Fission yield measurements and associated covariance data.

G.Kessedjian
LPSC
For the Lohengrin fission collaboration

Outlook

1- Fission yields : why do we need new measurements ?

2- Impact of the covariance in the decay heat for nuclear applications

3- Thermal neutron induced fission yields available

4- Self-normalization, systematic group, and ND Bases :
basic elements of correlations

example on measurements @ lohengrin

5- Proposal to increase the constraints on the decay heat

1- Fission yields : why do we need new measurements ?

➤Impact of fission yields in the actual and innovative fuel cycles

- Inventory of used fuel : isotopic composition
- Residual power : minor actinides and fission products
- Radiotoxicity of used fuel
- Experimental fuel studies : reaction cross sections and isotope yields are needed for comparison Calculation/ Experiment (C/E)
- Calculation/prediction of prompt γ rays emitted in a core

➤For fission process study

- Test the fission model predictions is necessary for the evaluations at different neutron energies
- Lack on dynamical aspect for fission process modelisation $\rightarrow Y(A,Z,\mathrm{E}^*,J\pi)$
  Spin distribution
  Search of signatures of the fission modes in the kinetic energy distributions
- Inconsistency between Models or evaluations and Experiments for heavy fragments and symmetric region
  → Nuclear charge Polarization

1- Fission yields : why do we need new measurements ?

➤Needs of new measurements

- Structure in mass and nuclear charge distributions (e.g. Fifrelin, neutron emission, γ prompt)
- Isotopic distributions near symmetric region ➤ Nuclear charge polarization
- Spin distributions of the fission fragments as a function of the excitation energy
  - ➢ e.g. modeling prompt γ emission

$$Y(A,Z,E_k,J,\pi)=Y(A)\cdot P(Z|A)\cdot P(E_k|A,Z)\cdot P(J,\pi|A,Z,E_k)$$

Mass Charge Kinetic energy spin ➤ distributions

➤Needs of details on the measurements

- Evaluation : No covariance available
- Mass = $\sum$ Isotope
- Variance(Mass) = $\sum$ Var(Isitope) > Var(major Isotope)

Not possible
Mass measurements are usually more available and precise than isotopic measurements

1- Fission yields : why do we need new measurements ?

➤ Needs of new measurements

- Structure in mass and nuclear charge distributions
  (e.g. Fifrelin, neutron emission, γ prompt)
- Isotopic distributions near symmetric region ➤ Nuclear charge polarization
- Spin distributions of the fission fragments as a function of the excitation energy
  - ➢ e.g. modeling prompt γ emission

$$Y(A,Z,E_k,J,\pi) = Y(A)\cdot P(Z|A)\cdot P(E_k|A,Z)\cdot P(J,\pi|A,Z,E_k)$$

Mass Charge Kinetic energy spin ➤ distributions

➤ Needs of details on the measurements

- Evaluation : No covariance available
- Mass = $\sum$ Isotope
- Variance(Mass) = $\sum$ Var(Isitope) + … + $\sum$ Cov (Isotopes)
  $>$ Var(major Isotope)

Not possible
Mass measurements are usually more available and precise than isotopic measurements

2- Impact of the covariance in the decay heat for nuclear applications

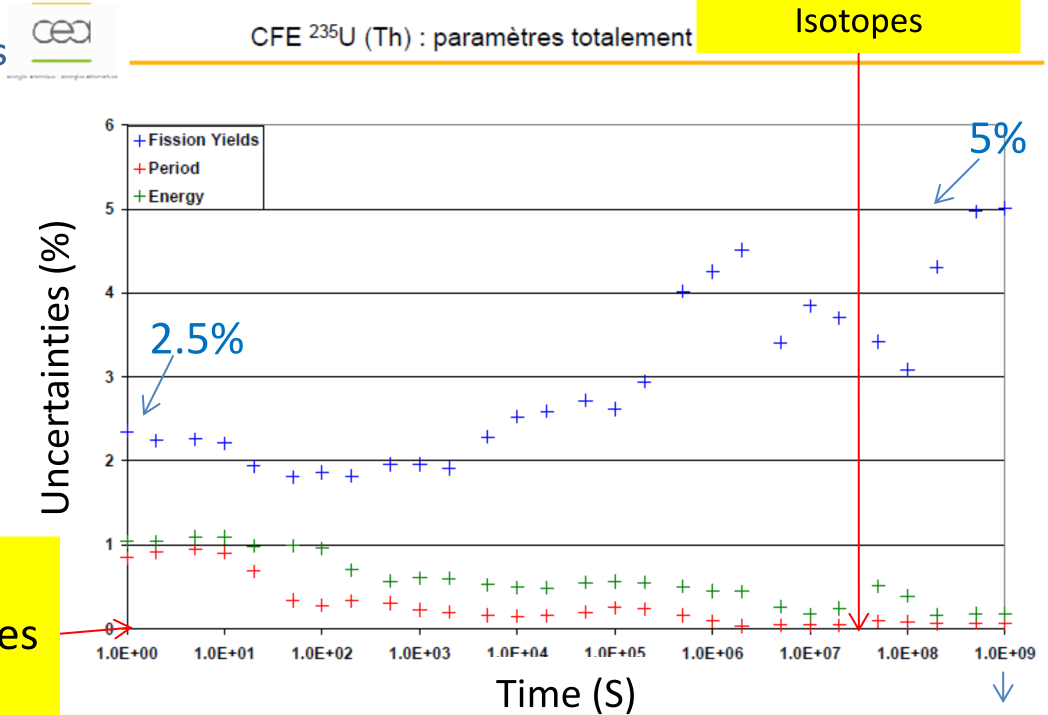
➤Sensitivities to residual power

→ Study realized by CEA/DEN in the framework of the reactor decay heat measurements ***after shutdown:*** only isotopes with a lifetime greeter than few seconds are considered ; usual data bases are used (ENSDF, JEFF, ENDF…)

→Assuming Independent measurements :

- fission yield uncertainties from 2.5% to 5%
- Energy (beta gamma) uncertainties from 1% to 0.2 %
- Period uncertainties from 1% to >0.1 %

Main contributors
~20
Isotopes



~ 100 most
Important Isotopes
after shutdown

J.Ch. Benoit, PhD Thesis CEA Cadarache
J.Ch. Benoit, O.Serot et al., Physor 2012.

2- Impact of the covariance in the decay heat for nuclear applications

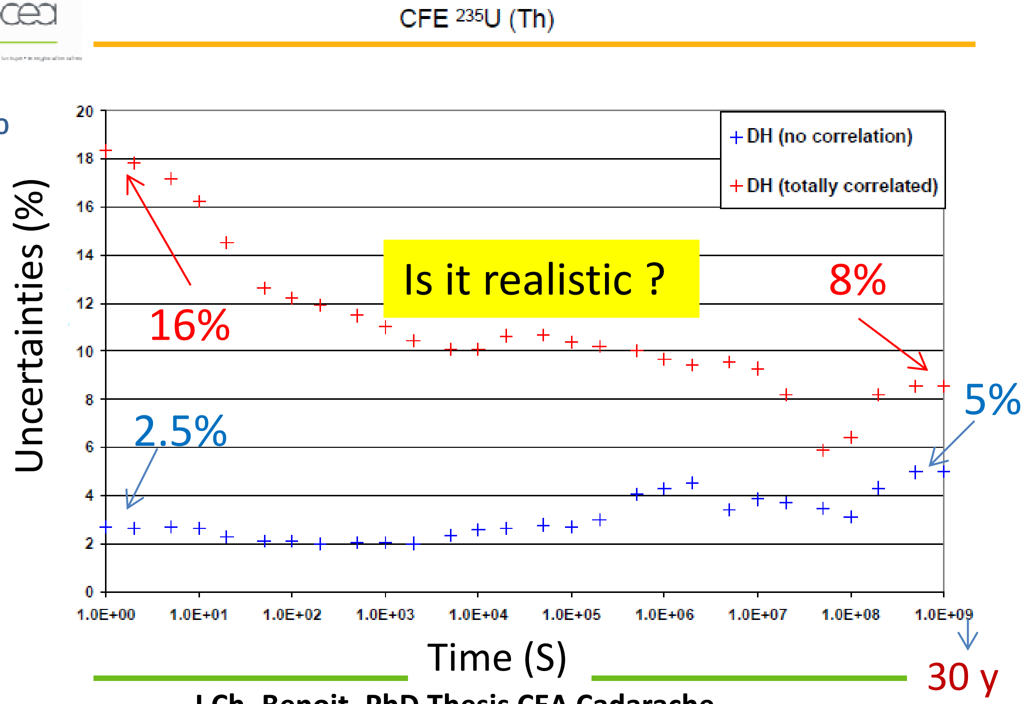
➤Sensitivities to residual power

→ Study realized by CEA/DEN in the framework of the reactor decay heat measurements ***after shutdown:*** only isotope with a lifetime greeter than few seconds are considered ; usual data bases are used (ENSDF, JEFF, ENDF…)

→ Assuming total correlations in fission yields data uncertainties from 8% to 16%

→Uncertainties due to the fission yields are greater than the mean β/γ energy released or the periods with a factor 2.5 to 80 according to :

- the full covariance of fission yields
-data bases available
- gamma ray energy deposition



J.Ch. Benoit, PhD Thesis CEA Cadarache
J.Ch. Benoit, O.Serot et al., Physor 2012.

