2 - \rho_{12} \Delta y_1 \Delta y_2) / \Delta y_1^2 + \Delta y_2^2 - 2\rho_{12} \Delta y_1 \Delta y_2 \\ \Delta y &= \Delta y_1^2 \Delta y_2^2 (1 - \rho_{12}^2) / \Delta y_1^2 + \Delta y_2^2 - 2\rho_{12} \Delta y_1 \Delta y_2 \end{aligned} \right\} \quad (8)$$

### 6.1.3.2. Simultaneous evaluation

One can distinguish between absolute (where ultimately the number of fissions and the number of fission product nuclei are determined) and relative (to a reference yield) fission yield measurements. Relative yields can be measured as ratio Ro or R-value Rv:

$$\left. \begin{aligned} Ro &= Y / Y_s \\ Rv &= (Y / Y_s)_E / (Y / Y_s)_T \end{aligned} \right\} \quad (9)$$

where Y is the yield to be measured, and subscripts S, E, T denote standard, energy E and thermal energy respectively. Y can be deduced from Ro or Rv using existing “standard” or rather “reference” yields. But the “reference” yields are not necessarily absolute standards, they may be measured in the same or another experiment.

To obtain reliable, consistent reference fission yields and their ratios or R-values, it is recommended to collect all absolutely measured data  $Y_i$  ( $i = 1, \dots, N$ ), their ratios  $Ro_{ij}$  or R-values  $Rv_{ij}$ , and evaluate them simultaneously. It does not matter whether the selected data are correlated or independent.

D.W. Muir has developed a method and the code **ZOTT** [6.1.7] for the simultaneous evaluation of correlated data. It uses partitioned correlated linear least squares analysis and is particularly appropriate for the updating of an existing data set, taking into account a relatively small number of supplementary measurements of these data themselves or of certain functions of the data, and make the matrix inversion reduced relative to solving the normal equations, while still retaining the minimum-variance guarantees.

We have used this method and adapted the code **ZOTT** for the evaluation of fission yield data. The adjusted optimum values are calculated employing the condition of minimum variance deviation, taking into account directly (differential) as well as indirectly (integral) measured data :

$$\left. \begin{aligned} \mathbf{a}' &= \mathbf{a} - \text{Cov}(\mathbf{a}, \mathbf{p}) \mathbf{G} \mathbf{P} \\ \mathbf{D}(\mathbf{a}') &= \mathbf{D}(\mathbf{a}) - \text{Cov}(\mathbf{a}, \mathbf{p}) \mathbf{G} \text{Cov}(\mathbf{p}, \mathbf{a}) \\ \text{Cov}(\mathbf{a}, \mathbf{p}) &= \text{Cov}(\mathbf{a}, \mathbf{b}) - \mathbf{D}(\mathbf{a}) \mathbf{R}^* \end{aligned} \right\} \quad (10)$$

where  $\mathbf{a}$  directly measured or differential quantities

$\mathbf{a}'$  adjusted or improved  $\mathbf{a}$

$\mathbf{p} = \mathbf{b} - \mathbf{R}\mathbf{a}$ , difference vector

$\mathbf{b}$  indirectly measured or integral quantities

$\mathbf{R}$  sensitivity or derivative matrix of  $\mathbf{b}$  to  $\mathbf{a}$

$\mathbf{G}^{-1} = \mathbf{D}(\mathbf{P}) = \text{Cov}(\mathbf{P}, \mathbf{P}) = \mathbf{D}(\mathbf{b}) - \text{Cov}(\mathbf{b}, \mathbf{a}) \mathbf{R}^* - \mathbf{R} \text{Cov}(\mathbf{a}, \mathbf{b}) + \mathbf{R} \mathbf{D}(\mathbf{a}) \mathbf{R}^*$

For application of the method and code, the key point is to construct the sensitivity matrix  $\mathbf{R}$ , which defines the relation between differential and integral quantities.  $\mathbf{R}$  is a matrix of  $m$  rows  $\times$   $n$  columns. Here  $n$  is the number of directly measured or differential quantities  $\mathbf{a}$ , i.e. “fundamental” quantities to be adjusted,  $m$  is the number of indirectly measured or integral quantities  $\mathbf{b}$ , the number of “duplicate” quantities.



To make the use of the code **ZOTT** more convenient for fission yield evaluation, some modifications have been made to the input data format [6.1.8]. Using the code, reasonable results have been obtained in the practical fission yield evaluation. This is illustrated by an example: The weighted means of thermal yields from  $^{235}\text{U}(\text{n},\text{f})$  are  $6.4475 \pm 0.0811$ ,  $6.1452 \pm 0.0679$ ,  $5.4388 \pm 0.1484$  for products  $^{95}\text{Zr}$ ,  $^{99}\text{Mo}$  and  $^{144}\text{Ce}$  respectively, the weighted means of the ratios are  $R(\text{Zr}/\text{Mo}) = 1.0540 \pm 0.0062$ ,  $R(\text{Zr}/\text{Ce}) = 1.2182 \pm 0.0107$ . For these data, the combined vector is

$$\mathbf{a}^* = 6.4475 \ 6.1452 \ 5.4388 \ 1.0540 \ 1.2182$$

its covariance matrix is

$$\mathbf{D}(\mathbf{a}) = \begin{pmatrix} 6.5772E-3 & & & & \\ & 4.6104E-3 & & & 0 \\ & & 2.2023E-2 & & \\ & 0 & & 3.8813E-5 & \\ & & & & 1.1449E-4 \end{pmatrix}$$

and the sensitivity matrix is

$$\mathbf{R} = \begin{pmatrix} 1 & -1 & 0 \\ 1 & 0 & -1 \end{pmatrix},$$

the adjusted vector and its error is:

$$\mathbf{a}'^* = 6.4764 \pm 0.0549 \ 6.1448 \pm 0.0516 \ 5.3278 \pm 0.0605 \ 1.0540 \pm 0.0058 \ 1.2156 \pm 0.0102$$

and the correlation coefficient matrix  $V_{\text{coe}}$  of vector  $\mathbf{a}'$  is:

$$\begin{array}{rcllclclcl} 1.000000E & + & 00 & 7.844954E & - & 01 & 6.766660E & - & 01 & 3.403997E & - & 01 & 9.480542E & - & 02 \\ 7.844954E & - & 01 & 1.000000E & + & 00 & 5.308412E & - & 01 & -3.160594E & - & 01 & 7.437438E & - & 02 \\ 6.766660E & - & 01 & 5.308412E & - & 01 & 1.000000E & + & 00 & 2.303368E & - & 01 & -6.688230E & - & 01 \\ 3.403997E & - & 01 & -3.160594E & - & 01 & 2.303368E & - & 01 & 1.000000E & + & 00 & 3.227172E & - & 02 \\ 9.480542E & - & 02 & 7.437438E & - & 02 & -6.688230E & - & 01 & 3.227172E & - & 02 & 1.000000E & + & 00 \end{array}$$

It can be seen that the correlation coefficients between adjusted thermal yields of Zr and Mo, Zr and Ce, Mo and Ce are 0.784, 0.677, 0.531 respectively. They are quite large, although the respective yields are independent before the adjustment. This is not due to a correlation between measurements, but introduced in the simultaneous evaluation process, where they affect each other via the included ratios.

Another noticeable feature is that some correlation coefficients are negative. They correspond to the correlation between the fission yield of  $^{99}\text{Mo}$  and the ratio  $R(\text{Zr}/\text{Mo})$ , as well as the fission yield of  $^{144}\text{Ce}$  and the ratio  $R(\text{Zr}/\text{Ce})$ . This is reasonable, as the ratios  $R(\text{Zr}/\text{Mo})$  and  $R(\text{Zr}/\text{Ce})$  become smaller with increasing Mo and Ce yields respectively.

A special application of the method and the ZOTT code is the calculation of the weighted mean of the correlated data. In this case one has to choose the sensitivity matrix as:

$$\mathbf{R}^* = (1 \ 1 \ \dots \ 1)$$

with as many '1' as there are input data.

We tested this application with the following example: below are four values of the  $^{99}\text{Mo}$  yield from  $^{235}\text{U}$  thermal fission measured at different laboratories:

$$6.16 \pm 0.12, \ 6.17 \pm 0.12, \ 6.14 \pm 0.16, \ 6.08 \pm 0.16,$$

Using the ZOTT code with the sensitivity matrix  $\mathbf{R}^* = (1 \ 1 \ 1 \ 1)$ , one gets for all calculated adjusted yields the same value  $6.1452 \pm 0.0679$ , which is equivalent to and is the same value as one gets for the simple weighted average calculated with the code **AVERAG** [6.1.9]. Also the calculated correlation coefficients of the output quantities are all 1, which is reasonable, as in this case, the input data and adjusted data are all the same physical quantity.

#### 6.1.3.3. Curve Fitting of Correlated Data

In the evaluation of experimental fission yields, the dependence of yields on neutron energy is an important topic to be studied. It was found that the dependence on energy is linear for some products, for others not. In the latter cases, the data need to be fitted with general curve fitting methods based on the least squares method. If the experimental data are correlated, as pointed out above, traditional curve fitting methods and programs cannot be used. In this case, the method and program **SPC**, developed by Liu et al. [6.1.10], can be applied.

**SPC** is a general spline fit program for multiple sets of correlated data. The fitting equation written in matrix form is

$$\mathbf{B}^*(\mathbf{W}-\mathbf{U}^*\mathbf{D}\mathbf{U})\mathbf{B}\mathbf{C}=\mathbf{B}^*(\mathbf{W}-\mathbf{U}^*\mathbf{D}\mathbf{U})\mathbf{Y}$$

where  $\mathbf{Y}$  is input data vector

$\mathbf{W}=\mathbf{V}_Y^{-1}$ , weight matrix,  $\mathbf{V}_Y$  covariance matrix of vector  $\mathbf{Y}$

$\mathbf{B}$  base spline matrix, defined by knots and order

$\mathbf{D}$  width matrix

$\mathbf{U}$  sum weight matrix for each set of data

$\mathbf{C}$  coefficient vector to be solved and the fit spline function  $\mathbf{S} = \mathbf{B}\mathbf{C}$ .

In the program, the knots of spline can be optimized automatically (one only needs to input the original knots), the order of spline base can be chosen according to the curve shape. The correlations among different data points within each set of data are taken into account (but it is supposed that there are no correlations among different data sets). Some methods are offered to deal with the so called PPP problem [6.1.11], which could occur in the correlative

data processing. The output of the fitted values and their covariance matrix are in the ENDF-6 format.

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## 6.2. AUXILIARY DATA FOR FISSION PRODUCT YIELD EVALUATIONS

### 6.2.1. Reanalysis of experimental measurements

In the study of discrepancies in the experimentally measured yields it is always best to go back to the original source of the data. Although such studies are unlikely to identify problems, occasionally the nuclear data used in the analysis of the measurement may have significantly changed. Some possible data types used for such a re-analysis are half-lives (determining fission product activity), gamma ray emission probabilities (determining photon production) and cross-sections (determining fission rates).

### 6.2.2. Parameters of fission product yield models

All fission product yield models depend upon other types of nuclear data. Chain yield models require knowledge of the average prompt neutrons emitted per fission. Fractional independent yield models require knowledge of the average prompt neutrons emitted per fission for each product mass. Models of isomeric splitting of yields require knowledge of the quantum states of both the fissioning compound nucleus and the fission products.

### 6.2.3. Adjustment of fission product yields

As described in Chapter 7, all modern yield evaluations use the physical conservation laws to adjust model fitting and evaluated data to agree with the well understood physical laws governing the fission process. Each evaluation team uses the conservation laws in different ways, by constraining mathematical fitting procedures and/or adjustment of the resultant data. The techniques are described more fully, for example, in Refs [6.2.1–6.2.6].

One principle conservation law used for these procedures is the constraint that nucleons are not created or destroyed. This requires knowledge of the neutron emission arising from the fission process to correct the nucleon balance after fission.

#### 6.2.4. Production of cumulative yield libraries

In producing cumulative yield libraries from the evaluated and adjusted independent yields it is necessary to use decay data (this is described in detail in Section 7.2.2.8). A complete description of the routes of decay and their branching is required for all fission products to produce accurate cumulative yield libraries.

#### 6.2.5. Testing of fission product yield evaluations

In testing fission product yield evaluations it is useful to consider methods that globally test the data against experiments, where the experiments and data calculations depend on a minimum of other nuclear data and mathematical approximations. Two such tests are the calculation of delayed neutron emission and the calculation of decay heat.

The total delayed neutron emission per fission can be calculated for a fissioning species as the product of the cumulative yield  $c(A,Z,I)$  and the delayed neutron branching fraction  $P_n$ , for each fission product  $(A,Z,I)$  summed over all delayed neutron emitting fission products. As well as calculating total delayed neutron emission, it is possible by using independent yields and decay data within inventory codes (eg. FISPIN and CINDER) to calculate delayed neutron emission as a function of time following either a fission pulse or a period of constant fission rate. Both types of calculations are described in Refs [6.2.6, 6.2.7].

The calculation of decay heat is another useful test of fission product yield evaluations as accurate measurements exist which require little nuclear data in their analysis, and the calculations are similar to those for delayed neutron emission. The only extra data required is the total energy of emitted particles per decay for gamma rays, electron and heavy particles (alpha particles and neutrons). From these data both time averaged and time dependent calculations can be carried out. An example of time dependent calculations is described in [6.2.6, 6.2.8].

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### 6.3. REFERENCE FISSION YIELDS

#### 6.3.1. Definition

Certain fission products that are easy to measure and/or have reliable fission yields are taken as internal reference nuclides in relative yield and R-value measurements, or used as monitors in nuclear industry. The fission yields (generally cumulative yields) of such fission products are referred to as reference yields.  $^{235}\text{U}$  thermal fission is an important reference fission reaction, and often the whole set of fission yields is used as reference.

#### 6.3.2. Application of reference fission yields

In fission yield evaluation and measurement, reliable and accurate reference yields are important as they determine the quality of recommended or measured yields. The fission products commonly used as internal reference nuclides in fission yield evaluation and measurement are shown in Table 6.3.1.

In nuclear industry, the yields of monitor fission products play an important role. For example, in reactor physics, monitor fission products are widely used for the determination of decay heat, burnup, loss of reactivity per cycle, and the energy release due to fission, in transmutation studies, shielding calculations, in neutron dosimetry, fuel handling and waste management, and in nuclear safety studies, etc. In these applications, about 60 monitor fission products are used, with half-lives ranging from very short to long, as can be seen in Table 6.3.2, and their yields are required.

Recently, CNDC made an intercomparison for the more important reference fission yields contained in the major data libraries (ENDF/B-VI, JENDL-3/FPY, JEF-2/FPY, CENDL/FPY, and BROND-2). Unfortunately that intercomparison showed that even the cumulative yields of  $^{235}\text{U}$  thermal fission are discrepant for about two thirds of all the yields compared (about 40 product nuclides for which experimental data were available have been compared as mentioned in Section 7.3, the description of the CENDL fission yield library). Typical examples are Kr, Pm, Sm, etc., which are included in Table 6.3.1. This means that presently the accuracies of the reference or monitor yields contained in these libraries are not satisfactory for practical applications. Table 6.3.3. shows that the discrepancies among these libraries for the reference fission yields of the most important monitor products are larger than the requested accuracy for reactor physics applications. Obviously, further evaluation efforts and measurements are required for the reference fission yields.

