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An in-depth analysis of Minor Actinide Fission Chambers Measurements in the FCA IX Experimental Programme.

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Abstract. FCA is a zero power facility located at Tokai in Japan. The FCA-IX experimental programme exhibits 7 different cores with a variety of different neutron spectra. This has been achieved through the use of different moderators and different Uranium enrichments (93% for most of them except FCA-IX-7 where it is 20%). Fission Chambers of seven different nuclides: ²³⁷Np, ²³⁸Pu, ²³⁹Pu, ²⁴²Pu, ²⁴¹Am, ²⁴³Am and ²⁴⁴Cm were used to measure ratios of fission reaction rate. Mass quantitative determinations of the electrodeposited nuclides were carried out through spectral analysis using a surface barrier silicon detector and a pulse height analyzer. The quantitative determination errors for electrodeposited nuclides were 3% for ²⁴⁴Cm and 1.5% for the other.

Uncertainties are calculated with COMAC V1 covariances associated for the JEFF32 library, with their own covariances for ENDF-BVII.1 and for JENDL4.0 libraries. The critical mass uncertainties are dominated by ²³⁵U capture contribution for JEFF3.1.1, JEFF3.2 and ENDF-BVII.1. However, uncertainties with JENDL4.0 are much lower due to a much reduced ²³⁵U capture contribution.

Calculated Uncertainty is mainly due to Nuclear Data Uncertainties and can be split into direct effect (Minor Actinide cross section) and indirect effect (through flux shape). The indirect effect is an important contribution to the total calculated effect (except for ²³⁸Pu): This is mainly due to ²³⁵U capture and that limits the possible feedback on the cross sections of minor actinides that have been measured through fission chambers. However, the fission rates of ²³⁷Np, of ²⁴³Am and ²⁴⁴Cm have a direct effect on significantly higher than those of the ²³⁵U and ²³⁹Pu in contrast to the ²³⁸Pu, ²⁴²Pu and the ²⁴¹Am. The use of criticality measurements of these 7 cores can help reduce the magnitude of the indirect effect of ²³⁵U and therefore allow closer access to effective fission sections of the measured actinide fission chambers. All data sets exhibit the same trends with ²⁴²Pu and ²⁴⁴Cm fissions too large by around 10% while ²⁴¹Am seems too small by 7%.

1 Introduction

FCA is a zero power facility located at Tokai in Japan. The FCA-IX experimental programme exhibits 7 different cores with a variety of different neutron spectra [1]. Fission Chambers of seven different nuclides: ²³⁷Np, ²³⁸Pu, ²³⁹Pu, ²⁴²Pu, ²⁴¹Am, ²⁴³Am and ²⁴⁴Cm were used to measure ratios of fission reaction rate [2]. This is a good opportunity for identify the correctness of Minor Actinides Fission cross sections.

2 FCA IX plant layout

FCA is a zero power facility located at Tokai in Japan. The FCA-IX experimental programme exhibits 7 different cores with a variety of different neutron spectra. This has been achieved through the use of different moderators and different Uranium enrichments (93% for most of them except FCA-IX-7 where it is 20%). In the following Figure 1, we can see the

normalized fission spectra as calculated with the TRIPOLI4 Monte Carlo code [3].

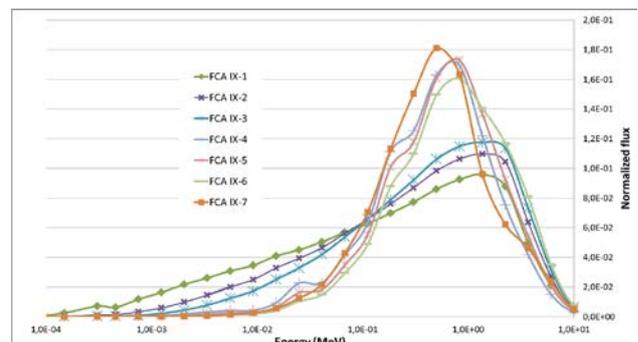


Figure 1. Normalised FCA-IX spectra

Given the relative similarities between most of the spectra, only FCA IX-1, FCA IX-6 and FCA IX-7 results have been reported in the following.

