Recent developments in fission product yield evaluation

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Summary

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- Motivations
- Fission Yields
  - What are they and how are they used in applied science?
  - How are they traditionally evaluated?
- Current models.
  - 5-Gaussians
  - Wahl Zp
  - Madland/England, Rudstam …
  - Ternary light charged particle emission.
- SG-37 -New experimental data and models.
  - Measurements $\Delta A>1$ $\Delta Z > 1$
  - New models (GEF code).
  - Covariance
  - Uncertainty propagation in applications
- ANDES developments

Motivations

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- Fission Yields are needed for the computer simulation of reactors, fuel cycles and waste management.
- They determine spent fuel inventories and resultant quantities such as decay heat, delayed neutron, anti-neutrinos, gamma-ray emission, and are important in understanding fuel performance.
- Current yield evaluations are adequate for current industrial applications, but future fuel cycles, reactors and potential new regulatory requirements require better data beyond thermal ^{235}U and ^{239}Pu, with rigorous uncertainties (including covariance for uncertainty propagation).

Governing equation

- The equation for any transmutation of materials in a neutron flux is governed by the production/destruction equation that Bateman developed in 1910 for radioactive decay and which was later extended to include neutron reactions.

$$\frac{dN_i}{dt} = -\lambda_i N_i + \sum_j \lambda_j N_j B_{j,i}$$
$$+ \sum_k N_k \sigma_{f,k} \phi Y_{k,i} - \sum_l N_i \sigma_{i,l} \phi + \sum_m N_m \sigma_{m,i} \phi$$

Here $Y_{k,i}$ is the independent yield of nuclide i from the fission of nuclide k.

Fission Yield Definitions (the small print)

Definitions

The **independent yield** y(A,Z,I) is the number of atoms of (A,Z,I) produced directly from one fission, but after the emission of prompt neutrons (but before any radioactive decay and hence the emission of delayed neutrons). It can be written as the product of 3 factors:

$$y(A, Z, I) = Y(A) \times f(A, Z) \times R(A, Z, I)$$

where the **sum yield** or **mass yield** Y(A) is the total of the independent yields (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: AEA-TRS-1015, James, Mills and Weaver 1990.

Fission Yield Definitions (the small print)

Definitions

The **cumulative yield** c(A,Z,I) of nuclide (A,Z,I) is the total number of atoms of that nuclide produced over all time after one fission. If the nuclide is stable the cumulative yield is the total number of atoms of that nuclide remaining per fission after all precursor decays (ignoring the effects of other nuclear reactions e.g. neutron capture). However, for a radioactive nuclide for which this is not the case, some atoms will have decayed before all have been produced.

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.

The **chain yield** Ch(A) is equal to the sum of all stable or long-lived cumulative yields for a given mass chain. It should be noted that the chain yield, Ch(A), and the sum or mass yield, Y(A), for a mass chain A may differ by a few per cent because the former applies after, and the latter before, delayed neutron emission.

From: AEA-TRS-1015, James, Mills and Weaver 1990.

Fission Yield Evaluation

What is nuclear data “evaluation”

- Can be described as the processing of giving value to a quantity or assessing quality ...

  But

  - Has to be consistent with measurements and constraints of physical laws within currently accepted knowledge.
  - Has to be consistent with best theory/models.
  - Has to be reported/distributed in a form that all can use easily (e.g. ENDF format).
  - Has to be tested against appropriate experiments.

Fission Yield Evaluation

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- Most available measurements are radiochemical and thus do not usually measure independent yields due to time to make measurements.
- Some particle identification method data used but only currently those uniquely defining A and Z.
- What is usually measured is the amount of a nuclide present, or its decay rate, after a period of time in which a number of fissions have occurred.
- Depending on the irradiation conditions and cooling time this can approximate the cumulative yield times by the number of fissions.
  i.e. if irradiation and cooling << half-life nuclide but >> half-lives of precursors.

Fission Yield Evaluation

- Current methods are data driven
  - Measurements used by preference, where available.
  - Measurements are difficult leading to many discrepant data.
  - Measurements are used to fit empirical/semi-empirical models to predict unmeasured values.
  - Often adjusted for physical constraints.
- Problems
  - Limited measurements ~15000 but mostly ^{235}U and ^{239}Pu thermal neutron induced fission.
  - What is measured is often not the most basic quantity.
  - Highly correlated quantities, need covariance to properly propagate uncertainties.

Data Analysis

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- UKFY3 based upon experimental data for yields of specific A, Z and I from >2000 references.
- Currently updating with 10 significant recent measurement reports (University of Manchester student Robert Frost). ~1100 new high resolution measurements

Dataset	Absolute	Ratio	Ratio of ratio	Total
UKFY3.6A (JEFF-3.1.1)	11887	1352	1471	14710
UKFY3.7 (Prelim)	12908	1441	1471	15820

Example of analysis

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- UKFY3/JEFF-3.1.1 ^{235}U thermal fission chain yield

99	RB	1188	6.400E-03	28.0		4.28	0.25	6.126E+00	(I)	0.9A
	SR	22003	1.090E-01	22.0					(E)	0.4
		1188	1.800E-01	33.0						
	NB(M)	21008	2.240E+00	5.8						
	MO	13382	5.900E+00	3.4	-1.17					
		13372	5.910E+00	20.0	-0.18					
		13054	5.932E+00	50.0	-0.07					
		13372	5.980E+00	20.0	-0.12					
		774	6.000E+00	9.0	-0.23					
		327	6.010E+00	3.5	-0.57					
		312	6.140E+00	3.0	0.08					
		213	6.160E+00	3.0	0.19					
		1151	6.170E+00	15.0	0.05					
		13395	6.200E+00	15.0	0.08					
		195	6.250E+00	3.0	0.69					
		1000	6.250E+00	4.0	0.51					
		341	6.350E+00	15.0	0.24					
	TC	628	6.140E+00	3.0	0.08					
	CHAIN	1098	6.040E+00	2.0	-0.79					
		248	6.140E+00	2.0	0.13					
		21531	6.320E+00	3.5	0.91					
		21054	6.350E+00	5.0	0.72					

From: Mills, Thesis 1995.

