

# Trends on Major Actinides from an Integral Data Assimilation

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**Abstract:** Nuclear data evaluations on major actinides can be improved by integral data assimilation. Appropriate integral measurements with reliable experimental techniques have been selected such as ICSBEP, IRPhE and MASURCA critical masses, PROFIL irradiation experiments and the FCA-IX experimental programme (critical masses and spectral indices). Highly reliable analyses are possible with the use of as-built geometries calculated with the TRIPOLI4 Monte Carlo code. The C/E values have been used in an integral data assimilation solving the Bayes equation. The trends on the JEFF3.1.1 <sup>235</sup>U capture cross section are quite consistent with recent differential measurements. Assimilation results suggest up to a 2.5% decrease for <sup>238</sup>U capture from 3 keV to 60 keV, and a 4-5% decrease for <sup>238</sup>U inelastic in the plateau region. For this energy range, uncertainties are respectively reduced from 3-4 to 1-2% and from 6-9% to 2-2.5% for <sup>238</sup>U capture and <sup>238</sup>U inelastic. Results on <sup>239</sup>Pu fission cross sections are included in posterior uncertainties. The increase trends on <sup>239</sup>Pu capture cross-section is of around 3% in the [2 keV-100 keV] energy range. For <sup>240</sup>Pu capture cross section, the increase is of around 4% in the [3 keV-100 keV] energy range and goes in the same direction as a recent evaluation.

## 1. INTRODUCTION

Five out of the six Generation IV concepts are Fast Reactors with breeding capability (they produce Pu out of depleted Uranium) and with ability to use possibility Pu and MA, that would otherwise be a waste (Pu from PWR-MOX not suitable for PWR).

In France, 2006 June 28 act requires the design of a generation of sodium fast reactors, those being the most mature Generation IV concept. It is the reason for which CEA with its industrial partners started the design of ASTRID<sup>1,2</sup> (Advanced Sodium Technological Reactor for Industrial Demonstration), a 600 MWe sodium-cooled fast reactor concept. Its construction should demonstrate the feasibility of a SFR design with enhanced safety and its ability to achieve a zero breeding gain hence enabling to use the entire Uranium ore and not only <sup>235</sup>U.

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Sodium Fast Reactors require the development and validation of scientific calculation tools as a proof of their characteristics used in their safety dossiers. Nuclear data, the input parameters of the neutronic codes, constitute the main source of uncertainty in neutronic calculations<sup>3</sup>. For instance, uncertainty (in pcm) on critical mass for the ASTRID core (using JEFF3.1.1 and its associated COMAC-V1 nuclear data covariances) amounts up to 1558 pcm. Major actinides are contributing significantly to this uncertainty as Table 1 illustrates.

**Table 1.** Uncertainty breakdown (in pcm) on critical mass for the ASTRID core (using COMAC-V1 nuclear data covariances).

Isotope	Fission	Capture	Elastic	Inelastic	NxN	Nu	Fission Spectrum	Total
<sup>16</sup> O	0	110	50	0	0	0	0	120
<sup>23</sup> Na	0	20	10	10	0	0	0	30
<sup>56</sup> Fe	0	80	30	50	0	0	0	100
<sup>238</sup> U	439	869	30	649	30	70	60	1168
<sup>238</sup> Pu	10	20	0	0	0	80	40	100
<sup>239</sup> Pu	699	190	10	60	10	100	210	759
<sup>240</sup> Pu	669	300	0	60	10	20	170	619
<sup>241</sup> Pu	70	90	0	10	0	70	150	200
<sup>242</sup> Pu	40	50	0	20	0	20	40	80
TOTAL	1069	859	60	649	30	170	320	1558

Existing fertile blankets with depleted U induces great sensitivities to <sup>238</sup>U cross-sections, notably inelastic. The use MOx fuel with Pu retrieved from PWR-MOx spent fuel induces high sensitivities not only to <sup>239</sup>Pu cross sections but also to other Pu isotopes cross sections (notably <sup>240</sup>Pu).

## 2. STRATEGY USED

Integral Data Assimilation (IDA) can contribute to nuclear data improvement if attention is being given to ways that minimize the possibility of creating compensating errors. This approach has been done in the past and it is important to learn from past integral data assimilation works to develop a strategy that can avoid important compensating errors.

### 2.1 Previous integral data assimilation works

Since the early seventies, several integral experiments assimilation work for fast reactor applications have been conducted.

