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Uncertainty Analysis Applied to Fuel Depletion Calculations

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This paper presents a newly developed uncertainty methodology, based on a statistical approach, for assessing uncertainties in lattice code predictions due to uncertainties in fuel description and depletion conditions. It has been applied to depletion calculations with CASMO-4 and experimental data from the ARIANE Programme to estimate the calculation uncertainties in nuclide concentration and other neutronic parameters at any time during the irradiation history. Results have shown that important information on the quality of the code's predictions can be obtained by analyzing the comparison of the code's estimates and their associated uncertainty, in the form of tolerance intervals, with experimental data and their reported errors.

KEYWORDS: uncertainty analysis, fuel depletion,CASMO-4, ARIANE

1. Introduction

The capabilities of the widely used lattice code CASMO-4 [1] to predict the isotopic composition of highly burnt MOX fuel were studied in a series of calculations that simulated the depletion history of several MOX fuel samples analyzed in the course of the ARIANE Programme [2, 3]. Comparison of the CASMO-4 estimates is discussed in a separate contribution [4]. Uncertainty analysis in neutronic applications has been generally based on sensitivity analysis making use of adjoint calculation techniques [5,6,7]. The present paper reports on a newly developed methodology, based on a statistical approach, for assessing uncertainties in lattice code predictions of the above type due to uncertainties in fuel description, viz. depletion conditions, initial isotopic content and fuel assembly dimensions. Applying this methodology, uncertainty propagation studies can be performed that quantify the uncertainties in the estimated nuclide compositions at any time during the irradiation history and compare them with the experimental uncertainties. This will enable conclusions to be drawn about the quality of the CASMO-4 calculational methods/data per se. The methodology also permits to obtain corresponding burn-up dependent uncertainty estimates for neutronic parameters such as k_{inf}, Σ_a, $\nu\Sigma_f$, etc. as a function of the uncertainties mentioned above.

2. Description of the Methodology

The methodology propagates uncertainties (stochastic quantities) in the variables describing the fuel and its depletion conditions to the nuclide concentrations as a function of burn-up, as well as at the time of measurement. It is based on the approach proposed by McKay et al [8], in which the uncertainties of a code's input and model variables are treated as stochastic quantities that, when deterministically processed by the code, generate output values which can also be treated and analyzed as random variables. In general terms, the code is a non-linear deterministic function, *Code(•)*, which transforms the vector of its input random variables (nominal values with stochastic uncertainty) $\boldsymbol{X} = (X_1,...,X_k)$ into the vector of

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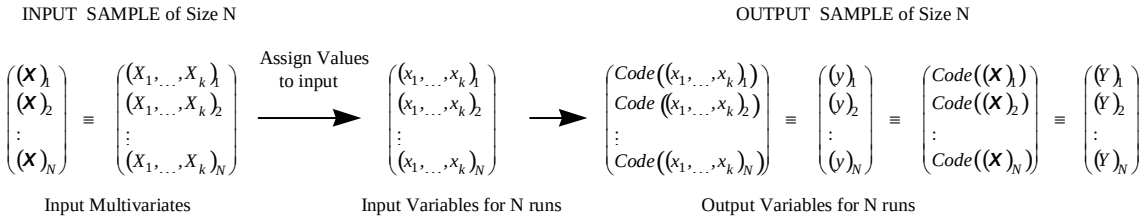
selected outputs $\boldsymbol{Y}$, thus propagating the stochastic nature of $\boldsymbol{X}$ to the code output, i.e. to $\boldsymbol{Y}$ (see Figure 1). Statistical sampling techniques, e.g. simple random sample, are employed to generate a series of code calculations with combinations of values of input and depletion variables randomly selected, which yield a sample of output values for the results of interest.

Probability density functions (PDFs) of the code results would contain all the information needed to compute their uncertainty, but such functions are usually not known. Since an analytical expression for *Code(•)* is not available, it is impossible to obtain an analytical expression for the PDFs of the output variables [8]. Thus, the only remaining alternative is to obtain as much information as possible about the properties of the PDFs from empirical distribution functions and estimators, such as mean, variances, quantiles, etc. The application of non-parametric statistical methods [9] for this purpose permits to extract important information about the code output uncertainties in terms of tolerance intervals with a certain probability content and level of confidence (statistically more appropriate to measure uncertainty than confidence intervals [8]). Tolerance intervals measure the uncertainty in the code's predictions by providing maximum and minimum values which bracket a certain fraction (e.g. 95%) of all possible output values that might result from the uncertainty in the input variables, and give a confidence level (e.g. 95%) for this statement.