3- Thermal neutron induced fission yields available

➤ Principal method used for fission yields measurements available :

- **Double Ionization chamber (IC) and Tof : 2E - 2V** :

  - mass resolution ($\sigma > 1\%$) Then mass measurements are naturally correlated
  - Complete mass range
  - For isotopic yields using IC in light mass region
    → charge de-convolution → Cov <0

- **Lohengrin spectrometer at ILL**

  - 1u resolution at 3 σ up to A ~ 150 – 160 according to the target
  - measurement over a complete mass range is impossible with a same target for mass → no complete data set → cross normalization
  - For isotopic yields by gamma spectroscopy, complete range
  - For isotopic yields using IC in light mass region
    → charge de-convolution → Cov <0

- **Radio-isotopic measurement :**

  Cumulated measurement over a long time > accumulation on the long life isotope
  → provide cumulated mass yields
  → no complete mass range

4- Self-normalization - systematic group - and ND Bases : basic elements of correlations

For all methods, the binary fission yields normalization are defined equal to 2

$$\sum_A Y(A) = 2 \Rightarrow Y(A) = \frac{N(A)}{\sum_A N(A)}$$ with $N(A)$ fission rate measurement for mass A

4- Self-normalization - systematic group - and ND Bases : basic elements of correlations

For all methods, the binary fission yields normalization are defined equal to 2

$$\sum_{A} Y(A) = 2 \Rightarrow Y(A) = \frac{N(A)}{\sum_{A} N(A)}$$ with $N(A)$ fission rate measurement for mass A

$$\sum_{A} Y(A) = 2 \Rightarrow Y(A_i) = \frac{N(A_i)}{\sum_{A_j} N(A_k)} = \frac{N(A_i)}{N(A_i) + \sum_{k \neq i} N(A_k)} \rightarrow Y_i = \frac{1}{1 + \frac{1}{N_i} \sum_{k \neq i} N_k}$$

4- Self-normalization - systematic group - and ND Bases : basic elements of correlations

For all methods, the binary fission yields normalization are defined equal to 2

$$\sum_A Y(A) = 2 \Rightarrow Y(A) = \frac{N(A)}{\sum_A N(A)}$$ with $N(A)$ fission rate measurement for mass A

$$\sum_A Y(A) = 2 \Rightarrow Y(A_i) = \frac{N(A_i)}{\sum_{A_j} N(A_k)} = \frac{N(A_i)}{N(A_i) + \sum_{k \neq i} N(A_k)} \rightarrow Y_i = \frac{1}{1 + \frac{1}{N_i} \sum_{k \neq i} N_k}$$

$$S_{Y_i;N_i} = 1 - Y_i > 0$$

$$S_{Y_i;N_{k \neq i}} = -Y_k < 0$$

$\longrightarrow$ Sensitivity of a fission yields to the fissions rates depend of fission yields

4- Self-normalization - systematic group - and ND Bases : basic elements of correlations

For all methods, the binary fission yields normalization are defined equal to 2

$$\sum_A Y(A) = 2 \Rightarrow Y(A) = \frac{N(A)}{\sum_A N(A)}$$

with $N(A)$ fission rate measurement for mass A

$$\sum_A Y(A) = 2 \Rightarrow Y(A_i) = \frac{N(A_i)}{\sum_{A_j} N(A_k)} = \frac{N(A_i)}{N(A_i) + \sum_{k \neq i} N(A_k)} \rightarrow Y_i = \frac{1}{1 + \frac{1}{N_i} \sum_{k \neq i} N_k}$$

$$S_{Y_i;N_i} = 1 - Y_i > 0$$

$$S_{Y_i;N_{k \neq i}} = -Y_k < 0$$

$\longrightarrow$ Sensitivity of a fission yields to the fissions rates depend of fission yields

$$\frac{Cov(Y_i;Y_j)}{Y_i \cdot Y_j} = \sum_k \underbrace{S_{Y_i;N_k} \cdot S_{Y_j;N_k}} \cdot \frac{Var(N_k)}{(N_k)^2} + 2 \cdot \sum_{k>l} \underbrace{S_{Y_i;N_k} \cdot S_{Y_j;N_l}} \cdot \frac{Cov(N_k;N_l)}{N_k \cdot N_l}$$

>0 if k≠i or l≠j

k=i & k≠j or k ≠ i & k=j ; <0
k=i=j ; >0
k ≠ i & k≠j ; >0

4- Self-normalization - systematic group - and ND Bases : basic elements of correlations

For all methods, the binary fission yields normalization are defined equal to 2

$$\sum_A Y(A) = 2 \Rightarrow Y(A) = \frac{N(A)}{\sum_A N(A)}$$

with $N(A)$ fission rate measurement for mass A

$$\sum_A Y(A) = 2 \Rightarrow Y(A_i) = \frac{N(A_i)}{\sum_{A_j} N(A_k)} = \frac{N(A_i)}{N(A_i) + \sum_{k \neq i} N(A_k)} \rightarrow Y_i = \frac{1}{1 + \frac{1}{N_i} \sum_{k \neq i} N_k}$$

$$S_{Y_i;N_i} = 1 - Y_i > 0$$

$$S_{Y_i;N_{k \neq i}} = -Y_k < 0$$

→ Sensitivity of a fission yields to the fissions rates depend of fission yields

$$\frac{Cov(Y_i;Y_j)}{Y_i \cdot Y_j} = \underbrace{\sum_k S_{Y_i;N_k} \cdot S_{Y_j;N_k} \cdot \frac{Var(N_k)}{(N_k)^2}}_{\text{Normalization}} + 2 \cdot \underbrace{\sum_{k>l} \underbrace{S_{Y_i;N_k} \cdot S_{Y_j;N_l}} \cdot \frac{Cov(N_k;N_l)}{N_k \cdot N_l}}_{\text{Systematic uncertainties}}$$

example : Lohengrin measurements

- Lohengrin mass separator

Lohengrin : selection with the mass on ionic charge ratios A/q and Kinetic energy on Ionic charge E/q

$$(A_1,E_1,q_1) \equiv (A_2,E_2,q_2) \equiv (A_3,E_3,q_3)$$

Setup:

- IC & $A/\Delta A|_{\text{Lohengrin}} = 400$
- Ge Clover

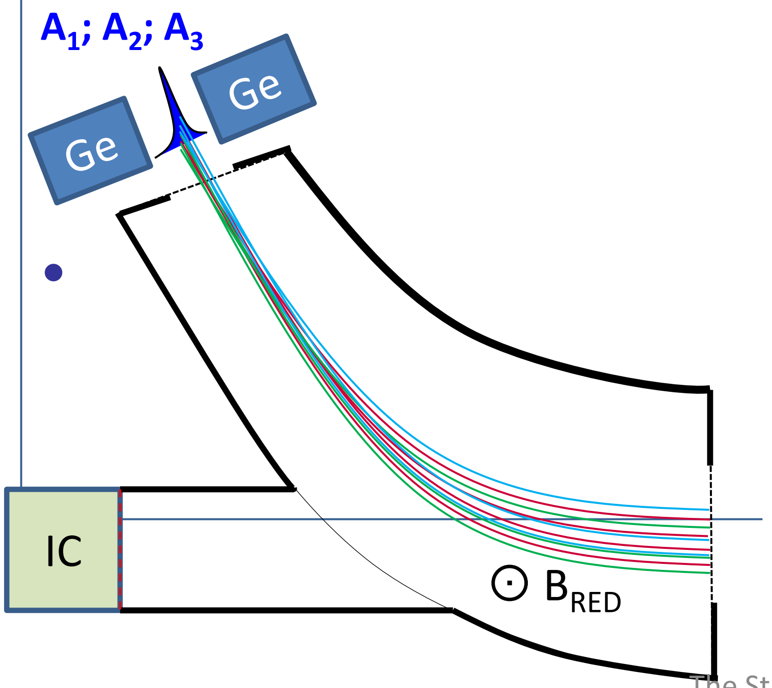
➤ mass yields up to A = 155 (at 3σ)

