Homogeneous Minor Actinide Transmutation in SFR: Neutronic Uncertainties Propagation with Depletion

L. Buiron and D. Plisson-Rieunier
CEA, Cadarache, France

Abstract

In the frame of next generation fast reactor design, the minimisation of nuclear waste production is one of the key objectives for current R&D. Among the possibilities studied at CEA, minor actinides multirecycling is the most promising industrial way achievable in the near-term. Two main management options are considered:

- *Multirecycling in a homogeneous way (minor actinides diluted in the driver fuel). If this solution can help achieving high transmutation rates, the negative impact of minor actinides on safety coefficients allows only a small fraction of the total heavy mass to be loaded in the core (~ few %).*
- *Multirecycling in hetereogeneous way by means of Minor Actinide Bearing Blanket (MABB) located at the core periphery. This solution offers more flexibility than the previous one, allowing a total minor actinides decoupled management from the core fuel. As the impact on feedback coefficent is small larger initial minor actinide mass can be loaded in this configuration.*

Starting from a breakeven Sodium Fast Reactor designed jointly by CEA, Areva and EdF teams, the so called SFR V2B, transmutation performances have been studied in frame on the French fleet for both options and various specific isotopic management (all minor actinides, americium only, etc.). Using these results, a sensitivity study has been performed to assess neutronic uncertainties (i.e coming from cross section) on mass balance on the most attractive configurations. This work in based on a new implementation of sensitivity on concentration with depletion in the ERANOS code package. Uncertainties on isotopes masses at the end of irradiation using various variance-covariance is discussed.

Introduction

The next nuclear reactors generation have to demonstrate their capacity to reduce the volume of their own radwaste but also those coming from the existing reactors (PWR). The minor actinides (MA) having the advantage of being fissile in fast neutron spectrum, most of GEN-IV reactors are thus the privileged tools to implement the transmutation of minor actinides. Among the many possible strategies, two main minor actinides management are being studied at CEA:

- The homogeneous mode, in which the actinides are diluted in fuel,
- The heterogeneous mode, in which the actinides are placed in dedicated assemblies in core or periphery (radial blankets) allowing, to decouple their management from the standard fuel.

Within this framework, it is necessary to evaluate the transmutation capacity of these two principal modes for current Sodium Fast Reactors (SFR) studied at CEA in the frame of the french fleet. The corresponding Minor Actinides transmutation rate level and corresponding consumption (or mass balance) are of major importance regarding industrial issues such as heavy nuclei inventories, fabrication and reprocessing material flows or evaluation of the size of the nuclear waste ultimate repository.

One of the main points of theses evaluations is the mass balance with irradiation which gives basic parameters for front end and back end of the whole fleet in the frame of closed fuel (multirecycling) cycle.

Beside the estimation of the level of "absolute" values, associated uncertainties have also to be evaluated for the whole set of relevant data. These estimations have to be performed for different core state such as end of cycle core for feedback coefficient or end of life for mass balance. This means that uncertainties have to be obtained not only at a fixed time but also have to be propagated all through irradiation. To do so, coupling both Boltzman and Bateman equations at sensitivities level is needed. This coupling is in progress in ERANOS code system and a first estimation of sensitivities on Bateman equation can be performed for mass balance purpose.

After a brief theoretical description of Boltzman/Bateman coupling in progress, the study presented in this paper focuses on transmutation performances and associated sensitivities with depletion for the SFR V2B core design [1] for which several fuel cycle scenarios involving minor actinides transmutation are available. Some uncertainties are presented using various covariance matrices available at CEA.

Bateman/Boltzman coupling strategy

Neutronic uncertainties are usually computed using the Perturbation Theory [2,3] approach allowing estimation of sensitivity factors for any linear and bilinear fonctionnal of the flux such as multiplication factor or reaction rates (derived from Boltzman equation only). These estimations usually used "static" hypothesis: the state of core (irradiated or not) is always considered at a specific time.

As macroscopic cross sections are a function of the irradiation time, the coupling of Boltzman and Bateman equations is required to have global uncertainties propagation with irradiation.

