Trends on major actinides from an integral data assimilation

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Abstract. The objective of this work is to revisit integral data assimilation for a better prediction of the characteristics of SFR cores.
ICSBEP, IRPhE and MASURCA critical masses, PROFIL irradiation experiments and the FCA-IX experimental programme (critical masses and spectral indices) with well-mastered experimental technique have been used. As calculations are performed without modelling errors (with as-built geometries) and without approximations with the TRIPOLI4 MC code, highly reliable C/E are achieved.
Assimilation results suggest a 2.5% decrease for ^{238}U capture from 3 keV to 60 keV, and a 4-5% decrease for ^{238}U inelastic in the plateau region. For this energy range, uncertainties are respectively reduced to 1-2% and to 2-2.5% for ^{238}U capture and ^{238}U inelastic respectively.
The increase trends on ^{239}Pu capture cross section of around 3% in the [2 keV-100 keV] energy range come from a low PROFIL ^{240}Pu/^{239}Pu ratio C/E. For ^{240}Pu capture cross section, the increase trend of around 4% in the [3 keV-100 keV] energy range goes in the same direction as the recent ENDF/B.VIII evaluation though at a much lower level.
The nuclear data uncertainty associated to SFR ASTRID critical mass is reduced to 470 pcm.

1 Introduction

Five out 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. Hence, CEA with its industrial partners started the design of ASTRID (Advanced Sodium Technological Reactor for Industrial Demonstration), a 600 MWe sodium-cooled fast reactor concept [1,2]. 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 ^{235}U.

Nuclear data uncertainties on major actinides as they stand in current nuclear data libraries still do not meet Generation IV core requirements [3]. 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 that is far too large. Major actinides are contributing significantly to this uncertainty.

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

2 Overall Approach

2.1. Introduction

Integral data assimilation 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.

2.2 Strategy used

To avoid deficiencies identified in previous integral data assimilation work, a more reliable nuclear data library was used JEFF3.1.1 for all isotopes [4] except for ^{23}Na for which JEFF3.2 was used [5]. The associated COMAC V1 covariances 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 [6].
Parametric studies have been done so as not be too dependent to the nuclear data covariance data that are still perfectible or even missing (COMAC V1 used).
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.
Attention is being given to ways that minimize compensating errors.

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Compensating errors between ^{238}U capture and ^{239}Pu fission (such as in CARNAVAL IV [7]) has been eliminated by using first only U-fueled experiments then adding Pu-fueled experiments.
The IDA has been using Bayesian inference [8] with a minimization of a cost function (1) in which $M_{\boldsymbol{\sigma}}$ is is the covariance matrix associated to nuclear data and $M_{\boldsymbol{E}}$ the one associated to integral experiments.

$$\chi^2_{GLS} = (\boldsymbol{\sigma} - \boldsymbol{\sigma}_{a\,priori})^T M_{\boldsymbol{\sigma}}^{-1} (\boldsymbol{\sigma} - \boldsymbol{\sigma}_{a\,priori}) + \big(\boldsymbol{E} - \boldsymbol{C}(\boldsymbol{\sigma})\big)^T M_{\boldsymbol{E}}^{-1} \big(\boldsymbol{E} - \boldsymbol{C}(\boldsymbol{\sigma})\big) \quad (1)$$

Solutions for nuclear data are given by equation (2) where S is the sensitivity matrix calculated with the ERANOS code system [9] while associated uncertainties are given by equation (3):

$$\boldsymbol{\sigma}' - \boldsymbol{\sigma}_{a\,priori} = M_{\boldsymbol{\sigma}}\,.\,S^T (M_{\boldsymbol{E}} + S.M_{\boldsymbol{\sigma}}\,.\,S^T)^{-1}.\ \Big(\boldsymbol{E} - \boldsymbol{C}\big(\boldsymbol{\sigma}_{a\,priori}\big)\Big) \quad (2)$$

$$M'_{\boldsymbol{\sigma}} = M_{\boldsymbol{\sigma}} - M_{\boldsymbol{\sigma}}\,.\,S^T (M_{\boldsymbol{E}} + S.M_{\boldsymbol{\sigma}}\,.\,S^T)^{-1}\, S.M_{\boldsymbol{\sigma}} \quad (3)$$