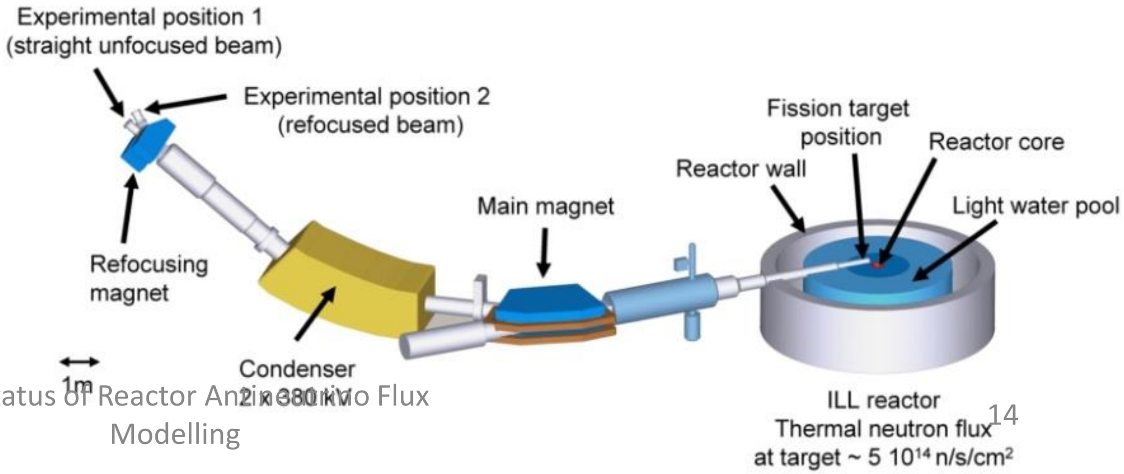
➤ Isotopic yields with γ spectrometry

➤➤ for low yields or low γ intensities,

signal/background ratio is too poor to obtain sufficient

accuracy





example : Lohengrin Mass measurements

- **Method : relative measurements** **(Same method for Isotopic yields)**

$$\left.\begin{array}{l} N(A) = \dfrac{Bu(t)\cdot \sum\limits_{q} \int\limits_{E} N(A,q,E)dE}{N(A,\overline{q},\overline{E})} \\ \sum\limits_{\text{Heavy}A} Y(A) = 1 \end{array}\right\} \Rightarrow Y(A) = \frac{N(A)}{\sum\limits_{\text{Heavy}A} N(A)}$$

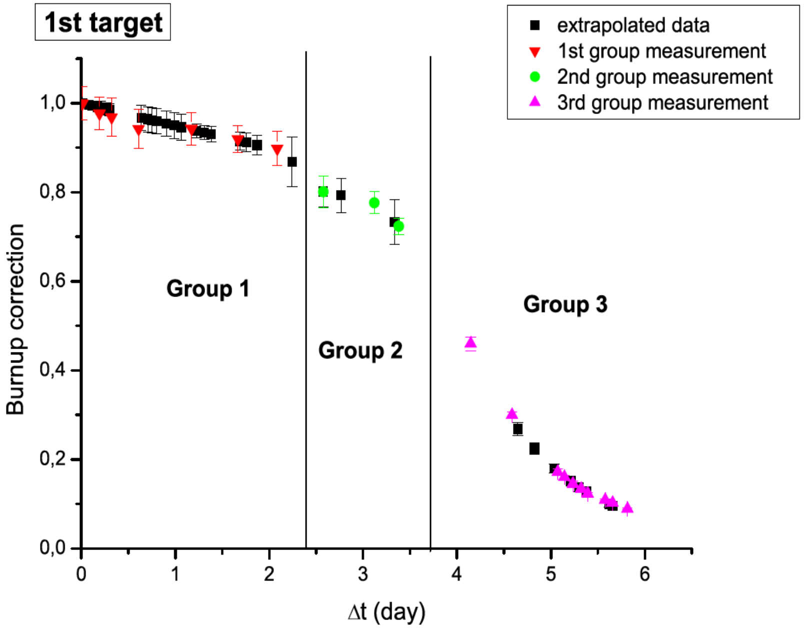
- **cross normalization** between measurements of each targets → systematic correction

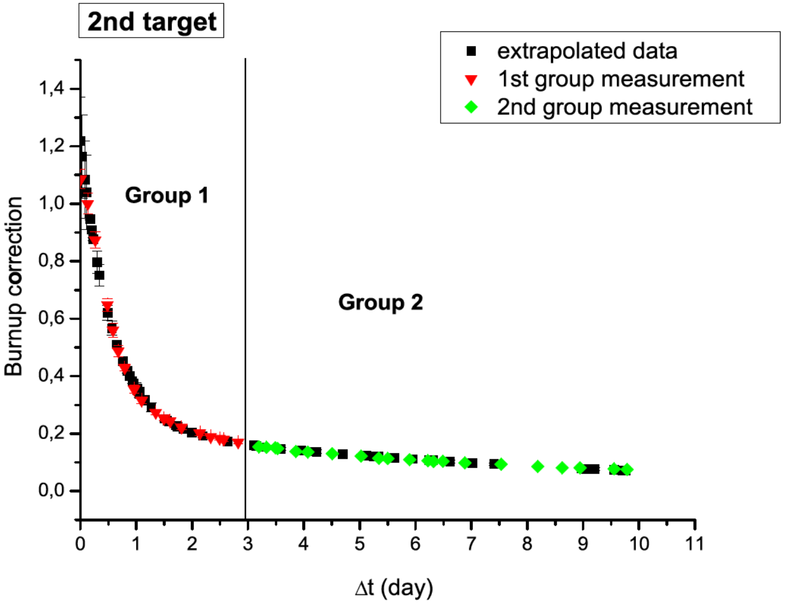
example : Lohengrin Mass measurements

- **Method : relative measurements** **(Same method for Isotopic yields)**

$$\left.\begin{array}{l} N(A)=\dfrac{Bu(t)\cdot\sum\limits_{q}\int\limits_{E}N(A,q,E)dE}{N(A,\overline{q},\overline{E})} \\ \sum\limits_{\text{Heavy}\,A}Y(A)=1 \end{array}\right\}\Rightarrow Y(A)=\frac{N(A)}{\sum\limits_{\text{Heavy}\,A}N(A)}$$

- **cross normalization** between measurements of each targets → systematic correction
- **burn up evolution Bu(t)**: target sputtering → systematic correction





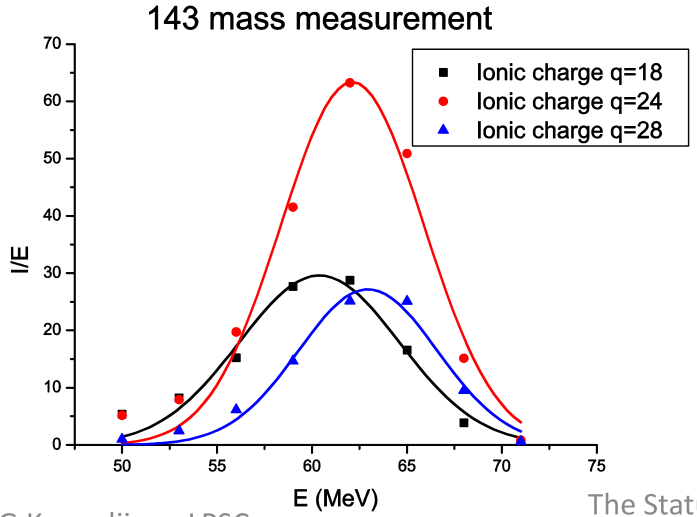
example : Lohengrin Mass measurements

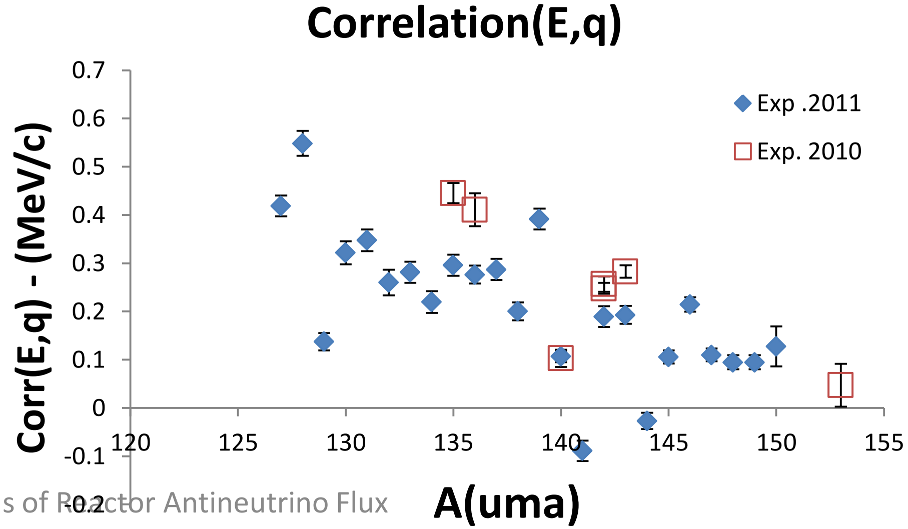
- **Method : relative measurements** **(Same method for Isotopic yields)**

$$\left.\begin{array}{l} N(A) = \dfrac{Bu(t)\cdot \sum\limits_{q} \int\limits_{E} N(A,q,E)dE}{N(A,\bar{q},\bar{E})} \\ \sum\limits_{\text{Heavy}A} Y(A) = 1 \end{array}\right\} \Rightarrow Y(A) = \frac{N(A)}{\sum\limits_{\text{Heavy}A} N(A)}$$