The inherent change on N_i, the concentration of isotope i, under irradiation by flux level ϕ is driven by the Bateman equation:

$$\frac{dN_i}{dt} = -\phi\sigma_i^a N_i + \sum_{j\neq i}\phi\sigma_j^c N_j - \lambda_i N_i + \sum_{j\neq i}\lambda_{j\to i} N_j \tag{1}$$

with σ_a and σ_c the absorption and capture cross sections respectively, and λ_i the decay constant of isotope i. Equation (1) can also be written as follows:

$$\frac{d\vec{N}}{dt} = (R\phi + D)\vec{N} = M\vec{N} \tag{2}$$

where N stands for isotopic vector and M the Bateman operator is splitted into cross sections operator *(R)* and decay constant operator *(D)*. The effective coupling is performed by building a functional [4] linking Boltzman equation, Bateman equation, direct and adjoint flux renormalisation by mean of Lagrange multipliers (N^+,Γ,Γ^+,P^+). The minimisation of this functional with respect to Lagrange multipliers leads to the most general form of the sensitivity S of an integral value T relative to parameter p:

$$S(T,p) = \frac{1}{T}\begin{pmatrix} \left\langle p\frac{\partial T}{\partial p}\right\rangle + \int\left\langle \vec{N}^+\left(\phi p\frac{\partial R}{\partial p}\right)\vec{N}\right\rangle dt + \\ \left\langle \Gamma^+\frac{\partial H}{\partial p}p\phi\right\rangle + \left\langle \Gamma p\frac{\partial H^+}{\partial p}\phi^+\right\rangle + \\ P^+\left\langle p\frac{\partial H}{\partial p}\vec{N}\phi\right\rangle \end{pmatrix} \tag{3}$$

where the brackets stand for integration on space, and energy and H is the Boltzman operator ($H=A-F/k$).

The first term in equation (3) is the traditional Boltzman sensitivity and second one is the Bateman sensitivity. The third and fourth ones are relatives to direct and adjoint flux normalisation, while the last one corresponds to power renormalisation.

In the second term appears N^+ which is solution to the adjoint form of the equation (1):

$$\frac{d\vec{N}^+}{dt} = -M^t \vec{N}^+ \tag{4}$$

For the "direct" concentration, the adjoint concentration N^+ plays a similar role as the adjoint flux toward the direct flux. For a final isotope i at some specific irradiation time, it represents the "weight" of the n previous isotopes ("fathers") which lead to i by nuclear reactions presents in the operator M. This adjoint depletion occurs backward in time from a final state ($t=t_F$) to the origin of the direct irradiation ($t=0$). The final adjoint concentration associated to the integral value T is given by the following formula:

$$\vec{N}^+(t_F) = \frac{\partial T}{\partial \vec{N}} \tag{5}$$

If T is a concentration or a linear combination of isotopic concentration $T=<u,N>$ with u the vector of individual isotopic weight, the adjoint concentration is u itself.

New developments in the ERANOS code system has been performed [5] in order to solve Bateman adjoint equation and to compute second term of the general sensitivity expressed in equation (3). In addition, initialisation of adjoint concentration is available for T corresponding to a linear isotopic concentration combination and also for multiplication factors such as single k_{eff} or $\Delta\rho=\Delta k/k^2$ (using Equivalent General Perturbation Theory assumptions). The last three terms of equation (3) imply estimation of parameters Γ, Γ^+ and P^+ that are solutions of source calculations (analogue to importance calculation) involving complex combination of direct and adjoint concentrations. Although implementation of such source is still in progress, functionalities were not available for this study. The uncertainty level ε of value T is evaluated using the well-known "sandwich" formula:

$$\varepsilon(T) = \sqrt{S^t(T) V S(T)} \tag{6}$$

where V is the cross sections variance-covariance matrix.

SFR core description

The design of the reference SFR 3 600 MWth MOX core (named SFR V2B hereafter) has been widely described in the past [1]. The design of the fuel assembly is based on a concept with large pins (10.73 mm diameter) and a spacer wire of small diameter (1 mm). The nominal performances of the core reach a burnup of 100 GWd/t with a residence time of fuel of approximately 2000 EFPD and a null internal breeding gain (IBG), this last point being a design goal. As the tightening of the pin bundle in the fuel assembly requires a cladding material which does not swell significantly to reach the expected burnup, an advanced ferritic steel (ODS) is consider.