3 Integral Data Assimilation Studies

3.1. Uranium configurations

The Uranium configurations chosen for the exercise are MASURCA R2, MASUSCA 1B, FCA IX-1 to 7, GODIVA and FLATTOP ^{235}U. 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. Critical masses C/Es calculated for different configurations display great dispersion in results. Nuclear Data uncertainties (of the order of 1000 to 2000 pcm) are far beyond experimental uncertainties (Figure 1).

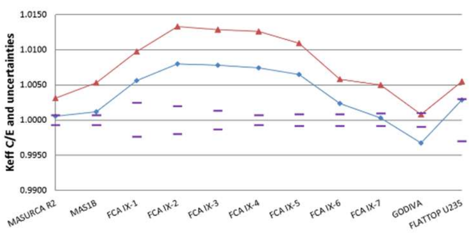

Figure 1: Critical mass C/E and uncertainties for Uranium configurations (JEFF libraries)

Use of integral experiments to identify which reaction and isotopes are responsible for this dispersion (assimilation using Bayesian Inference with CONRAD).

3.2 Plutonium configurations

The Plutonium configurations chosen are MASURCA 1A', MASURCA ZONA2, ZPPR10A, ZPR6/7, ZPR6/7 high ^{240}Pu, BFS 82-2, MASURCA PRE-RACINE 2A & 2B, SNEAK 7A & 7B , JEZEBEL ^{239}Pu & ^{240}Pu, FLATTOP ^{239}Pu. Also, FCA-IX fission chambers C/Es of ^{237}Np, ^{241}Am, ^{243}Am, ^{238}Pu, ^{242}Pu, ^{244}Cm have been used as well as PHENIX PROFIL irradiated samples C/E on ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{237}Np. Critical masses C/Es calculated for different configurations display some dispersion in results. We used as a starting point JEFF3.1.1 + ^{23}Na JEFF3.2. Nuclear data uncertainties (of the order of 1000 to 2000 pcm) are far beyond experimental uncertainties (Figure 2), which justify the use of IDA method.

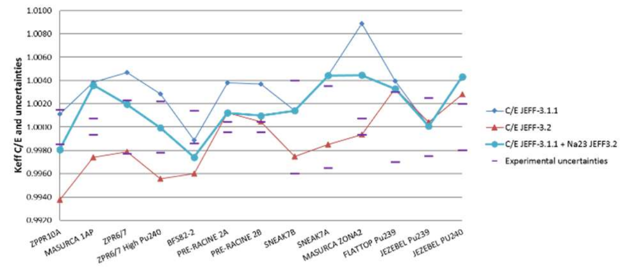

Figure 2: Critical mass C/E and uncertainties for Plutonium configurations (JEFF libraries)

3.3 Trends for ^{235}U Capture

As seen on Figure 3, the trends were a -27 to -33 % decrease at 1-2 keV (end of RRR) which is in agreement with Danon measurements (2017)[10] and a +8 to +10% increase in the 10-100keV region (URR) which is in agreement with Jandel measurements (2012) [11]. Uncertainties are significantly reduced.

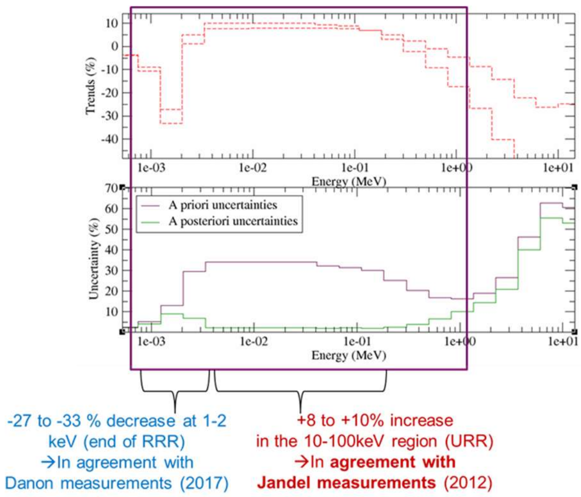

Figure 3: Trends for ^{235}U capture from JEFF3.1.1 evaluation

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

3.4 Trends for ^{238}U Inelastic and Capture

For ^{238}U inelastic and capture cross sections, a comparison of ^{235}U-^{238}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}U capture from 3 keV to 60 keV, and a 4-5% decrease for ^{238}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}U capture and ^{238}U inelastic.