Mass distribution

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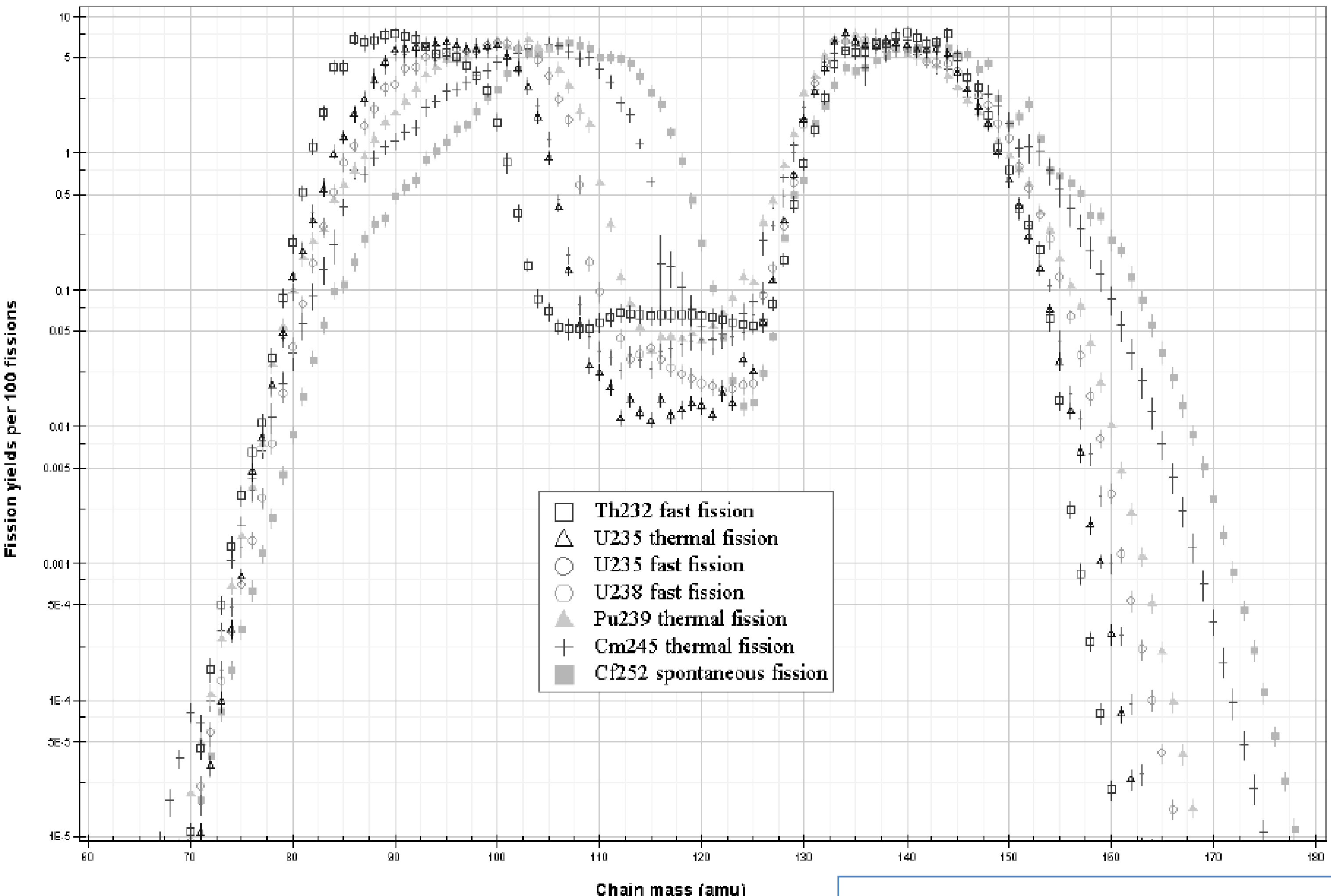

From: JEFF report 20, 2009.

Models –
Mass Distribution

Historically empirical fitting of Gaussian distributions have been used to model chain yields (mass yields). Work in the 1960's showed that the best results were obtained using 5 Gaussians. Due to physical constraints there are only 7 free parameters to fit:

$$
\begin{aligned}
Y(A) = {} & \frac{N_1}{\sigma_1\sqrt{2\pi}}\left[e^{-\left(\frac{(A-\bar{A}-D_1)^2}{2\sigma_1^2}\right)} + e^{-\left(\frac{(A-\bar{A}+D_1)^2}{2\sigma_1^2}\right)}\right] \\
& + \frac{N_2}{\sigma_2\sqrt{2\pi}}\left[e^{-\left(\frac{(A-\bar{A}-D_2)^2}{2\sigma_2^2}\right)} + e^{-\left(\frac{(A-\bar{A}+D_2)^2}{2\sigma_2^2}\right)}\right] \\
& + \frac{N_3}{\sigma_3\sqrt{2\pi}}\, e^{-\left(\frac{(A-\bar{A})^2}{2\sigma_3^2}\right)}
\end{aligned}
$$

with $N_3=2(1-N_1-N_2)$.

Models – Charge distribution (Wahl Zp)

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The independent yield is calculated as the integral of a normal distribution

$$FI(A,Z) = \frac{1}{2}F(A,Z)N(A)(\mathrm{erf}(V) - \mathrm{erf}(W))$$

where

$$V = \frac{Z(A) - Z_p(A) + 0.5}{\sigma_Z(A)\sqrt{2}} \qquad W = \frac{Z(A) - Z_p(A) - 0.5}{\sigma_Z(A)\sqrt{2}}$$

F(A,Z) describes the odd-even effect and N(A) is a normalization constant

The integral of a normal distribution between a and b is given by:

$$\frac{1}{2}\left(erf\left(\frac{a}{\sqrt{2}}\right) - erf\left(\frac{b}{\sqrt{2}}\right)\right)$$

Models –
Charge distribution (Wahl Zp)

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$Z_p(A)$ is the most probable charge for mass A

This is calculated as the "unchanged charge distribution" corrected for prompt neutron emission with a term describing the variation of the charge offset ΔZ

In the heavy mass peak this is:

$$Z_p(A_H) = A'_H \frac{Z_f}{A_f} + \Delta Z(A'_H)$$

In the light mass peak, by conservation of mass and charge

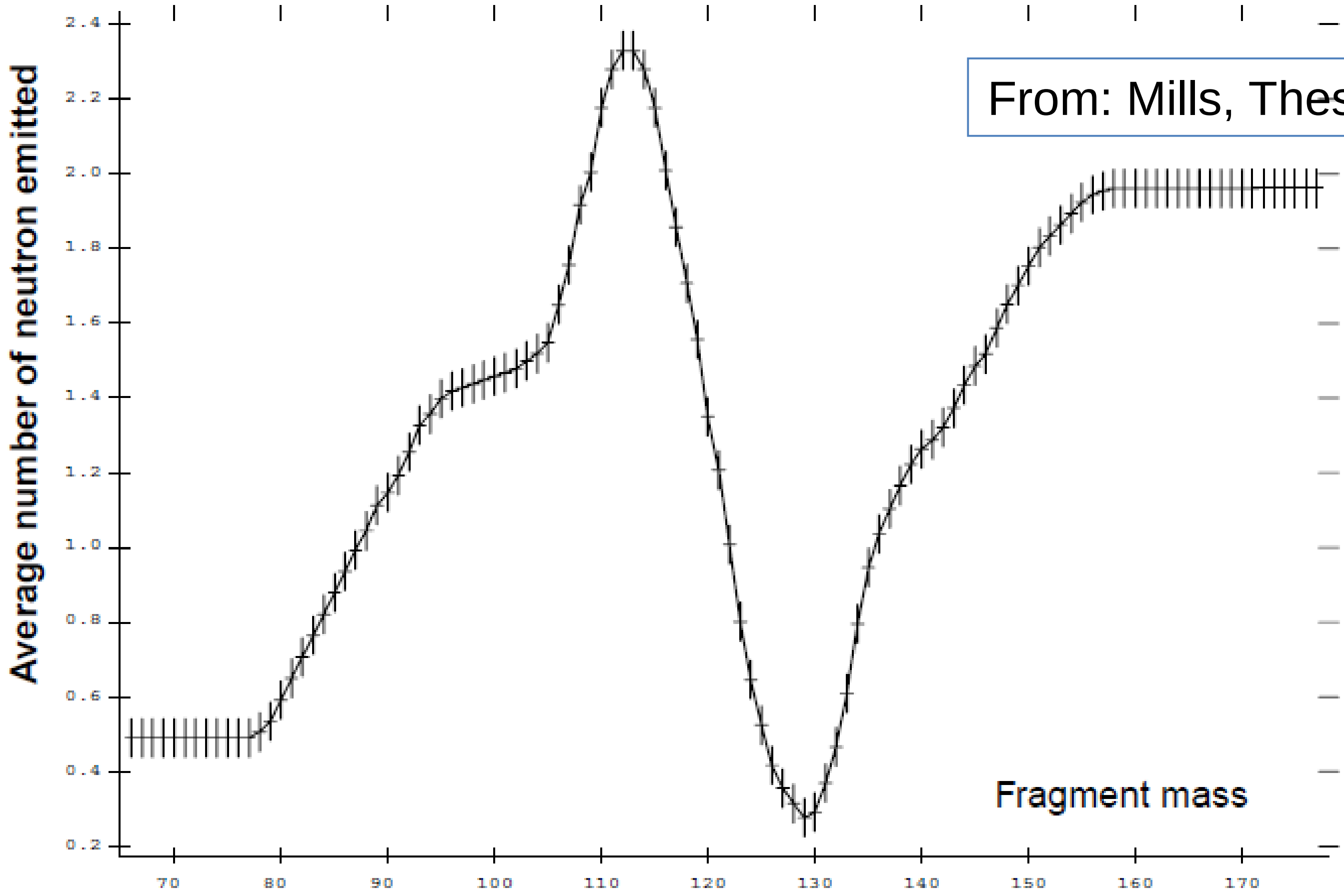
$$Z_p(A_L) = A'_L \frac{Z_f}{A_f} + \Delta Z(A'_{Hc}) \qquad A'_{Hc} = A'_f - A'_L$$

The mass after prompt neutron emission is A' and the mass before is A. Thus

$$A' = A - \nu(A)$$

Models –
Charge distribution (Wahl Zp)

The average neutron emission per fragment mass, $\nu(A)$, can be calculated from the method of Terrell using measured mass yields.