The reference core design is presented briefly in Table 1. The core configuration ensures self-breeding without the help of fertile blankets.

Table 1: Nominal performances of the reference core SFR V2B

SFR V2 B – main characteristics	
Power (MWth/MWe)	3600/1450
Power density (W/cm^3)	207
Number of fuel elements (inner/external)	453 (267/186)
Life time	5 × 410 = 2050 EFPD
Pu | 239PuEq content (%) E1 | E2 (%)	15.80 | 10.96 14.65 | 17.44
HN Pu+AM inventory (t)	12.5 (incl. 0.095 Am241)
SFR V2 B – main Performances	
LHR MAX (W/cm)	420
$\Delta\rho$ cycle (pcm) / (pcm/EFPD)	-450/-1.1
GBGnst (BOC/ EOC)	-0.001/-0.004
GBG_{eff}	0.000
BU_{moy} /BU_{max} (GWd/t)	99/139
Damage Max (dpa NRT Fer)	148

Calculation hypothesis

Minor actinides configurations

Two specific scenarios relative to the French fleet are retained as bases for the transmutation evaluations [6]:

- Scenario *F2A*: homogeneous multirecycling of all actinides in homogeneous mode on the basis of SFR V2B core. Here the minor actinides content is adjusted with respect to a mixt PWR/SFR fleet inventory
- Scenario *F1G*: heterogeneous multirecycling of all actinides in heterogeneous mode on the basis of SFR V2B core with minor actinide bearing blankets. Here the minor actinides $(MA)O_2$ content in the $UO_2^+(MA)O_2$ mixture loaded in the blankets is always set to 20%.

These scenarios take into account the transition between a fleet constituted of 100% PWR (beginning of the SFR deployment) and a 100% SFR fleet (equilibrium). Here, we focus on homogeneous mode.

Figure 1: Time evolution of the MA content in initial loading, "scenario F2A"

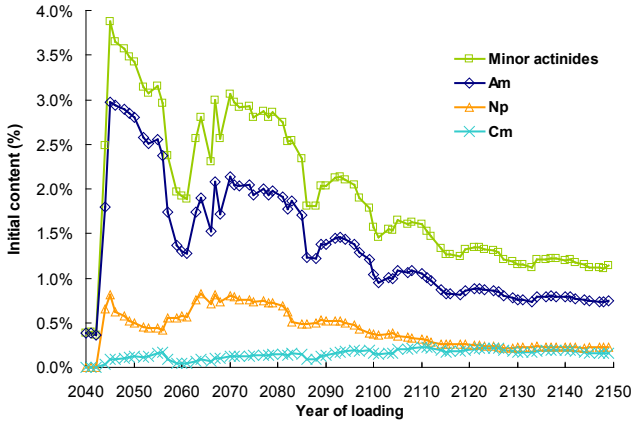

Figure 1 shows the time evolution of the minor actinide content necessary to ensure the transition between a PWR fleet and a SFR fleet in the frame of the F2A scenario. This content presents a maximum around 3.88% corresponding to the beginning of the transient of SFR deployment and does stabilise around 1.20%. The evolution of the composition shows that the contribution of curium increases regularly to be stabilised after a few decades. One can see that the MA content peak occurs at the very beginning of the SFR deployment. The content quickly increases from residual content (~0.4%) to nearly 4.0%. In this case it is interesting to investigate sensitivity and uncertainty levels relative mass balance.

To do so, a parametric study on the level of the MA content is one carried out from reference case (MOX) up to the maximum value of the scenario (4%). Minor actinides isotopic composition used here are presented in Table 3.

Table 3: Homogeneous mode minor actinides isotopic content

Element	Isotope	Mass content (%)
Uranium	U235	0.25
	U238	99.75
Plutonium	Pu238	3.57
	Pu239	47.39
	Pu240	29.66
	Pu241	8.23
	Pu242	10.37
	Am241	0.78

Depletion scheme

Calculations were performed using the ECCO/ERANOS code package using the JEFF3.1 library [7]. The transport option (Bistro S8) has been used for all calculations with a 33-group working library which has been generated from a 1968-group master library. For comparison, fine-group (1968 groups) ECCO cell calculations were also performed, and provide an accurate description of the reaction thresholds and resonances. The fine-group calculation has been performed for the 2-dimensional description of a sub-assembly. Because the broad 33-group library has been generated from this master fine group library, it has been found that differences on the core parameters are small.