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3 Criticality

Experimental values for criticality are given by JAEA [1]. Results of the analysis using TRIPOLI4 (T4) [3] and JEFF3.2 [7] are given in the following Table 1:

Table 1: E-C values for FCA IX critical masses using TRIPOLI4 & JEFF3.2

	FCA IX-1	FCA IX-6	FCA IX-7
Experiment	1.00340	1.00250	1.00220
□ exp (pcm)	240	60	70
T4 JEFF32 Heterogeneous	1.01438	1.01081	1.01122
□ T4 (pcm)	7	7	6
E-C (pcm)	-1079	-820	-890
□ (pcm)	250	92	99

One can see that there is a significant bias between JEFF3.2 calculations and experiments. E-C as well as uncertainties on Keff have been calculated for different libraries. These use COMAC V1 covariances [6] for the JEFF32 library [7] and their own covariances for ENDF-BVII.1 [8] and for JENDL4.0 [9] (Table 2).

Table 2: E-C values for FCA IX critical masses using different libraries and associated covariances

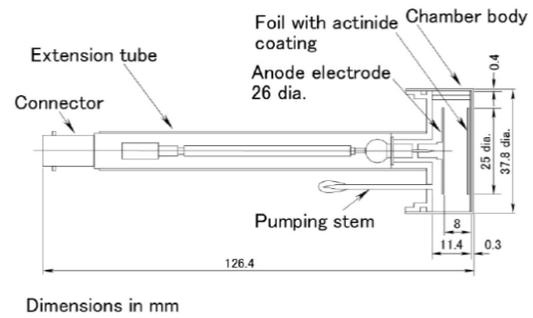
E-C (pcm)	FCA IX-1	FCA IX-6	FCA IX-7
JENDL4.0	-1079	-307	-290
ND Uncertainty	724	606	811
ENDF-BVII.1	-548	-706	-1022
ND Uncertainty	2670	1789	1822
JEFF3.2	-1079	-820	-890
ND Uncertainty	2639	2143	2334

The critical mass uncertainties are dominated by ^{235}U capture contribution for both JEFF3.2 and ENDF-BVII.1. However, uncertainties with JENDL4.0 are, in general, much lower due to a much reduced ^{235}U capture contribution. Since C-E values are quite low with both JEFF3.2 and ENDF-BVII.1, a ^{235}U capture seems the major suspect in the too high calculated reactivity level. Current trends on criticalities advocate for an increase of ^{235}U capture for JEFF3.2 and ENDF B VII.1. An assimilation technique will help identify which energy region is concerned.

4 Reaction Rates

Fission Chambers of seven different nuclides: ^{237}Np , ^{238}Pu , ^{239}Pu , ^{242}Pu , ^{241}Am , ^{243}Am and ^{244}Cm were used to measure ratios of fission reaction rate. Mass quantitative determinations of the electrodeposited nuclides were carried out through spectral analysis using a surface barrier silicon detector and a pulse height analyzer [2]. The quantitative determination errors for electrodeposited nuclides were 3% for ^{244}Cm and 1.5% for the other. Figure 2 shows how these fission chambers are designed.

Figure 2: FCA-IX fission chambers



The detailed analyses of the Minor Actinide fission chambers measurements performed during the FCA-IX programme have been done using ERANOS2.3 [4]. Associated to the C/E values are the experimental uncertainty and the calculated one. This last one is dominated by nuclear data uncertainties and has been obtained with the sandwich formula using covariance matrices associated to JEFF3.2 libraries (COMAC V1) [6], and their own ones for ENDF-BVII.1 and JENDL4.0 libraries. Sensitivities were obtained with the GPT method [5] in ERANOS [4] (Table 3).

Table 3: C/E values for FCA IX fission rate ratios using JEFF3.2

JEFF3.2	FCA IX-1	FCA IX-6	FCA IX-7
IS_F37/F49	0.976	0.956	0.953
Exp Unc	0.022	0.019	0.024
Calc Unc	0.041	0.04	0.052
Total Unc	0.046	0.044	0.057
IS_F48/F49	1.144	0.978	0.987
Exp Unc	0.035	0.026	0.026
Calc Unc	0.018	0.02	0.023
Total Unc	0.039	0.033	0.035
IS_F42/F49	1.117	1.089	1.081
Exp Unc	0.022	0.019	0.024
Calc Unc	0.034	0.03	0.047
Total Unc	0.041	0.035	0.053
IS_F51/F49	0.954	0.927	0.909
Exp Unc	0.022	0.024	0.024
Calc Unc	0.031	0.027	0.05
Total Unc	0.038	0.036	0.055
IS_F53/F49	0.948	0.928	0.929
Exp Unc	0.022	0.024	0.024
Calc Unc	0.091	0.091	0.099
Total Unc	0.093	0.094	0.102
IS_F64/F49	1.092	1.13	1.091
Exp Unc	0.036	0.038	0.037
Calc Unc	0.052	0.053	0.067
Total Unc	0.063	0.065	0.077

This way of doing helps tracking the origin of nuclear data uncertainty in the calculation of measured fission rate ratios.