- **cross normalization** between measurements of each targets → systematic correction
- **burn up evolution Bu(t)**: target sputtering → systematic correction
- **kinetic energy distribution E_k**
- **Ionic charge distribution q**

-**(E_k ,q) correlation**

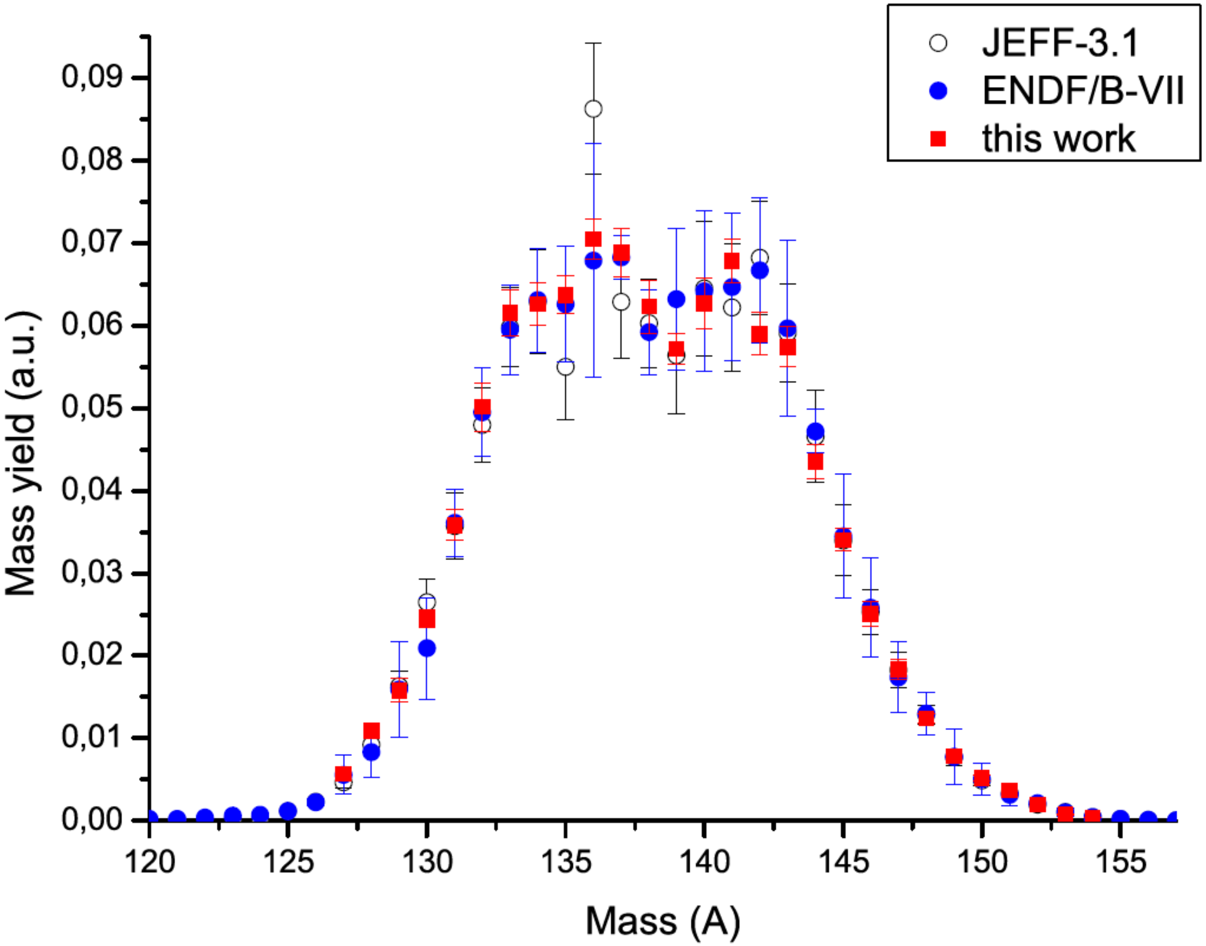

143 mass measurement


Correlation(E,q)

example : Lohengrin Mass measurements

e.g. $^{233}U(n_{th},f)$ mass yields in heavy mass region :

→ partial results using 2 targets



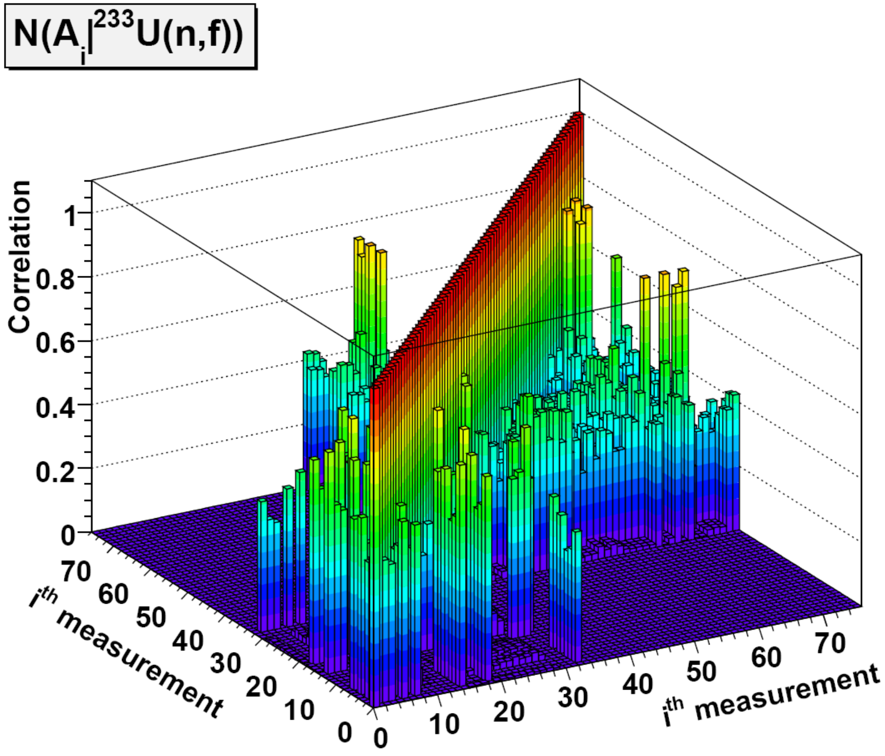
PhD thesis
F. Martin

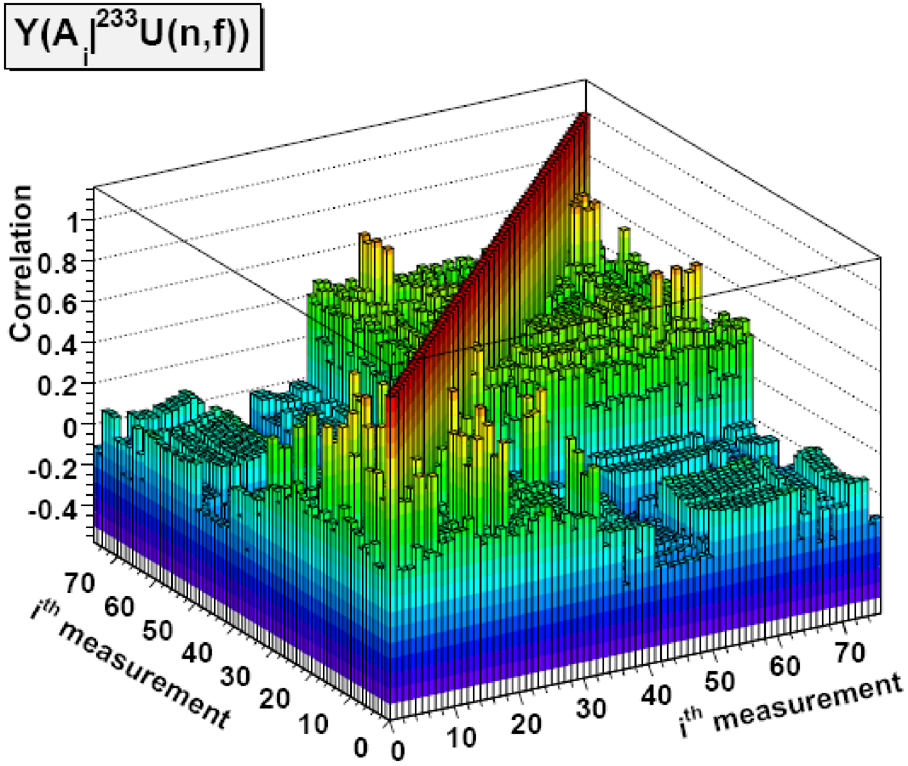
example : Lohengrin Mass measurements

e.g. $^{233}U(n_{th},f)$ mass yields in heavy mass region :
→ partial results using 2 targets

$$Corr(Y_i;Y_j) = \frac{Cov(Y_i;Y_j)}{\sigma(Y_i)\cdot\sigma(Y_j)}$$

- Correlation matrix between the N_{Ai} (number of events)
- Correlation matrix between the Y_{Ai} (mass yields)





example : Lohengrin Mass measurements

Questioning on the data sets :