For the core geometry model of the core, a cylindrical (RZ) model is adopted. Validity of the RZ model was assessed against the results of heterogeneous (hexagonal-Z) calculation using the TGV/VARIANT 3-dimensional nodal transport code.

Heavy nuclei depletion calculations are performed with an extended chain form Th230 up to Cf252 isotope for a total of 33 heavy nuclei. A simplified model is used for fission product description: a set of 15 lumped fission product for major isotopes (U isotopes, Pu isotopes, Am isotopes, Np237, Cm243, Cm244, Cm245) for which an effective capture cross section has been settled to be representative of real isotopic fission yield distribution.

Core depletion is performed using a simplified averaged irradiation scheme, so mass balance will be estimated between beginning of life (full core loading) and end of life (full core unloading). Using this hypothesis leads to quite good accuracy on core average mass balance at the end of irradiation since it preserves total core fluence.

Covariance data

Uncertainties calculation needs a set of variance-covariance covering the widest range of isotopes and associated cross-sections. As we only focus isotopic mass balance we have to deal with actinides isotopes and the main cross sections that appear in the Bateman equation: capture, fission and (n,xn).

At international level, several teams devoted to nuclear data work on cross section evaluation and associated variance-covariance data. In the frame of GENIV reactor design, CEA is involved in that process and does produce new measurements and/or evaluations on some energy range of interest for both LWR and Fast Reactors.

In the present work, two set of variance-covariance data were used:

- BOLNA [8] from the OECD NEA/WPEC Subgroup 26
- COMAC V0.1 [9] from the CEA work analysis.

The frame of COMAC data set is to provide accurate and up to date variance-covariance data for JEFF3.1 cross section library (and the new JEFF3.2 to come) for main isotopes (heavy nuclei, coolant and structures' material used in both LWR and FR). As this work is an iterative process, accuracy on data coming from measurements (analytic, differential and integral experiments) and models continuously improve.

Here, the goal is not to discriminate between the two sets but to illustrate their impact on relevant parameters such as mass balance, neutronic feedback coefficients, etc.

Mass balance and uncertainties for homogeneous mode

Mass balance

Table 5 displays the mass balance between begin and end of irradiation for minor actinides. Transmutation rate r and associated consumption c are estimated via the following simple formulas:

$$r = \frac{mass_{Unloaded} - mass_{Loaded}}{mass_{Loaded}}$$
$$c = \frac{mass_{Loaded}}{P_{core}^{th}\Delta t} \times r \qquad (7)$$

Positive values stand for isotopic creation while negative ones stand for isotopic disappearance.

For MOX case, the minor actinides content resumes to lonely Am241 coming from Pu241 decay between manufacturing and loading. In this case, the core exhibits net production of minor actinides for neptunium (Np), americium (Am) and curium (Cm).

For 1% MA content initially loaded the minor actinides mass balance at end of irradiation is quite close to zero. Although there is a MA equilibrium, the core shows a net production of curium while the Am and Np initial mass are reduced by 10% and 30% respectively.

From 1% to 4%, although the Np and Am transmutation rates show no linear increase, the corresponding mass consumption is linear.

The main isotopes that contribute to minor actinides final mass are:

- Np237 produced by (n,2n) reaction from U238
- Am241 produced by Pu241 decay

- Am243 coming from captures on Pu242 and Am241
- Cm244 coming mainly from capture on Am243

Table 5: Minor actinides mass balance and transmutation performance, homogeneous mode