TABLE 6.3.1. FISSION PRODUCTS WHOSE FISSION YIELDS ARE USED AS REFERENCE

Nuclide	Half-life	Nuclide	Half-life	Nuclide	Half-life
<sup>83g</sup> Kr	stable	<sup>85m</sup> Kr	4.480 h	<sup>85g</sup> Kr	10.72 y
<sup>86</sup> Kr	stable	<sup>87</sup> Kr	76.3 m	<sup>88</sup> Kr	2.84 h
<sup>89</sup> Sr	50.55 d	<sup>90</sup> Sr	28.6 y	<sup>91g</sup> Y	58.51 d
<sup>95</sup> Zr	64.02d	<sup>95m</sup> Nb	3.61 d	<sup>95g</sup> Nb	34.97 d
<sup>95</sup> Mo	stable	<sup>96</sup> Zr	stable	<sup>99</sup> Mo	66.0 h
<sup>100</sup> Mo	stable	<sup>101</sup> Ru	stable	<sup>102</sup> Ru	stable
<sup>103</sup> Ru	39.26 d	<sup>105g</sup> Rh	35.36 h	<sup>106</sup> Ru	371.63 d
<sup>106g</sup> Rh	29.80 s	<sup>111g</sup> Ag	7.45 d	<sup>115m</sup> Cd	44.6 d
<sup>115g</sup> Cd	53.46 h	<sup>125</sup> Sb	2.73 y	<sup>131</sup> I	8.04 d
<sup>131m</sup> Xe	11.9 h	<sup>131g</sup> Xe	stable	<sup>132</sup> Te	78.2 h
<sup>132</sup> Xe	stable	<sup>133m</sup> Xe	2.188 d	<sup>133g</sup> Xe	5.245 d
<sup>133</sup> Cs	stable	<sup>134g</sup> Xe	stable	<sup>134g</sup> Cs	2.062 y
<sup>135</sup> I	6.61 h	<sup>135m</sup> Xe	15.29 m	<sup>135g</sup> Xe	9.09 h
<sup>136</sup> Xe	stable	<sup>136</sup> Cs	13.16 d	<sup>137</sup> Xe	3.818 m
<sup>137</sup> Cs	30.17 y	<sup>138</sup> Xe	14.08 m	<sup>140</sup> Ba	12.746 d
<sup>140</sup> La	40.272 h	<sup>141</sup> Ce	32.501 d	<sup>141</sup> Pr	stable
<sup>143</sup> Ce	33.0 h	<sup>143</sup> Nd	stable	<sup>144</sup> Ce	284.4 d
<sup>144g</sup> Pr	17.28 m	<sup>144</sup> Nd	stable	<sup>145</sup> Nd	stable
<sup>146</sup> Nd	stable	<sup>147</sup> Nd	10.98 d	<sup>147</sup> Pm	2.6234 y
<sup>148</sup> Nd	stable	<sup>148m</sup> Pm	41.29 d	<sup>148g</sup> Pm	5.370 d
<sup>149</sup> Pm	53.08 h	<sup>149</sup> Sm	stable	<sup>151</sup> Pm	28.40 h
<sup>151</sup> Sm	90 y	<sup>153</sup> Sm	46.7 h	<sup>153</sup> Eu	stable
<sup>154g</sup> Eu	8.8 y	<sup>155</sup> Eu	4.96 y	<sup>156</sup> Eu	15.19 d
<sup>161</sup> Tb	6.90 d				

TABLE 6.3.2. MONITOR FISSION PRODUCT YIELDS REQUIRED FOR REACTOR PHYSICS APPLICATIONS

Nuclide	BU PWR <sup>a</sup>	BU FBR <sup>b</sup>	Monitoring	Delayed Ns	Capture <sup>c</sup>	Radiotoxicity
<sup>140</sup> La			xxx			
<sup>92</sup> Sr			xxx			
<sup>103</sup> Ru		xxx	xxx		xxx	
<sup>95</sup> Zr			xxx			
<sup>144</sup> Ce			xxx			
<sup>106</sup> Ru			xxx			
<sup>137</sup> Cs			xxx			
<sup>134</sup> Cs			xxx		xxx	
<sup>154</sup> Eu			xxx			
<sup>135</sup> Xe	xxx					
<sup>103</sup> Rh	xxx	xxx				
<sup>143</sup> Nd	xxx	xxx				
<sup>131</sup> Xe	xxx	xxx				
<sup>133</sup> Cs	xxx	xxx				
<sup>149</sup> Sm	xxx	xxx				
<sup>99</sup> Tc	xxx	xxx				xxx
<sup>152</sup> Sm	xxx					

Nuclide	BU PWR <sup>a</sup>	BU FBR <sup>b</sup>	Monitoring	Delayed Ns	Capture <sup>c</sup>	Radiotoxicity
<sup>147</sup> Pm	xxx	xxx				
<sup>151</sup> Sm	xxx	xxx				
<sup>153</sup> Eu	xxx	xxx				
<sup>145</sup> Nd	xxx	xxx				
<sup>155</sup> Eu	xxx					
<sup>154</sup> Eu	xxx					
<sup>109</sup> Ag	xxx	xxx				
<sup>155</sup> Gd	xxx					
<sup>95</sup> Mo	xxx	xxx				
<sup>147</sup> Sm	xxx					
<sup>150</sup> Sm	xxx					
<sup>101</sup> Ru	xxx	xxx				
<sup>148m</sup> Pm					xxx	
<sup>148</sup> Pm					xxx	
<sup>156</sup> Eu					xxx	
<sup>136</sup> Cs					xxx	
<sup>79</sup> Se						xxx
<sup>93</sup> Zr						xxx
<sup>126</sup> Sn						xxx
<sup>129</sup> I						xxx
<sup>135</sup> Cs		xxx				xxx
<sup>105</sup> Pd		xxx				
<sup>107</sup> Pd		xxx				
<sup>97</sup> Mo		xxx				
<sup>102</sup> Ru		xxx				
<sup>104</sup> Ru		xxx				
<sup>91</sup> Zr		xxx				
<sup>137</sup> I				xxx		
<sup>89</sup> Br				xxx		
<sup>94</sup> Rb				xxx		
<sup>90</sup> Br				xxx		
<sup>88</sup> Br				xxx		
<sup>85</sup> As				xxx		
<sup>138</sup> I				xxx		
<sup>98m</sup> Y				xxx		
<sup>95</sup> Rb				xxx		
<sup>139</sup> I				xxx		
<sup>97</sup> Br				xxx		
<sup>93</sup> Rb				xxx		
<sup>99</sup> Y				xxx		
<sup>91</sup> Br				xxx		
<sup>135</sup> Sb				xxx		

burnup determination for <sup>a</sup> pressurized water reactor (PWR) <sup>b</sup> fast breeder reactor (FBR)  
<sup>c</sup> capture effect on decay heat.

TABLE 6.3.3. COMPARISON BETWEEN MEEK & RIDER, ENDF/B-VI AND JEF-2 FOR SOME REFERENCE YIELDS

U235(T)									
nuclide	Meek & Rider (M&R)		ENDF/B-VI (B-6)		JEF-2 (J-2)		differences (%)		
	CY <sup>a</sup>	ΔCY <sup>a</sup> (abs)	CY <sup>a</sup>	ΔCY <sup>a</sup> (abs)	CY <sup>a</sup>	ΔCY <sup>a</sup> (abs)	B-6/M&R	J-2/M&R	J-2/B-6
106Rh	4.03E-03	5.65E-05	4.02E-03	5.62E-05	4.07E-03	1.53E-04	-0.43	0.85	1.28
133Cs	6.69E-02	9.36E-04	6.70E-02	2.34E-04	6.62E-02	6.98E-04	0.16	-1.10	-1.26
134Cs	1.33E-07	1.86E-09	7.71E-08	4.93E-08	2.03E-07	7.60E-08	-42.03	52.71	163.44
135Cs	6.54E-02	9.15E-04	6.54E-02	4.18E-02	6.58E-02	1.47E-03	0.02	0.65	0.63
137Cs	6.22E-02	8.71E-04	6.19E-02	3.09E-04	6.24E-02	5.39E-03	-0.49	0.41	0.90
144Nd	5.50E-02	7.70E-04	5.50E-02	1.92E-04	5.46E-02	3.81E-04	-0.03	-0.69	-0.66
145Nd	3.93E-02	5.50E-04	3.93E-02	1.38E-04	3.93E-02	3.12E-04	0.04	0.01	-0.03
148Nd	1.67E-02	2.34E-04	1.67E-02	5.86E-05	1.68E-02	1.72E-04	0.03	0.11	0.09
154Eu	1.30E-08	1.82E-10	1.94E-09	1.24E-09	3.17E-09	1.21E-09	-85.08	-75.63	63.31
140Ba	6.21E-02	8.70E-04	6.21E-02	6.21E-04	6.28E-02	7.49E-04	0.04	1.04	0.99
92Sr	5.92E-02	8.28E-04	5.94E-02	8.31E-04	5.99E-02	3.25E-03	0.37	1.18	0.81

U235(F)									
nuclide	Meek & Rider (M&R)		ENDF/B-VI (B-VI)		JEF-2		differences (%)		
	CY <sup>a</sup>	ΔCY <sup>a</sup> (abs)	CY <sup>a</sup>	ΔCY <sup>a</sup> (abs)	CY <sup>a</sup>	ΔCY <sup>a</sup> (abs)	B-6/M&R	J-2/M&R	J-2/B-6
144Nd	5.26E-02	1.4	5.27E-02	3.69E-04	4.89E-02	9.95E-04	0.1184	-7.172	-7.282

### 6.3.3. Evaluation for some reference yields

An evaluation of reference fission yields has been carried out at CNDC as a CRP task. The results are presented in Table 6.3.4. The evaluation method for reference fission yields is the same as for other fission yields. It should be emphasized that the fission yields used as reference are usually obtained from measurements rather than by model calculation, especially if adequate experimental data are available. Results from absolute yield measurement should be evaluated first as internal reference, if possible.

TABLE 6.3.4. SOME RESULTS OF REFERENCE YIELDS

#### a. <sup>235</sup>U thermal fission

<sup>99</sup> Mo	6.150000E+00	4.920000E-02
<sup>140</sup> Ba	6.321430E+00	4.705610E-02
<sup>148</sup> Nd	1.671650E+00	2.873459E-03
<sup>95</sup> Mo	6.537096E+00	5.437335E-02
<sup>100</sup> Mo	6.257285E+00	5.594396E-02
<sup>95</sup> Zr	6.479100E+00	5.040000E-02
<sup>96</sup> Zr	6.237431E+00	1.936396E-01
<sup>83</sup> Kr	5.433502E-01	5.769334E-03
<sup>85</sup> Kr	2.840601E-01	8.824119E-04
<sup>85m</sup> Kr	1.360382E+00	4.989226E-02
<sup>86</sup> Kr	1.971164E+00	1.540071E-02
<sup>87</sup> Kr	2.595844E+00	9.200242E-02
<sup>88</sup> Kr	3.561856E+00	1.691554E-01
<sup>89</sup> Sr	4.719514E+00	2.876379E-01
<sup>90</sup> Sr	5.737442E+00	3.667082E-02
<sup>101</sup> Ru	5.203938E+00	6.048889E-02
<sup>102</sup> Ru	4.319135E+00	4.964242E-02
<sup>143</sup> Ce	5.990197E+00	3.550890E-02
<sup>156</sup> Eu	1.204900E-02	7.625000E-04
<sup>144</sup> Ce	5.328000E+00	5.160000E-02
<sup>147</sup> Nd	2.283900E+00	3.570000E-02
<sup>161</sup> Tb	9.204700E-05	1.270000E-05

#### b. <sup>235</sup>U 14.8 MeV fission

<sup>95</sup> Zr	.....5.3062E+00	1.448E-01
<sup>99</sup> Mo	.....5.2179E+00	1.693E-01
<sup>144</sup> Ce	.....3.3290E+00	1.003E-01
<sup>147</sup> Nd	.....1.6447E+00	4.640E-02
<sup>156</sup> Eu	.....5.6665E-02	3.651E-03
<sup>161</sup> Tb	.....5.1257E-03	6.422E-04

#### c. <sup>238</sup>U 14.0 MeV fission

<sup>95</sup> Zr	.....4.8329E+00	1.166E-01
<sup>99</sup> Mo	.....5.6491E+00	1.232E-01
<sup>144</sup> Ce	.....3.6818E+00	1.731E-01
<sup>147</sup> Nd	.....2.0489E+00	6.120E-02
<sup>156</sup> Eu	.....1.1020E-01	4.100E-03
<sup>161</sup> Tb	.....8.2510E-03	8.000E-04

#### d. <sup>238</sup>U 14.8 MeV fission

<sup>95</sup> Zr	.....4.9907E+00	8.720E-02
<sup>99</sup> Mo	.....5.6256E+00	9.500E-02
<sup>144</sup> Ce	.....3.8143E+00	1.127E-01
<sup>147</sup> Nd	.....2.0784E+00	4.310E-02
<sup>156</sup> Eu	.....1.0830E-01	4.400E-03
<sup>161</sup> Tb	.....8.2400E-03	6.500E-04



## Chapter 7

### EVALUATED FISSION YIELD FILES

#### 7.1. FISSION PRODUCT YIELD EVALUATION FOR THE USA EVALUATED NUCLEAR DATA FILES

##### 7.1.1. Historical background

For reliable and consistent nuclear calculation, it is desirable to have an accurate set of nuclear data. The United States National Nuclear Data Center at Brookhaven National Laboratory has published such an Evaluated Nuclear Data File (ENDF/B-VI). The General Electric Company has published evaluated fission yield data in a document NEDO-12154 [7.1.1]. This fission yield evaluation, conducted at Los Alamos since 1981, has been greatly expanded in cooperation with the Fission Yield and Decay Data Subcommittee of CSEWG (Cross Section Evaluation Working Group) for inclusion in the ENDF/B files, and with contributions from the IAEA CRP yields program.

Table 7.1.1 shows the areas expanded specifically for each version of those files, the last version containing contributions from the IAEA CRP. There were intermediate evaluations between 1978 and 1994 that were supplied to the CRP and to others on request, but distribution was very limited.

TABLE 7.1.1. EVALUATION EVOLUTION

EVALUATION	PRE-ENDF	ENDF/B-IV	ENDF/B-V	ENDF/B-VI
Report	NEDO-12154	NEDO-12154-1	NEDO-12154-2E	ENDF-349
Year	1972	1974	1978	1994
Fission Reactions	10	10	20	60
Cumulative Yields	YES	YES	YES	YES
Independent Yields	NO	YES	YES	YES
Isomer Ratios	NO	NO	YES	YES
Odd-Even Pairing	NO	YES	YES	YES
Delayed Neutrons	NO	NO	YES	YES
Charge Balance	NO	YES	YES	YES
Ternary Fission	NO	NO	YES	YES
References	812	956	1119	1575
Input Values	6000	12400	18000	36000
Final Yields	11000	22000	44000	132000

The latest ENDF/B-VI version includes about 132 000 yields and their uncertainties in 60 sets of about 1100 values for independent yields before delayed neutron (DN) emission by 271 precursors [7.1.2], and a like number of cumulative yields after DN emission. The sets include 60 fission reactions of heavy nuclides including several fission neutron energies. The models used, data sources, evaluation methods, and the integral tests were largely described in [7.1.3] and, of course, in the main report, ENDF-349. [7.1.4], some tests made are also discussed.

### 7.1.2. General Approach

Not every one of the 1153 independent fission product yields required for each fissionable nuclide has been measured. All of the unmeasured values have been calculated from the best available models. All chain yields are normalized to 100% for the light and heavy mass peaks separately. Within each mass, a Gaussian charge distribution has been assumed about the most probable charge,  $Z_p$ , with a constant standard deviation of 0.560 charge units, except where individual charge dispersion widths have been evaluated by others such as J.K. Dickens [7.1.5] and A.C. Wahl [7.1.6]. Odd-even proton and neutron pairing effects are superimposed. All  $Z_p$  values have been adjusted within their limits of error to obtain an acceptable proton balance from the sum of binary and ternary fission yields.

The direct yields to the metastable and ground state isomers are nuclear spin dependent. These direct yields are apportioned to each isomer (where the spins are known) by the model of Madland [7.1.7]. Delayed neutron emission is treated by including a proper treatment for fission yields of delayed neutron emitters. As a result all independent fission yields are now appropriately given before delayed neutron emission, and all cumulative fission yields are given after delayed neutron emission in the traditional manner. All recommended yields are the result of weighted averages of experimental and model values. A systematic error of 2% is combined with the reported random error of each absolute fission yield in which a systematic error has not been included by its author. Mass spectrometric measurements have been assigned errors no smaller than 0.5% relative.

Radiochemical measurements, because of the uncertainties in absolute accuracy's of decay schemes and counting efficiencies, have been assigned errors no smaller than 20% for all measurements made before 1955, and 10% for those made since 1955. A few discrepant measurements were rejected by traditional statistical criteria such as the Dixon Range Test. Discrepant values are retained in the published input files but not used. Measured yields always dominate model values, including values for isomeric states.

The evaluations use the  $Z_p$  distribution model modified by  $Z$  and  $N$  pairing and variable sigma. These values are based on their preliminary evaluation of chain yields for ENDF/B-VI ([7.1.8] and subsequent private cooperation with A.C. Wahl). A small adder is applied to each  $Z_p$  to maintain charge balance over the entire yield set. Where isomerism occurs, the Madland [7.1.7] model is used if spins are known, or a 50/50 split otherwise. Only charge and mass balance over the entire yield set is imposed. The  $Z_p$  (and  $A'_p$ ) models have no theoretical basis but have been reasonably successful (see [7.1.3] and other papers, e.g. by Wahl, James and Mills, Lammer in the same proceedings of the "Specialists Meeting on Fission Product Nuclear Data". We also present most of the data testing results for the US evaluations and more detail on model parameters in this document.)

### 7.1.3. Discussion of Errors

The chain yields in the peak regions are known to about 1% in the well known measured fissionable nuclides, and to about 14% in the less well measured fissionable nuclides. The chain yields in the wings and valley are less well known and uncertainties range from about 8 to 30%. The U235T yields are strikingly better known. Table 7.1.2 looks at the makeup of the better known U235T chain yield errors. It shows that these smaller errors are the result of many determinations rather than abnormally small errors assigned to individual measurements. The average individual measurement is about  $\pm 3\%$  but in some cases about an average of 20 measurements result in the deviation of the mean being smaller than  $\pm 1\%$ .

TABLE 7.1.2. INDIVIDUAL MEASUREMENT ERRORS FOR MOST ACCURATE U235T FP MASSES

MASS	INDIV. SIGMA, %	N	UNWEIGHTED SIGMA OF MEAN, %	WEIGHTED SIGMA OF MEAN, %
83	1.94	9	0.6	0.5
85	2.58	36	0.4	0.4
86	1.85	12	0.5	0.5
131	4.30	30	0.8	0.4
132	3.84	25	0.8	0.4
133	3.94	21	0.9	0.4
134	3.09	18	0.7	0.4
135	3.20	16	0.8	0.4
136	1.59	10	0.5	0.4
137	3.23	17	0.8	0.5
143	2.09	17	0.5	0.4
144	3.07	25	0.6	0.4
145	2.02	18	0.5	0.4
146	2.11	19	0.5	0.4
147	3.32	20	0.7	0.7
148	2.45	27	0.5	0.4
150	2.00	17	0.5	0.5
MEAN	2.74	20	0.6	0.4

Because of the difficulty in obtaining data free of systematic errors and information such as shown in Table 7.1.2, it is concluded that current yield recommendations should not be given accuracy assignments smaller than about  $\pm 0.4\%$  in the ENDF files.

The treatment of errors is complicated by such factors as: (a) the range of yields varying over 12 orders of magnitude; (b) small uncertainties on many measured chain yields compared to large uncertainties in model values; and (c) the need to have independent yield uncertainties consistent with cumulative yields (see [7.1.9] for a discussion of the correlation method). The distribution of yields along any mass chain is roughly a Gaussian about the estimated  $Z_p$  value and this is only about 1.5 charge units wide (FWHM). Therefore, the direct yields vary greatly from the most probable value at any given mass, and of course, greatly over the entire mass range; this variation contributes to the evaluation difficulty. There are other problems associated with decay branchings in evaluating uncertainties.