$(\boldsymbol{X})_i$: represents sample number i of the k input variables.
$Code((\boldsymbol{X})_i)$: represent the application of *Code* to sample $(\boldsymbol{X})_i$

Fig.1 Sampling Process Scheme: k Input Variables, Sample of Size N

The size of the sample necessary for a given probability content and confidence level of tolerance intervals defined by the *maximum* and *minimum* values of the output sample can be obtained from non-parametric statistical analysis, following the approach of Glaesser et al. [10,11] for applications to thermal-hydraulic analysis. These authors followed a rigorous approach to minimum sample size, as discussed in Ref. [9], based on the general formula obtained by Noether [12]

$$\alpha \geq \sum_{i=0}^{r+m-1} \binom{n}{i} (1-q)^i \, q^{n-i} \,, \tag{1}$$

where $(1-\alpha)$ is the confidence of the tolerance interval, q is the proportion of the population within the interval, n is the size of the sample (the unknown), and r and m are numbers related to the values of the sample that bound the tolerance interval. The most used values are $r+m=1$, one-side tolerance limits, and $r+m=2$, two-sided tolerance limits. Solution of this equation for n, when q and $(1-\alpha)$ are known, yields the minimum sample size. In Ref. [9], tables are given with the solution for various $((1-\alpha), q)$ pairs and for the cases $r+m=1$ *and* $r+m=2$. For $r+m=2$, Eq. (1) is known as the Wilks Formula [13,14]: $\alpha \geq (1-n)\, q^n + n\, q^{n-1}$. For instance, for two-sided tolerance limits of 95% for the population $(q = 0.95)$ and for 95% confidence $(1-\alpha = 0.95)$, the minimum sample size, n, is ***93.*** The parameters q and $(1-\alpha)$ are also referred to usually as β and γ, respectively, so that one talks about a *tolerance interval* with $\beta = .95$ and $\gamma = .95$. Alternatively, it is also possible to obtain statistical importance and local and global

sensitivity measures with the methodology described, such as the Spearman's and Kendall's coefficients, or simple or partial rank correlation coefficients (SRCCs, PRCCs) for non-linear systems [8,9], in the case of both the scalar and burn-up dependent uncertainty determination (see Figure 4). These, however, are not discussed in this paper.

3. Description of the Calculation Procedure

3.1 Fuel Sample Characteristics

The fuel sample analyzed (Sample BM5) belonged to a MOX fuel rod in a 14x14 fuel assembly irradiated for 6 cycles (20-25) at the Swiss Beznau-1 NPP. The MOX enrichment was 5.5 weight-per-cent (*w/o)* Pu [2,3] (*with respect to heavy metal*). The fuel assembly was shuffled in the core during re-loading outages, thus experiencing different neutron flux conditions that tended to diminish the influence of inter-assembly neutron flux changes on the sample burn-up. The total average rod burn-up was calculated as 52 MWd/kgHM, that of the sample being assessed as 58.9 MWd/kgHM.

3.2 CASMO-4 Depletion Model

The uncertainty analysis made use of an equivalent 1-D pin-cell model with seven radial fuel zones [4], developed according to the information in Refs.[2, 3]. The initial isotopic composition of the sample was based on the measurements made by the different radio-chemical laboratories participating in the ARIANE Programme. Detailed irradiation histories for the CASMO-4 depletion model were built from data given in Refs. [2,3] for sample power density, coolant temperature and density, fuel temperatures, and boron content as a function of burn-up. The 70-group library J2LIB of CASMO-4 [1], based on JEF2.2 data, was employed (nuclide production and decay).