	MOX	1%	2%	3%	4%
Initial Load (kg)					
Np237	0	154	309	463	618
Np239	0	0	0	0	0
Np	0	154	309	463	618
Am241	95	472	945	1417	1889
Am242m	0	1	2	3	4
Am243	0	91	182	273	364
Am	95	564	1129	1693	2258
Cm242	0	0	0	0	0
Cm243	0	0	1	1	1
Cm244	0	12	24	37	49
Cm245	0	4	8	12	16
Cm246	0	0	1	1	1
Cm	0	17	34	51	67
M.A	95	736	1472	2207	2943
Final unload (kg)					
Np237	30	98	171	247	325
Np239	10	11	10	10	10
Np	40	108	182	257	335
Am241	176	288	489	696	909
Am242m	10	20	36	52	68
Am243	180	196	241	287	334
Am	365	503	765	1035	1312
Cm242	10	17	29	40	52
Cm243	1	2	4	5	7
Cm244	69	86	119	152	184
Cm245	8	13	19	26	32
Cm	89	119	174	228	280
M.A	495	731	1121	1520	1927
Transmutation rate (%)					
Np		-29.8	-41.2	-44.5	-45.8
Am	285	-10.9	-32.2	-38.9	-41.9
Cm		+609	+416	+350	+316
M.A	420	-0.68	-23.8	-31.1	-34.5
Mass balance (kg/TWeh)					
M.A	+5.6	-0.1	-4.9	-9.6	-14.2

Final concentration sensitivity

In order to estimate uncertainties, one first needs to get sensitivity coefficients. For mass balance, as stated in Equation (3), Bateman sensitivity combines direct and adjoint concentrations and derivative of Bateman equation relative to cross section. Table 6 shows cross section dependence of sensitivity (integrated over energy and space) for the main final minor actinides isotopic mass for MOX and 4% MA content cases. Only absolute values greater than 0.05 are shown.

Table 6: Minor actinides final concentration sensitivity

Final Np237 concentration				
Isotope	Nxn			
	MOX		4%	
U238	+0.895		+0.075	
	Capture		Fission	
	MOX	4%	MOX	4%
U235	+0.059			
U236	+0.064			
U238	-0.063			
Np237	-0.301	-0.570	-0.061	-0.120
Final Am241 concentration				
Pu240	+0.240			
Pu241	-0.058		-0.302	
Am241	-0.065	-0.098	-0.061	-0.097
Final Am243 concentration				
Pu241	+0.071			
Pu242	+0.883		-0.058	
Am243	-0.320			
Final Cm244 concentration				
Pu241	+0.052			
Pu242	+0.916	+0.233		
Am243	+0.764	+0.602		
Cm244	-0.126	-0.186	-0.059	-0.091

For Np237, initial concentration in MOX case is null and final concentration comes from (n,2n) reaction on U238 and thus the corresponding sensitivity is close to 1. For 4% MA content case, the initial mass is greater than was is produced in the MOX case at end of irradiation so the final mass balance is almost no sensitive to (n,xn) cross section. For both cases, the final Np237 concentration/mass is also sensitive to its own capture cross section with negative values.

For Am241, the production process involves decay from Pu241. In the MOX case, as the initial Am241 is quite small, the final concentration show high sensitivity level to cross sections that "impact" Pu241 concentration during irradiation: neutron capture on Pu240 and fission of Pu241.

For 4% MA content, as for Np237 case, the initial Am241 content is by far greater that what is produced by MOX case, the sensitivity on Pu240 capture cross section decrease from 0.24 to less than 0.05.

The same effect can be observed for Am243 and Cm244 isotopes: if the initial concentration is small, sensitivity of the final concentration to "ancestor isotopes" exhibits high level.

Final concentration/mass uncertainties

Tables 7 shows uncertainty levels for BOLNA and COMAC variance-covariance data set respectively. On general level, COMAC results show improvements of uncertainties values for main minor actinides value the whole range of MA content tested here. Relative uncertainties tend to reduce with MA content since "coupling" of sensitivity between isotopes far from each other in the depletion chain becomes small.

Table 7: Final concentration/mass uncertainties for BOLNA and COMAC data (%), homogeneous mode

Content	MOX		2%		4%	
Isotope	BOLNA	COMAC	BOLNA	COMAC	BOLNA	COMAC
Np237	5.07	5.57	2.17	5.17	2.06	5.07
Am241	3.50	1.13	1.32	0.41	1.04	0.29
Am242m	6.96	5.01	7.51	6.07	7.52	6.10
Am243	19.83	4.30	12.64	3.37	9.33	3.11
Cm242	3.39	1.26	1.53	0.83	1.36	0.77
Cm243	16.87	28.40	18.96	27.37	19.20	27.37
Cm244	20.98	7.48	9.78	7.88	7.20	7.99
Cm245	24.52	35.25	15.16	29.73	14.74	28.93

COMAC values lead to lower uncertainties than BOLNA for the main americium isotopes whose mass is still important at the end of irradiation: Am241 and Am243.