#### 7.1.4. Some Final Comments

For anyone not having access to the internet, the full report [7.1.4] can be obtained from the authors on compressed diskettes. That report is itself a condensation of an effort that has extended over about 28 years. The report is large because of the extensive references and the inclusion of all evaluated data, known measurements (including values we feel are in error and not used) and inclusion of all distribution parameters. The main text of the report is actually small, having been refined over several earlier publications. (The 1575 references add considerably to its length.) Therefore, the present summary primarily serves the purpose of alerting interested parties of the open existence of the internet report.

The ENDF/B-VI evaluated fission yields have been further extrapolated to include additional unmeasured low yield nuclides and renormalised by a maximum of 0.23% to a total of 200% for both fission product yield peaks combined. Some additional isomeric states are included and some mass chains are extended; these do not alter the basic evaluation in [7.1.4]. The resulting independent and cumulative fission yields are available as ENDF files from U.S.A. National Nuclear Data Center, Brookhaven National Laboratory, Upton, New York 11973. **Appendix A.2** (Tables 1-6) contains the mass chain yields for the 60 fission reactions evaluated. These are not easily obtained from the ENDF/B-VI files but are included in the LA-UR-93-3106 PostScript report.

The authors appreciate the cooperation of the scientific community over many years in assisting the ENDF/B-VI effort, in particular Dr. W. B. Wilson, and would also like to thank members of this IAEA CRP for their various inputs. It was not possible to use all suggestions because of limited support and time constraints, and because unfortunately all funding for continued yield evaluation in the US ceased in 1993.

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## 7.2. THE UK/JEF FISSION YIELD LIBRARIES

### 7.2.1. Historical background

In the UK the evaluation of fission product yields for use in computer libraries was pioneered by Crouch at Harwell; see for example Crouch [7.2.1]. The libraries he produced were named Crouch 1, 2, and 3. After his retirement, the work was continued at Winfrith, first on an interim library, Crouch 4, [7.2.2], and then on UKFY1, produced by Banai et al. [7.2.3], which was adopted by the first stage, JEF1, of the Joint Evaluated File.

A detailed comparison between different libraries was made in an earlier paper, presented at a meeting on decay heat at Studsvik [7.2.4].

The latest complete UK library, UKFY2, which has been adopted for JEF2, is a considerable advance on any of the earlier UK libraries (although, of course, it builds on them). Many more fissioning systems were included, the database of measurements was brought up-to-date, a new analysis of fractional independent yields had been made, and the method of fitting the yields to constraints arising from conservation laws was improved. The library produced from this evaluation is in the ENDF-6 format [7.2.5]. Both independent (pre-delayed neutron emission) and cumulative (post-delayed neutron emission) yields are available, together with their standard deviations. For a given fission reaction, the yields are correlated by the fitting process: a prescription is available for the production of their covariance matrices. This work was funded jointly by the Central Electricity Generating Board, British Nuclear Fuels plc, and the United Kingdom Atomic Energy Authority; the UKAEA's contribution was mostly through its Underlying Research Programme. The work was carried out jointly by the University of Birmingham and the UKAEA at Winfrith Atomic Energy Establishment.

The UKFY2 evaluation was described in full in three reports [7.2.6-7.2.8]. The methodology of the evaluation is described in full in [7.2.6]. We have produced tables of evaluated, unadjusted chain and independent yields, which are given in a report [7.2.7]. A detailed description of the remaining discrepancies are given in [7.2.8]. The complete library in ENDF-6 format is available from the NEA Data Bank, along with the FITFYS subroutine and some of the input data files. Several remaining problems were identified or emphasized by this work. A few are listed below.

- (1) There were considerable gaps in the data, especially in charge distributions, but also even in chain yields for the more important fission reactions, and obviously for nearly all fission products from the higher actinides. There also remained some significant discrepancies between measurements, including some for chain yields from the thermal neutron induced fission of  $^{235}\text{U}$ . More measurements are needed, but in the meantime improved extrapolation techniques, preferably based on sound theory, were needed. We noted that it is necessary to use extrapolation both for unmeasured yields for fairly well investigated fission reactions, and for most yields for some reactions which are almost entirely unstudied.
- (2) More measurements were needed to test and improve the existing empirical formulae.
- (3) It was still not clear how yields vary with incident neutron energy. This was particularly uncertain for independent yields and for ternary fission products.

These problems were felt to need further study. British Nuclear Fuels plc then funded a continuation of this work toward a new evaluation, UKFY3. This work was carried out within the framework of this CRP.

This paper describes the UKFY2 evaluation, then describes the advances made in the development of UKFY3.

### 7.2.2. UKFY2

Many of the problems encountered in this work are found to a greater or lesser extent in any evaluation, and so some of the techniques developed may be of wider interest. To be generally useful, any data library must be as accurate and as complete as possible. Accuracy requires an up-to-date and complete compilation of experimental results, and a statistically sound method of treating the measurements and of considering the inevitable discrepancies between some of them. Completeness requires considerable care in interpolation and extrapolation to fill the equally inevitable gaps where there are no measured data at all. Since a user needs to know the accuracy of the data, it is also necessary to estimate as carefully as possible the standard deviations of the evaluated data and the correlations between them. This last task is sometimes the longest and hardest. The database, evaluation methods, and the interpolation of data and parameters are considered below.

Fission product yield nuclear data should satisfy some simple conditions arising from physical conservation laws of nucleon number and of charge. Different evaluating teams use these conditions differently: either to adjust the data, or, if they have not been used for adjustment, as tests subsequent to the evaluation process. There are essentially four such conditions:

- (1) The yields, apart from those of the relatively rare light products from ternary fission, should sum to 2.
- (2) There should be conservation of nucleon number.
- (3) There should be overall conservation of charge.
- (4) There should be detailed conservation of charge giving equal yields of complementary elements. (This condition is weakened slightly by the occurrence of ternary fission.)

UK evaluations have applied successively more constraints. For example, Crouch's library published in 1977 [7.2.1] used the first two conditions as constraints and the others as tests, while the present evaluation, like its immediate predecessor UKFY1 [7.2.3], used all four as constraints. Further, as the yields are constrained using a least-squares method, it is a straightforward matter to obtain a covariance matrix.

Up to this stage in our evaluation process, little account has been taken of the occurrence of isomers among the fission products, since all the techniques (with a slight exception in the initial treatment of measured chain yields) apply to the total independent yield of a nuclide, including all isomers. The experimental values of the ratios of independent yields of isomeric to ground states was reviewed. The method of Madland and England [7.2.9] was used to calculate ratios for which there were no measurements.

The calculation of cumulative from independent yields is then described. This requires data on branching ratios of radioactive decays, including  $P_n$  values for delayed neutron precursors, and these were all obtained from the preliminary JEF2 decay data library, supplemented where necessary by the theoretical values of Klapdor [7.2.10].

The need to complete the UKFY2 evaluation within a fairly short period had meant that several quite important topics have had to be glossed over. Equally, other matters requiring further investigation have arisen in the course of the work. These were later investigated as part of the UKFY3 evaluation.

#### 7.2.2.1. Fissioning systems

It was felt desirable to base the selection of fission reactions in the UK libraries on objective criteria. Consequently a series of calculations were made with the inventory code FISPIN, described by Burstall [7.2.11], and its 1988 libraries for both thermal and fast reactors. For the former, initial fueling by enriched uranium, recycled uranium, mixed plutonium/uranium and thorium/uranium, were individually considered. The ratings and irradiations applied to the calculations were greater than actually achieved in practice at present, but were values regarded as feasible within the foreseeable future. Reactions were regarded as important if they contributed more than 0.1% of the fission rate at any time. Thus, it is thought that the derived list is more than adequate for some time to come. It does, of course, depend on the initial fuel compositions and on the assumed capture and fission cross-sections for the higher actinides; it is acknowledged that some of these latter may be considerably in error.

In addition, the library includes fission of  $^{232}\text{Th}$ ,  $^{233}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  by "high energy" (about 14 MeV) neutrons; these reactions were in the earlier UK libraries. We have also considered the yields from the spontaneous fission of  $^{242}\text{Cm}$  and  $^{244}\text{Cm}$  which are important as sources of neutrons in reactors and in fuel handling, and of  $^{252}\text{Cf}$ , which is important as a standard.

The complete set is given in Table 7.2.1. The reactions considered by the burnup calculations are given in three sub-sets, in the three left-hand columns. These are distinguished by the value of the maximum fission rate percentage due to the nuclide in question at any time during the irradiation. (The range in which the percentage falls is indicated in the column heading). Clearly, the required accuracy of yields is greater if the percentage fission rate is greater; hence we consider that nuclides in column 1 need the most careful treatment, followed by those in column 2, and then by those in column 3.

#### 7.2.2.2. Definitions and notation

The atomic number and mass number of the fissioning nucleus are denoted by  $Z_f$  and  $A_f$  respectively. For the neutron-induced fission of a nuclide of mass number  $A_{\text{target}}$ ,  $A_f = A_{\text{target}} + 1$ , while for spontaneous fission,  $A_f$  is the mass number of the fissioning nuclide.

A fission product nuclide is specified symbolically by the triplet (A,Z,I), where A and Z are respectively the mass number and atomic number, and I indicates the isomeric state (I = 0 for the ground state, I = 1, 2 for the 1<sup>st</sup>, 2<sup>nd</sup> excited state). If a fission product has no isomers, or if we are referring to the sum of yields for all its isomers, we use the doublet (A,Z).

TABLE 7.2.1. THE 39 FISSIONING SYSTEMS IN UKFY2

Maximum fraction of fission rate			
>10%	1-10%	0.1-1%	sf
nuclides: 5	2	12	3
* $^{233}\text{U}$ TFH * $^{235}\text{U}$ TFH * $^{238}\text{U}$ FH * $^{239}\text{Pu}$ TF * $^{241}\text{Pu}$ TF	* $^{240}\text{Pu}$ F $^{245}\text{Cm}$ TF	* $^{232}\text{Th}$ FH $^{234}\text{U}$ F $^{236}\text{U}$ F $^{237}\text{Np}$ TF $^{238}\text{Np}$ TF $^{238}\text{Pu}$ TF $^{242}\text{Pu}$ F $^{241}\text{Am}$ TF $^{242\text{m}}\text{Am}$ TF $^{243}\text{Am}$ TF $^{243}\text{Cm}$ TF $^{244}\text{Cm}$ TF	$^{252}\text{Cf}$ $^{242}\text{Cm}$ $^{244}\text{Cm}$

\* Nuclides in UKFY1 [7.2.3] and previous UK libraries [7.2.1,7.2.2].

sf spontaneous fission.

T thermal neutron fission.

F fast neutron fission.

H 14 MeV ('high') neutron fission.

The independent yield  $y(A,Z,I)$  is the number of atoms of  $(A,Z,I)$  produced directly from one fission, after the emission of prompt neutrons (but before the emission of delayed neutrons). It can be written as the product of 3 factors:

$$y(A,Z,I) = Y(A) f(A,Z) R(A,Z,I)$$

where the sum yield  $Y(A)$  is the total independent yield (before delayed neutron emission) of all fission products of mass number  $A$ ;  $f(A,Z)$  is the fractional independent yield of all isomers of  $(A,Z)$ ; and  $R(A,Z,I)$ , the isomeric yield ratio, is the fraction of  $(A,Z)$  produced directly as isomer  $I$ .

From the definition, it follows that:

$$\sum_Z f(A,Z) = 1 \quad \text{for all } A$$

$$\sum_I R(A,Z,I) = 1 \quad \text{for all } (A,Z)$$

so that

$$Y(A) = \sum_{Z,I} y(A,Z,I) \quad \text{for all } A$$

The usefulness of these formulae derives from the fact that, with the exception of delayed neutron ( $\beta^-n$ ) emission and the few very long-lived  $\alpha$  decays, all the radioactive decays of fission products are  $\beta^-$  or  $\beta^+$ , or isomeric transitions, and in none of these is  $A$  altered. Thus, to a very good approximation, the fission products can be considered as belonging to distinct mass chains.



The cumulative yield  $c(A,Z,I)$  of  $(A,Z,I)$  is the total number of atoms of that nuclide produced over all time after one fission. If the nuclide is stable and at the end of a mass chain, the cumulative yield is the total number of atoms remaining per fission, and is termed the chain yield  $Ch(A)$ . Similarly, for a nuclide with a much longer half-life than any of its precursors,  $c(A,Z,I)$  is very nearly equal to the amount of it produced at a time short compared to its half-life but long compared to those of its precursors. However, for a radioactive nuclide for which this is not the case, some atoms will have decayed before all have been produced, so that at no time will there actually be  $c(A,Z,I)$  atoms per fission present.

An equivalent definition that is more useful is the following: immediately at the end of an "infinite" irradiation at the rate of 1 fission per second,  $c(A,Z,I)$  is the rate of decay of  $(A,Z,I)$  if that nuclide is radioactive, or its rate of production if it is stable. Consequently cumulative yields are useful in computing total fission product decay energies and delayed neutron emission rates.

The sum yield  $Y(A)$  and the chain yield  $Ch(A)$  for a mass chain  $A$  may differ by a few per cent, because the former applies before, and the latter after, delayed neutron emission. Tables of the calculated differences for each chain of some fissioning systems are given in [7.2.6]. It is sometimes difficult to decide which of these two quantities has been measured; this is an area to which more study should be devoted in future evaluations. Further discussion of cumulative yields can be found below.

### 7.2.2.3. Databases and data collection

The database used for the UKFY2 evaluation consisted of 3 files, containing data measured absolutely, relatively, and by "ratio of ratio" methods.

The most straightforward of these are the absolute measurements, in which the yield of a nuclide per fission is measured. However, this method requires knowledge of the number of fissions, which can be difficult to determine practically.

In the second type, a nuclide with a yield that is assumed to be accurately known is used to estimate the number of fissions. Thus, the unknown yield is determined relative to the standard yield.

The third type of measurement uses the "ratio of ratio" or "R-value" method. This assumes that the yields of both the fission product of interest ( $x$ ) and a monitor fission product ( $r$ ) are known from a reference fission reaction (indicated by the subscript 2). Then, if the yield of the monitor  $r$  is also known from the fission reaction of interest (subscript 1), the yield of  $x$  from reaction 1 can be found from:

$$y_1^x = y_2^x \left[ \frac{A_1^x \times A_2^r}{A_2^x \times A_1^r} \right] \frac{y_1^r}{y_2^r}$$

where  $y$  is a yield and  $A$  an activity, which is the quantity actually measured.

The term in brackets in the above equation is the 'R-value' and is made up of the measured activities determined in a simultaneous standard reaction irradiation. The other components of the right hand side must be determined absolutely or assumed. In this

evaluation measured yields only, and not predictions of unmeasured yields, are used to convert 'R-value' or relative measurements to absolute values.

The evaluation proceeded in two iterative stages. In the first stage, only absolute measurements (i.e. of the first type described above) were used. In the second, the two types of relative measurements were included as well, using for the standard yields values obtained in the first stage. Because these standards are, by definition, known accurately, their values will hardly be changed in the second stage and so further iterations are not required.

Experimental fission yield data were collected from three computer readable sources:

- i. Crouch's fission yield database, which was used in the evaluations [7.2.1] and [7.2.3].
- ii. The international database EXFOR described in [7.2.12]; the last update for UKFY2 was received in November 1989.
- iii. A database of recent references, produced after a thorough search of recent literature.

We believe the combined UKFY2 database to be complete up to 1988, and to contain some results published in 1989. It has 39% more items of data than that used by Crouch in 1977 [7.2.1] (12137 compared with 7448), including 60% more chain yield measurements.

The measurements were examined to remove duplications and to ensure consistency of isomeric states. In the evaluation, discrepant data were extensively examined to remove entry errors and to increase unacceptably low estimates of error. The experimentalists' uncertainties were adjusted in the previous UK evaluations to be always greater than 5% [7.2.1], unless significant justification was present in the reference. Considering the more accurate measurements now possible with mass separators this limit has now been reduced to 1%, for this type of measurement. However, a discrepant data point was down-weighted, if it was significantly different from other measurements, and the discrepancy could not be resolved by deeper study of the references (see below). Only in extreme cases were discrepant measurements entirely deleted from the UKFY2 database, and this was recorded in the reference database.

#### *7.2.2.4. Evaluation method*

##### *7.2.2.4.1. General treatment of measurements*

We have already mentioned a change in the treatment of reported uncertainties for mass-spectrometric measurements. For convenience, the revised rules used in attributing input uncertainties are stated here. Throughout this work all uncertainties are quoted at the 1 standard deviation level.

- (1) For recent mass-spectrometric measurements, the quoted uncertainty was used provided it was not less than 1%. Otherwise, 1% was used, unless there was good justification for the published value in the reference.
- (2) For other types of measurement, the quoted uncertainty was used provided it was not less than 5%. Otherwise, 5% was used, unless there was good justification for the published value in the reference.

- (3) Mass-spectrometric results quoted without any estimate of uncertainty were given a standard deviation of 10%.
- (4) Other results quoted without any estimate of uncertainty were given a standard deviation of 15%.

These standard deviations were then used to obtain weighted means in the following way. For  $n$  measurements  $y_i \pm \sigma_i$ ,  $1 \leq i \leq n$ , the mean is:

$$Y = \sum w_i y_i / W$$

where

$$W = \sum w_i$$

and the  $i^{\text{th}}$  weight is:

$$w_i = 1 / \sigma_i^2$$

The internal and external standard deviations of the mean are respectively:  $\sigma_I = 1 / \sqrt{W}$ , and

$$\sigma_E = \sqrt{\sum_{i=1}^{i=n} w_i (y_i - \bar{y})^2 / W(n-1)}$$

The value of  $\chi^2$  is:

$$\chi^2 = \sum_{i=1}^{i=n} w_i (y_i - \bar{y})^2$$

with  $n-1$  degrees of freedom if there are no constraints between the  $y_i$ . A useful quantity for indicating the consistency of the data is the "Birge factor":

$$R_B = \sqrt{\chi^2 / (n-1)}$$

It will be seen that  $\sigma_E = R_B \sigma_I$ .