Among them, the statistical adjustment method (called BARACCA) was used to produce two CARNAVAL libraries (III and IV)<sup>4</sup> with the objectives of answering needs for PHENIX and SUPER-PHENIX respectively. For the adjustment, for each nuclear data, equiprobable probabilities were associated to an interval of possible values. This choice was done because assuming that the discrepancy in C/E values for integral experiments came mainly from normalization issues when performing microscopic measurements. Because, there were no constraints in the a priori multigroup cross sections other than the boundary of the initial interval, work resulted in compensating errors between <sup>238</sup>U capture cross sections and <sup>239</sup>Pu fission cross section.

ERALIB1<sup>5</sup> is a library resulting from an integral data adjustment whose experimental database includes 355 integral data (critical masses, spectral indices, buckling etc.) coming from 71 distinct experimental programs. Prior variances-covariances matrices associated to

nuclear data were produced from expert judgements. The improvements made on the assimilation process include:

- The use of Gaussian distributions for multigroup nuclear data to perform adjustments based on Bayes Inference using Generalized Least Square Methods.
- The use of statistical indicators ( $\chi^2$  criteria) to identify inconsistent integral data, which need to be eliminated from the experimental database.

However, the experimental database included many unreliable measurements and some of covariance were lacking physical meaning (especially for  $^{23}\text{Na}$ ). This led in particular to discrepant sodium void worth.

## 2.2 Current Strategy

To avoid deficiencies identified in previous Integral Data Assimilation (IDA) work, a more reliable nuclear data library was used JEFF3.1.1<sup>6</sup> for all isotopes with the exception of  $^{23}\text{Na}$  for which JEFF3.2 was used<sup>7</sup>. The associated COMAC V1 covariance<sup>8</sup> were used as they mimic the evaluation process itself.

Only reliable integral experiments have been selected. They are ICSBEP, IRPhE and MASURCA critical masses, PROFIL irradiation experiments and the FCA-IX experimental programme (critical masses and spectral indices). Highly reliable experiment analyses are now possible avoiding method approximation and using as-built geometries as it is possible with the TRIPOLI4 Monte Carlo code<sup>9</sup>.

Marginalization technique has been used for light and structural isotopes for which approximations (anisotropy, secondary neutron energy distribution) in the integral data assimilation technique are rather high. A test on whether Prompt Fission Neutron Spectrum (PFNS) needs to be fitted or not (such as in ERALIB1). It appears from this test that PFNS is a major source of uncertainty and requires to be fitted<sup>12</sup>.

Attention has been given to ways that minimize compensating errors. Compensating errors between  $^{238}\text{U}$  capture and  $^{239}\text{Pu}$  fission (such as in CARNAVAL IV) has been eliminated by using first only U-fueled experiments then adding Pu-fueled experiments.

The IDA has been using Bayesian inference<sup>10</sup> with a minimization of a cost function (1) in which  $M_\sigma$  is the covariance matrix associated to nuclear data and  $M_E$  the one associated to integral experiments.

$$\chi_{GLS}^2 = (\boldsymbol{\sigma} - \boldsymbol{\sigma}_{a\text{ priori}})^T M_\sigma^{-1} (\boldsymbol{\sigma} - \boldsymbol{\sigma}_{a\text{ priori}}) + (\mathbf{E} - \mathbf{C}(\boldsymbol{\sigma}))^T M_E^{-1} (\mathbf{E} - \mathbf{C}(\boldsymbol{\sigma})) \quad (1)$$

Solutions for nuclear data are given by equation (2) where  $S$  is the sensitivity matrix calculated with the ERANOS code system<sup>11</sup> while associated uncertainties are given by equation (3):

$$\boldsymbol{\sigma}' - \boldsymbol{\sigma}_{a\text{ priori}} = M_\sigma \cdot S^T (M_E + S \cdot M_\sigma \cdot S^T)^{-1} \cdot (\mathbf{E} - \mathbf{C}(\boldsymbol{\sigma}_{a\text{ priori}})) \quad (2)$$

$$M'_\sigma = M_\sigma - M_\sigma \cdot S^T (M_E + S \cdot M_\sigma \cdot S^T)^{-1} S \cdot M_\sigma \quad (3)$$