Notably, the simultaneous use of FLATTOP-^{235}U/GODIVA and FLATTOP-^{239}Pu/JEZEBEL enables us to discriminate contributions from ^{238}U and fissile isotopes. GODIVA is a bare ^{235}U sphere while FLATTOP-^{235}U is surrounded by a $U_{depleted}$ blanket. The same occurs for JEZEBEL and FLATTOP-Pu (Figure 4).

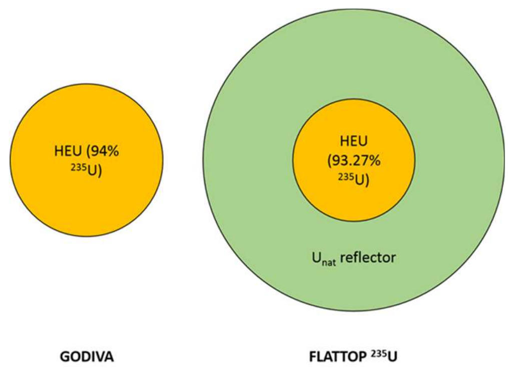

Figure 4: GODIVA bare core and U_{nat} reflected FLATTOP ^{235}U core

The simultaneous use of GODIVA and FLATTOP-^{235}U and JEZEBEL ^{239}Pu and FLATTOP-^{239}Pu is relevant to reassess ^{238}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.

In the plateau region (~1 MeV to 6 MeV), a 5-6% decrease is suggested for ^{238}U inelastic (Figure 5).

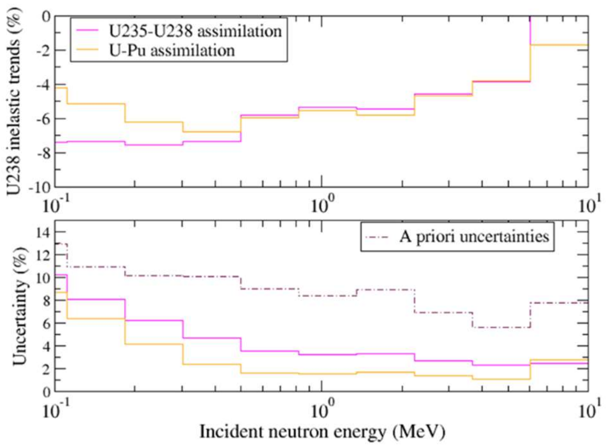

Figure 5: ^{238}U inelastic trends for different assimilations

U-Pu assimilation results are compared to ^{235}U & ^{238}U assimilation results. In the plateau region (~1 MeV to 6 MeV), a 5-6% decrease is suggested. Results from the two assimilations are consistent, which means compensating errors with Plutonium cross sections are negligible.

This decrease trend is in agreement with recent evaluations (see suggested value compared to CIELO and JEFF-3.3 ones on Figure 6; Posterior uncertainties for assimilation results are in dotted line).