If  $R_B \leq 1$ , then the data are consistent and  $\sigma_E \leq \sigma_I$ . If  $R_B > 1$ , then the data are inconsistent (though not necessarily significantly so) and  $\sigma_E > \sigma_I$ .

From  $\chi^2$  and the number of degrees of freedom, the probability of the data being consistent can be calculated. Those with a probability of less than 10% were listed in a set of discrepancy tables, values less than 1% and 0.01% being flagged for special consideration. In these cases, the data were checked for transcription errors between the quoted values in the references and the values in the database. Next, the relevant papers were studied to see if there were reasons for renormalizing or down-weighting the measurements. Usually, obvious corrections such as these could not be found; after all, any published results are subject to much checking and review. the amount of detailed study was in any case limited by the quantity of data. Consequently, after references had been checked, an automatic down-weighting procedure was applied to the data to reduce major discrepancies. This technique was based upon the normalized residual

$$R_i = (y_i - \bar{y}) \sqrt{\frac{w_i W}{W - w_i}}$$

which should be normally distributed with a mean of zero and a standard deviation of unity.  $R_i^2$  is equal to the decrease in  $\chi^2$  that would occur if the  $i^{\text{th}}$  measurement were removed. The  $|R_i|$  were constrained to be less than 2.5, for all yields with more than two measurements, by down-weighting the measurement with the largest value. If the maximum  $|R_i|$  was greater than 2.5 the  $w_i$  was adjusted to give  $R_i$  equal to 2.5, and then the averaging process was repeated. This procedure reduces the weight of points far from the mean, thus reducing the effect of discrepant data on the weighted mean. The theoretical justification for these definitions and statements is given in [7.2.13]; see also the appendix of [7.2.6].

The tables of evaluated yields in [7.2.7] clearly indicate where down-weighting has been employed. Both internal and external standard deviations of the mean are quoted and the larger of the two recommended.

Automatic down-weighting by the above method is not advised if there are only two measurements, which are mutually discrepant but of similar precision. Indeed, it is of doubtful use in considering any clearly bimodal distribution of data, which indicates systematic differences probably due to different experimental techniques that should be investigated in depth.

We would like to emphasize that the automatic down-weighting described here (or any alternative method) should not be used in preference to a detailed analysis of the original measurements. It is only when the latter is fruitless, or, as in the present case, time prevents more than a partial study of the literature, that an automatic procedure should be considered in an evaluation, and then its use should be clearly indicated.

Separate unadjusted evaluations were made for chain, fractional independent, and fractional cumulative yields. From these evaluations, sets of mean chain yields, fractional independent and cumulative yields with standard deviations were produced that could be used as input in the fitting of semi-empirical models as described below.

#### 7.2.2.4.2. Chain yields

Measurements were included both of a chain yield itself and of cumulative yields of nuclides close enough to the end of the decay chain and with sufficiently small independent yields for their cumulative yields to be good approximations to the chain yield. As several chains have nuclides with isomers near the stable end, a typical decay scheme of the form shown in Figure 7.2.1 was considered:

Nuclide C is either stable, or its cumulative yield can be assumed to be equal to the chain yield Y. Nuclide B has two isomers: a ground state, g, with cumulative yield G and an excited state, m, with a significant half-life ( $> 0.1$  second usually) with cumulative yield M. A fraction  $a$  of the excited state decays are by  $\beta^-$  emission to C, the remainder are by internal transition to the ground state.

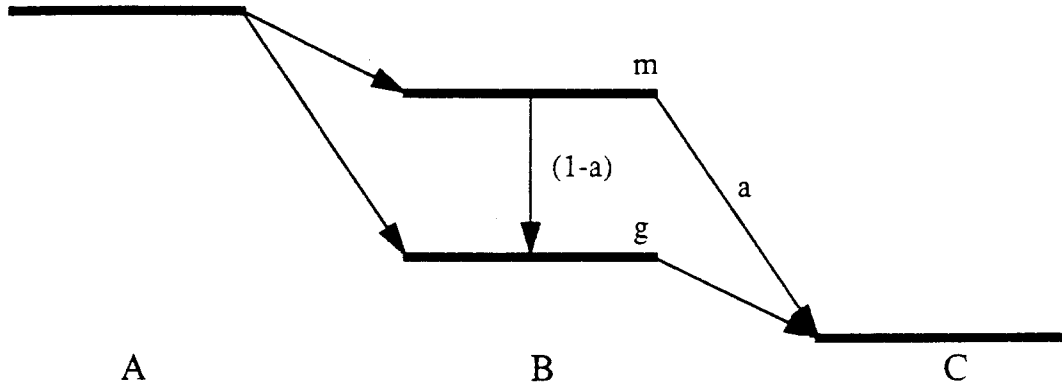


FIG. 7.2.1. Typical decay scheme at the end of a decay chain.

Measurements of M, G and Y were first considered separately, and then their means adjusted by least-squares to fit the condition:

$$Y = G + aM$$

which holds if the independent yield of C is negligible.

If the unadjusted means and standard deviations are indicated by bars, then the adjusted chain yield is given by:

$$Y = \bar{Y} - (\bar{Y} - \bar{G} - a\bar{M})\bar{\sigma}_Y^2 / D$$

with standard deviation  $\sigma_Y$  given by:

$$\sigma_Y^2 = \bar{\sigma}_Y^2 (\bar{\sigma}_G^2 + a^2 \bar{\sigma}_M^2) / D$$

where

$$D = \bar{\sigma}_Y^2 + \bar{\sigma}_G^2 + a^2 \bar{\sigma}_M^2$$

and where  $\sigma_Y$ , etc are the unadjusted standard deviations of Y, etc determined by the methods of the previous sub-section. (Any uncertainty in the branching fraction a is ignored, in order to ensure consistency with the decay data.)

The value of  $\chi^2$  for the fit is:

$$\chi^2 = (\bar{Y} - \bar{G} - a\bar{M})^2 / D$$

with 1 degree of freedom. If  $\chi^2 > 1$ , then the standard deviation  $\sigma_Y$  was multiplied by the Birge factor  $R_B = \sqrt{\chi^2}$ .

#### 7.2.2.4.3. Fractional independent and cumulative yields

For nuclides away from the stable ends of mass chains, measurements may be of either fractional or of absolute yields: the former being obtained if the same experiment also determines the chain yield. For the present purpose, fractional yields were required, so absolute values in the database were converted by dividing by the evaluated (unadjusted) mass

yield. The latter was either the experimental mass yield average (see above) or obtained from a fit to the mass yield distribution (see below).

#### 7.2.2.5. *Interpolation and extrapolation to fill gaps*

##### 7.2.2.5.1. General

To produce independent and cumulative yield libraries for reactor and decay heat calculations, all significant chain yields and fractional independent yields need to be known.

About 120 chain yields are required (mass 60 to 180) for these purposes. As even the system with the best coverage of measurements,  $^{235}\text{U}$  thermal, has only 111 measured chains and some systems (for example  $^{243}\text{Am}$ ) have none, a model or method of prediction is required for chain yields. Similarly, if fractional independent yields greater than  $10^{-8}$  are considered to be important, approximately 900 are required, whereas even for  $^{235}\text{U}$  less than 250 have been measured.

There are several semi-empirical models and methods of interpolation or extrapolation for fractional independent and chain yields which can be used.

##### 7.2.2.5.2. Chain yields

Three techniques were tried for filling gaps in chain yields:

- (1) For a given fissioning system, interpolation of log (chain yield) as a function of chain mass, i.e. the standard mass-yield curve. This technique was the simplest, but could only be used effectively for a small gap of one or two missing yields, as the function is rapidly varying and has a varying slope, and clearly could not be used for systems with no experimental data at all.
- (2) For a given mass chain, interpolation of log (chain yield) as a function of the effective fissioning mass, i.e. the mass of the fissioning system minus the mean number of prompt neutrons  $\bar{\nu}_p$ . Some of these graphs show slowly varying trends, which could be used to produce estimates of unmeasured chain yields. However for most chain masses the data are sparse or have large uncertainties, which makes interpolation difficult.
- (3) The fitting to the mass-yield curve by a set of Gaussians, as proposed by Musgrove et al [7.2.14] and used more recently by Dickens [7.2.15]. This technique is based upon the apparent similarity of the chain yield distributions to Gaussian distributions. This method is the most versatile of the three, as it can be readily used to predict yields from systems on which few or even no measurements have been made. Details of it and of its present application are given in the next sub-Section.

##### 7.2.2.5.3. Five Gaussian fit to chain yields

We have followed Musgrove et al [7.2.14] in using five Gaussians (viz. two for each peak of the mass distribution, and one for near-symmetric fission). More recently theoretical

justification for such a multiple Gaussian fit has been attempted, see for example Brosa et al. [7.2.16]. The 15 parameters (strength, mean and width of each Gaussian) were reduced to 8 by assuming symmetry and requiring a total yield of 2. This model gives the chain yield  $Y(A)$  as:

$$Y(A) = \frac{N_1}{\sigma_1 \sqrt{2\pi}} \left[ e^{-(A-\bar{A}-D_1)^2/2\sigma_1^2} + e^{-(A-\bar{A}+D_1)^2/2\sigma_1^2} \right] \\ + \frac{N_2}{\sigma_2 \sqrt{2\pi}} \left[ e^{-(A-\bar{A}-D_2)^2/2\sigma_2^2} + e^{-(A-\bar{A}+D_2)^2/2\sigma_2^2} \right] \\ + \frac{N_3}{\sigma_3 \sqrt{2\pi}} e^{-(A-\bar{A})^2/2\sigma_3^2}$$

where  $N_i$  is the coefficient of the  $i^{\text{th}}$  Gaussian and  $\sigma_i$  its width parameter,  $\bar{A}$  is the mean mass of the distribution and  $D_i$  is the separation of the  $i^{\text{th}}$  Gaussian peak from  $\bar{A}$ .

Because the chain yields sum to 2,  $N_3 = 2(1 - N_1 - N_2)$ .

The evaluated chain yields were fitted to this model, and Table 7.2.2 gives the values of the parameters for systems for which there were sufficient data for satisfactory fits to be obtained. The fits for  $^{235}\text{U}$  thermal and fast,  $^{233}\text{U}$  thermal, and  $^{239}\text{Pu}$  thermal are shown in Figs 2–5 of [7.2.6]. Such a representation by only 5 Gaussians, of course, smoothes the distribution, removing the small fluctuations that are observed experimentally.

To extend the 5-Gaussian representation to systems with sparse data, the parameters already obtained, in Table 2, were plotted against fissioning mass and charge, and fitted by linear or quadratic functions of  $A_f$ . (No systematic trend with  $Z_f$  could be detected.) Then new 5-Gaussian fits were made to the chain yield data, allowing only one of the parameters to vary in turn. Linear or quadratic fits were then made to each of the varied parameters, with the functions of  $A_f$ . After all parameters had been fitted in this manner, the whole process was repeated. After two iterations the refitting procedure failed to produce any improvement in  $\chi^2$ . The functions thus obtained were:

$$\bar{A} = \frac{A_f - \bar{v}_p}{2} \quad [\text{fixed by nucleon conservation}] \\ N_1 = 0.0003846 A_f + 0.6215 \quad [\text{fast}] \\ N_1 = 0.0010563 A_f + 0.4579 \quad [\text{thermal}] \\ \sigma_1 = 0.2017 A_f - 42.906 \\ D_1 = 27.1 - 0.67832(A_f - 230) + 0.013664(A_f - 230)^2 \\ N_2 = 0.286 \\ \sigma_2 = 0.1125 A_f - 24.375 \\ D_2 = 19.9 - 0.595(A_f - 230) + 0.00125(A_f - 230)^2 \\ \sigma_3 = 12.0$$

TABLE 7.2.2. FITTED PARAMETERS FOR THE 5 GAUSSIAN MODEL

Actinide	$\bar{A}$	$N_1$	$\sigma_1$	$D_1$	$N_2$	$\sigma_2$	$D_2$	$\sigma_3$
$^{233}\text{U}$ Thermal	115.86	0.7116	4.230	24.63	0.2855	3.048	17.11	13.79
$^{235}\text{U}$ Thermal	116.90	0.7158	4.298	24.02	0.2823	2.423	16.63	9.250
$^{239}\text{Pu}$ Thermal	118.50	0.7100	5.587	21.34	0.2887	2.630	15.27	2.237
$^{229}\text{Th}$ Thermal	114.03	0.7146	3.081	27.80	0.2832	2.335	21.73	11.22
$^{232}\text{Th}$ Fast	115.49	0.7079	3.522	26.45	0.2823	2.472	19.10	11.80
$^{235}\text{U}$ Fast	116.79	0.7081	4.516	23.76	0.2868	2.503	16.42	13.09
$^{238}\text{U}$ Fast	118.02	0.7102	5.031	22.85	0.2852	2.131	15.85	9.525
$^{232}\text{Th}$ High	114.65	0.6989	4.451	24.88	0.1164	4.264	18.78	11.17
$^{233}\text{U}$ High	115.12	0.5586	5.959	23.12	0.2190	5.243	19.27	11.78
$^{235}\text{U}$ High	116.01	0.6241	5.535	23.26	0.1824	3.208	16.00	11.99
$^{238}\text{U}$ High	117.42	0.6605	5.962	22.32	0.1921	2.938	15.81	11.80

Although this is a simple model and the goodness of fit to the parameters only moderate, reasonable results were obtained when chain yields were calculated for the systems in Table 7.2.2 using the predicted (instead of the fitted) Gaussian parameters and compared with experimental data. In [7.2.6], figures show "predicted" and evaluated measured chain yields for  $^{235}\text{U}$  thermal and fast chain yield distributions. Quite good agreement was also obtained with evaluated measured chain yields for other systems which had not been included in Table 7.2.2; [7.2.5] shows the comparisons for  $^{236}\text{U}$  fast,  $^{238}\text{U}$  fast, and  $^{237}\text{Np}$  thermal and fast.

The yields calculated from the formulae were then plotted with measured yields against  $(A_f - \bar{v}_p)$  as in Interpolation Method (ii) (above). The present method gave good agreement in some cases but many comparisons were inconclusive; either the uncertainties were too large or there were too few points to test the prediction. However, a definite trend away from the prediction occurs in the region of masses 89-100 for  $(A_f - \bar{v}_p) < 232$ . There, a straight line fitted experimental data better than the curve from the derived 5 Gaussians.

Therefore chain yields for the evaluation were first taken from the experimental averages. Then short gaps were filled by interpolation as noted in Interpolation Method (i). Larger gaps were filled using the 5 Gaussian model with parameters from Table 7.2.2 if appropriate, or obtained from the above equations if not. The predictions from this model were modified before being used to produce complete data sets. First, the central Gaussian was not allowed to contribute outside the two main peaks (to avoid unrealistic flaring of the distribution at very low yields). Secondly the predictions were renormalized with the experimental data. This renormalization considered the two cases of (a) the wings of the distribution and (b) gaps in the experimental data. The predictions in the wings, up to the first experimental point, were renormalized by a constant factor so that for this point the predicted yield equaled the measured value. In gaps, predicted yields at each end were fitted to measured values by making the logarithm of the normalization factor vary linearly with mass across the gap. Finally, for  $(A_f - \bar{v}_p) < 232$  and for  $89 \leq A \leq 100$ , straight line fits of  $\log Y$  against  $(A_f - \bar{v}_p)$  were used in preference to the other methods of interpolation.



This procedure gave a complete set of chain yields for each fissioning system based as firmly as possible on experimental data. Graphs of these chain yields are given in [7.2.6] for some of the 39 fissioning systems considered.

#### 7.2.2.5.4. Fractional independent yields

The fractional independent yields can be fitted by either the  $A_p'$ , or the  $Z_p$  models, described by Wahl [7.2.17,7.2.18] and the references therein; these represent the fractional independent yields as Gaussian distributions in mass and charge respectively, modified by an odd-even effect. We concentrated on the  $Z_p$  model and have used the same parameterization as Wahl. An earlier attempt to fit each chain by distributions with individual parameters failed, because many of the chains had such sparse or inaccurate data that the resulting parameters were clearly unphysical. Consequently, it was necessary to restrict the number of fitted quantities by requiring, like Wahl, that the Gaussian parameters for the different mass chains be relatively simple functions of mass number.

The fractional independent yields  $f(A,Z)$ , (summing over isomers), are modeled by:

$$f(A,Z) = \frac{1}{2} F(A,Z) \times N(A) \left\{ \operatorname{erf} \left[ \frac{Z - Z_p(A) + \frac{1}{2}}{\sigma_z \sqrt{Z}} \right] - \operatorname{erf} \left[ \frac{Z - Z_p(A) - \frac{1}{2}}{\sigma_z \sqrt{Z}} \right] \right\}$$

where  $F(A,Z)$  gives the odd-even effect,  $N(A)$  is a normalization constant to ensure summation to 1 for each mass, and  $Z_p$ , and  $\sigma_z$  are the mean and width of the Gaussian distribution without the odd-even effect.

$Z_p$  is equal to the unchanged charge distribution (UCD) prediction for most probable charge; corrected by a term  $\Delta Z(A')$ . i.e. for the high mass peak

$$Z_p(A_H) = A'_H \frac{Z_f}{A_f} + \Delta Z(A'_H)$$

and for the low mass peak :

$$Z_p(A_L) = A'_L \frac{Z_f}{A_f} - \Delta Z(A_f - A'_L).$$

Here,  $A'$  is the mean fragment mass before prompt neutron emission:  $A' = A - \bar{\nu}_p(A)$  where  $\bar{\nu}_p(A)$  is the mean number of prompt neutrons from that fragment. (Strictly the argument of  $\bar{\nu}_p$  should be  $A'$ , but approximating it with  $A$  causes little error.) For  $^{235}\text{U}$  thermal the values for  $\bar{\nu}_p(A)$  were taken from [7.2.17], and for other reactions the same function was used but renormalized to give the appropriate total  $\bar{\nu}_p$ .