- If all sets correspond to a complete measurement

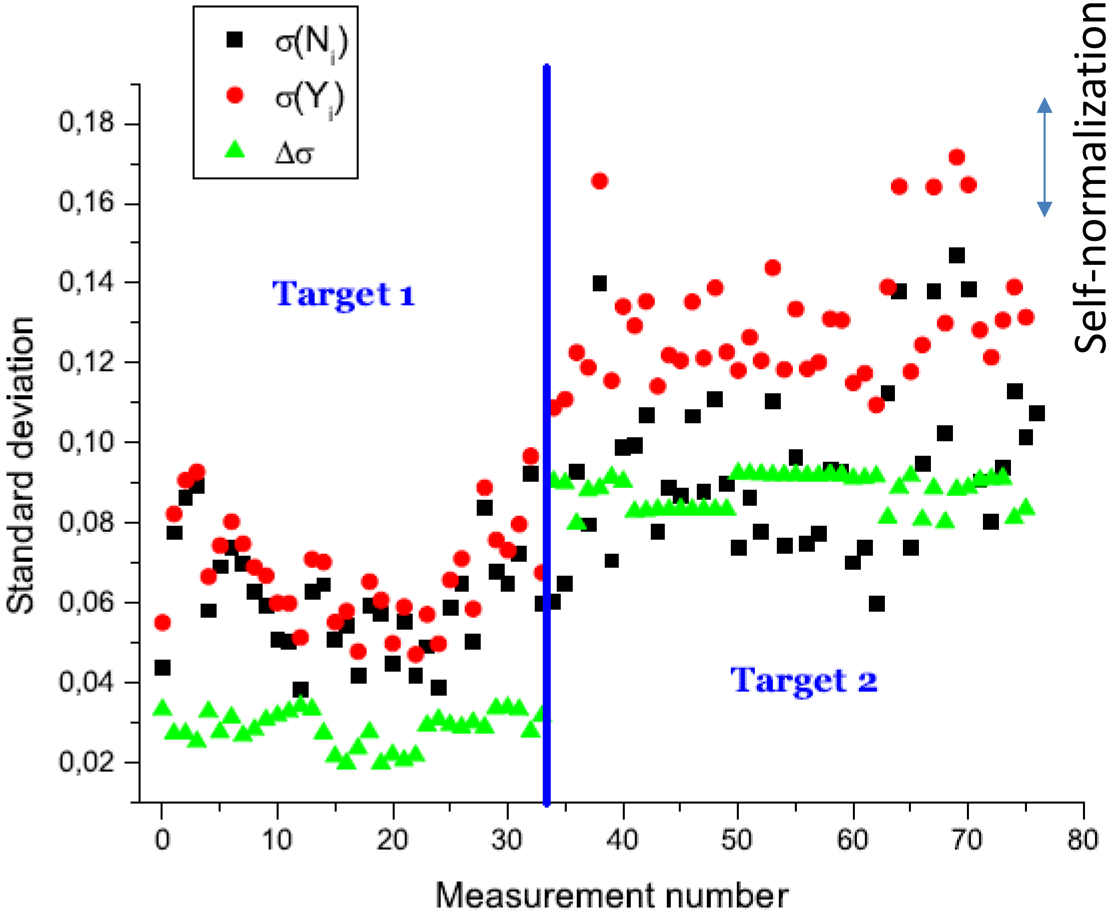
→Yields and covariance are defined set by set

- If the data sets are not complete

→ need a cross normalization

→ covariance terms between elements of different sets exist

→ induce a positive contribution on the fission yields variances



5- Proposal to increase the constraints on the decay heat

Complete the β/γ measurements per fissioning nucleus with differential β/γ decay heat measurements per mass A in order to :

- Increase the number of measurements to test calculations
- Change the systematic uncertainties of the experiment
- measure directly

$$P(t;\beta;\gamma|A) = f\,(t;Y(A,Z);E_{\beta;\gamma};\lambda)$$

Yields – Energy released – Periods

- Intermediate measurements between TAGS and elementary fission curve (integral measurement per actinide)

- Measurement in a time range starting from 1-2 µs

5- Proposal to increase the constraints on the decay heat

Coupled Lohengrin ⊗ Gas Filled Magnet spectrometers

→ Goal : quasi Isobaric beam

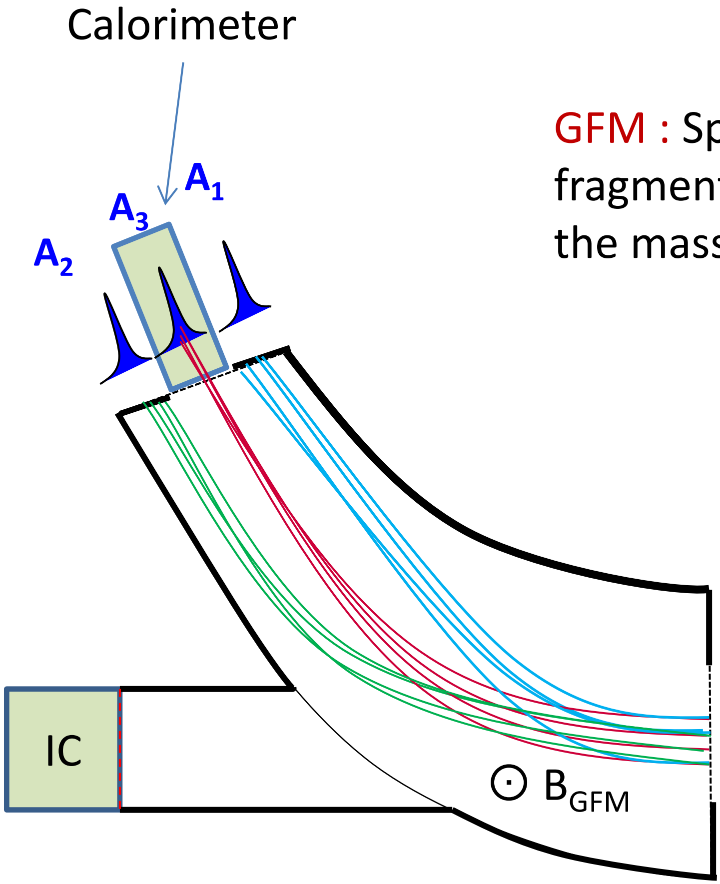
Lohengrin : selection with the mass on ionic charge ratios A/q and Kinetic energy on Ionic charge E/q

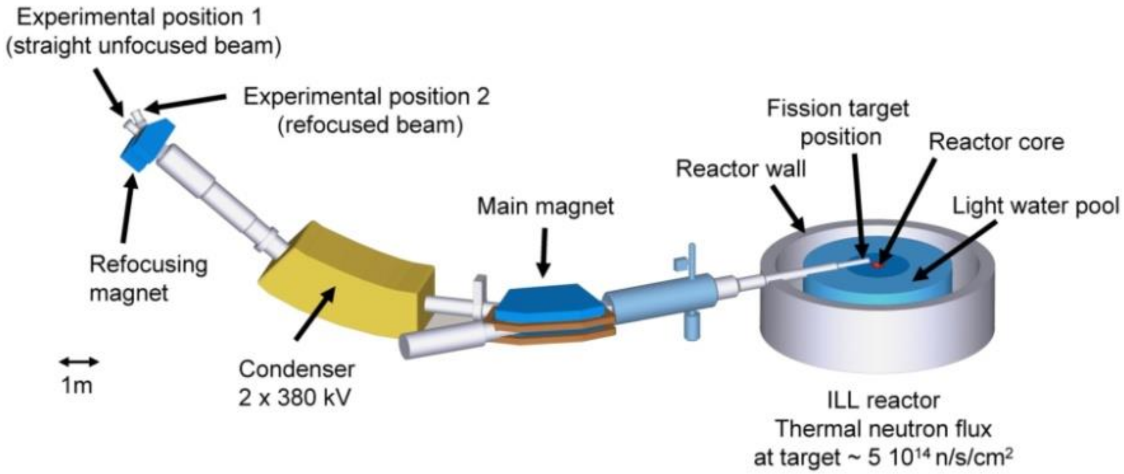
$$(A_1,E_1,q_1)\equiv (A_2,E_2,q_2) \equiv(A_3,E_3,q_3)$$