Models –
Charge distribution (Wahl Zp)

The odd-even effect is modelled as F(A,Z) using two parameters $\overline{F}_N$ and $\overline{F}_Z$:

F(A,Z)	proton number Z	neutron number N
$\overline{F}_Z\overline{F}_n$	even	even
$\frac{\overline{F}_Z}{\overline{F}_n}$	even	odd
$\frac{\overline{F}_n}{\overline{F}_Z}$	odd	even
$\frac{1}{\overline{F}_Z\overline{F}_n}$	odd	odd

From: Mills, Thesis 1995.

Models-
Isomeric splitting of yields

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- In JEFF-3.1.1 there exists 388 nuclides with two long-lived isomeric states and 21 with three or more.
- Only small number of measurements.
- The main predictive model available is that of Madland and England which assumes the fragments with a spin near a long-lived isomer would preferentially feed that isomer.
  Model used in UKFY2/JEF-2.2.
- Rudstam proposed a modification that included energetic feasibility that affects a few isomers.
  Results used in UKFY3/JEFF-3.x.

Models-
Ternary fission

- The emission of protons, deuterons, tritons, alpha particles and other light fragments up to 30 amu have been observed from fission.
- The most common emission is an alpha particle, second tritons, …
- In UKFY2/JEF-2.2 empirical relationships were used to determine unmeasured yields.
- In UKFY3/JEFF-3.x improved model results published by Serot et al at ND2004 were used.

Cumulative Yields- Q matrix

- Given the individual decay branches for all nuclides in the decay paths from one nuclide to a distant daughter it is possible to calculate the fraction of j that decays to i

$$Q_{j,i} = \sum_{allpaths} \left(\prod_{each j \to i} B_{j,j+1} B_{j+1,j+2} \ldots B_{i-1,i} \right)$$

- If $Q_{i,i}$ is defined as 1 and $Q_{k,i}$ =0 (i.e.. where k does not decay to i), the cumulative yield can be calculated from the independent yield.

$$Y_i^c = \sum_j Y_j^i Q_{j,i}$$

From: Mills, Thesis 1995.

Adjustment to physical constraints

Constraint	Equation (From: Mills, Thesis 1995.)
Conserve fragment number (50<A<200)	$\sum_{A} Y(A) = 2.0$
Conserve fragment number in upper (and lower peak)	$\sum_{A > \frac{A_f}{2}} Y(A) = 1$
Conserve mass	$\sum_{A} A Y(A) = A_f - \overline{\nu_p} - A_{LCP}$
Conserve charge	$\sum_{ZA} Z \; f(A,Z) \; Y(A) = Z_f - Z_{LCP}$
Complementary yield pairs (where $Z_f = Z_1 + Z_2$)	sum (Y(A) F(A,Z_1)) = sum(Y(A) F(A,Z_2))
or for a given Z then	$\sum_{A} f(A,Z) Y(A) = \sum_{A} f(A, Z_f - Z) Y(A) \quad \text{for all } Z < \frac{Z_f}{2}$

Validation of evaluated files

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- Check of consistency of files (format)
- Check physical consistency/conservations
- Delayed neutron emission
  Calculated from Y^c + P_n values only.
- Decay heat pulses
  Y^i + ENDF formatted decay data using code.
- Decay heat from PWR/BWR assemblies
  Y^i + ENDF formatted decay data using code.
- Chemical Analysis of fuel
  PWR, BWR, AGR, MAGNOX etc. (NEA EGADSNF)
  Y^i + ENDF formatted decay data using code.

Summary of WPEC subgroup 37

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- Setup in 2013 under the NEA Working Party on International Evaluation Collaboration to investigate improving fission yield evaluation methods
- Consists of 3 tasks:
  - 1: Document and compare existing methodologies.
  - 2: Insights, new measurements and models to understand and reconcile discrepancies.
  - 3: Possible new fission product data, format and covariance data for applications.
- Note to evaluate new data types e.g. measurements with resolution in ΔA, $\Delta Z > 1$ need more experimental information than just result!

  PLEASE RECORD/MAKE AVAILABLE TO EXFOR!

GEF code

- The following slides are taken from work by Karl-Heinz Schmidt, Beatriz Jurado and Charlotte Amouroux
  - GEF - an approach to fission based on fundamental laws of physics and mathematics combined with empirical fitting of ~50 parameters
    (to model most systems between ^{220}Th and ^{262}Rf with excitation energies <20MeV)
  - Now includes second chance fission and isomeric yields.
  - Validation of GEF results
  - Covariances from GEF
- See preprint and website for further details
  http://www.cenbg.in2p3.fr/-GEF,354-
  http://www.khs-erzhausen.de/GEF.html

  or JEFF report 24 (2014)

Concept of the GEF model

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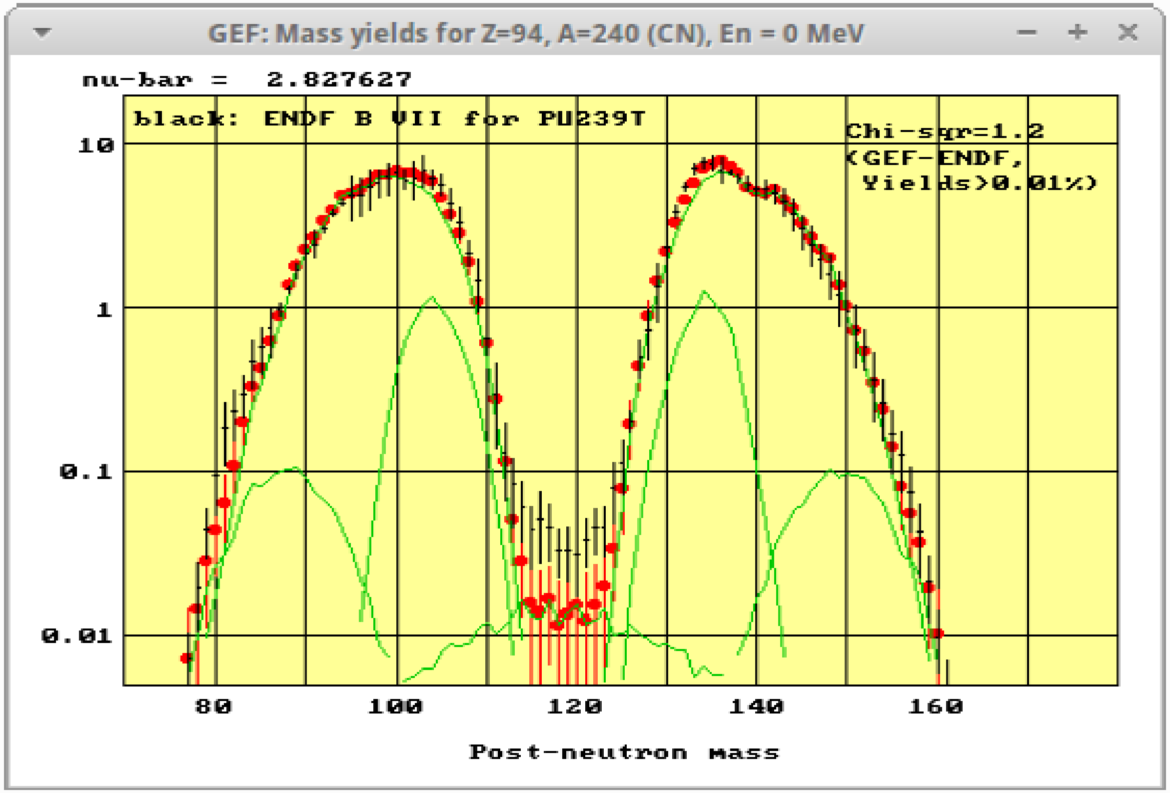
- **General approach using global theoretical models and considerations on the basis of universal laws of physics and mathematics.**
  - Topological properties of a continuous function in multi-dimensional space ($\rightarrow$ Fission barriers.)
  - Condensation of matter at low T ($\rightarrow$ Level densities.)
  - Evolution of quantum-mechanical wave functions in systems with complex shape ($\rightarrow$ Fragment shells.)
  - Memory effects in the dynamics of stochastic processes ($\rightarrow$ Dynamical freeze-out.)
  - Influence of the Second Law of thermodynamics on the evolution of open systems. ($\rightarrow$ Energy sorting.)