## 3. INTEGRAL DATA ASSIMILATION STUDIES

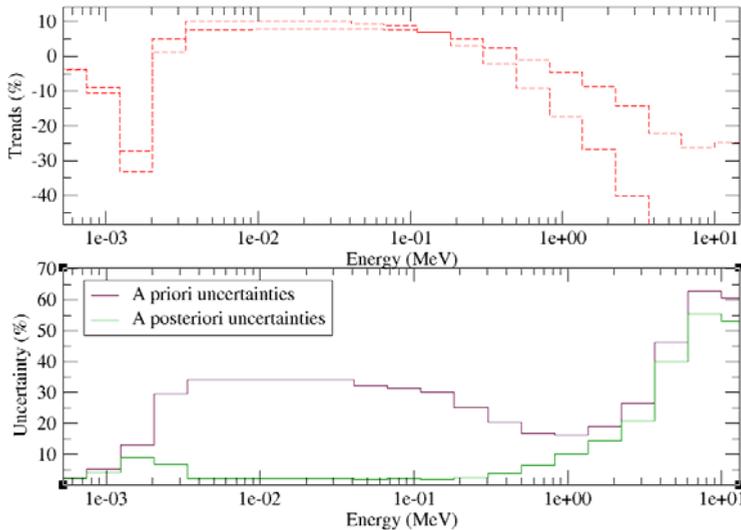
### 3.1 Integral experiments

The Uranium configurations chosen for the exercise are MASURCA R2, MASUSCA 1B, FCA IX-1 to 7, GODIVA and FLATTOP U235. The Plutonium configurations chosen are MASURCA 1A', MASURCA ZONA2, ZPPR10A, ZPR6/7, ZPR6/7 high Pu240, BFS 82-2, MASURCA PRE-RACINE 2A & 2B, SNEAK 7A & 7B, JEZEBEL Pu239 & Pu240, FLATTOP Pu239. Also, FCA-IX fission chambers C/Es of Np237, Am241, Am243, Pu238, Pu242, Cm244 have been used as well as PHENIX PROFIL irradiated samples C/E on U238, Pu238, Pu239, Pu240, Pu241, Pu242, Am241, Np237. C/E values display a great dispersion

in results. Nuclear data uncertainties (of the order of 1000 to 2000 pcm) are far beyond experimental uncertainties, which justify the use of IDA method.

### 3.2 Trends for $^{235}\text{U}$ Capture

As seen on Figure 1, the trends were a -27 to -33 % decrease at 1-2 keV (end of RRR) which is in agreement with Danon measurements (2017)<sup>13</sup> and a +8 to +10% increase in the 10-100keV region (URR) which is in agreement with Jandel measurements (2012)<sup>14</sup>. Uncertainties are significantly reduced.



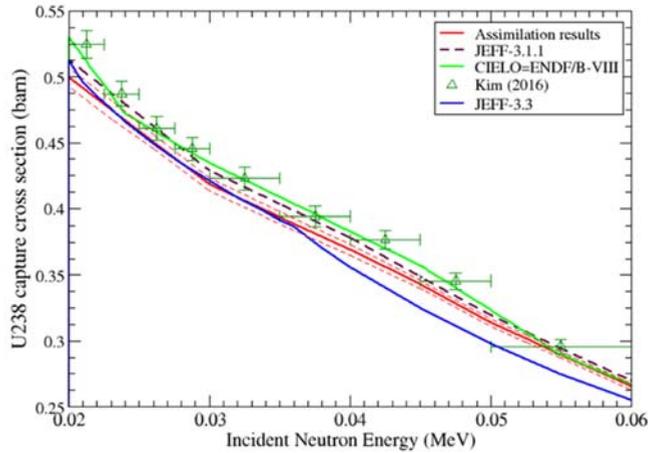
**Figure 1:** Trends on  $^{235}\text{U}$  capture

The consistency of integral data assimilation results on  $^{235}\text{U}$  capture cross section with recent differential measurements encourages us to rely on the simultaneous use of PROFIL C/E and various U235 enriched critical masses to reassess capture cross sections.

### 3.3 Trends for $^{238}\text{U}$ Capture and Inelastic

For  $^{238}\text{U}$  inelastic and capture cross sections, a comparison of  $^{235}\text{U}$ - $^{238}\text{U}$  assimilation results and U-Pu assimilation results allows us to conclude that the trends proposed for these cross sections are not the result of compensating errors with Pu nuclear data. Assimilation results suggest up to a 2.5% decrease for  $^{238}\text{U}$  capture from 3 keV to 60 keV, and a 4-5% decrease for  $^{238}\text{U}$  inelastic in the plateau region. For these energy range, uncertainties are respectively reduced from 3-4 to 1-2% and from 6-9% to 2-2.5% for  $^{238}\text{U}$  capture and  $^{238}\text{U}$  inelastic. The simultaneous use of GODIVA and FLATTOP-235U and JEZEBEL 239Pu and FLATTOP-239Pu is relevant to reassess  $^{238}\text{U}$  inelastic cross sections. Their critical mass C/E are greatly affected by the reflecting properties of the FLATTOP core depleted Uranium blankets very sensitive to this cross section.