GFM : Spatial dispersion of fission fragments according to the mass A and Nuclear charge Z

$$\left| \begin{array}{l} B\cdot\rho \propto A\cdot\left\langle \frac{v(Z)}{q(Z)}\right\rangle_{\mathrm{Gaz,P}} \\ B\cdot\rho \propto \frac{A}{Z^{1/3}} \end{array} \right.$$

[1]

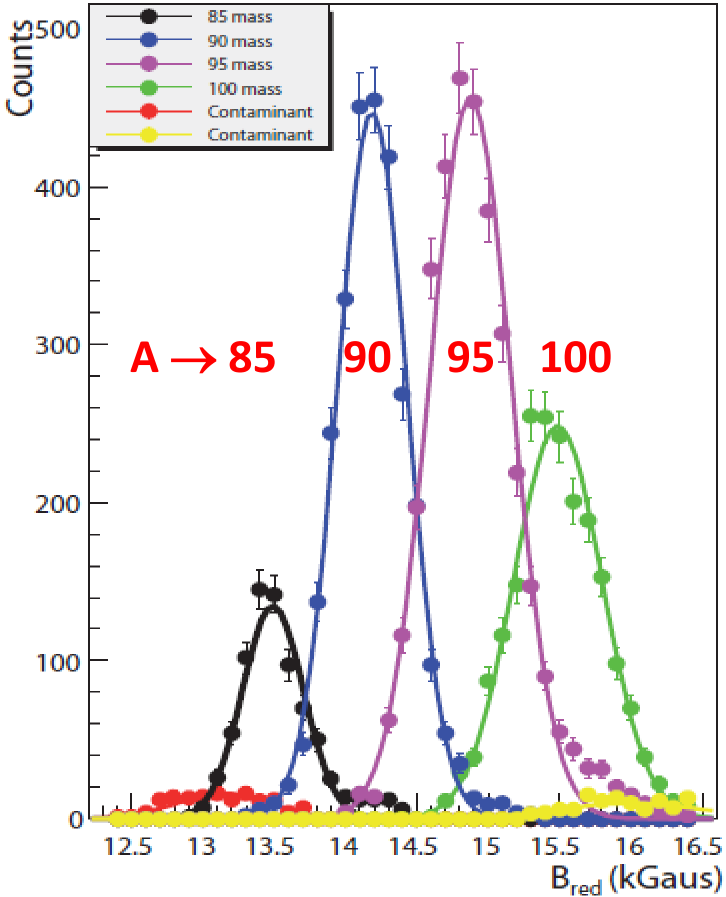




5- Proposal to increase the constraints on the decay heat

Available ^{4}He Gas Filled Magnet spectrometer@ Lohengrin

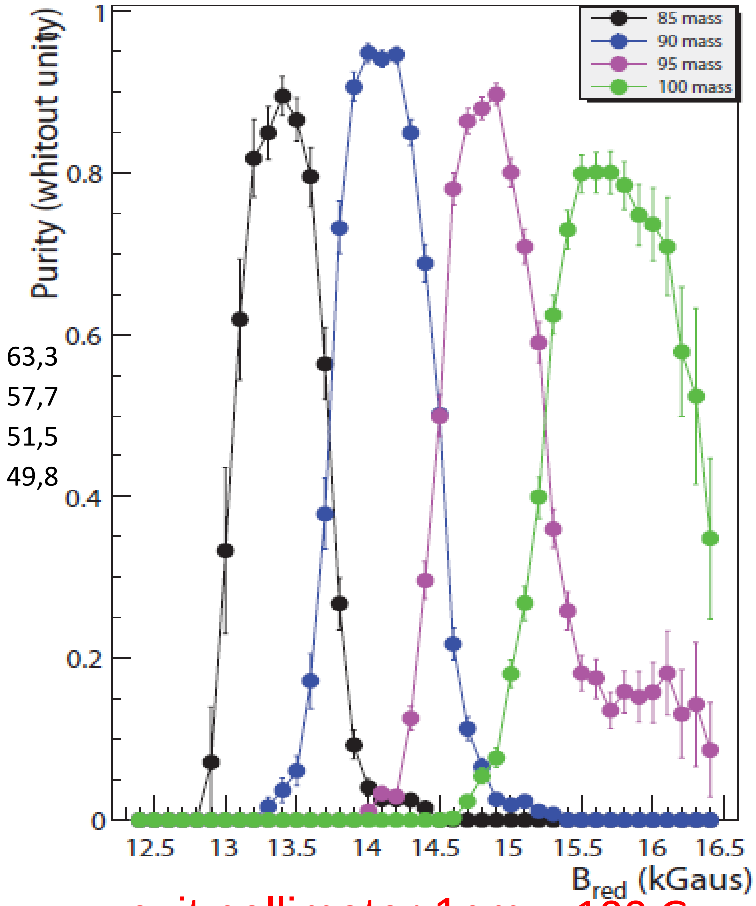
Mass separation with GFM



A → 85 90 95 100

A/ΔA → 63 ; 58 ; 52 ; 50

Evolution of Purity of a mass with Bred



63,3

57,7

51,5

49,8

exit collimator 1cm ≡ 100 Gauss

Conclusion

- Due to the competition between normalization and systematic uncertainties, correlation matrix doesn’t get extreme values (1 or -1) but could have huge structures around zero

- Nevertheless, the structures induce coherence between mass and isotopic uncertainties and then error compensations in calculations.

- To increase the precision on the decay heat, it could be interesting to complete the differential and integral measurements with semi-differential measurements.
→ Coupled Lohengrin ⊗ Gas Filled Magnet spectrometers and β/γ calorimeter

Thus, we will increase the number of comparisons and identify the isobaric chains where TAGS are required

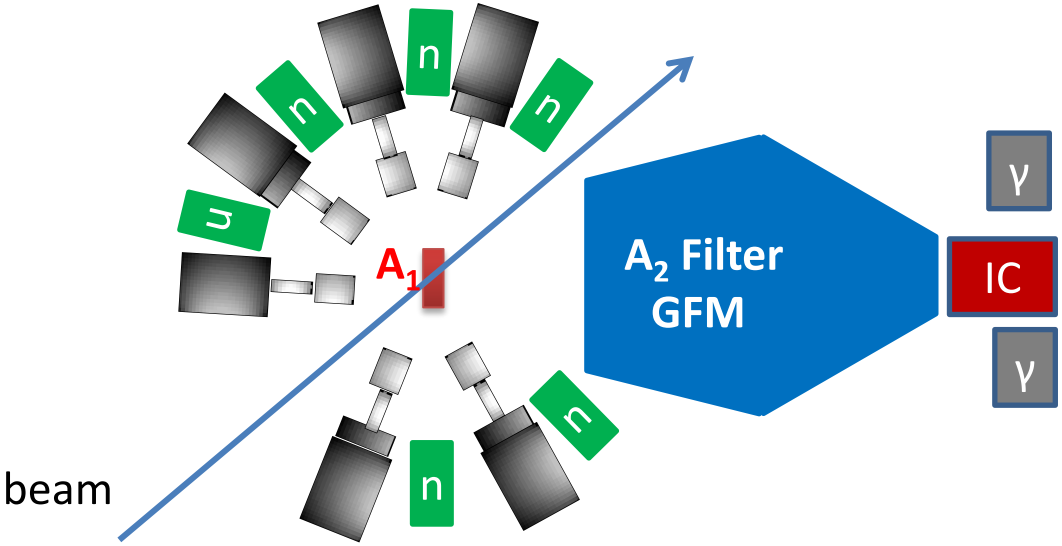
Perspectives @ ILL

FIPPS Project dedicated to fission induced prompt particle study.

- Project of new instrument, complementary to the Lohengrin facility
  → Nuclear structure and nuclear fission studies

- n/gamma detectors coupled to a fission fragment filter

- Goal of the filter :
  - Characterize the complementary mass ($<A_2>$, E_k)
  - Clean the gamma spectrum to identify the discrete gamma rays of (A_1, Z_1)



Collaboration for the measurement campaign @Lohengrin :

C. Sage, G. Kessedjian, A. Chebboubi, A. Bidaud, A. Billebaud, N. Capellan,
S. Chabod, O. Méplan
LPSC, UJF, INP, Grenoble

H. Faust, U. Köster, A. Blanc, P. Mutti,
ILL, Grenoble

O. Litaize, O. Serot, D. Bernard
CEA/Cadarache

A. Letourneau, S. Panebianco, T. Materna, C. Amouroux
CEA/Saclay

X. Doligez, ***IPN, Orsay***

PhD thesis : F.Martin, C. Amouroux

Backup

1- Fission yields : why do we need new measurements ?

Pourquoi des rendements > puissance Beta gamma (t)
    intérêt puissance residuelle
    intérêt spectre beta > neutrino
        total beta /gamma emission per fissioning nucleus

1.Thermal neutron induced fission : Lohengrin ???