From: JEF-DOC/1571, Schmidt et al, 2014.

Example: Mass distribution of the thermal neutron fission of ^{239}Pu



Green lines: Fission channels from GEF

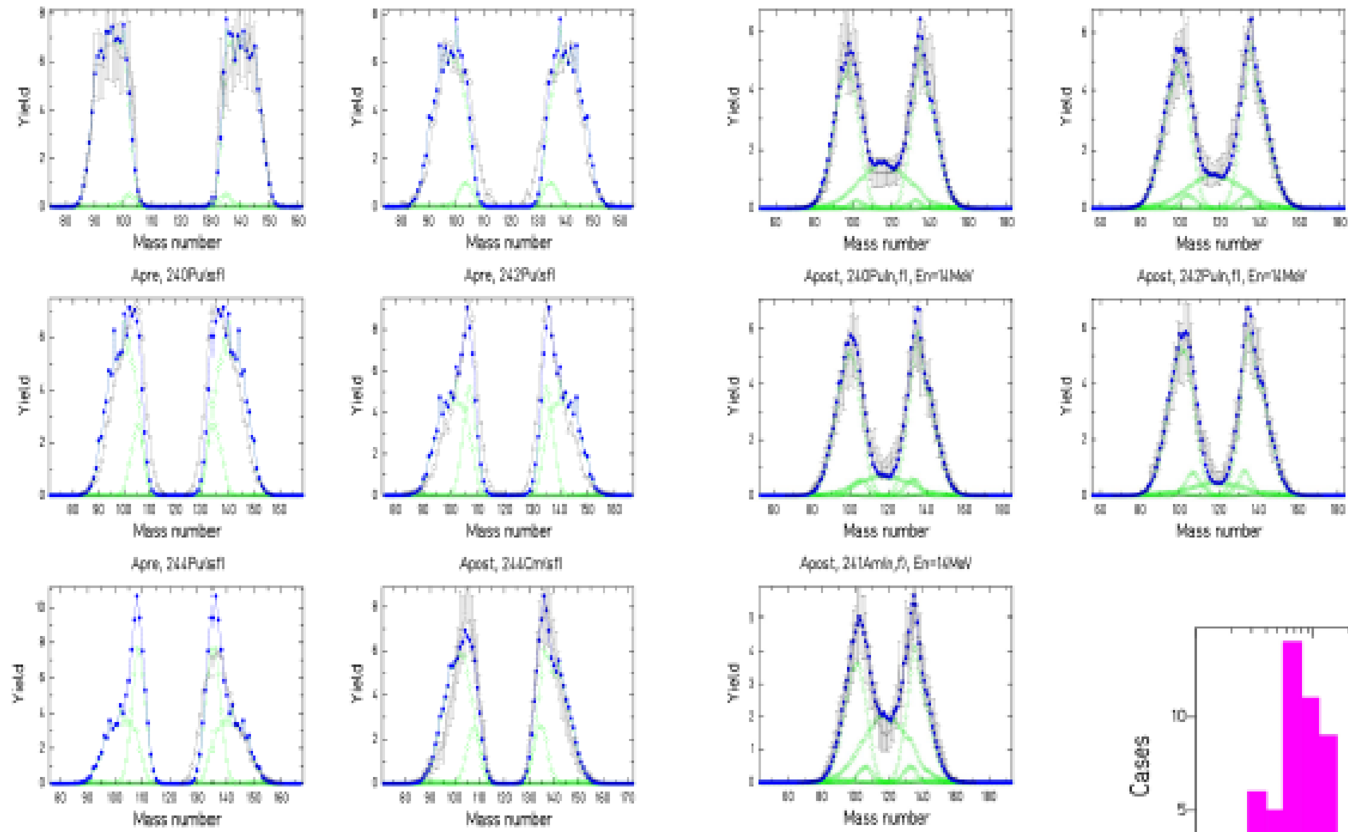
Red points and red error bars: Mass distribution from GEF

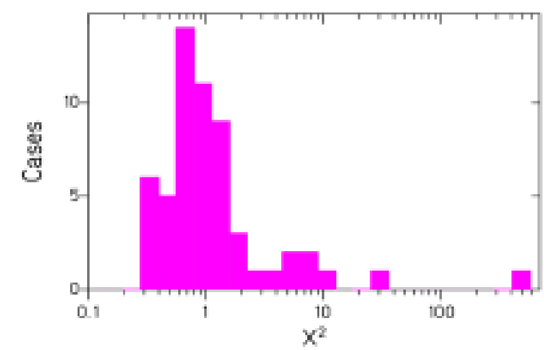
Black error bars: ENDF/B-VII

Principle parameters (given by the model, simplified):
<A>, sigma_A, Y(A) of each fission channel determined by position, width, depth of the fission valleys (12 parameters).

From: JEF-DOC/1570, Schmidt et al, 2014.