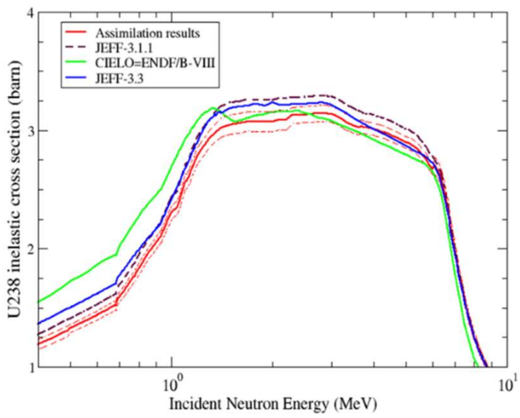

Figure 6: Assimilation Results compared to major evaluations for ^{238}U inelastic

Because Prompt Fission Neutron Spectrum (PFNS) lacks some differential measurements [12], variances are large. Since integral experiments available cannot distinguish between ν and χ (PFNS) the possibility of having compensating errors is high. A test on whether PFNS needs to fitted or not (such as in ERALIB1 [13]). It appears that PFNS is a major source of uncertainty and requires to be fitted. However, results depend very much in the associated covariance data. These should reflect the way the PFNS has been evaluated. In this work, the PFNS covariance is assumed to have a Madland shape, which is challenged. The finding trends are going in the opposite direction to most recent evaluations which are not based on this Madland shape. However, this does not affect the magnitude of trends on ^{238}U capture cross sections (Figure 7) when PFNS is fitted but differ on whether ^{235}U and ^{238}U PFNS is fitted or not through assimilation.

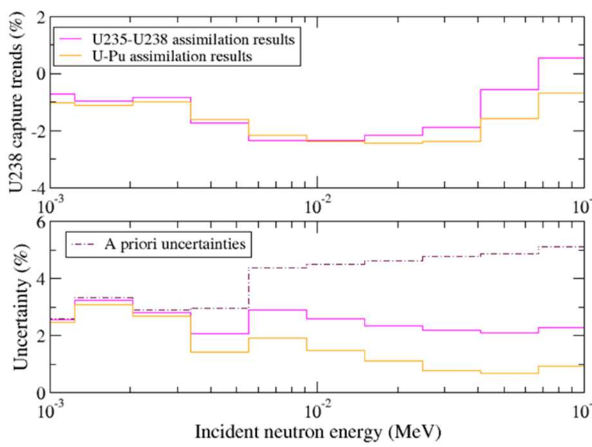

Figure 7: ^{238}U capture trends for different assimilations

Assimilation results for ^{238}U capture are compared with differential measurements [14] and with the "a priori" JEFF-3.1.1, CIELO and JEFF-3.3 evaluation (Figure 8). Posterior uncertainties for assimilation results are in dotted line.

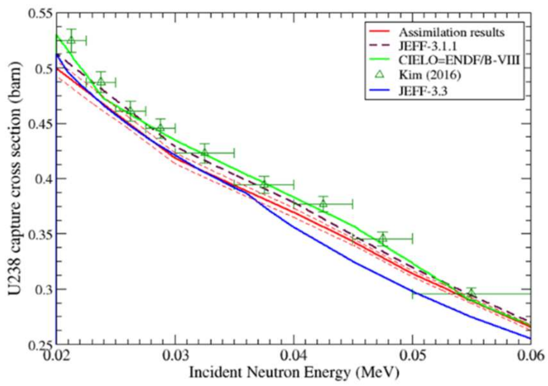

Figure 8: Assimilation Results compared to major evaluations for ^{238}U capture

3.5 Trends for Pu isotopes

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

Table 2. Trends on Pu isotope cross sections

Cross section	Trends (%)	Energy range	Experiments	Comments
^{239}Pu fission	<1% (+/- 1.7% or less)	0.1 keV- 500 keV	Pu-fueled critical masses	Trends included in posterior unc.
^{239}Pu capture	from -1.4 to -4.8 % (+/- 2-5%)	0.04 keV - 300keV	PROFIL and critical masses	
^{240}Pu fission	**from - 7.5 to -9.6% (+/-8-11%)**	**0.75 keV-100 keV**	**Mostly JEZEBEL- ^{240}Pu**	**Risk of compensating errors with multiplicity and PFNS. Trends are close to posterior uncertainties.**
	from -3.5 to -4.6% (+/- 4-7%)	**1 keV-3.7 MeV**		
^{240}Pu capture	from 2.5 to -4.2 % (+/- 2-4.5%)	0.7 keV - 100 keV	PROFIL	
^{242}Pu fission	**- 10% (+/- 3.5%)**	**500 keV- 4 MeV**	**FCA-IX IS**	**Underestimated prior uncertainties (~2% in the plateau region)**
^{242}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		

These trends will be detailed in the following paragraphs.