The fact that these cores are all enriched Uranium introduce an indirect effect due to the capture cross section of ^{235}U and that limits the possible feedback on the cross sections of minor actinides that have been measured through fission chambers. However, the fission rates of ^{237}Np , of ^{243}Am and ^{244}Cm have a direct effect significantly higher than those of the ^{235}U and ^{239}Pu in contrast to the ^{238}Pu , ^{242}Pu and the ^{241}Am for which information is diluted.

One can illustrate this with use of different covariance sets.

The use of COMAC V1 covariances for FCA IX $^{237}\text{Np}/^{239}\text{Pu}$ reaction rate ratio using the JEFF3.2 library gives the following results (Table 4).

Table 4: Uncertainty breakdown for FCA IX-1 $^{237}\text{Np}/^{239}\text{Pu}$ reaction rate ratio using JEFF3.2

COMAC V1	CAPTURE	ELASTIC	INELASTIC	NU	FISSION	N,XN	TOTAL
Fe54	0.006	0.016	0.008	-	-	0.000	0.019
Fe56	0.040	0.071	0.128	-	-	0.000	0.152
Cr52	0.003	0.003	0.010	-	-	0.000	0.011
Ni58	0.012	0.012	0.007	-	-	0.000	0.018
Ni60	0.005	0.005	0.003	-	-	0.000	0.008
Co	0.007	0.425	0.243	-	-	-	0.489
Np237	0.000	0.000	0.000	0.000	2.911	0.000	2.911
Pu239	0.000	0.000	0.000	0.000	1.588	0.000	1.588
U235	2.256	0.192	0.374	0.007	0.276	0.002	2.312
U238	0.282	0.074	0.117	0.015	0.126	0.000	0.339
TOTAL	2.274	0.478	0.479	0.016	3.330	0.002	4.089

^{237}Np and ^{239}Pu are direct contributions to the total uncertainty while ^{235}U is an indirect effect to the total uncertainty. The situation is quite different for the same reaction rate ratio $^{237}\text{Np}/^{239}\text{Pu}$ when using JENDL4.0 library (Table 5).

Table 5: Uncertainty breakdown for FCA IX-1 $^{237}\text{Np}/^{239}\text{Pu}$ reaction rate using JENDL4.0

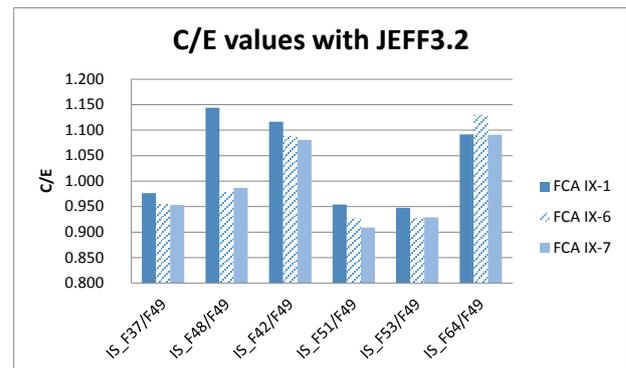
JENDL4.0	CAPTURE	ELASTIC	INELASTIC	NU	FISSION	N,XN	TOTAL
Fe56	0.047	0.085	0.490	-	-	0.000	0.500
Cr52	0.001	0.012	0.029	-	-	0.000	0.031
Ni58	0.005	0.008	0.016	-	-	0.000	0.019
Ni60	0.001	0.002	0.007	-	-	0.000	0.007
Np237	0.000	0.000	0.000	0.000	1.554	0.000	1.554
Pu239	0.000	0.000	0.000	0.000	0.538	0.000	0.538
U235	0.395	0.054	0.626	0.007	0.642	0.005	0.981
U238	0.080	0.102	0.066	0.016	0.010	0.004	0.147
TOTAL	0.406	0.146	0.798	0.017	1.766	0.006	1.985

One can notice that in this case, ^{235}U contribution to the total uncertainty is much reduced and that one can now derive clear trends on ^{237}Np fission cross-sections.