3.3 Depletion Calculation

The calculations depleted the fuel sample up to 58.9 MWd/kgHM according to the irradiation history, with depletion intervals less or equal to 1 MWd/kgHM, in order to preserve the accuracy of the "predictor-corrector" solution procedure used by CASMO-4. The calculations were extended to the time of measurement for each of the nuclides listed in Table 2, in order to account for the decay during the fuel cool-down period.

3.4 Uncertainty Analysis Procedure

The application of the uncertainty methodology described above to depletion calculations required the assignment of uncertainty measures in the form of probability density functions (PDFs) to all the input variables considered. Irradiation history variables were assumed to vary 1% with a uniform PDF about their given values (according to the detailed irradiation history) for the 6 cycles (20 to 25) of the sample irradiation in Beznau-1. PDFs for pellet dimensions, density, Zr homogenization for the pin cell and experimental determination of the sample initial isotopic inventory were assumed normal, and their parameters obtained from fuel manufacture's data and the ARIANE Programme Report. Figure 2 shows, as examples, the PDFs for ^{239}Pu initial concentration and the uniform fuel-temperature uncertainty assumed during irradiation. Geometric, fuel density and homogenized Zr-content uncertainties have been considered in terms of normal-3σ (99% probability content) values. Simple random sampling of these PDFs produced a set of input variable values that were used to perform 100 different CASMO-4 depletion calculations, which is above the value of ***93*** [9, Table A6] determined from the Wilks formula for a two-sided *tolerance interval* of $\beta = 95\%$ probability content and $\gamma = 95\%$ confidence level.

Table 1 Uncertainty Data for the 18 Input and Irradiation History Variables Considered

Name of Variable	Units	Distribution	Min	Max	95% Unc	Nominal	Variance	Std.Dev.
Fuel Temperature	*K*	*Uniform*	*-1.0%*	*1.0%*		*0.000*		
Moderator Temperature	*K*	*Uniform*	*-1.0%*	*1.0%*		*0.000*		
Boron Concentration	*ppm*	*Uniform*	*-1.0%*	*1.0%*		*0.000*		
Coolant Density	*g/cm^3*	*Uniform*	*-1.0%*	*1.0%*		*0.000*		
Power Density	*W/gM*	*Uniform*	*-1.0%*	*1.0%*		*0.000*		
Fuel Density	*g/cm^3*	*Normal*	*10.2180*	*10.5180*	*0.150*	*10.368*	*2.5000E-03*	*5.0000E-02*
Gap Thickness	*cm*	*Normal*	*0.0026*	*0.0168*	*0.00705*	*0.0097*	*5.5277E-06*	*2.3511E-03*
Clad Thickness	*cm*	*Normal*	*0.0542*	*0.0692*	*0.00745*	*0.0617*	*6.1696E-06*	*2.4839E-03*
Equivalent Pin Pitch Radius	*cm*	*Normal*	*0.8147*	*0.8569*	*0.00841*	*0.8358*	*4.9396E-05*	*7.0283E-03*
Equivalent Zr Content	*g/cm*	*Normal*	*0.1032*	*0.1157*	*1.893%*	*0.1095*	*4.2925E-06*	*2.0718E-03*
U235 Concentration in Fuel 4	*w/o*	*Normal*	*0.2116*	*0.2250*	*2.050%*	*0.21830*	*5.0067E-06*	*2.2376E-03*
U234 Concentration in Fuel 4	*w/o*	*Normal*	*0.0016*	*0.0022*	*10.010%*	*0.00189*	*8.9481E-09*	*9.4595E-05*
U238 Concentration in Fuel 4	*w/o*	*Normal*	*93.6436*	*94.9164*	*0.450%*	*94.280*	*4.4999E-02*	*2.1213E-01*
Pu238 Concentration in Fuel 4	*w/o*	*Normal*	*0.0317*	*0.0347*	*3.050%*	*0.03317*	*2.5588E-07*	*5.0584E-04*
Pu239 Concentration in Fuel 4	*w/o*	*Normal*	*3.6039*	*3.6661*	*0.570%*	*3.63500*	*1.0732E-04*	*1.0360E-02*
Pu240 Concentration in Fuel 4	*w/o*	*Normal*	*1.2651*	*1.2869*	*0.570%*	*1.27600*	*1.3225E-05*	*3.6366E-03*
Pu241 Concentration in Fuel 4	*w/o*	*Normal*	*0.3574*	*0.3636*	*0.570%*	*0.36050*	*1.0556E-06*	*1.0274E-03*
Pu242 Concentration in Fuel 4	*w/o*	*Normal*	*0.1442*	*0.1466*	*0.570%*	*0.14540*	*1.7172E-07*	*4.1439E-04*