The  $N(A)$  can be calculated to better than 1 part in  $10^5$  from:

$$\frac{1}{N(A)} = \frac{1}{2} [F(A, \text{even } Z) + F(A, \text{odd } Z)] + [F(A, \text{even } Z) - F(A, \text{odd } Z)] \xi(A)$$

where

$$\xi(A) = \frac{2}{\pi} e^{-\pi^2 \sigma_z^2 / 2} \cos[\pi Z_p(A)]$$

Note that the separate sums of the even  $Z$  and the odd  $Z$  fractional independent yields are respectively

$$N(A) F(A, \text{even } Z) [\frac{1}{2} + \xi(A)] \quad \text{and} \quad N(A) F(A, \text{odd } Z) [\frac{1}{2} - \xi(A)]$$

The factor  $F(A, Z)$  giving the combined proton and neutron odd-even effect is defined as follows (here  $N = A - Z$  is the neutron number):

Even $A$ , Even $Z$ (Even $N$ )	$F(A, Z) = \bar{F}_Z \bar{F}_N$
Even $A$ , Odd $Z$ (Odd $N$ )	$F(A, Z) = 1 / (\bar{F}_Z \bar{F}_N)$
Odd $A$ , Even $Z$ (Odd $N$ )	$F(A, Z) = \bar{F}_Z / \bar{F}_N$
Odd $A$ , Odd $Z$ (Even $N$ )	$F(A, Z) = \bar{F}_N / \bar{F}_Z$

The correction term to  $Z_p$ ,  $\Delta Z(A')$ , is composed of linear terms, as shown in Figure 7.2.2 below representing the heavy mass distribution.

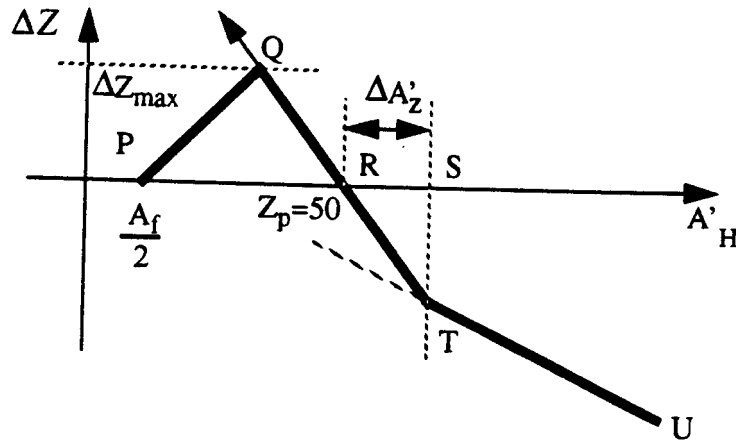


FIG. 7.2.2. Assumed Variation of  $\Delta Z(A')$  with  $A'$ .

Around the high mass peak,

$$\Delta Z(A'_H) = \Delta Z(A'_H = 140) + \left[ \frac{\delta \Delta Z}{\delta A'_H} \right] [A'_H - 140]$$

and around the low mass peak

$$\Delta Z(A'_L) = \Delta Z(A'_H = 140) + \left[ \frac{\delta \Delta Z}{\delta A'_H} \right] [A_f - A'_L - 140]$$

where the value of  $\left[ \frac{\delta \Delta Z}{\delta A'_H} \right]$  is constant.

These equations represent the straight line TU in Figure 7.2.2.

Around symmetrical fission the  $\Delta Z(A')$  and  $\sigma_z$  undergo rapid changes described in Wahl's references [7.2.17, 7.2.18]. Wahl [7.2.18] defines the variations with 3 further parameters  $\sigma_{50}$ ,  $\Delta Z_{\max}$  and  $\Delta A'_Z$ .

$\Delta Z(A')$  is assumed to be zero at symmetry  $A_f$  and at  $Z_p = 50$  (points P and R respectively). Between these points,  $\Delta Z(A')$  attains a positive maximum  $\Delta Z_{\max}$  at point Q.

The point R is determined by the value  $A'_H$  for which  $Z_p = 50$  (a closed shell for protons), if the correction term is ignored (i.e.  $\Delta Z = 0$ ). The point S is a distance  $\Delta A'_Z$  along the  $A'_H$  axis from R. T, which is vertically below, is then fixed by this value of  $A'_H$  and by the equations above of the line TU. T and R define a straight line which crosses the horizontal line  $\Delta Z = \Delta Z_{\max}$  at the point Q. Q and the point P of symmetrical fission define the final straight line of the set.

The Gaussian width  $\sigma(A)$  has either of two values. Near symmetry, between the points P and S (i.e. for  $1/2A_f \leq A'_H < A'_H(Z_p = 50) + \Delta A'_Z$ ), and in the corresponding region for light masses,  $\sigma(A) = \sigma_{50}$ . For other masses, i.e. over the regions of the chain yield peaks,  $\sigma(A) = \sigma_Z$ .

The  $Z_p$  model was initially fitted to our weighted average fractional independent and cumulative yield sets using a general non-linear least squares procedure (NAG routine E04FCF). Errors (1 standard deviation) were calculated from the  $\chi^2$  and the covariance matrix output by the routine. To simplify the fitting, the cumulative yield weighted averages were converted to sum yields. This conversion used our chain yields and the recommended experimental  $P_n$  values from [7.2.19], by removing additions to and losses from the chain due to delayed neutron emission. The converted sum yields were thus just the sum of the previous independent yields in the relevant chain and the independent yield of the nuclide itself.

The input yields to the least-squares fit were constrained to have uncertainties of 5% or greater, to stop a few highly-weighted values dominating the fit. As small yields in a chain are difficult to measure, a lower limit was set on the fractional yields that were fitted, at the point where the reduced  $\chi^2$  rapidly diverged.

For  $^{235}\text{U}$  thermal it was possible to fit all the parameters: however, for other reactions it was not possible to fit parameters  $\sigma_{50}$ ,  $\Delta Z_{\max}$  or  $\Delta A'_Z$  as there are no significant data in the region of near symmetric fission. Therefore the  $^{235}\text{U}$  thermal values for these parameters were used and only the 5 other parameters varied in the fits.

When this was first attempted [7.2.20] many systems were fitted. However most of the data were fractional cumulative yields near 1.0 in value (i.e. for nuclides near the stable ends of chains). Also, it was not certain whether complete  $P_n$  values were available for some chains with small yields. It was therefore decided to ignore the fractional cumulative yields. This left only 4 systems with sufficient data to be fitted, but the uncertainties on the derived parameters were considerably reduced. The results for the 4 fission reactions are given in Table 7.2.3.

TABLE 7.2.3. PARAMETERS FOR THE WAHL  $Z_p$  MODEL

Parameter	$^{233}\text{U}$ T	$^{235}\text{U}$ T	$^{239}\text{Pu}$ T	$^{233}\text{U}$ F
$\Delta Z(A'=140)$	-0.511	-0.523	-0.4854	-0.3758
$\pm$	0.013	0.012	0.0076	0.0205
$\delta\Delta Z/\delta A'$	-0.0153	-0.0078	-0.0143	-0.01883
$\pm$	0.0027	0.0017	0.0014	0.00446
$\overline{\sigma_Z}$	0.5735	0.5460	0.5506	0.5206
$\pm$	0.0104	0.0112	0.0080	0.0238
$\overline{\sigma_{50}}$	(0.35)	0.35	(0.35)	(0.35)
$\pm$		0.02		
$\overline{F_Z}$	1.267	1.2776	1.144	1.121
$\pm$	0.031	0.026	0.015	0.036
$\overline{F_N}$	1.1075	1.077	1.0509	1.002
$\pm$	0.025	0.022	0.0139	0.031
$\Delta A'_Z$	(0.941)	0.941	(0.941)	(0.941)
$\pm$		0.260		
$\Delta Z_{\max}$	(0.693)	0.693	(0.693)	(0.693)
$\pm$		0.238		
reduced $\chi^2$	3.8	2.61	1.86	2.08
minimum fit used	0.005	0.05	0.05	0.1
number of measurements	132	145	81	42

Values in brackets are derived from fits for  $^{235}\text{U}$  T only (for other systems data are insufficient to fit these parameters and the  $^{235}\text{U}$ T values are assumed)

$$\text{reduced } \chi^2 = \left\{ \frac{1}{\text{no. of points}} \sum_i \left[ \frac{(\text{EXPT}_i - \text{CALC}_i)}{\sigma_i^2} \right]^2 \right\}^{0.5}$$

For  $^{235}\text{U}$  thermal, probably the most measured system, there was a total of 145 values after data for isomers had been combined. Of these, 47 were discrepant. However the agreement between fit and measurement is good for most masses, particularly for results for the low mass peak where considerable data is available from recently-developed mass separator measurements.

It is difficult to extrapolate this model to other systems, due to the small group of systems fitted (3 thermal and 1 fast) and the large associated errors in the parameters. The  $\sigma_Z$  are similar and show no apparent trend, so the average of 0.554 was used. The  $F_Z$  shows a trend in fissioning charge and energy, and we assume a value of 1.0 at 14 MeV. Using the values for two atomic numbers (92 and 94) and 2 energies (thermal and fast), we assume:

$$\begin{aligned} \overline{F_Z}(\text{thermal \& spont.}) &= \begin{cases} 1.2723 - 0.06415(Z_f - 92) & \text{if } Z_f \leq 96 \\ 1 & \text{if } Z_f \geq 97 \end{cases} \\ \overline{F_Z}(\text{fast}) &= 0.88 \overline{F_Z}(\text{thermal}) \\ \overline{F_Z}(\text{high energy}) &= 1.0 \end{aligned}$$

For  $\bar{F}_N$  the value becomes 1.0 for fast and 14 MeV energies, the thermal values showing a charge dependency:

$$\bar{F}_N(\text{thermal\& spont.}) = 1.0758 - 0.01245(Z_f - 92)$$

The parameters  $\Delta Z(A' = 140)$  and  $\frac{\delta \Delta Z}{\delta A'_H}$  are small additions to the UCD estimate and thus means of the fitted values were chosen as being reasonably adequate, that is  $-0.494$  and  $-0.01266$  respectively.

The fractional independent data for all the systems of interest were then produced using the parameters either from the fit where appropriate or from the above extrapolations. For most mass chains, the agreement between calculated and measured data is moderately good: the exceptions are sufficiently few for them to be studied in detail in the next round of evaluation.

#### 7.2.2.5.5. Ternary yields

The main requirement in UKFY2 for ternary yields was for yields for  $^4\text{He}$  and  $^3\text{H}$  for all the systems. The yield of  $^4\text{He}$  at different incident neutron energies shows little discernible difference from that for thermal neutron fission for  $^{233}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , and therefore the yields were considered energy independent. This disagrees with Thomas and Whetstone's work described by Madland and Stewart [7.2.21] which suggested a linear dependence on the excitation energy of the system. The graphs in [7.2.6] show possible structure but within the uncertainties of the current measurements.

For extrapolation to nuclides for which there were no ternary yield measurements, two methods described in [7.2.21] were tried. These fit the yield as linear functions of respectively (i)  $Z_f^2/A_f$  and (ii)  $(4Z_f - A_f)$ . Figures 24 and 25 in [7.2.6] show the data for  $^4\text{He}$  plotted against these functions. Choosing  $Z_f^2/A_f$  as the independent variable gave the better fit and thus this was used for prediction of missing yields. The least-squares line is given by:

$$Y(^4\text{He}) = 0.0647 Z_f^2/A_f - 2.1292$$

where a reasonable estimate of the uncertainty in  $Y(^4\text{He})$  is  $\pm 20\%$ .

For  $^3\text{H}$  both variables were tried but it is difficult to justify anything other than a constant ratio of  $^3\text{H}$  to  $^4\text{He}$  yields. Therefore,

$$Y(^3\text{H}) = 0.06554 Y(^4\text{He})$$

was used for unmeasured tritium yields. A reasonable estimate of the uncertainty in the predicted  $Y(^3\text{H})$  is  $\pm 25\%$ .

The energy dependence of  $^3\text{H}$  yields remains uncertain, there being considerable discrepancies between the few measurements available. Tritium yield against energy for  $^{235}\text{U}$  and  $^{239}\text{Pu}$  include considerable discrepancies. Most of the differences are due to the method of separating ( $\alpha$  particles and tritons; early work considered only the energy of the fragment,

but more recent work by, for example, Ouasti [7.2.22] measured also the energy loss, allowing different particles to be more precisely distinguished. Ouasti's results show very little variation with energy, but no measurements were made between thermal energy and 0.5 MeV, which is the most important region for averaging over a "fast" neutron spectrum.

#### 7.2.2.6. Fitting to constraints

Using the evaluation methods described above, a library in ENDF-5 format [7.2.23] was produced. However, as was pointed out above, there are constraints that must apply to the yields due to the conservation of nucleons and charge:

$$\begin{aligned}\sum_A Y(A) &= 2 \\ \sum_A A \times Y(A) &= A_f - \bar{\nu}_p \\ \sum_{ZA} Z \times f(A, Z) \times Y(A) &= \sum_A Z_p(A) \times Y(A)\end{aligned}$$

The first two conditions together imply the third.

Also there should be equality of yields of complementary elements:-

$$\sum_A f(A, Z) \times Y(A) = \sum_{A'} f(Z_f - Z, A') \times Y(A') \quad \text{for all } Z < Z_f/2$$

These relationships ignore ternary fission, which introduces errors of less than 1%.

As in the previous evaluation UKFY1, the first three constraints were applied for all A. However, whereas in UKFY1 the fourth was applied for the 15 pairs of elements nearest symmetry, in UKFY2 this was applied to those pairs of elements with the greatest yields. The number of pairs was increased until the reduced  $\chi^2$  did not excessively exceed unity or, where reduced  $\chi^2$  was always greater than 1.0 the number of pairs was chosen to be that which gave a minimum value.

The values of  $\bar{\nu}_p$ , used in the fitting are given in [7.2.6]. The number of elements pairs fitted and the subsequent  $\chi^2$  per degrees of freedom and summation of fitted element yields are given in Table 6 of [7.2.6].

The details of the least-squares fit are given in the Appendix to [7.2.6]. It gives, of course, not only the adjusted yields for each fissioning system but also their covariance matrix. The diagonal terms of each matrix give the standard deviations which are required by the ENDF-5 format, but the complete matrices (39 symmetric matrices each of order approximately  $900 \times 900$ ) would be too large to store in the library. However the fitting program FITFYS is quick enough for it to be reasonably incorporated as a sub-routine in any program that needs the covariances; the authors can supply both the code and the input (unadjusted) yields and uncertainties if requested.

The adjustments in both chain and independent yields were mostly less than one standard deviation. Figures 31-34 of [7.2.6] show examples of the adjustments in chain yields relative to the experimental (and predicted) chain yields, and Figs 35 and 36 of Ref. [7.2.6]

display the distributions of the adjustments relative to the standard deviations for all chain yields and all fractional independent yields respectively. Except for a few extreme cases in the latter, with changes greater than 2 standard deviations, the adjustments will be seen to be relatively small.

It is worth noting that the formulae for interpolation and extrapolation of chain and fractional independent yields described above are designed to satisfy the first three constraints; consequently it was generally the case that greater adjustments were needed for those fissioning systems that had more measured yields and thus more experimental noise in the data.

#### 7.2.2.7. Isomeric yields

Except in the initial interrogation and averaging of the database, the evaluated yields have been those of complete nuclides and we have ignored the division of the independent yield of a nuclide between its isomers. To allow for such a division for a nuclide with excited state m and ground state g, we define the isomeric yield ratio  $r(A,Z)$ ;

$$r(A,Z) = \frac{y(A,Z,m)}{y(A,Z,m) + y(A,Z,g)}$$

Where possible, experimental data were used for the  $r(A,Z)$ . These are of two types. Preference was given to direct measurements of the ratio, as these are free of normalization problems. Failing these, the independent yield evaluations contained a number of pairs of isomers for each of which there was an averaged value, so that the above equation could be used to calculate  $r$ . However, measurements of either kind were found to be sparse, so that recourse had to be made to theory. We used the method of Madland & England [7.2.9], as that requires minimal information (the spins of the excited and ground states) about the nuclide; even that was sometimes uncertain, but generally we relied on the spin data in a preliminary version of JEF2.

Nuclides with 3 isomeric states (including the ground state) were considered using an extension of the Madland & England model [7.2.9].

#### 7.2.2.8. The calculation of cumulative yields

The concept of the cumulative yield  $c(A,Z,I)$  of the nuclide  $(A,Z,I)$  has been introduced already, where two equivalent definitions were given. Although these definitions may be applied with no difficulty to short-lived fission products, there is some ambiguity with the decay products of nuclides having very long half lives. First, it is reasonable to ignore the very long-lived  $\alpha$  decays ( $^{144}\text{Nd}$  [half-life =  $2.4 \times 10^{15}$  years] and  $^{147}\text{Sm}$  [half-life =  $10^{11}$  years]), as these will lead to an apparent approximate doubling of the cumulative yields of the daughters ( $^{140}\text{Ce}$  and  $^{143}\text{Nd}$ ) which, because of the times involved, is absurd. Other similarly long-lived  $\beta$ -decays should also have been removed but are still in the present library. These however are easily detected; greater problems arise with half-lives of say 1-100 years, i.e. of the same order as irradiation or storage times. Because of these problems we would recommend that cumulative yields should be used only rarely; the safer method is to use independent yields with an inventory code that treats correctly all relevant types of decay, and also permits the

consideration of neutron capture. As has already been stated, cumulative yields are useful in some special applications: in delayed neutron calculations, only short-lived nuclides are of importance. In calculations of total energy emission, corrections should be made for the contribution emitted after long decay times.