Mass distributions from SF and 14 MeV neutrons



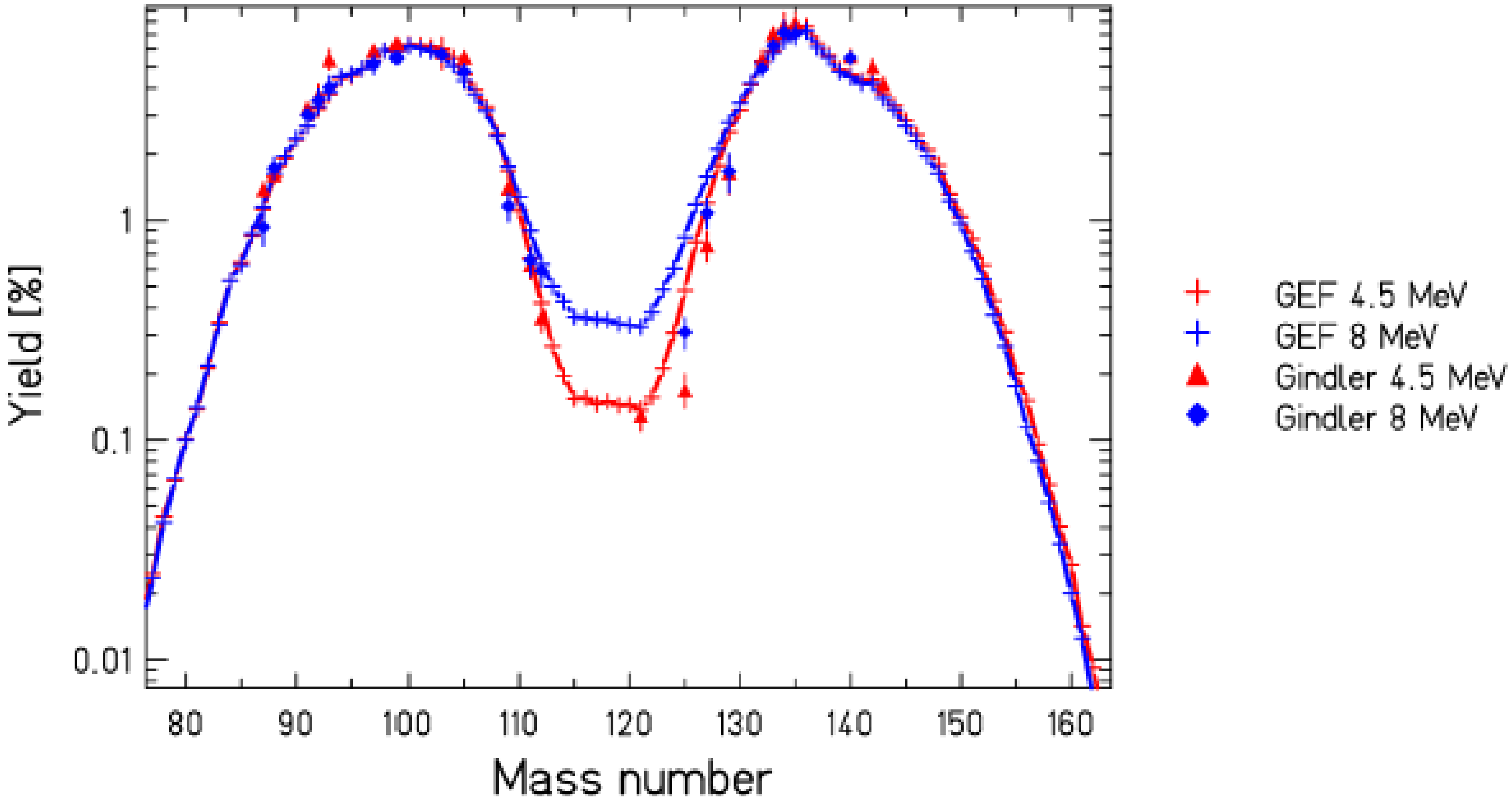


Overall good agreement with ENDF/B-VII

Total: 59 cases

From: JEF-DOC/1572, Schmidt et al, 2014.

Mass distributions of ^{239}Pu fission with 4 and 8 MeV neutrons



From: JEF-DOC/1572, Schmidt et al, 2014.

Nuclide distributions
Example from $^{235}U(n_{th},f)$

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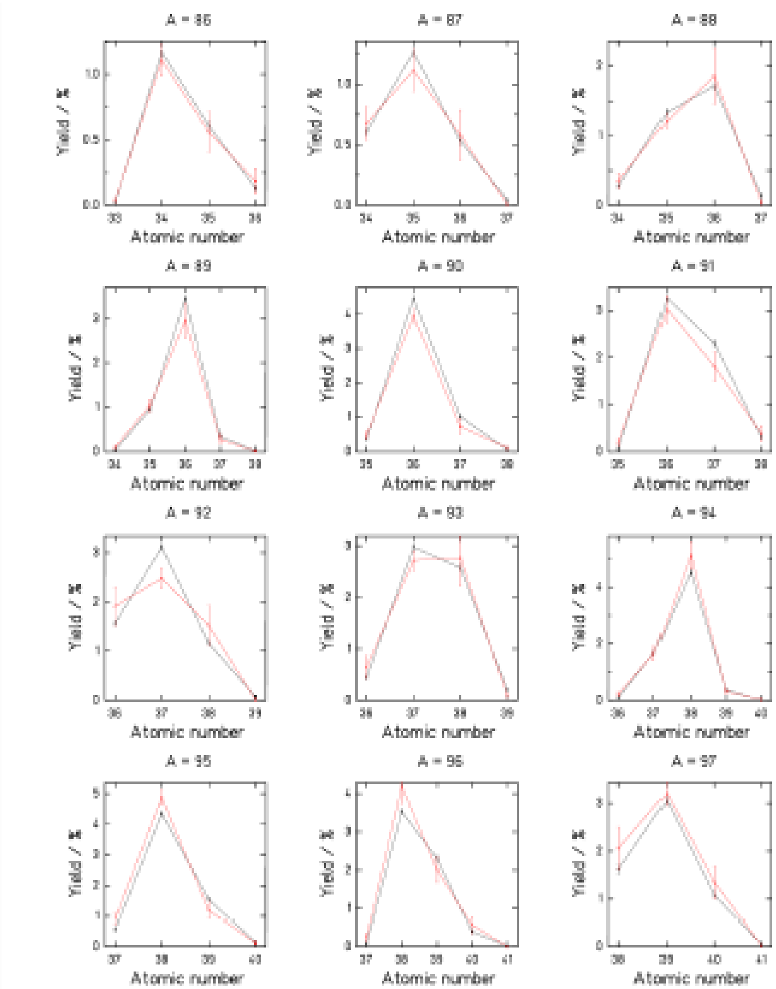

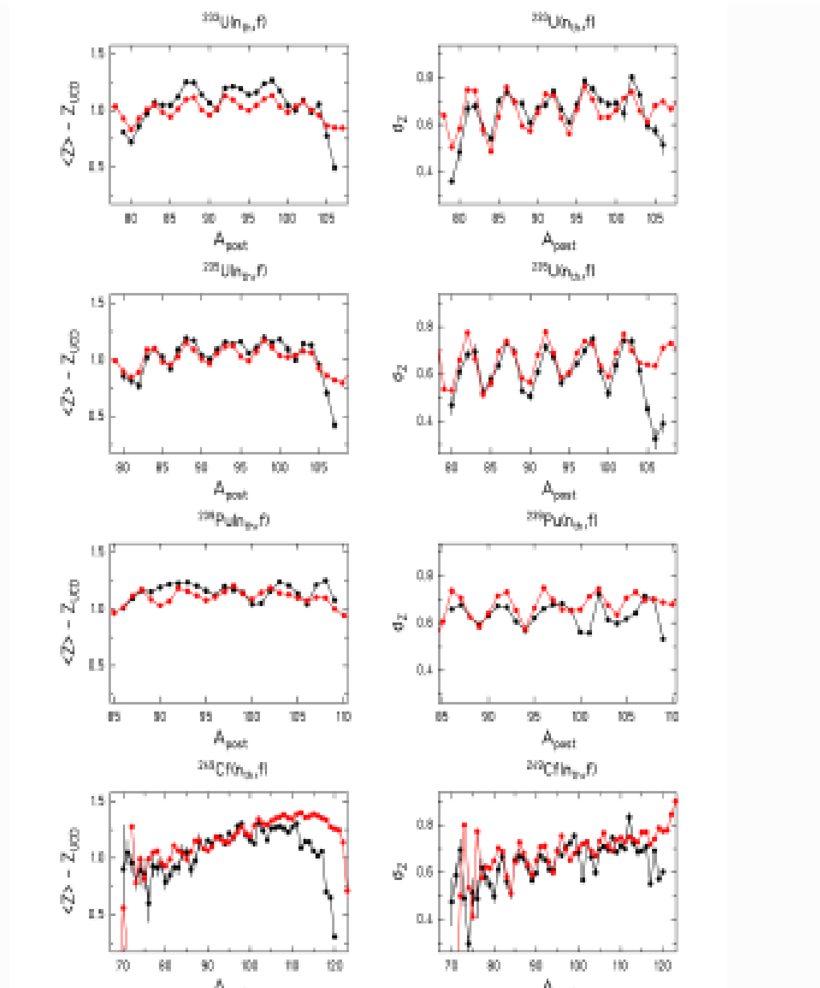

From: JEF-DOC/1572, Schmidt et al, 2014.