3.5.1 ^{239}Pu fission

Results on ^{239}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% (Figure 9). There is no differential measurement able to reach that type of uncertainty unless some breakthrough in experimental techniques. This means that IDA process will be needed in a way or another.

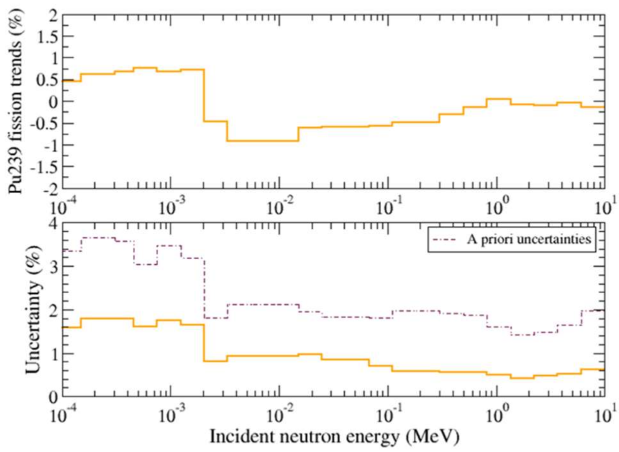

Figure 9: ^{239}Pu fission trends and uncertainty reduction

3.5.2 ^{239}Pu capture

The increase trends on ^{239}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.
Assimilation results do not go in the same direction as the JEFF-3.3 evaluation but are more in agreement with Mosby [15] (Figure 10).

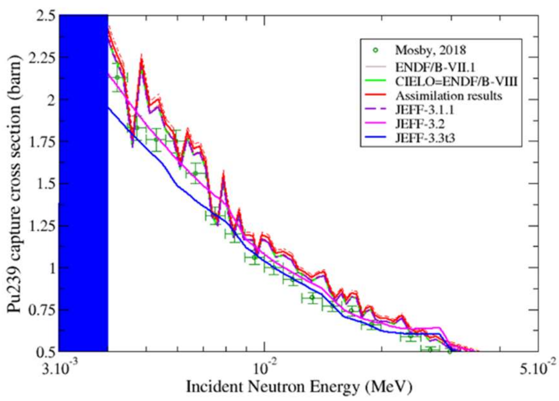

Figure 10: ^{239}Pu capture assimilation results compared to different evaluations

3.5.2 ^{240}Pu fission

For ^{240}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 (Figure 11).

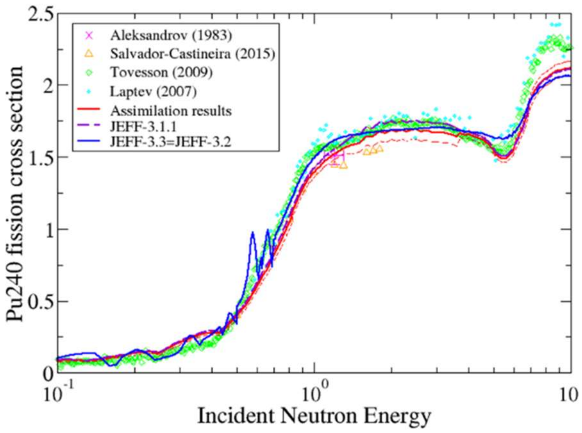

Figure 11: ^{240}Pu fission assimilation results compared to different evaluations

Assimilation results are not consistent with measurements from Tovesson [16] and Laptev [17]. Measurements from Salvador-Castiñeira [18] push towards an even higher decrease of this cross section above the threshold.

3.5.3 ^{242}Pu fission

C/E values for FCA-IX spectral index ^{242}Pu/^{239}Pu ratio are highly overestimated (C/E~1.12). Integral data are in better agreement with Tovesson measurements [16] and Salvador-Castiñeira [19] ones than Weigmann [20] and Matei [21] ones (Figure 12).