From the definition given of cumulative yields,

$$c(A, Z, I) = y(A, Z, I) + \sum_{(A', Z', I')} b(A', Z', I' \rightarrow A, Z, I) \times c(A', Z', I')$$

where  $b(A', Z', I' \rightarrow A, Z, I)$  is the fraction of decays of  $(A', Z', I')$  that go to  $(A, Z, I)$ .

As has already been explained, the rare and slow  $\alpha$  decays are ignored in this context. This leaves two complications that prevent a cumulative yield equaling simply the sum of independent yields up to and including the nuclide under consideration. First, the occurrence of isomers splits decay chains. Secondly,  $(\beta^-, n)$  decays destroy to a small extent the independence of individual mass chains. If it were not for these delayed neutron decays, the cumulative yield of the stable nuclide at the end of a mass chain would equal the sum of independent yields along the chain; their presence means that there are small differences between the two, which were tabulated first for the earlier UKFY1 evaluation in [7.2.4] and for UKFY2 in [7.2.6]. In using the above equation, note must also be taken of the small fraction of fission products that decay by other modes which change the mass of the nucleus.

All these types of decay can be considered in sequence if we number each fission product so that each decay goes to a nuclide with a larger number than the parent. This entails:

- (1) Ordering by decreasing mass number  $A$ .
- (2) Ordering  $\beta^-$  nuclides by increasing  $Z$ , and  $EC/\beta^+$  nuclides by Stable nuclides decreasing  $Z$ . come last in each chain.
- (3) Ordering ground states after first isomeric states after second isomeric states, etc.

Then, if we denote nuclides numbered in such a sequence by  $i, j, \dots$  the above equation becomes:

$$c_i = y_i + \sum_j b_{j \rightarrow i} \times c_j$$

or in matrix form (with a bar denoting a transposed matrix):

$$\mathbf{c} = \mathbf{y} + \bar{\mathbf{b}}\mathbf{c}$$

Since the only non-zero values of  $b_{j \rightarrow i}$  are those for which  $j < i$ , this equation can be readily used to compute cumulative from independent yields. However, the calculation of the variances (and covariances) of the cumulative yields from those of the independent yields is more complicated. From the above

$$(\mathbf{I} - \bar{\mathbf{b}})\mathbf{c} = \mathbf{y}$$

or

$$\mathbf{c} = \mathbf{Q}\mathbf{y}$$

where

$$\mathbf{Q} = (\mathbf{I} - \bar{\mathbf{b}})^{-1}$$

Then if a change  $\delta y$  in  $y$  produces a change  $\delta c$  in  $c$ ,



$$\langle \delta c \times \delta \bar{c} \rangle = Q \langle \delta y \times \delta \bar{y} \rangle \bar{Q}$$

In particular, the variance of  $c_i$  is given by:

$$\langle \delta c_i^2 \rangle = \sum_{j,k} Q_{i,j} \times Q_{i,k} \langle \delta y_j \times \delta y_k \rangle$$

The covariances of the independent yields  $y_i$  are calculated by the adjustment program FITFYS described in the appendix of [7.2.6]. The matrix  $Q$  can be calculated from radioactive decay branching ratios; its definition in above implies that it is a lower triangular matrix with  $Q_{ij} = 0$  if  $j > i$ , diagonal terms  $Q_{ii} = 1$ , and, for  $j < i$ ,

$$Q_{ij} = \sum_{k=j+1}^{k=i} b_{j \rightarrow k} Q_{ik}$$

If, for a given value of  $i$ ,  $Q_{ij}$  is calculated successively for  $j$  the values of  $Q_{ik}$  on the right hand side of this equation will already be known. At most 30 nuclides will contribute to the variance of  $c_i$  so it suffices to calculate and store  $Q_{i(i-m)}$ , where  $m = \text{Min}(i, 30)$ .

#### 7.2.2.9. Production of the UKFY2 and JEF2.2 libraries

Following the completion of UKFY2 in 1990, the library was submitted to the JEF Technical Working groups. In July 1993, a revision of the library was produced using the decay data branching ratios from the final JEF2.2 decay data. However, in producing this library it became apparent that some short-lived nuclides were not present in the JEF2.2 decay data, some of these contributed greater than 10% of the yields in some mass chains for some fissioning systems. Thus a correction term was applied to adjust the independent yields such that a calculation of the chain yields would arrive at the recommended chain yield values.

### 7.2.3. UKFY3

The UKFY3 evaluation and associated studies are fully described in [7.2.24]. This section briefly summarizes the main differences between the UKFY2 and UKFY3 evaluations. The reader is directed to [7.2.24] for a fuller explanation of the studies undertaken and the results and conclusions of these.

#### 7.2.3.1. Experimental database

The experimental database was increased in size from the 12958 measurements in UKFY2 (1989) to 14710 for UKFY3 (1993). The major sources of new data were new data added to CINDA and EXFOR (see Section 3.1), especially important was the addition by V. McLane of the NNDC, Brookhaven, USA of references from [7.2.25].

Continued checking of the UK Database and comments in private communications from Rider and Wahl (as part of the CRP) removed many remaining discrepancies from the Database.

#### 7.2.3.2. Analysis of data

For UKFY3 the same general analysis procedures were applied as for UKFY2. This included the use of the normalised residual technique to down-weight discrepant data.

However, to use as much of the data as possible a recursive procedure was applied to the data. For UKFY2 the analysis was applied to the yield measurements reported per fission. Then these values were used to convert relative and ratio of ratio data to per fission. For UKFY3 this procedure was repeated until no further data could be converted and all the recommended yield values had converged.

### 7.2.3.3. Chain yields

In UKFY3 the 5 Gaussian model was used, and the new experimental data fitted. The results are shown in Table 7.2.4. An improvement to the method was to include the symmetrical fission coefficient,  $N_3$ , in the fit and to calculate  $N_2$  from the conservation laws. This gave better fits in the valley region, as the  $N_3$  parameter was being fitted rather than calculated from the difference of much larger terms whose uncertainties were greater than the value of  $N_3$ .

In the production of complete chain yield sets the method was refined. Firstly the experimental data was used. Then either the fits in Table 7.2.4 with a 15% uncertainty or the  $A_f$  functions developed for UKFY2 were used with a 30% uncertainty. To reduce discontinuities in the data the fits were normalized to the available experimental data. At the wings of the distribution the calculations were normalized to the first experimental point. In gaps in the distribution the calculations were adjusted by multiplying by a normalization factor, the log of which was linearly interpolated across the gap.

TABLE 7.2.4. PARAMETERS OF FIVE GAUSSIAN MODEL FITTED FOR UKFY3 DATA

System	$\bar{A}$	$\sigma_3$	$N_3$	$D_1$	$\sigma_1$	$N_1$	$D_2$	$\sigma_2$
U232T	114.95	7.793	0.12348	25.622	3.3131	0.58928	18.165	2.5814
U233T	115.77	12.403	0.0035518	25.173	4.0843	0.63013	18.106	2.8934
U235T	116.90	9.2523	0.0017926	24.020	4.2980	0.71541	16.63	2.4233
Np237T	117.65	11.999	0.0019580	23.021	4.8979	0.71223	16.347	2.2646
Pu238T	118.07	14.013	0.057234	22.344	4.3587	0.52996	15.465	2.5792
Pu239T	118.56	5.0556	0.0026530	22.019	5.4481	0.62909	15.529	2.8419
Th232F	115.55	12.583	0.011459	25.777	3.7309	0.76299	18.748	2.3108
U233F	115.73	12.583	0.010323	25.149	4.117	0.60318	18.054	3.2895
U235F	116.90	9.2473	0.0048303	24.02	4.2996	0.71242	16.629	2.4223
U236F	117.26	12.357	0.0093219	23.506	4.6746	0.69363	16.698	2.6646
U238F	118.25	11.091	0.0020110	22.573	5.0631	0.70937	15.923	2.3411
Np237F	117.62	5.0262	0.0031789	22.984	5.1300	0.68104	16.316	2.822
Pu239F	118.53	13.11	0.0084215	22.054	5.5834	0.59505	15.557	3.0172
Pu240F	118.98	11.908	0.0054060	21.687	5.5375	0.64734	15.205	2.7563
Am241F	119.41	18.042	0.010871	21.271	6.1518	0.68763	14.821	3.0097
Th232H	114.63	17.959	0.26599	25.116	4.9097	0.64460	19.822	2.1401
U233H	115.06	8.7537	0.15569	24.879	6.0685	0.23376	20.061	5.7029
U235H	115.78	10.625	0.14799	23.901	5.722	0.43167	18.751	5.3988
U238H	117.29	11.605	0.14141	22.626	5.9625	0.63919	15.935	3.0685
Np237H	117.00	14.988	0.22453	22.93	5.8876	0.53814	16.800	5.0347
Pu239H	117.67	19.364	0.28772	22.256	6.5284	0.47125	14.868	3.7989
Pu240H	117.88	13.295	0.10671	21.765	6.6275	0.54892	14.996	4.4052
Pu242H	118.78	13.091	0.16488	20.819	6.3737	0.67228	14.811	2.3789
Am241H	118.32	12.168	0.26029	21.034	7.6984	0.52527	14.551	4.043

#### 7.2.3.4. Ternary yields

The new data and methods used for UKFY3 are fully described in Section 4.4.

#### 7.2.3.5. Fractional independent yields

As for UKFY2, the  $Z_p$  model of Wahl [7.2.17] was used to fit the new experimental data. Private communications from Wahl showed that in the UKFY2 work the  $DA'_Z$  parameter had been incorrectly interpreted. Fortunately, when the model and programs were corrected the differences in calculated yields and goodness of fit were not significant.

The increased data in the UKFY3 database allowed six fissioning systems to be fitted. The results are given in Tables 7.2.5 and 7.2.6.

To produce complete sets of fractional independent yields for the above six fissioning systems the above parameters were used.

For other systems it is necessary to interpolate or extrapolate. The parameters  $\frac{\delta\Delta Z}{\delta A'_H}$ ,  $\bar{\sigma}_Z$ ,  $\bar{\sigma}_{50}$ ,  $\bar{F}_N$ ,  $\Delta A'_Z$ , and  $\Delta Z_{\max}$  did not show significant variation, and thus weighted means of  $-0.01$ ,  $0.5$ ,  $0.4$ ,  $1.06$ ,  $0.7$  and  $0.6614$  respectively were used. The other two parameters,  $\bar{F}_Z$  and  $\Delta Z(A' = 140)$ , did show significant variation.

$\bar{F}_Z$  appeared to be constant at  $1.05$  for fast and high energy fission, but for thermal fission was approximated by:

$$\bar{F}_Z = 1.29 - 0.07(Z_f - 92) \quad \bar{F}_Z \geq 1.0$$

In a similar fashion  $\Delta Z(A' = 140)$  shows variation with nuclear charge, which can be approximated for different fissioning systems by the following:

$$\begin{aligned} \Delta Z(A' = 140) &= -0.55 + 0.03 (Z_f - 92) \quad \text{for thermal} \\ &= -0.42 + 0.03 (Z_f - 92) \quad \text{for fast} \\ &= -0.23 + 0.03 (Z_f - 92) \quad \text{for high energy} \\ &= -0.55 + 0.03 (Z_f - 92) \quad \text{for spontaneous} \end{aligned}$$

#### 7.2.3.6. Independent yield isomeric splitting

Although considerable improvements have been reported to the CRP in the modeling of isomeric splitting, e.g. Rudstam [7.2.26] and more recent private communications, this requires two parameters for each fissioning system. So far these parameters have only been determined for  $^{235}\text{U}$  thermal neutron fission and there is no current method of determining their values for other systems. Also the experimental measurements contain a considerably number of discrepancies. Thus it was decided to use the Madland & England model [7.2.9] with the currently available JEF2.2 decay data to produce the independent isomeric splitting factors for UKFY3, as was used for UKFY2.

TABLE 7.2.5. PARAMETERS FROM FITTING  $^{235}\text{U}$  AT THERMAL, FAST AND HIGH ENERGIES

Parameter	U235T	U235F	U235H
$\Delta Z(A' = 140)$	$-0.5329 \pm 0.009$	$-0.418 \pm 0.1$	$-0.229 \pm 0.1$
$\frac{\delta \Delta Z}{\Delta A'_H}$	$-0.0066 \pm 0.0001$	$-0.022 \pm 0.001$	$-0.010 \pm 0.001$
$\overline{\sigma}_Z$	$0.5368 \pm 0.0042$	$0.503 \pm 0.002$	$0.620 \pm 0.009$
$\overline{\sigma}_{50}^a$	$0.3449 \pm 0.023$	(0.3449)	$0.169 \pm 0.005$
$\overline{F}_Z$	$1.294 \pm 0.024$	$1.046 \pm 0.02$	$1.05 \pm 0.02$
$\overline{F}_N$	$1.077 \pm 0.02$	$1.046 \pm 0.02$	$1.06 \pm 0.02$
$\Delta A'_Z{}^a$	$0.70 \pm 0.43$	(0.7)	(0.7)
$\Delta Z_{\max}{}^a$	$0.6614 \pm 0.64$	(0.6614)	(0.6614)
reduced $\chi^2$	2.69	2.02	2.29
minimum fity	—	0.05	—
number of measurements	210	48	30

<sup>a</sup> Values in brackets could not be fitted directly and the U235 thermal induced neutron fission results were used with these kept constant during the fit.

TABLE 7.2.6. PARAMETERS FROM FITTING  $^{233}\text{U}$ ,  $^{239}\text{Pu}$  AND  $^{229}\text{Th}$  YIELDS FOR THERMAL NEUTRON FISSION

Parameter	U233T	Pu239T	Th229T
$\Delta Z(A' = 140)$	$-0.573 \pm 0.01$	$-0.493 \pm 0.05$	$0.713 \pm 0.01$
$\frac{\delta \Delta Z}{\Delta A'_H}$	$-0.014 \pm 0.003$	$-0.013 \pm 0.002$	$-0.011 \pm 0.001$
$\overline{\sigma}_Z$	$0.571 \pm 0.004$	$0.566 \pm 0.002$	$0.5221 \pm 0.0002$
$\overline{\sigma}_{50}^a$	$0.47 \pm 0.07$	$0.566 \pm 0.002$	(0.3449)
$\overline{F}_Z$	$1.259 \pm 0.02$	$1.049 \pm 0.02$	$1.60 \pm 0.02$
$\overline{F}_N$	$1.054 \pm 0.02$	$1.051 \pm 0.02$	$1.07 \pm 0.02$
$\Delta A'_Z{}^a$	(0.7)	(0.7)	(0.7)
$\Delta Z_{\max}{}^a$	(0.6614)	(0.6614)	(0.6614)
reduced $\chi^2$	3.16	2.13	1.67
minimum fity	0.05	0.05	0.05
number of measurements	100	85	48

<sup>a</sup> Values in brackets could not be fitted directly and the U235 thermal induced neutron fission results were used with these kept constant during the fit.

#### 7.2.3.7. Future work on UKFY3

In the preliminary version of UKFY3, produced in 1993, similar procedures to UKFY2 were used with the above described improvements. There remains the problem of missing decay data for some significant nuclides for which adjustments had to be made. The update to the JEF2.2 decay data for JEFF3 will include the addition of this missing data (evaluation by Dr. A.L. Nichols and funded by BNF plc.). When this is completed the UKFY3 library will be reprocessed using the new decay data.

#### 7.2.4. Conclusions

Recent UK work in the area of fission product yield evaluation has been described. The chief remaining task is the testing of the new libraries by comparison with others, and also by calculation of decay heat and of delayed neutron emission rates. Some preliminary tests have been carried out and are described in [7.2.24].

### ACKNOWLEDGEMENTS

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### 7.3. THE CENDL FISSION YIELDS LIBRARY

#### 7.3.1. History of the CENDL library

The evaluation of fission yields was started by Wang Dao et al. in China in 1976 and led to the production of a preliminary file called CENDL fission yield library (CENDL/FPY) in 1987. The file contains 10 fission reactions:  $^{235}\text{U}(\text{T})$ ,  $^{235}\text{U}(\text{F})$ ,  $^{235}\text{U}(\text{H})$ ,  $^{238}\text{U}(\text{F})$ ,  $^{238}\text{U}(\text{H})$ ,  $^{239}\text{Pu}(\text{T})$ ,  $^{239}\text{Pu}(\text{F})$ ,  $^{241}\text{Pu}(\text{T})$ ,  $^{233}\text{U}(\text{T})$ , and  $^{232}\text{Th}(\text{F})$ , each of them including 1170 fission product nuclides, of which 152 are stable. Among the unstable ones there are 192 isomers. For every fission product nuclide, both independent and cumulative yields are given together with their uncertainties. The CENDL/FPY was in the ENDF-5 format.

After Wang Dao retired in 1994, his work was continued by Liu Tingjin and Liang Qichang. Since then, more than 10 important fission yields for  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^{239}\text{Pu}$  fission induced by thermal, fission spectrum, about 14 MeV and other mono-energetic neutrons have been evaluated, the evaluation of some others is in progress. Furthermore, CENDL/FPY is being improved and a new version will be produced based on the new evaluations.

### 7.3.2. Evaluation of experimental data

#### 7.3.2.1. Data sources

The sources of experimental fission yield data for evaluation in CENDL/FPY are the measurement database generated by Meek&Rider in 1980, the EXFOR database maintained by IAEA/NDS, and recently published literature.

#### 7.3.2.2. Analysis of data

For the fission product yield evaluation, one of the steps required before the processing is the analysis of experimental data to produce a set of “best estimate” recommended values. This analysis sometimes led to the yield adjustments or/and estimates of their errors.

##### 7.3.2.2.1. Yield adjustments

Depending on what corrections and adjustments have to be made, the experimental data are classified as absolute yields, relative yields, and R-values.