Isomeric yields

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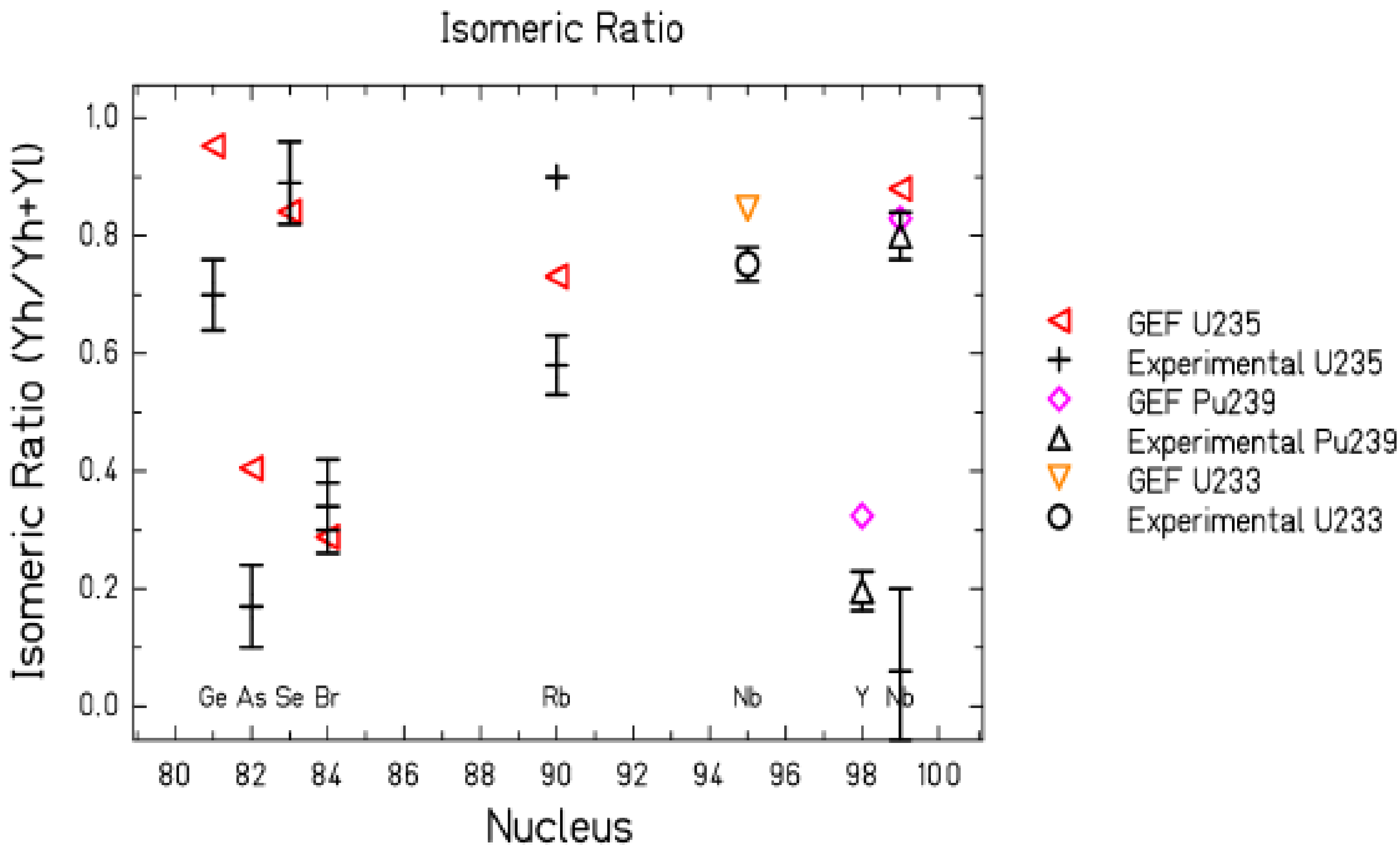

From: JEF-DOC/1572, Schmidt et al, 2014.

Uncertainties and covariances

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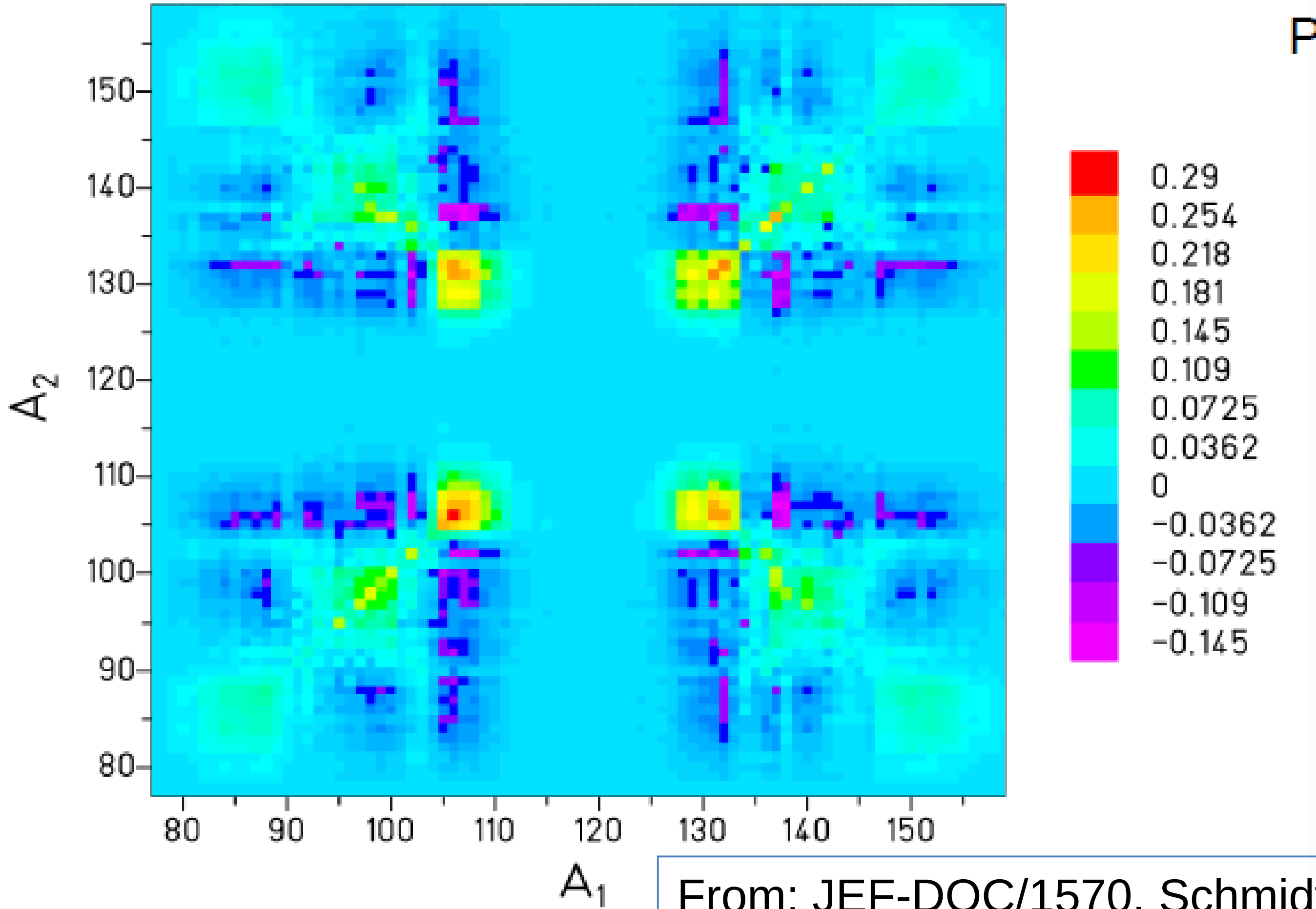
- Calculations with perturbed parameters
- Unavoidable coupling between the parameters by normalization of yields to 200%.
- Best values and uncertainties of parameters from CHI-squared of all systems.
- Resulting multi-variant distributions (distributions of mass yields Y(A1) vs. Y(A2)).
- Deduce uncertainties and covariances.

$$\mathrm{cov}(x,y)=\sum_{i=1}^{N}\frac{(x_i-\bar{x})(y_i-\bar{y})}{N}$$

From: JEF-DOC/1570, Schmidt et al, 2014.

Full covariance matrix



From: JEF-DOC/1570, Schmidt et al, 2014.

Determined by the inner logic of the model.
Basically different from experimental covariances!

Fission Yield Covariance

FY covariance data generation:

- Great efforts have been committed to develop methodologies for correlation generation (full covariance matrices) for FY data.