2-Mesure de rendement > auto-normalisation ou non ! Independant de JEFF
    Mass
    Isotopic   dependant de eval ou de mesures Y(A)
    dependant de la manip, des choix d'analyses
        eg SOFIA mass resolution sig = 0.4
            Z resolution sig = 0.6
        Mass and charge matrice is statistical deconvolution

covariance $\neq$ systematic uncertainty

3- Impact des cov dans l'écal de la puissance résiduelle

4- Option pour contraindre les incert : mesure de puissance residuelle par ligne isobarique produit par la fission
    Mesures de TAGS > isotope/isotope
    GFM >   Mass per mass /fissioning isotope
    FIPPS

1- Fission yields : why do we need new measurements ?

➤ Impact of fission yields in the actual and innovative fuel cycles

→ Inventory of used fuel : isotopic composition
→ Residual power : minor actinides and fission products
→ Radiotoxicity of used fuel
→ Experimental fuel studies : reaction cross sections and isotope yields are needed to comparison Calculation/ Experiment (C/E)
→ Calculation/prediction of prompt γ rays emitted in a core

Main contributors
~20
Isotopes

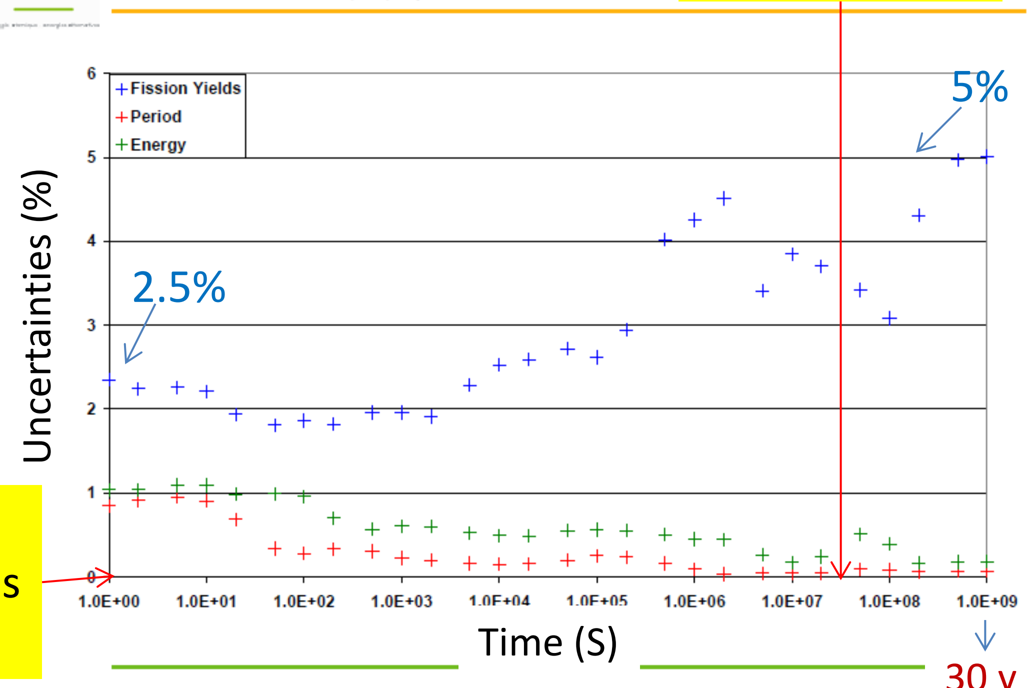
➤ Sensitivities to residual power

→ Independent measurements : uncertainties from 2.5% to 5%

→ Total correlations in data uncertainties from 8% to 16%

→Uncertainties due to the fission yields are greater than the mean β/γ energy released or the periods with a factor 2.5 to 800.

CFE ^{235}U (Th) : paramètres totalement



30 y

≥ 100
Important Isotopes
after shutdown

J.Ch. Benoit, PhD Thesis CEA Cadarache
J.Ch. Benoit, O.Serot et al., Physor 2012.

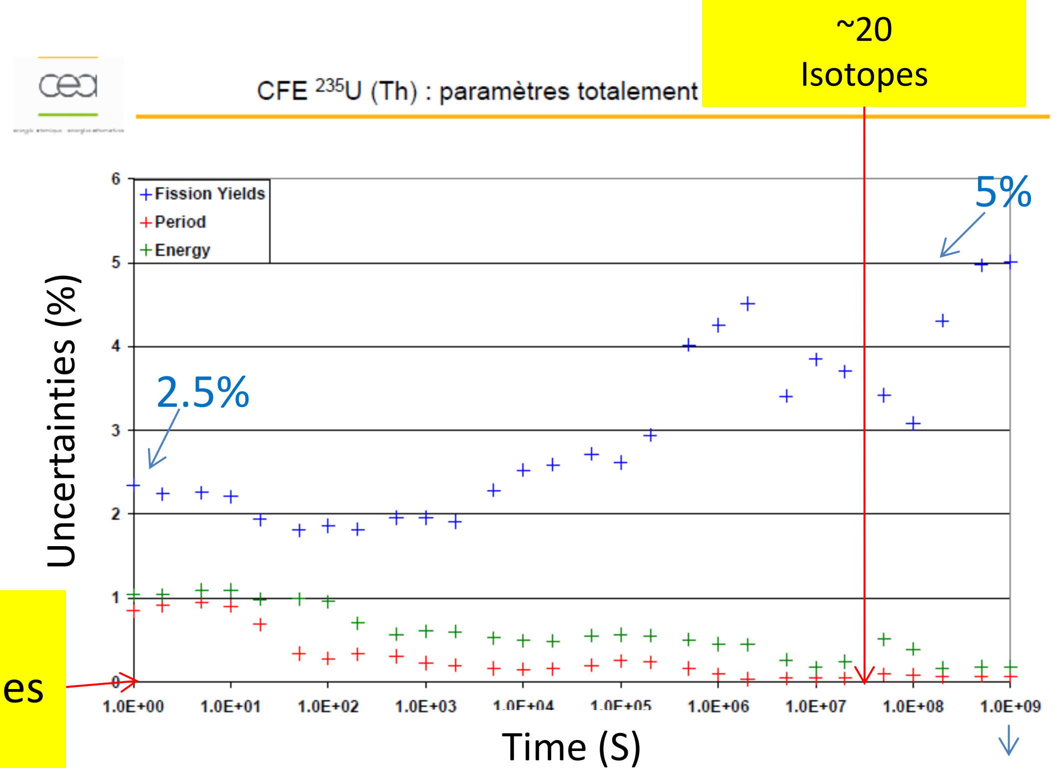
2- Impact of the covariance in the decay heat for nuclear applications

➤Sensitivities to residual powe

→ Independent measurements
uncertainties from 2.5% to 5

→ Total correlations in data
uncertainties from 8% to 16

→Uncertainties due to the fiss
yields
are greater than the mean β/γ
released or the periods with a
2.5 to 800.

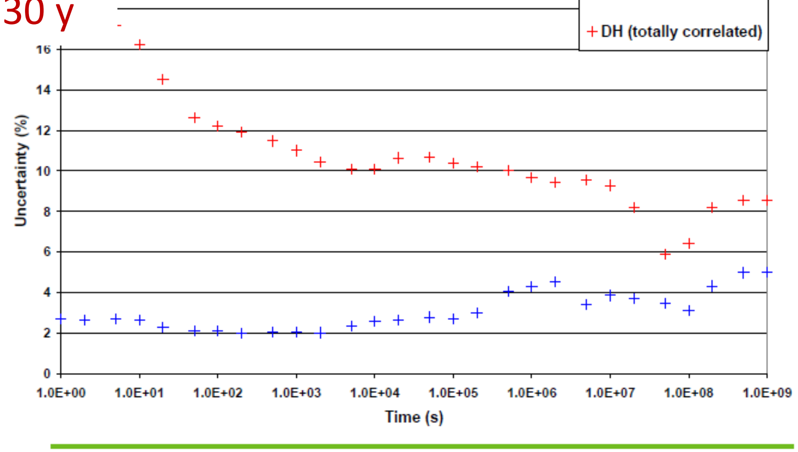


)
sotopes
down

30 y

J.Ch. Benoit, PhD Thesis CEA Cadarache
J.Ch. Benoit, O.Serot et al., Physor 2012.