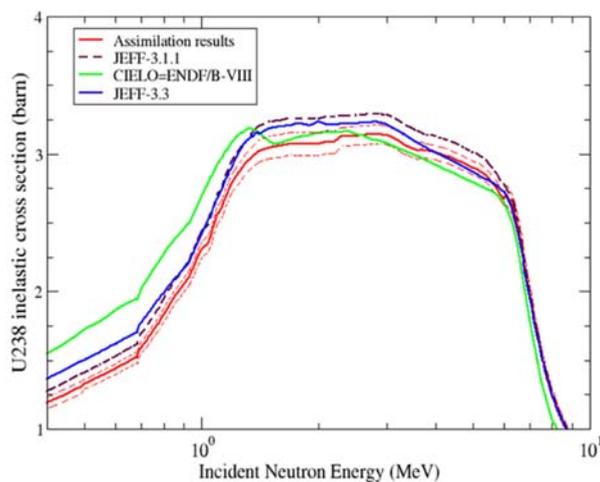
Assimilation results for  $^{238}\text{U}$  capture are compared with differential measurements<sup>15</sup> and with the "a priori" JEFF-3.1.1, CIELO and JEFF-3.3 evaluation (Figure 2). Posterior uncertainties for assimilation results are in dotted line.



**Figure 2:** Assimilation Results compared to major evaluations for  $^{238}\text{U}$  capture

Because Prompt Fission Neutron Spectrum (PFNS) lacks some differential measurements, variances are large. Since integral experiments available cannot distinguish between  $\nu$  and  $\chi$  (PFNS) the possibility of having compensating errors is high. Hence, a parametric study has been conducted with  $^{235}\text{U}$  and  $^{238}\text{U}$  PFNS fitted or not through assimilation. The magnitude of trends on  $^{238}\text{U}$  capture cross sections differ on whether  $^{235}\text{U}$  and  $^{238}\text{U}$  PFNS is fitted or not through assimilation.

In the plateau region ( $\sim 1$  MeV to 6 MeV), a 5-6% decrease is suggested for  $^{238}\text{U}$  inelastic. This decrease trend is in agreement with recent evaluations (see suggested value compared to CIELO and JEFF-3.3 ones on Figure 3; Posterior uncertainties for assimilation results are in dotted line).



**Figure 3:** Assimilation Results compared to major evaluations for  $^{238}\text{U}$  inelastic

Notably, the simultaneous use of FLATTOP-U235/GODIVA and FLATTOP-Pu239/JEZEBEL enables us to discriminate contributions from U238 and fissile isotopes. Indeed, GODIVA is a bare U235 sphere while FLATTOP-U235 is surrounded by a  $U_{\text{depleted}}$  blanket. The same occurs for JEZEBEL and FLATTOP-Pu.

### 3.4 Trends for Pu isotopes

Summary of trends on Pu isotope data, which can have a significant impact for the ASTRID core, are given in Table 2.

**Table 2.** Trends on Pu isotope cross sections

Cross section	Trends (%)	Energy range	Experiments	Comments
$^{239}\text{Pu}$ fission	<1% (+/- 1.7% or less)	0.1 keV- 500 keV	Pu-fueled critical masses	Trends included in posterior unc.
$^{239}\text{Pu}$ capture	from -1.4 to -4.8 % (+/- 2-5%)	0.04 keV - 300keV	PROFIL and critical masses	
$^{240}\text{Pu}$ fission	<b>from - 7.5 to -9.6% (+/-8-11% )</b>	<b>0.75 keV-100 keV</b>	<b>Mostly JEZEBEL- <math>^{240}\text{Pu}</math></b>	<b>Risk of compensating errors with multiplicity and PFNS. Trends are close to posterior uncertainties.</b>
	<b>from -3.5 to -4.6% (+/- 4-7%)</b>	<b>1 keV-3.7 MeV</b>		
$^{240}\text{Pu}$ capture	from 2.5 to -4.2 % (+/- 2-4.5%)	0.7 keV - 100 keV	PROFIL	
$^{242}\text{Pu}$ fission	- 10% (+/- 3.5%)	500 keV- 4 MeV	FCA-IX IS	<b>Underestimated prior uncertainties (~2% in the plateau region)</b>
$^{242}\text{Pu}$ capture	from -3 to -9% (+/- 2-5%)	3 keV - 300 keV	Trends are driven by PROFIL (C/E~1.14).	IDA trends have an unreliable energy breakdown due to Covariances
	around -20 % (+/- 12%)	0.4 keV- 3 keV		

Results on  $^{239}\text{Pu}$  fission cross sections are included in posterior uncertainties which have been significantly reduced through the IDA process (from 3.5-2% to 2.5-0.5%). There is no differential measurement able to reach that type of uncertainty. This means that IDA process will be needed in a way or another. The increase trends on  $^{239}\text{Pu}$  capture cross-section is of around 3% in the [2 keV-100 keV] energy range. From 4keV up to 30 keV, the ENDF/B-VIII evaluation corresponds to JEFF-3.1.1. For  $^{240}\text{Pu}$  capture cross section, the increase is of around 4% in the [3 keV-100 keV] energy range and goes in the same direction as the recent ENDF/B.VIII evaluation though at a much lower level.