In the design of the input sample based on the data in Table 1, it was necessary to include the inter-dependencies between some of the depletion parameters, i.e. fuel temperature and power density, moderator temperature and coolant density, and moderator temperature and power density. An analysis of the irradiation history of BM5 showed a clear statistical correlation between these variables, since they are physically related during plant operation. This was taken into account when the sample of input variables was generated by including in the sampling procedure information about the correlation in terms of regression coefficients that quantified the statistical correlations observed.

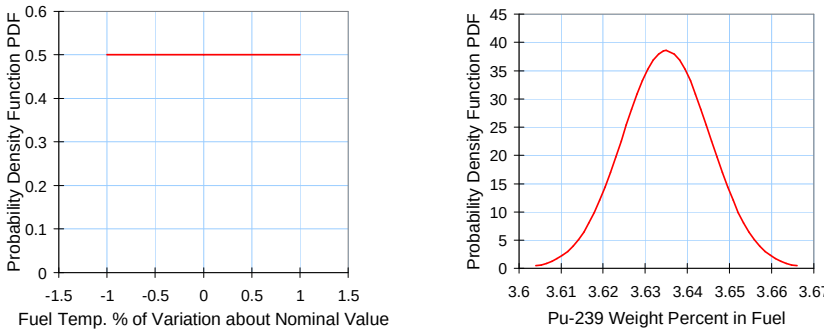


Fig.2 Probability Density Functions Quantifying Uncertainty in Fuel Temperature Value during Irradiation (*Uniform*) and ^{239}Pu Initial *w/o* in Fuel (*Normal*)

Figure 3 displays the results of the sampling process for the initial concentration of ^{235}U and ^{239}Pu. One hundred values were generated for each of the 18 input variables, and one CASMO-4 depletion calculation performed for each set of 18 variable values. Pearson's, Kendall's and Spearman's [9] correlation matrices for the sample showed that no spurious correlations appeared between input variables that were assumed independent. It is important to mention that the initial composition was normalized so that the sum of the fractions of individual Pu and U isotopes with respect to the total Pu and U content was 1.0, respectively, for each sample generated. Initial Am content, negligible in comparison to Pu and U, was left as a free variable to complete the total heavy metal (HM) content in the fuel sample. Figure 3 shows, again as example, the results of the sampling procedure for the fuel temperature and the initial ^{239}Pu concentration.

The processing of input uncertainty data, execution of CASMO-4 and processing of output information have been automated and used in conjunction with the statistical analysis package SUSA [15] to produce the statistical measures of the uncertainty in the output nuclide densities shown in Table 2.

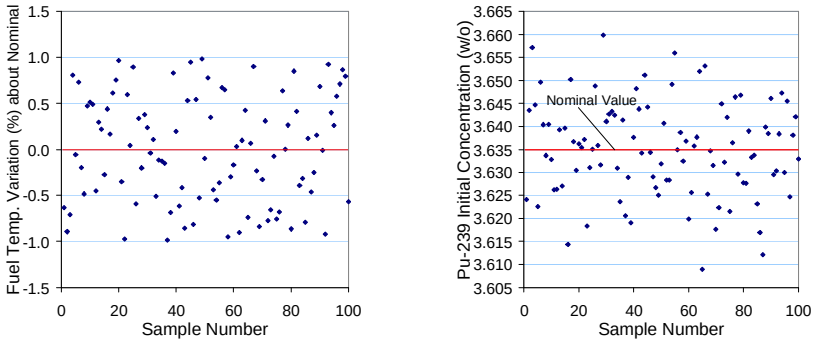


Fig.3 Results of the Sampling Process (100 Samples) for Fuel Temperature Value during Irradiation (*Uniform PDF*) and ^{239}Pu Initial *w/o* in Fuel (*Normal PDF*)

4. Results and Discussion

The application of the present methodology produced quantitative uncertainty estimates in the form of two-side tolerance intervals, as well as mean values, for the isotopic contents and other neutronic parameters as a function of burn-up. An example can be seen in Figure 4. It shows the results for the concentration of ^{239}Pu and the assembly k_{inf} as a function of burn-up.