Evaluation : No covariance avalaible
Mass = ∑Isitope
Variance(Mass) = ∑ Var(Isitope) +
∑ Cov (Isotope)



example : Lohengrin measurments

- Lohengrin mass separator

Lohengrin : selection with the mass on ionic charge ratios A/q and Kinetic energy on Ionic charge E/q

$$(A_1,E_1,q_1) \equiv (A_2,E_2,q_2) \equiv (A_3,E_3,q_3)$$

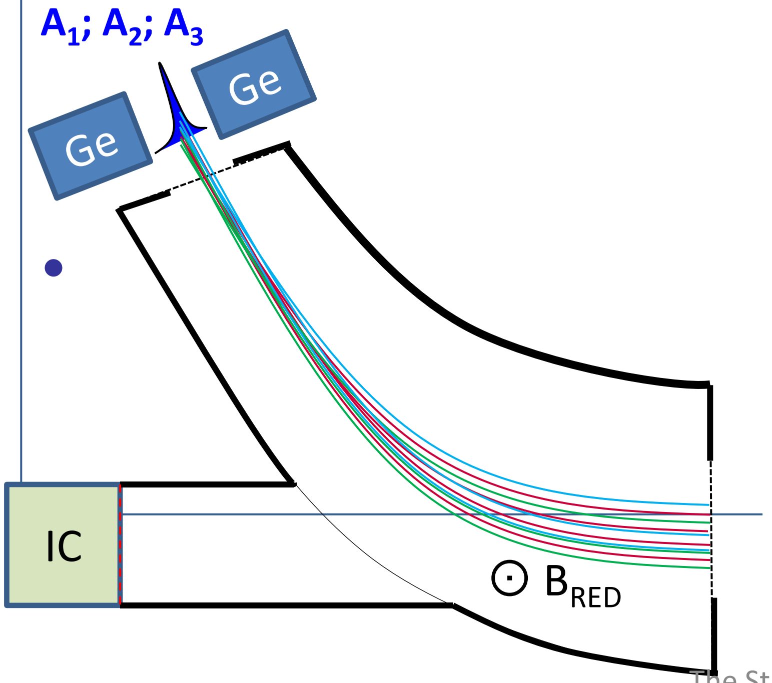
Setup:

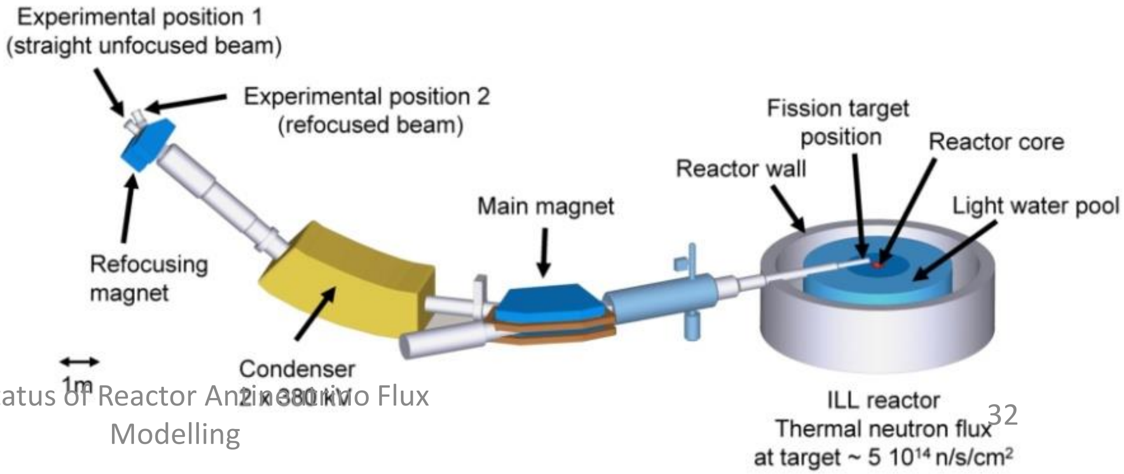
- IC & $A/\Delta A|_{\text{Lohengrin}} = 400$
- Ge Clover

➤ mass yields up to A = 155 (at 3σ)

➤ Isotopic yields with γ spectrometry

➤➤ for low yields or low γ intensities, signal/background ratio is too poor to obtain sufficient accuracy





example : Lohengrin measurments

- **Method : relative measurements**

$$\left.\begin{array}{l} N(A) = \dfrac{Bu(t)\cdot \sum\limits_{q} \int\limits_{E} N(A,q,E)dE}{N(A,\overline{q},\overline{E})} \\ \sum\limits_{A} Y(A) = 2 \end{array}\right\} \Rightarrow Y(A) = \frac{N(A)}{\sum\limits_{A} N(A)}$$

- **cross normalization** : inerrant problem of the fission yield measurements > sum ≡2 !

    - Even if no systematic exist in the determination of the FF rates, the yields are self-correlated

        > few percents (3 - 5% for 5% precision of rate N(A)

    - Partial measurements : normalization to the evaluations

        > history dependent > time dependent

            > few percents (5-15%) if experimental data are not de-normalized for the evaluations!

            > Raw data dependence > information on experimental methods

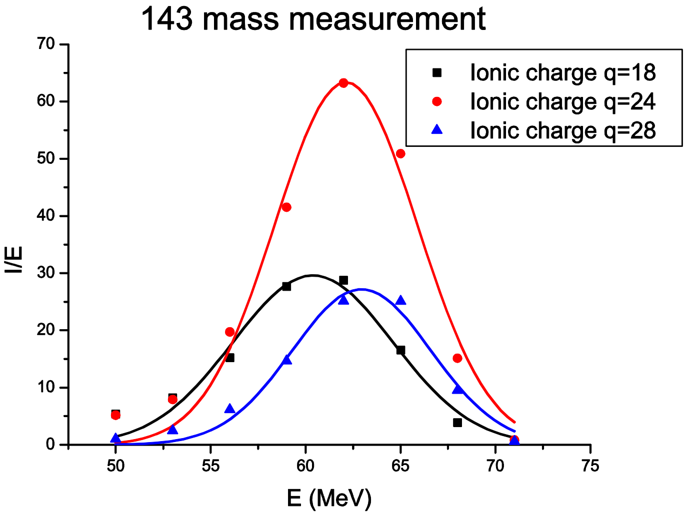
            > intrinsic normalization of the method

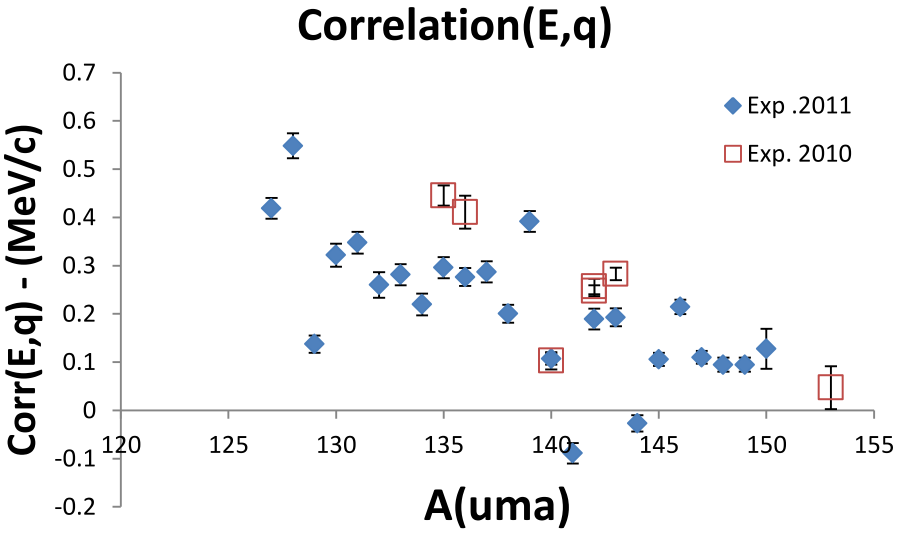
-**burn up evolution Bu(t)**: target sputtering > the dependence of the target (production) and target thickness used > At least few percent (1-5%) according the target

1-Lohengrin facility : method and limits

- **kinetic energy distribution E_k :**
  - > no models due to the dependence of the target made
  - > At least few percents according to the full description or not (0.-3%)
- **Ionic charge distribution q** :
  - > not completely at low charge (10-16)> limit of the Lohengrin electric fields
  - > electron conversion depend of the nuclear structures of Isotopes
    - > At least few percent (1-3%)

-(E_k ,q) correlation > At least few percent (3-5%) if no measurement mass per mass





1-Lohengrin facility : method and limits

- **Uncertainties are-they independent ?**

  - Kinetic energy distribution > Yes if complete E_k distribution is detailed
  > No if assumption on the E_k distribution (tail)
  - Ionic charge distribution > not completely due to the low ionic charge
  - Burnup > No !
  - cross normalization > Never !!!

- **Limit of precision on the final yields Y(A)** : 5 to 10 % if there are not assumptions on the method of measurement. In the available data set, only few isotopes in the *light fragment region* have been studied

- **New measurements** :

  - complete range mass – complete distributions > independent of existing data
  > consequence : beam time for this kind of measurements !
  - Covariance matrix is not a problem, it is the solution
  > Variance-Covariance builds the coherence in data set

What is a true measurement ? Eigen value of covariance matrix of the measurements !

$^{233}U(n_{th},f)$ mass yields : analysis

High fission rate :

Self sputtering => apparent target thickness reduction

Evolution monitored by repeated 136/21/E scans