For Np237, the MOX cases are driven by U238(n,xn) uncertainty level which is similar for BOLNA and COMAC. Beyond MOX, Np237 uncertainty is driven by Np237 capture data for which COMAC contribution is twice BOLNA a first order.

Results for Cm243 and Cm245 show a net increase for COMAC values. For main (quadratic) contributions to the Cm243, the uncertainty levels are:

- Cm242 capture: 28% for COMAC compared to 12% for BOLNA.
- Cm243 fission: 3% for COMAC compared to 14% for BOLNA.

The same analysis on Cm245 shows different contribution level from its “fathers” namely Pu242, Cm244 for COMAC and BOLNA data.

This first set of results have to be analysed more in depth to provide feedback on nuclear data evaluators on specific isotope, cross section and energy range to decrease uncertainty level. These results have been provided to fuel cycle scenario in order to evaluate impact on global parameter such as mass inventory, radiotoxicity reduction, etc.

Conclusions

Minor actinides transmutation issue is one of the main concerns of the fourth generation reactors design. Uncertainties on mass balance are needed in order to see the impact on core conception (viability) as well as on fuel cycle back end and front end. In this scope nuclear uncertainty data propagation with depletion has been made available in the frame of the ERANOS deterministic code system by means of Bolztman/Bateman coupling.

Minor actinides mass uncertainties were estimated using COMAC variance-covariance data. Considering homogeneous management using SFR V2B core these uncertainties are ranging 1% to 5% for Americium and Neptunium isotopes. Larger values are obtained for Curium isotopes. Some work still have to be done at the evaluation level to reduce these values to comply with acceptable uncertainty level required by scenarios.

References

[1] P. Sciora et al. (2009), "A break even oxide fuel core for an innovative french sodium-cooled fast reactor: neutronic Studies Results", Proceedings of GLOBAL 2009, Paris, France.

[2] Gandini, A., G. Palmiotti and M. Salvatores (1986), "Equivalent generalised perturbation theory (EGPT)", Annals of Nuclear Energy, Vol. 13, Issue 3, pages 109-114.

[3] Gandini, A. (1975), "Time-Dependent Generalised Generalised Perturbation Methods for Burnup Analysis", Technical Report CNEN RT/FI(75)4, Comitato Nazionale per l'Energia Nucleare, Rome, Italy.

[4] Williams, M.L. (1979), "Development of Depletion Perturbation Thoery for Coupled Neutron/Nuclide Fields Feasibility", ANS Nuclear Science and Engineering, Vol. 70, Number 1, pages 20-36.

[5] Rimpault, G. et al. (2002), "The ERANOS code and data system for fast reactor neutronic analyses", Proceedings of the PHYSOR 2002 International Conference on the New Frontiers of Nuclear Technology: Reactor Physics, Safety and High-Performance Computing: the ANS 2002 RPD Topical Meeting, 7-10 October 2002, Seoul, Republic of Korea.

[6] Coquelet-Pascal, C. et al. (2011), "Comparison of Different Scenarios for the Deployment of Fast Reactors in France – Results Obtained with COSI", GLOBAL 2011: Toward and Over the Fukushima Daiichi Accident, Paper No. 360942, 11-16 December 2011, Makuhari Messe, Chiba, Japan.

[7] Koning, A. et al (2006), "The JEFF-3.1 Nuclear Data Library," Nuclear Energy Agency Data Bank, OECD, JEFF Report 21, NEA No. 6190, Paris, France.

[8] Rochman, D., M. Herman, P. Oblozinsky, S.F. Mughabghab (2007), "Prelimnary Cross Section and ν-bar Covariances for WPEC Subgroup 26", BNL Report: BNL-77407-2007-IR, Brookhaven National Laboratory, United States.

[9] Archier, P., G. Noguère, C. De Saint Jean, A.J.M Plompen, C. Rouki (2013), "New JEFF-3.2 Sodium Induced Cross-sections Evaluation for Neutron Fast Reactors Applications: from 0 to 20 MeV", The 2013 International Conference on Nuclear Data for Science and Technology (ND2013), 4-8 March 2013, New York, United States, Nuclear Data Sheets (2014), Vol. 118, pages 140-143.