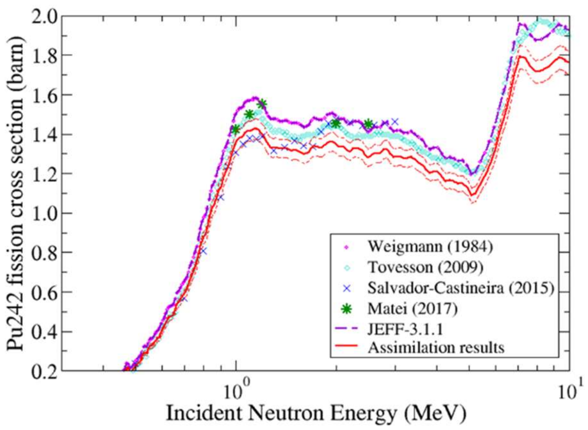

Figure 12: ^{240}Pu fission assimilation results compared to different evaluations

3.5.4 ^{242}Pu capture

Trends are driven by a single integral information: PROFIL (C/E~1.14 with JEFF-3.1.1) and follow prior uncertainties. IDA trends hence have an unreliable energy breakdown with prior uncertainty greatly underestimated below 4keV (beginning of the URR) (Figure 13).

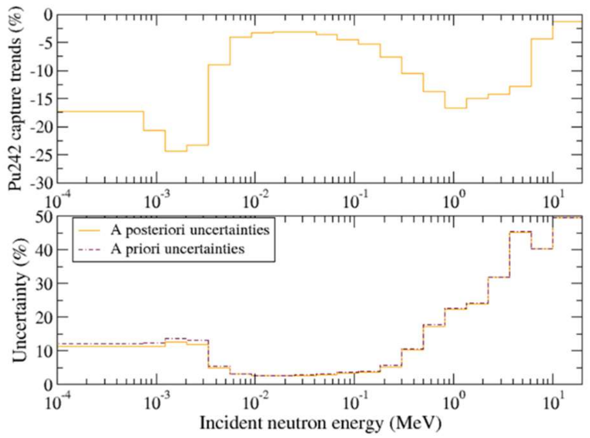

Figure 13: ^{242}Pu capture trends and uncertainty reduction

The trend goes in the same direction than Buckner [22], though as a much lower level (Figure 14).

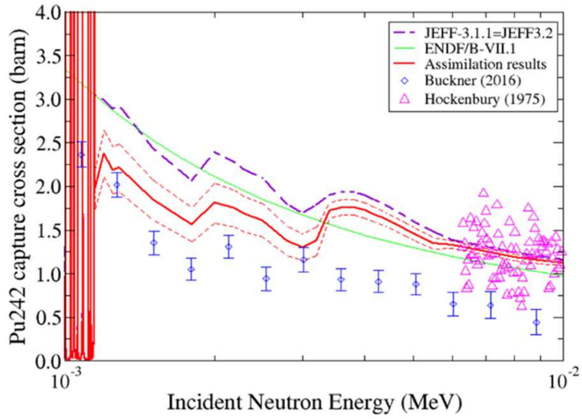

Figure 14: ^{242}Pu capture assimilation results compared to different evaluations

4. Impact on the ASTRID CFV Core

There is a significant impact of ^{238}U capture and inelastic on the critical mass of the ASTRID core but also ^{239}Pu, ^{240}Pu and ^{242}Pu fission cross-sections (Table 3). With a prior value for the critical mass of 1.02688 (1.02908 with JEFF-3.^{1}.1 for all isotopes except ^{23}Na for which JEFF3.2 is used: Impact of using ^{23}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}Pu and ^{240}Pu fission cross-sections.

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}U capture cross section are quite consistent with recent differential measurements.

IDA results suggest also a 2.5% decrease for ^{238}U capture from 3 keV to 60 keV, and a 4-5% decrease for ^{238}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}U capture and ^{238}U inelastic.

The increase trend on ^{239}Pu capture cross section is of around 3% in the [2 keV-100 keV] energy range. For ^{240}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}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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