Absolute yields are defined as  $Y_i = N_i/N_f$ , where  $Y_i$  and  $N_i$  are the fission yield and number of atoms of an investigated nuclide  $i$  respectively;  $N_f$  is the number of fissions in the irradiated sample. Usually it is difficult to adjust the absolutely measured yields, and they even don't require or are not amenable to adjustments. Hence, the error reported is the only parameter that can be adjusted.

The relative yield is defined as  $RY_i = Y_i/Y_s$ , where  $Y_i$  and  $Y_s$  are the absolute yields of investigated and reference nuclides, respectively. Relative yields can be adjusted by using well known reference yields in order to obtain absolute fission yields; or absolute fission yields obtained from relative yields can be readjusted by using well known reference yields, if the original reference yields used by the authors are given.

An R-value is defined as  $R = (Y_i/Y_s)_1/(Y_i/Y_s)_2$ , where  $Y$  is the absolute yield;  $i$  refers to the investigated nuclide, and  $s$  to the reference nuclide; 1 denotes the investigated fission reaction, and 2 the reference fission reaction. (Revised) absolute fission yields can be obtained from published R-values by using a set of well known up-to-date (reference) fission yields; or published absolute yields obtained from unpublished R-values can be recalculated by using a set of up-to-date (reference) fission yields if the original ones used by authors are given.

Furthermore, decay corrections can be made for all types of measured yields by using reliable up-to-date half-lives, if those used by the author(s) or the raw data as well as the relevant experimental details are given.

#### 7.3.2.2.2. Error estimates

The experimental error is very important for the analysis and evaluation of data.

Measurers report the errors in a variety of ways. Usually the fission yield value is reported together with a overall uncertainty that includes all error contributions from the measurement. However, sometimes only standard deviations corresponding to the precision of the measurements, or even no errors at all are given. Therefore have the reported uncertainties sometimes to be adjusted to represent reasonable estimates of the overall errors.

The error of a relative value (either relative yield or R-value), was statistically combined with the errors in the absolute yields of the reference nuclides to give an overall error of the absolute yield.

If an absolute yield is published only with a random error, a 2% upper limit of inherent systematic error was combined with the reported one.

For different measurement techniques, the following lower limits were adopted for the overall errors:

- 20% for Geiger-counter era measurements, before 1955;
- 10% for NaI(Tl) detector era measurements between 1955 and 1965;
- 5% for Ge(Li) detector era measurements made after 1965: direct  $\gamma$  ray spectrometry, radiochemical and other special methods;
- 2% for mass spectrometric measurements;
- 30% for estimated values.

If no errors were reported, an overall error of three times the lower limit for the measuring technique involved was assigned.

If the reported positive and negative errors were different, the smaller value plus two-thirds of the difference was used.

In some cases, particularly for more recent measurements, the preset error limits are not appropriate. For example the accuracy of the direct  $\gamma$  ray spectrometry method was limited in [7.3.1] to  $\pm 10\%$  at best. However, since 1975, higher accuracies like 3% [7.3.2] have been achieved in some measurements; for this kind of data, their accuracies were directly adopted.

#### 7.3.3.2.3. Data processing

After the analysis and processing of experimental data, adjusted values were obtained for all these data, including independent and cumulative yields.

For a given group of experimental data, e.g. the cumulative yield of a certain fission product for a given incident neutron energy and fissioning nuclide, first duplicate data were eliminated. Then outliers (values that obviously deviate from the rest), which are frequently present, were identified and statistical tests were applied to decide about their inclusion.



For a set of  $n$  measurements  $x_i$  and their assessed standard deviations  $\Delta x_i$ , the weighted averages and their errors are calculated as follows:

The weighted average yield is:

$$y_w = \frac{\sum_{i=1}^n \frac{1}{(\Delta x_i)^2} x_i}{\sum_{i=1}^n \frac{1}{(\Delta x_i)^2}}$$

The internal ( $\sigma_1$ ) and external ( $\sigma_2$ ) standard deviations are, respectively:

$$\sigma_1 = \sqrt{\frac{1}{\sum_{i=1}^n \frac{1}{(\Delta x_i)^2}}}$$

and

$$\sigma_2 = \sigma_1 \times \varepsilon = \sigma_1 \sqrt{\frac{\sum_{i=1}^n \left( \frac{x_i - y_w}{\Delta x_i} \right)^2}{n-1}}$$

A useful test of the consistency of the data is the  $\chi^2$  test :

$$\chi^2 = \sum_{i=1}^n \left( \frac{x_i - y_w}{\Delta x_i} \right)^2$$

The weighted mean yield and  $\sigma_2$  were taken as the recommended value and its error.

Eventually, an evaluated experimental data file containing independent and cumulative yields was obtained, which was to be combined (see 7.3.4) with the yields obtained from model calculation, using the method of error propagation.

### 7.3.3. Model calculation of yields

#### 7.3.3.1. Calculated independent yields

Empirical charge distribution models derived from experimental data relate the systematics of nuclear charge distribution in fission to parameters of simple mathematical functions. Two empirical models have been developed [7.3.3,7.3.4]. The  $Z_p$  model was adopted for our work for historical reasons.

For a given mass chain of mass number A, the fractional independent yield of an isobar of atomic number Z is calculated by using Gaussian charge dispersion formula,

$$f(A, Z) = \frac{1}{\sigma\sqrt{2\pi}} \int_{Z-0.5}^{Z+0.5} e^{-\frac{(Z-Z_p(A))^2}{2\sigma^2}} dZ \quad (1)$$

where  $\sigma$  is the Gaussian width parameter and  $Z_p(A)$  is the most probable charge. For CENDL,  $\sigma$  was taken to be constant  $0.56 \pm 0.06$  [7.3.5] irrespective of mass number and fissioning system. The  $Z_p(A)$  values were taken from [7.3.6], they were calculated from the following equation:

$$Z_p(A) = Z_p(A, \text{standard}) + \Delta Z_p(\text{ternary}) + \Delta Z_p(A),$$

where:

$Z_p(A, \text{standard})$  is the standard value for  $^{235}\text{U}$  thermal fission [7.3.7];

$\Delta Z_p(\text{ternary})$  is the correction term for ternary fission [7.3.8], the values of which depend on fissioning systems, as shown in Table 7.3.1.

$\Delta Z_p(A)$  is the correction term for non  $^{235}\text{U}$  thermal fission [7.3.7], which is further decomposed into the following three terms:

$$\Delta Z_p(A) = a(Z_c - 92) + b(A_c - 236) + c(E^* - 6.52)$$

where  $Z_c$  and  $A_c$  are the charge and the mass number, respectively, of the compound nucleus, and  $E^*$  is the excitation energy in MeV.

TABLE 7.3.1. CORRECTION FACTOR FOR TERNARY FISSION

Fissioning system	$\Delta Z_p(\text{Ternary})$	$Z_c$	$A_c$	$E^*(\text{MeV})$
$^{235}\text{U}(\text{T})$	0.05062	92	236	6.52
$^{235}\text{U}(\text{F})$	-0.10021	92	236	8.31
$^{235}\text{U}(\text{HE})$	-0.06241	92	236	21.26
$^{238}\text{U}(\text{F})$	-0.00860	92	239	6.57
$^{238}\text{U}(\text{HE})$	-0.10089	92	239	19.52
$^{239}\text{Pu}(\text{T})$	0.01846	94	240	6.50
$^{239}\text{Pu}(\text{F})$	-0.06287	94	240	8.30
$^{241}\text{Pu}(\text{T})$	-0.08924	94	242	6.27
$^{233}\text{U}(\text{T})$	0.03989	92	234	6.81
$^{232}\text{Th}(\text{F})$	0.01299	90	233	6.56

TABLE 7.3.2. VALUES OF THE COEFFICIENTS a AND b

Mass Region	a	b
Light peak products	$0.414 \pm 0.016$	$-0.143 \pm 0.007$
Valley products	$0.500 \pm 0.030$	$-0.165 \pm 0.020$
Heavy peak products	$0.547 \pm 0.010$	$-0.188 \pm 0.004$

TABLE 7.3.3. VALUES OF THE COEFFICIENT c

Mass No.	c	Mass No.	c
66–74	$0.0200 \pm 0.0030$	116	$0.0341 \pm 0.0068$
76	$0.0200 \pm 0.0025$	118	$0.0371 \pm 0.0067$
78	$0.0200 \pm 0.0025$	120	$0.0402 \pm 0.0064$
80	$0.0200 \pm 0.0020$	122	$0.0433 \pm 0.0061$
82	$0.0200 \pm 0.0016$	124	$0.0464 \pm 0.0056$
84	$0.0200 \pm 0.0016$	126	$0.0495 \pm 0.0050$
86	$0.0200 \pm 0.0016$	128	$0.0505 \pm 0.0041$
88	$0.0196 \pm 0.0012$	130	$0.0509 \pm 0.0031$
90	$0.0189 \pm 0.0011$	132	$0.0500 \pm 0.0025$
92	$0.0181 \pm 0.0011$	134	$0.0472 \pm 0.0024$
94	$0.0171 \pm 0.0010$	136	$0.0416 \pm 0.0021$
96	$0.0160 \pm 0.0010$	138	$0.0328 \pm 0.0016$
98	$0.0157 \pm 0.0009$	140	$0.0276 \pm 0.0014$
100	$0.0159 \pm 0.0013$	142	$0.0241 \pm 0.0012$
102	$0.0173 \pm 0.0017$	144	$0.0217 \pm 0.0013$
104	$0.0190 \pm 0.0023$	146	$0.0203 \pm 0.0016$
106	$0.0211 \pm 0.0030$	148	$0.0195 \pm 0.0019$
108	$0.0233 \pm 0.0037$	150	$0.0195 \pm 0.0023$
110	$0.0257 \pm 0.0046$	152	$0.0195 \pm 0.0027$
112	$0.0283 \pm 0.0057$	154	$0.0195 \pm 0.0031$
114	$0.0311 \pm 0.0062$	156	$0.0195 \pm 0.0035$
		158–172	$0.0195 \pm 0.0039$

The values of the coefficients a, b, and c are shown in Tables 7.3.2 and 7.3.3.

Considering the existence of isomeric [7.3.9] and odd-even pairing [7.3.10, 7.3.11] effects of fission product nuclides, the calculated fractional independent yields from Eq. (1) should be modified as follows :

- (1) The odd-even effects in proton- and neutron-pairing cause deviations from the Gaussian charge dispersion curve shape described by Eq. (1). Let  $F_x(A,Z)$  be a correction factor for the odd-even effect, then the actual fractional independent yield is approximated by

$$Y(A,Z) = F_x(A,Z) f(A,Z) \quad (2)$$

where  $F_x(A,Z)$  is expressed as :

$$\begin{aligned}
 F_x(A,Z) &= 1 + (p + n), \text{ for even } A - \text{even } Z \\
 &= 1 + (p - n), \text{ for odd } A - \text{even } Z \\
 &= 1 - (p - n), \text{ for odd } A - \text{odd } Z \\
 &= 1 - (p + n), \text{ for even } A - \text{odd } Z
 \end{aligned}$$

where  $p$  is the proton-pairing term and  $n$  is the neutron-pairing term. The magnitudes of both parameters, which are shown in Table 7.3.4., depend on the fissioning system.

- (2) All the adjusted fractional independent yields in a given mass chain are normalized to the corresponding chain yield in order to obtain the absolute independent yield of every isobar.

TABLE 7.3.4 VALUES OF PARAMETERS FOR PAIRING EFFECTS

Fissioning system	$p$	$n$
$^{235}\text{U(T)}$	$0.228 \pm 0.034$	$0.044 \pm 0.034$
$^{235}\text{U(F)}$	$0.151 \pm 0.179$	$0.029 \pm 0.039$
$^{235}\text{U(HE)}$	$0.015 \pm 0.016$	$0.003 \pm 0.004$
$^{238}\text{U(F)}$	$0.329 \pm 0.479$	$0.063 \pm 0.100$
$^{238}\text{U(HE)}$	$0.018 \pm 0.019$	$0.003 \pm 0.004$
$^{239}\text{Pu(T)}$	$0.171 \pm 0.206$	$0.033 \pm 0.044$
$^{239}\text{Pu(F)}$	$0.124 \pm 0.143$	$0.024 \pm 0.031$
$^{241}\text{Pu(T)}$	$0.206 \pm 0.256$	$0.040 \pm 0.055$
$^{233}\text{U(T)}$	$0.210 \pm 0.264$	$0.041 \pm 0.056$
$^{232}\text{Th(F)}$	$0.327 \pm 0.469$	$0.063 \pm 0.098$

### 7.3.3.2. Calculated cumulative yields

In this library, all the decay chains were taken from the JNDC Nuclear Data Library of Fission Products [7.3.12].

For a specified decay chain - containing the decay branchings of beta decays, positron emissions, isomeric transitions, and delayed neutron emissions, on the basis of the well-known chain yield and the above-mentioned independent yields of all the isobars - the cumulative yields were calculated by:

- (1) deducing the cumulative yield of the initial nuclide by subtracting the independent yields of all the isobars except the initial one from the chain yield;
- (2) calculating the cumulative yield of each isobar by adding its independent yield to the cumulative yield of the precursor, starting with the initial chain member.

For every one of 1170 fission product nuclides of a fissioning system, both independent and cumulative yields were calculated, not only those which have no experimental data. This way, two calculated yield data files, one each for independent and cumulative yields, were created, which were combined with the evaluated experimental data described in Section 7.3.2, using statistical and error propagation methods.

### 7.3.4. Production of recommended values for CENDL/FPY

#### 7.3.4.1. Recommended independent yields

Calculated independent yields were assigned large (100%) errors. Where evaluated experimental independent yields exist, they were statistically combined with the calculated independent yields by taking reciprocal variances as weights, and the associated errors were calculated as described in Section 7.3.2. Where no evaluated experimental independent yields exist, the calculated independent yields and their errors were directly adopted.

After application of the above treatments to every fission product nuclide, a preliminary recommended independent yield data file was created; then, the independent yields in a given decay chain were normalized to the corresponding well-known chain yield so that the recommended independent yields were finally obtained.

#### 7.3.4.2. Recommended cumulative yields

Once the recommended independent yields had been obtained as described above, the preliminary recommended cumulative yields were calculated in the same manner as the calculated cumulative yields (Section 7.3.3.2.), and the absolute error limits given in 7.3.2.2.2. were adopted. The procedure for obtaining the recommended cumulative yields by combining the calculated cumulative yields with the evaluated experimental ones was the same as that for the recommended independent yields.

#### 7.3.4.3. Format

Both recommended independent and cumulative yields were converted into ENDF-5 format. The resultant library was checked to ensure that it conformed with the appropriate ENDF-5 conventions and physical constraints, by using the ENDF-6 utility codes (consisting of CHECKER, FIZCON, PSYCHE, etc.) developed by NNDC/BNL.

### 7.3.5. Tests and comparisons

Several integral tests and comparisons have been made [7.3.13]. First, the decay powers of fission products of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  thermal fission were calculated with the CINDER 10 code using the CENDL yields decay times of  $\sim 0\text{--}10^{13}$  seconds and a short irradiation ( $10^{-4}$  s). The results were compared to calculations using the ENDF/B-VI yields. In these calculations, the decay half-lives and branching fractions are based on ENDF/B-V data, and the decay energies are based partly on modifications made by JNDC. The graphical comparisons over the important cooling time ( $\leq 10^3$  s) of interest in the hypothetical loss-of-coolant accident are shown in Figs 7.3.1-7.3.4. For  $^{239}\text{Pu}$  thermal fission, the American Nuclear Society (ANS) Draft Decay-Heat Standard [7.3.14] is also included for comparison (Fig. 7.3.4).

In the above comparisons, a least squares fit was used to produce the  $\alpha$ ,  $\lambda$  pairs [7.3.15]. For the  $^{235}\text{U}$  thermal fission decay power ( $\beta+\gamma$ ), a fit within 1.5% at all times was obtained.

Table 7.3.5 compares the prompt neutron yields, delayed neutron yields, average charges with ENDF/B-VI data.

Recently the CENDL/FPY fission yields were compared to those from ENDF/B-VI, JENDL-3/FPY, JEF-2/FPY, and BROND-2 for about 40 important fission products from  $^{235}\text{U}$  thermal fission, where experimental data were available.

The conclusions from this inter-comparison can be summarized as follows:

- (1) The cumulative fission yields in all 5 libraries are basically in agreement for 13 fission products, that is about one third of all nuclides compared, but about two thirds of the nuclides compared are discrepant. The trends of the yield curves also show differences, especially for the product nuclides at the two wings of the mass distribution. Typical examples are  $^{88}\text{Kr}$ ,  $^{151}\text{Pm}$  etc. In general, there are more discrepancies for yields induced by fission spectrum neutrons.
- (2) Generally speaking, the data for independent fission yields are even more discrepant. The discrepancies among the absolute values comprise often one or more orders of magnitude, up to a maximum of 4 orders of magnitude. Typical examples are  $^{109}\text{Pd}$ ,  $^{111}\text{Ag}$  etc. The differences are not only in the absolute values, but also in the curve shapes (reflecting relative yields), typical examples being  $^{103}\text{Ru}$ ,  $^{133}\text{I}$ , etc.
- (3) Usually, the dependence of fission yields on incident neutron energy is assumed to be linear,  $y(E) = aE + b$ , or logarithmic-linear,  $\ln(y) = aE + b$ , particularly for products in the peak regions of the mass distribution. However, this could not be confirmed for many nuclides, even in the peak regions. Examples among the more important fission products are  $^{99}\text{Mo}$ ,  $^{133}\text{I}$ ,  $^{141}\text{Ce}$ ,  $^{147}\text{Nd}$  and  $^{156}\text{Eu}$ . The dependence is even more complicated in the keV resonance region.
- (4) Generally, the data from CENDL/FPY and JENDL-3/FPY agree better.