  Methodologies proposed at the kick-off meeting of WPEC-SG37 (May 2013), based on:

  - **Perturbation theory** applied to the "Five Gaussians and Wahl's models" (*Musgrove et al., 1973; Wahl, 1988*), proposed by *Pigni et al. (2013)*.
  - **Monte Carlo parameter perturbation** using the GEF code (*Schmidt and Jurado, 2010*), presented by Schmidt (*2013*).
  - **Bayesian/general least-squares (GLS) method**, where the IFY covariance matrix is updated with information on the chain yields as proposed by *Kawano and Chadwick (2013)*, and previously applied by *Katakura (2012)*.
    - A variation of this proposal, with IFYs covariance matrix updated with CFYs ones is described and reported by UPM/SCK (*L. Fiorito et al., 2014*)

From: Carlos J. Díez , private communication, 2014.

Covariance Matrix Evaluation for Independent Fission Yields

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N. Terranova[1], O. Serot[2], P. Archier[2], C. De Saint Jean[2], M. Sumini[1]

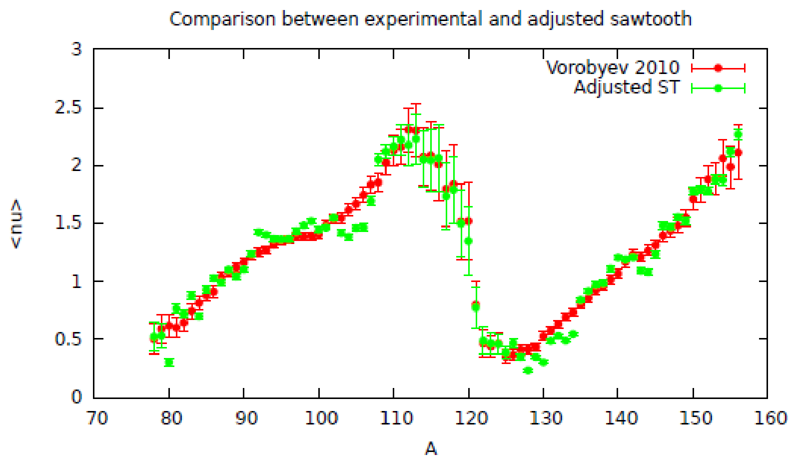
[1]Dipartimento di Ingegneria Industriale (DIN), Università di Bologna, Italy

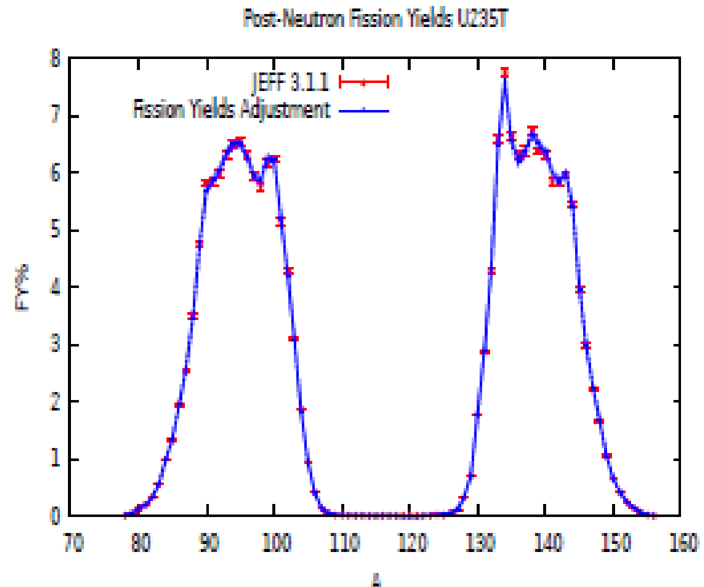
[2]CEA, DEN, Cadarache, F-13108 Saint Paul les Durance, France

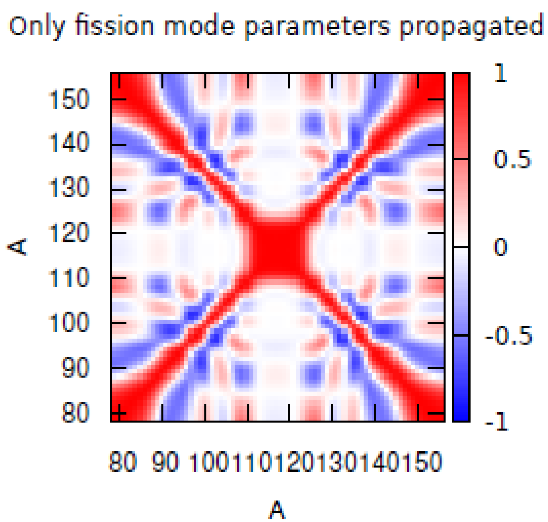
- They have developed a method able to represent faithfully JEFF 3.1.1 evaluations for mass fission yields in the CONRAD system.
- The adjustment of the parameters for the pre-neutron fission modes and the saw-tooth curve has given acceptable results.
- Preliminary correlation information have been produced for mass fission yields, considering only statistical uncertainties.
- For future to consider systematic uncertainties, charge and isomeric yields

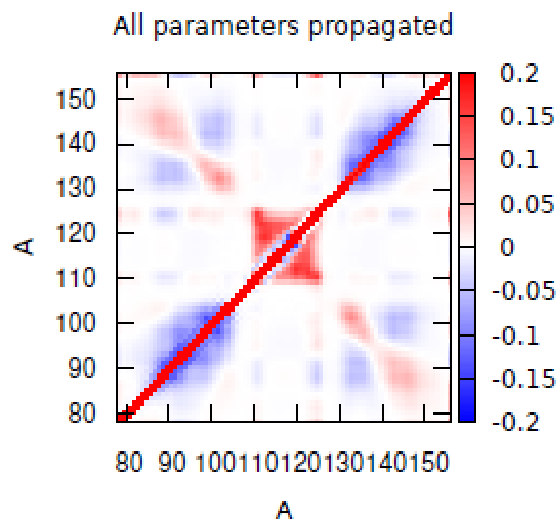
From: Terranova et al, private communication, 2013.

Covariance Matrix Evaluation for Independent Fission Yields



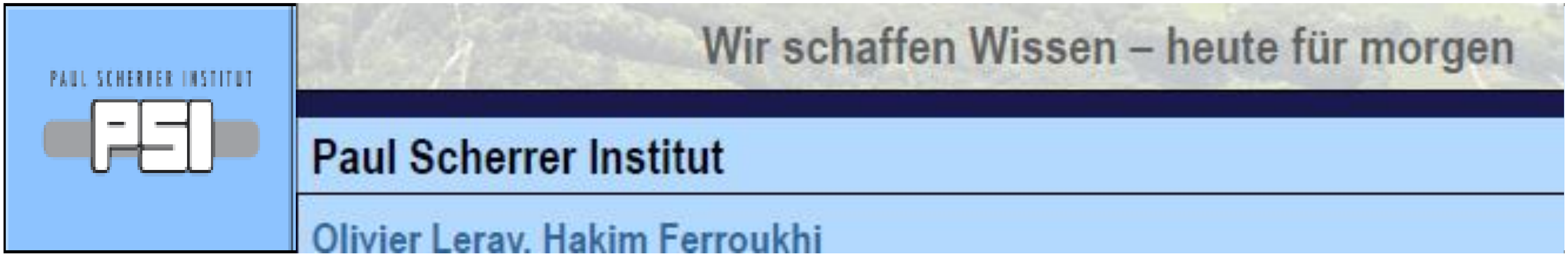






From:
Terranova et al,
private communication,
2013.