Given this, one can derive trends on cross sections being measured.

The C/E values for FCA IX fission rate ratios using JEFF3.2 are presented in the following Figure 3.

Figure 3: C/E values on FCA-IX fission rate ratios using JEFF3.2



Similarly, the C/E values for FCA IX fission rate ratios using JENDL4.0 and ENDF-BVII.1 are presented respectively in Figures 4 and 5.

Figure 4: C/E values on FCA-IX fission rate ratios using JENDL4.0

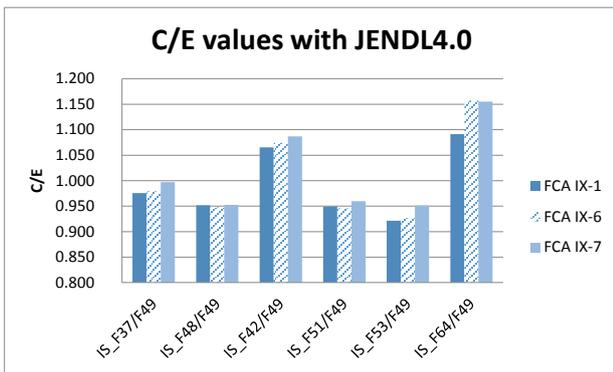
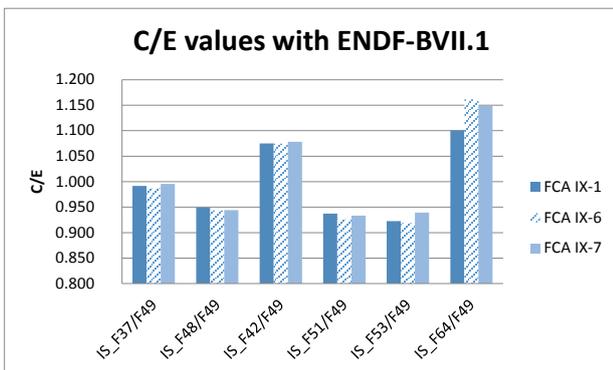


Figure 5: C/E values on FCA-IX fission rate ratios using ENDF-BVII.1



All data sets exhibit the same trends with ^{242}Pu and ^{244}Cm fission cross sections too large by around 10% while ^{241}Am fission cross section seems too small by 7%. Fission cross sections of ^{237}Np and ^{243}Am seems correct, while statements on ^{238}Pu fission cross section has to wait an integral data assimilation work before drawing any conclusion.

5 Conclusions

7 FCA-IX cores offer a variety of neutron spectrum thanks to different moderators and enrichments. FCA-IX-1 to FCA-IX-6 cores exhibit a 93% ^{235}U enrichment while FCA-IX-7 has 20% ^{235}U enrichment.

The fact that these cores are all Uranium enriched cores introduces an indirect effect due to ^{235}U capture cross section which somehow limits feedbacks on Minor Actinide cross sections being measured through fission chambers at least for ^{238}Pu , ^{242}Pu and ^{241}Am . This conclusion comes from ^{235}U covariances as they are associated to JEFF3.2 (named COMAC V1) and ENDFB VII.1 files. The situation is quite different for JENDL4.0 for which ^{235}U covariances are smaller by a factor 3. The use of experimental keff for these 7 cores can help reducing the magnitude of the ^{235}U indirect effect of

JEFF3.2 and ENDF-BVII.1 and hence will allow having a refine access to MA fission cross sections though measured fission chambers.

The JEFF3.2 dominant uncertainty contributions are for each individual fission ratios from the nuclide itself (direct effect), ^{239}Pu (direct effect as used at the denominator) and ^{235}U (indirect effect mostly capture but also inelastic and fission). For JEFF3.2, ^{237}Np , ^{243}Am and ^{244}Cm have a direct effect significantly larger than the other 2 which means that the measurement can be used to validate their fission cross sections.

All data sets exhibit the same trends with ^{242}Pu and ^{244}Cm fissions too large by around 10% while ^{241}Am seems too small by 7%. Although the 7 FCA-IX cores have different neutron spectra, this conclusion stands for all cores.

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