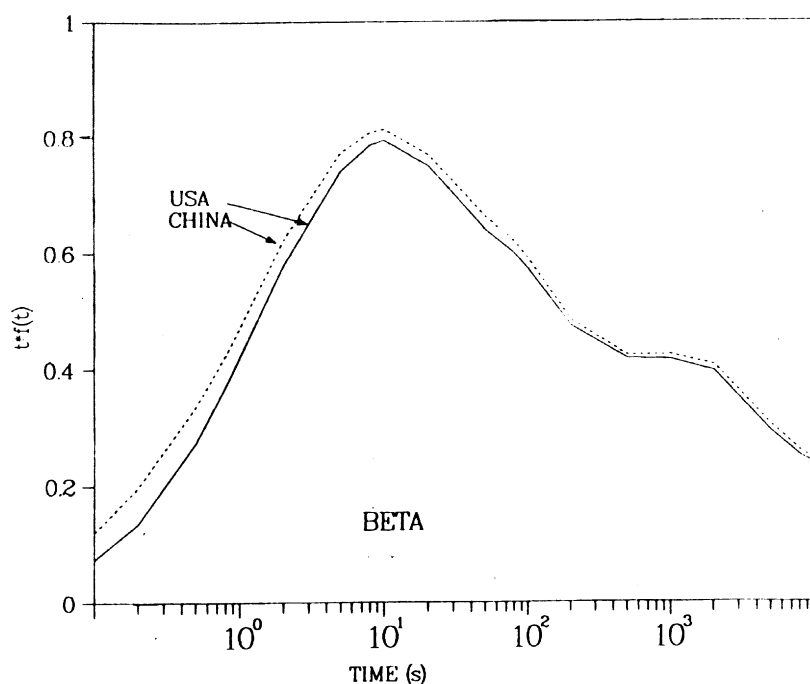


FIG. 7.3.1  $^{235}\text{U}(T)$  beta decay power comparison.

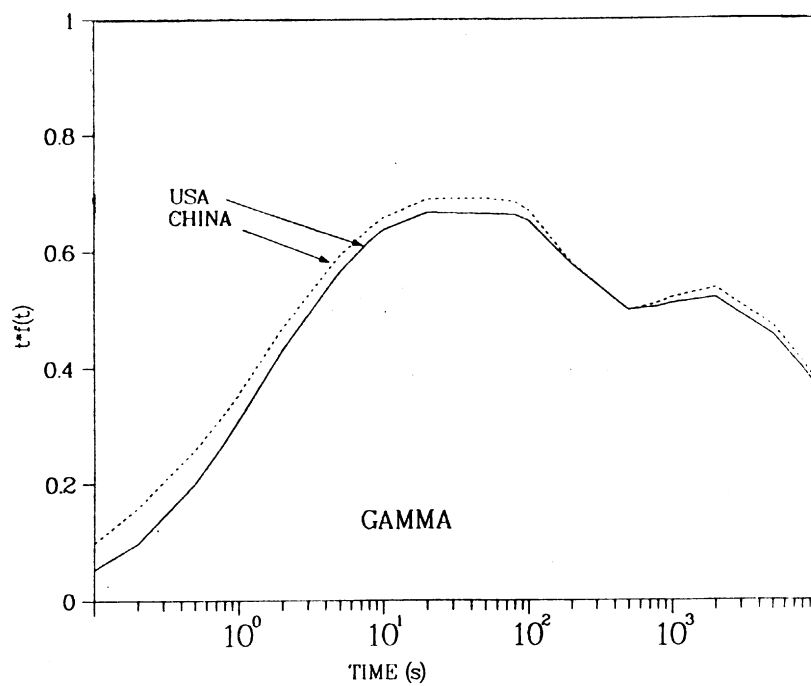


FIG. 7.3.2  $^{235}\text{U}(T)$  gamma decay power comparison.

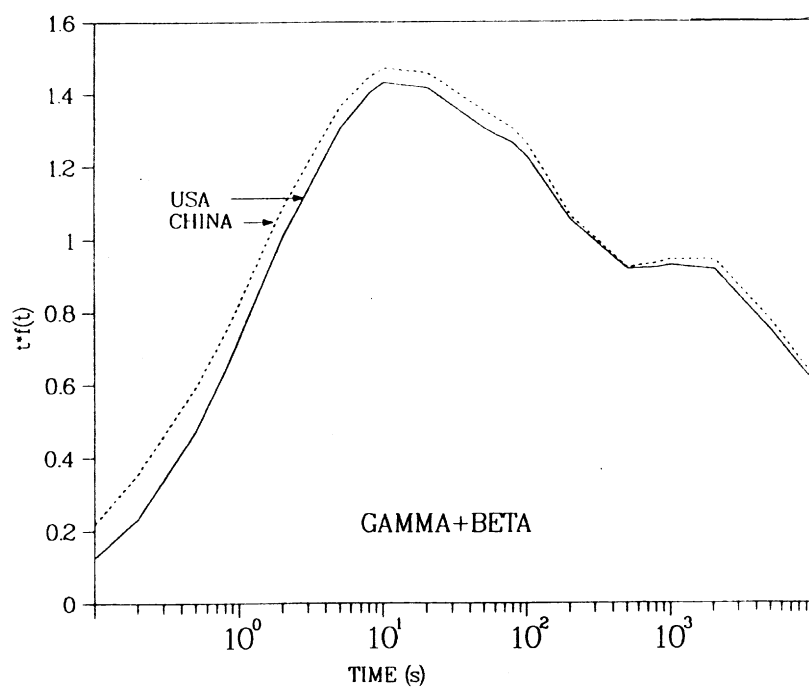


FIG. 7.3.3  $^{235}\text{U}(T)$  gamma + beta decay power comparison.

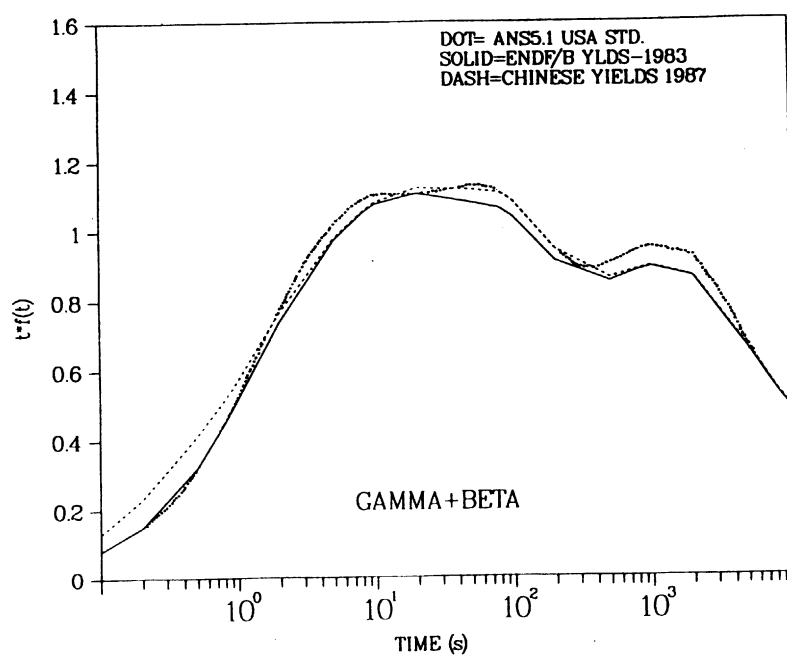


FIG. 7.3.4  $^{239}\text{Pu}(\text{T})$  gamma + beta decay power comparison.

TABLE 7.3.5 COMPARISON OF NEUTRON YIELDS

Fissioning nuclide	$\nu_{\text{total}}$		$\nu_d$ per 100 fissions		CENDL yields		
	CENDL yields	ENDF/B-VI yields <sup>a</sup>	CENDL yields	ENDF/B-VI yields <sup>a</sup>	$\Sigma Z$	$\Sigma N$	$\Sigma A$
$^{235}\text{U}(\text{T})$	2.5240	2.6402 (2.44)	2.0440	1.7845 (1.67)	92.05	141.42	233.48
$^{235}\text{U}(\text{F})$	2.3797	2.4356 (2.48)	2.4879	2.0583 (1.67)	91.94	141.68	233.62
$^{235}\text{U}(\text{H})$	4.1845	4.3582 (4.40)	1.2201	1.0870 (0.90)	92.06	139.75	231.82
$^{238}\text{U}(\text{F})$	2.924	2.8799 (2.47)	3.1272	4.0480 (4.40)	92.01	144.07	236.08
$^{238}\text{U}(\text{H})$	4.5236	4.4508 (4.43)	2.6310	2.7624 (2.60)	91.93	142.55	234.48
$^{239}\text{Pu}(\text{T})$	2.9567	2.8989 (2.89)	1.0005	0.7588 (0.65)	93.98	143.06	237.04
$^{239}\text{Pu}(\text{F})$	2.7827	2.8941 (2.95)	0.6787	0.683 (0.65)	94.01	143.21	237.22
$^{241}\text{Pu}(\text{T})$	3.0900	2.9322 (2.96)	1.6539	1.405 (1.62)	93.87	145.04	238.91
$^{233}\text{U}(\text{T})$	2.2470	2.5088 (2.50)	1.1787	0.9719 (0.74)	92.13	139.62	231.75
$^{232}\text{Th}(\text{F})$	2.8363	2.3128 (2.02)	4.9439	5.645 (5.27)	89.81	140.36	230.16

<sup>a</sup> preliminary ENDF/B-VI yields;

Values given in parentheses are taken from the ENDF/B-V evaluation and are based on measurements.



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### 7.4. YIELD EVALUATIONS FOR MODEL DEVELOPMENT

The methods that have been developed for evaluation and treatment of nuclear-charge distribution data [4.1.3] include options for:

- a) FI-IN and FC-CU interconversions using Y(A) data sets,
- b) not using data judged to be unreliable, but keeping and listing them for future evaluation,
- c) assigning evaluator's errors to be used in calculations instead of experimental errors judged to be too low,
- d) deriving FI from FC,
- e) averaging yields for each fission product,
- f) creating files of data for least-squares analysis,
- g) normalizing ( $\Sigma FI = 1.00$ ) and calculating the average  $Z$ ,  $\bar{Z}$ , and the root mean square, RMS, for each A, when there are sufficient data.

The model-independent  $\bar{Z}$  and RMS values proved useful for deriving and testing  $Z_p$  and  $A'_p$  model functions for individual fission reactions [4.1.3].

Options a, b, c, d, and f have been applied [4.2.5, 4.2.6] (see also Section 4.2) to new experimental data and to data obtained from other evaluations [4.1.4, 4.1.5] to create data files for least squares analysis. The model parameters derived for a number of fission reactions

showed systematic trends that could be represented by equations, which allowed yield estimates to be made for many fission reactions [4.2.5, 4.2.6] (see also Section 4.2).

The several data files for each of the 12 fission reactions that have been investigated (Section 4.2) can be combined and treated by the options described above. Results of model calculations and comparisons with experimental data and  $\bar{Z}$  and RMS could be then presented in tables and graphs similar to those in reference [4.1.3]. The current  $Y(A)$  data sets (see Section 4.1), or improved versions, should be used for option a) and for derivation of  $\bar{v}_A$  values used in both the  $Z_p$  and  $A'_p$  model calculations. Also, data for still other fission reactions could be obtained from the other evaluations [4.1.4, 4.1.5 and 4.2.13] and/or from the literature and could be evaluated to give a larger database.

## CONTENTS OF THE CD-ROM

*The enclosed CD-ROM contains products of the CRP work comprising additional valuable information, namely the computer program YCALC for downloading directly to a PC, and appendices with tables which were considered too bulky for inclusion in the printed book.*

*THE YCALC program is a PC version of Wahl's programs for calculating chain, cumulative or independent yields as described in Sections 4.1 and 4.2. Both programs give the same results, but YCALC is more convenient for PC users.*

**Appendix A** presents some tables of evaluated fission yields as provided by evaluators.

*One task of the CRP was the inter-comparison of evaluated fission yield files with the goal to reveal discrepancies among evaluations as well as among experimental data and to identify (ranges of) fission yields that need to be measured. In the course of the CRP, several such studies were undertaken, and the results are presented in **Appendix B**.*

### DESCRIPTION OF CONTENTS

#### **Program YCALC** (G. Grommes, H.O. Kling, H.O. Denschlag)

*A PC program for calculating mass yields, fractional and absolute independent and cumulative yields for 12 fission reactions. Results can be displayed as tables or graphs. The instructions for installing the program are given separately of YCALC\_install.doc.*

#### **Appendix A: Tables of evaluated fission yields**

##### A.1. Fission product yields for U235T, U238F, U238H and Pu239T (A.C. Wahl)

*A tabulation of fission product yield values and percent errors that have been derived from Wahl's evaluation of fractional independent yields.*

##### A.2. Chain yields from the ENDF/B-VI file (T.R. England and B.F. Rider)

*60 sets of chain yields derived from the ENDF/B-VI evaluated yield file.*

#### **Appendix B: Inter-comparison of evaluated fission yield files**

*Inter-comparison of evaluated fission yield files was one of the CRP tasks. Several inter-comparisons have been made which are all included on the CD-ROM.*

##### B.1. Comparison of chain yields for 11 fission reactions (A.C. Wahl)

*Comparison of ENDF/B-VI and UKFY3 with experimental results*

##### B.2. Comparison of ENDF/B-VI and JEF-2.2 fission yield libraries for U235T, U238F, Pu239T and Pu239F (F. Storrer and D. Rochman)

*Independent yields from ENDF/B-VI, JEF-2.2 and their ratios are presented.*

### B.3. Comparison of chain yields and discrepancy tables (R.W.Mills)

*Inter-comparisons of chain yields for 6 fission reactions are presented as figures. Furthermore, 69 tables list deficient (discrepant, unmeasured, or with only one measurement) chain yields where further measurements are required.*

### B.4. A comparison of ENDF/B-VI and UKFY3 mass chain yields (J.M. Campbell, T.R. England and W.B.Wilson)

*Tabulation of chain yields for 70 fission reactions from ENDF/B-VI and UKFY3 together (when available).*

## FILES STORED ON THE CD-ROM

A_cover.doc	Word file	Appendix A cover page
A1_intro.doc	Word file	Appendix A1: introductory text
A1_table.txt	ASCII file	Appendix A1: yield tables
A2_all.pdf	pdf file	Appendix A2 (complete)
A2_all.ps	Postscript file	Appendix A2 (complete)
B_cover.doc	Word file	Appendix B cover page
B1_intro.doc	Word file	Appendix B1: introductory text
B1_table.txt	ASCII file	Appendix B1: inter-comparison tables
B2_intro.doc	Word file	Appendix B2: introductory text
B2_PU9F.xls	Excel file	Appendix B2: <sup>239</sup> Pu fast independent yields
B2_PU9T.xls	Excel file	Appendix B2: <sup>239</sup> Pu thermal independent yields
B2_U5T.xls	Excel file	Appendix B2: <sup>235</sup> U thermal independent yields
B2_U8F.xls	Excel file	Appendix B2: <sup>238</sup> U fast independent yields
B3_all.doc	Word file	Appendix B3 (complete)
B4_intro.pdf	pdf file	Appendix B4: introductory text
B4_table.pdf	pdf file	Appendix B4: inter-comparison tables
Ycalc	program package	to setup the programs for running YCALC
YCALC_install.doc	Word file	Description of YCALC program and instructions for installing and executing it.

## ABBREVIATIONS

A	nuclear mass
$A_f$	mass of fissioning (compound) nucleus
$A'_H$	most probable heavy fragment mass of a mass yield distribution in fission
$A'_L$	most probable light fragment mass of a mass yield distribution in fission
$A'_p$	most probable mass of an isotopic mass yield distribution in fission
amu	atomic mass unit
BNL	Brookhaven National Laboratory, Upton, New York, USA
BROND	Russian Evaluated Nuclear Data Library
CENDL	Chinese Evaluated Nuclear Data Library
CINDA	Computer Index (of bibliographic references for) Neutron Data
CNDC	Chinese Nuclear Data Committee
DN	delayed neutron(s)
ENDF	Evaluated Nuclear Data File = name of format and file of the US.
ENSDF	Evaluated Nuclear Structure and Decay data File
EXFOR	Exchange Format = name of format for the exchange of experimental data and of data library itself (see Section 3.1.1.2)
FPY	fission product yield(s)
FPND	fission product nuclear data
FWHM	full width at half maximum (of a distribution)
INIS	International Nuclear Information System
JEF	Joint Evaluated File of the OECD countries
JEFF	Joint Evaluated Fission and Fusion Project (File) of the OECD countries
JENDL	Japanese Evaluated Nuclear Data Library
LCP	light charged particles
NEA-DB	Nuclear Energy Agency Data Bank
NNDC	National Nuclear Data Center, Brookhaven, Upton, NY, USA
OECD	Organisation for Economic Co-operation and Development
$P_n$ (-value)	neutron emission probability for delayed neutron precursors
R-value	result of a “ratio of ratios” fission product yield measurement
UKAEA	United Kingdom Atomic Energy Authority
UKFYx	United Kingdom Fission Yield file, version x
WRENDL	World Request List for Nuclear Data measurements
Z	nuclear charge
$Z_c, Z_f$	charge of fissioning (compound) nucleus
$Z_p$	most probable charge of an isobaric charge yield distribution in fission
$\bar{\nu}$	( $\bar{\nu}$ = average) number of ( $\nu_p$ = prompt) neutrons emitted in fission
$\sigma$	width of a (Gaussian) distribution (related to FWHM), or measurement error (1 standard deviation), or neutron reaction cross-section



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### **Research Co-ordination Meetings**

Vienna, Austria: 2–4 October 1991, 21–23 April 1993,  
17–19 October 1994, 14–18 October 1996