Status on fission yield perturbation methodology at PSI using CASMO-5

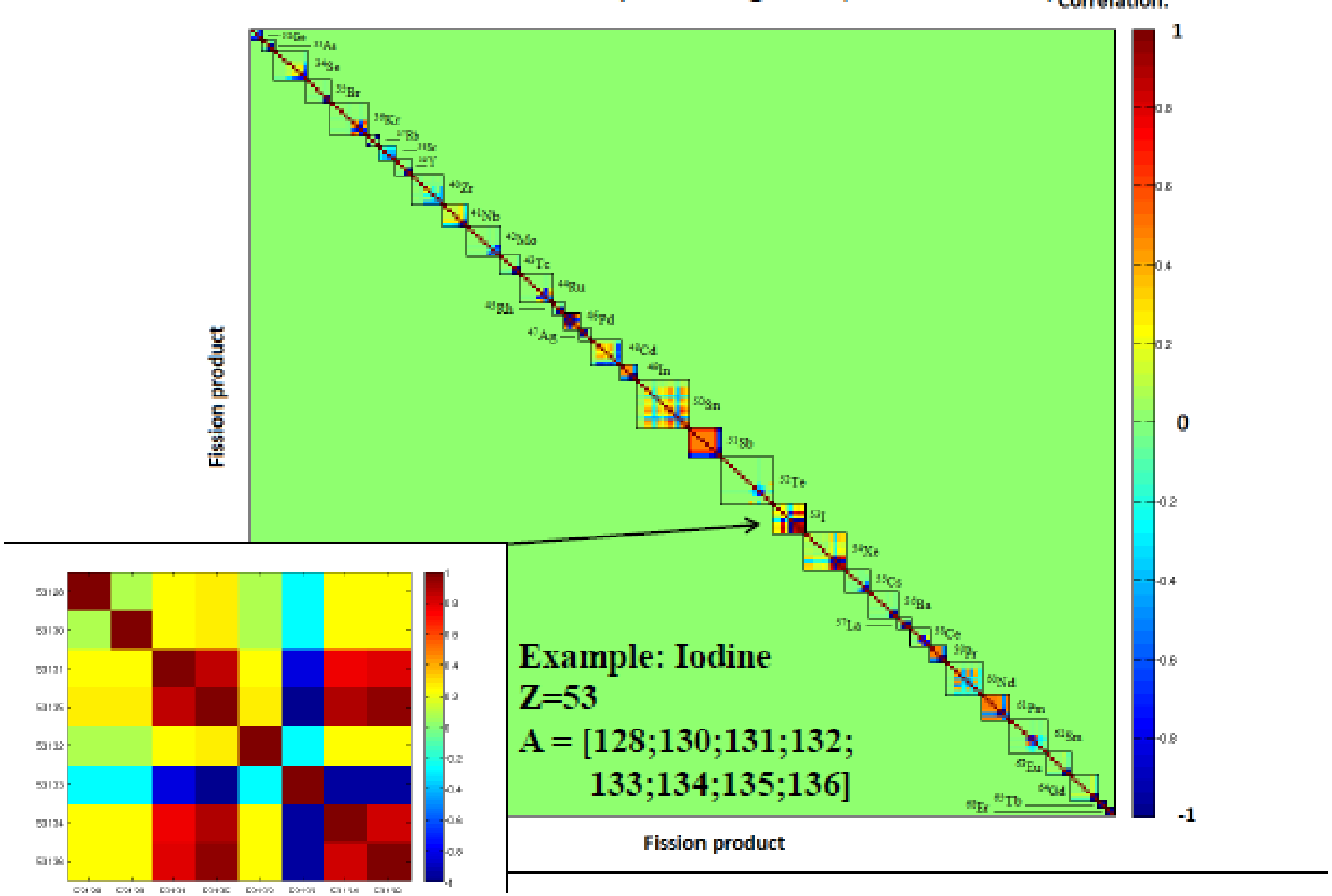


- Recent developments of the SHARK-X tool using
  - Direct perturbation (brute force) and Statistical Sampling
  - Adjustment of FY data in CASMO to force physical constraints
  - Considers problems with PDF (-ve parameters)
- Motivation is to calculate uncertainties on decay heat, isotopic composition and reactivity.

From: Leray et al, private communication, 2014.

Status on fission yield perturbation methodology at PSI using CASMO-5

Fission Yield Correlation matrix for U^{235} (215 daughters, 35 elements)



From: Leray et al, private communication, 2014.

EC co-funded project ANDES

Accurate Nuclear Data for Energy Sustainability

- Designed to address the nuclear data needs associated to the new reactors and new fuel cycles supported by SNETP (Strategic Nuclear Energy Technology Platform), in its strategic research agenda and in the ESNII proposal (European Sustainable Nuclear Industry Initiative). Includes:
  - improvement of uncertainties/covariance's in evaluation
  - validation of present/new data libraries with integral experiments
- Work continuing in CHANDA project

  Solving Challenges in Nuclear Data for the Safety of European Nuclear Facilities

ANDES Workpackage 2
ACAB (UPM) used as testbed

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- Koning/Rochman (NRG): “TMC” cross-section evaluation and TENDL
- Leeb (TUW) GENUS evaluation of activation cross-sections with covariance from experiments using a Bayesian approach.
- Cabellos/Diez/Mills (UPM): Generating fission yield and decay data covariance data using a range of methods (direct analysis, automated sensitivity matrices approaches).
- Diez (UPM): Calculation of a wide range of inventories for fast reactor systems with ACAB and compared to other methodologies (e.g. routes in SCALE and TMC).

IMPACT OF THE FISSION YIELD COVARIANCE DATA IN BURN-UP CALCULATIONS

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Table 3. Uncertainty in number density (in %) for some important fission products at 60 GWd/MTU. Fission Yield source of uncertainty (standard deviation) is taken from ENDF/B-VII.1.

Nuclide	Corr.	No corr.	GRS XSUSA	Nuclide	Corr.	No corr.	GRS XSUSA
^{79}Se	3.5	16.0	-	^{142}Nd	0.8	3.5	-
^{90}Sr	0.8	6.2	-	^{143}Nd	0.4	6.5	5.9
^{95}Mo	0.5	8.4	7.9	^{144}Nd	0.2	3.9	-
^{99}Tc	0.8	10.0	9.5	^{145}Nd	0.4	7.1	6.7
^{101}Ru	0.7	4.6	-	^{146}Nd	0.7	10.8	-
^{106}Ru	1.2	13.7	-	^{148}Nd	0.8	13.7	13.0
^{103}Rh	1.1	12.1	-	^{147}Pm	0.6	10.3	-
^{109}Ag	10.9	17.8	-	^{147}Sm	0.5	9.4	-
^{125}Sb	4.2	19.1	-	^{149}Sm	0.6	12.2	10.6
^{129}I	2.7	20.7	-	^{150}Sm	0.6	10.3	-
^{135}I	2.8	4.3	-	^{151}Sm	0.7	11.7	-
^{131}Xe	0.4	6.9	-	^{152}Sm	0.6	11.3	8.8
^{135}Xe	0.4	5.1	-	^{151}Eu	0.7	12.1	-
^{133}Cs	0.3	3.4	1.7	^{153}Eu	0.8	9.9	-
^{134}Cs	0.3	3.0	-	^{154}Eu	0.8	10.4	-
^{135}Cs	0.3	3.4	-	^{155}Eu	1.0	9.5	-
^{137}Cs	0.5	1.5	1.7	^{155}Gd	1.0	10.5	8.8
^{139}La	0.9	3.2	-	^{156}Gd	1.2	9.0	-
^{144}Ce	0.2	8.0	-	^{157}Gd	1.3	9.5	-
				^{158}Gd	2.3	11.3	-

i) No correlation between fission products (ΔFYs/No corr.)

ii) FYs including correlations for ^{235}U and ^{239}Pu taken from Katakura methodology (ΔFYs/Corr.)

iii) GRS calculation

From: O. Cabellos, D. Piedra, Carlos J. Diez, JEF/DOC-1566, 2014.

Conclusions

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- From an evaluator’s perspective there is a renaissance in fission yield measurement and theory/models.
- There is also new needs driving these studies.
- Considerable activity has been applied to understanding uncertainty and covariance.
- These ideas need to be further developed and applied in:
    - Fission Yield Evaluations
    - Codes that use these data

Lots of interesting work ahead!

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