### 4. IMPACT ON THE ASTRID CFV CORE

There is a significant impact of  $^{238}\text{U}$  capture and inelastic on the critical mass of the ASTRID core but also  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{242}\text{Pu}$  fission cross-sections. With a prior value for the critical mass of 1.02688 (1.02908 with JEFF-3.1.1 for all isotopes except  $^{23}\text{Na}$  for which JEFF3.2 is used: Impact of using  $^{23}\text{Na}$  evaluation from JEFF-3.2 instead of JEFF-3.1.1 on the ASTRID core reactivity: -220 pcm.) the IDA conducted led to a posterior value of 1.02255 (-435 pcm). The Integral Data Assimilation reduces uncertainties associated to nuclear data significantly with, in particular, a significant reduction on  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  fission cross-sections.

**Table 3.** Impact of the nuclear data trends (in pcm) on the critical mass of the ASTRID core and associated uncertainty (a priori uncertainties using COMAC-V1 nuclear data covariance are also given).

all values in pcm	Impact on $k_{\text{eff}}$	Posterior associated uncertainty contribution	Prior associated uncertainty contribution
U238_CAPTURE	249	180	869
U238_NU	0	40	70
U238_FISSION_SPECTRUM	12	40	60
U238_ELASTIC	-7	30	30
U238_INELASTIC	342	60	649
Pu238_CAPTURE	6	10	20
Pu238_FISSION	-82	20	10
Pu239_CAPTURE	-96	20	190
Pu239_FISSION	-143	120	699
Pu239_NU	-49	40	100
Pu239_FISSION_SPECTRUM	-37	50	210
Pu240_CAPTURE	-100	40	300
Pu240_FISSION	-323	170	669
Pu240_NU	-2	10	20
Pu240_FISSION_SPECTRUM	-66	90	170
Pu241_CAPTURE	7	70	90
Pu241_FISSION	-8	60	70
Pu241_NU	-6	60	70
Pu241_FISSION_SPECTRUM	-29	130	150
Pu242_CAPTURE	99	30	50
Pu242_FISSION	-197	20	40
<b>Total actinides (pcm)</b>	<b>-435</b>	<b>490</b>	<b>1558</b>

## 5. CONCLUSION AND PERSPECTIVES

Integral Data Assimilation (IDA) in this work has proved to be efficient in identifying the sources of possible normalization problems in the differential measurements. This achievement is mainly due to the progresses in identifying the different sources of uncertainties whether they are from nuclear data evaluations themselves (through covariance) or from integral experiments (either for their set up or their modelling).

The trends on the JEFF3.1.1  $^{235}\text{U}$  capture cross section are quite consistent with recent differential measurements.

IDA results suggest also a 2.5% decrease for  $^{238}\text{U}$  capture from 3 keV to 60 keV, and a 4-5% decrease for  $^{238}\text{U}$  inelastic in the plateau region. For this energy range, uncertainties are respectively reduced from 3-4 to 1-2% and from 6-9% to 2-2.5% for  $^{238}\text{U}$  capture and  $^{238}\text{U}$  inelastic.

The increase trend on  $^{239}\text{Pu}$  capture cross section is of around 3% in the [2 keV-100 keV] energy range. For  $^{240}\text{Pu}$  capture cross section, the increase is of around 4% in the [3 keV-100 keV] energy range and goes in the same direction as the recent ENDF/B.VIII evaluation though at a much lower level.

As perspectives for future works, there is a need of more differential measurements and more reliable nuclear data covariance.

For instance, IDA has identified the lack of differential measurements, in particular for prompt fission neutron spectrum or  $^{238}\text{U}$  inelastic and parametric studies have shown that

nuclear data covariance data with better reliability are required (for PFNS and some capture cross sections).

There is also a request to have covariance associated to anisotropy of scattering and distribution of secondary energy neutrons in particular for light and structural isotopes. This will enable to improve the IDA by incorporating all sources of uncertainties. Also adding more integral experiments will help increasing the reliability of the IDA, specifically if these are defined to target a given spectrum or nuclide.

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