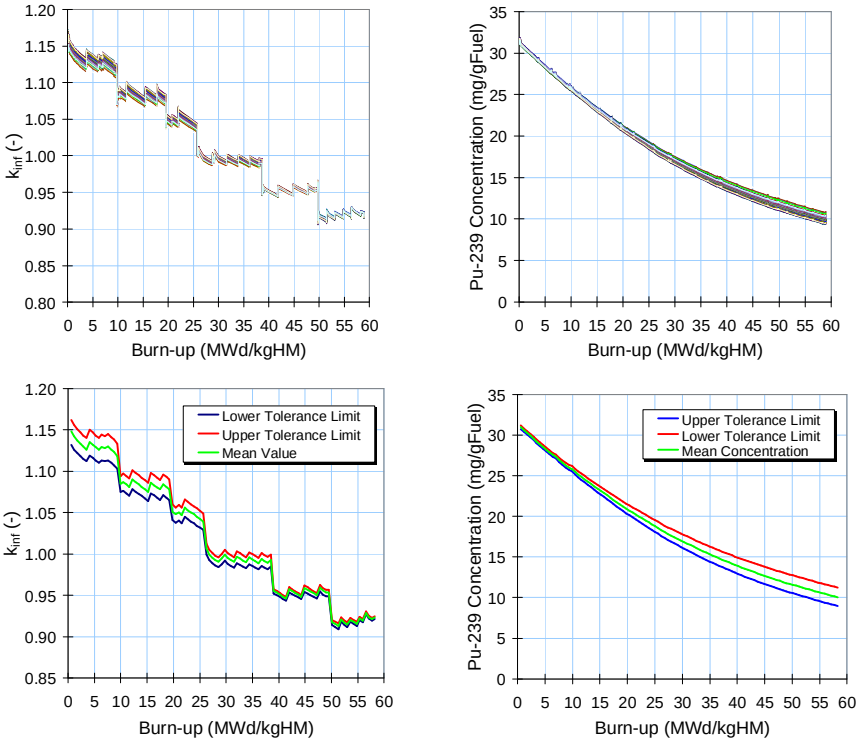


Fig.4 Results of CASMO-4 Calculations 25 to 75 *(Upper Plots)* for Assembly k_{inf} and ^{239}Pu Concentration with [β =.95, γ = .95] tolerance intervals, and Mean Values (*Lower Plots*)

The upper plots display the results of runs 25 to 75 of the 100 performed, a subset of the total output sample. The lower plots show the uncertainties in the values of these two output variables as the input-variable uncertainties of Table 1 are propagated through the CASMO-4 calculations. The results are produced after non-parametric statistical methods have been applied with SUSA to the output sample, in order to generate the *tolerance intervals* as a function of burn-up. The mean values are bracketed by the upper and lower tolerance interval limits for $\beta = .95$ and $\gamma = .95$.

In Figure 4, one can see that the uncertainty in the content of ^{239}Pu grows with burn-up, since its depletion is affected by the uncertainty in its initial concentration, by the uncertainty in its build-up and in that of nuclides that produce ^{239}Pu through (n,γ) reactions, viz. mainly ^{238}U. These reactions are influenced by uncertainties in the neutron spectrum, which are dependent on the uncertainties in irradiation parameters, and geometry of the fuel assembly (water-to-metal ratio). It should then be expected that as the burn-up proceeds, these uncertainties accumulate and tend to increase the uncertainty in the isotopic content of ^{239}Pu. A similar behavior was observed with other nuclides. In contrast, Figure 4 also shows that the uncertainty in k_{inf} is reduced with burn-up. k_{inf} is a 'global' parameter for the fuel assembly, which depends on the overall neutron balance and, as such, on the concentrations of all the nuclides present during the burn-up, these in turn being dependent on the depletion conditions, e.g. boron content, fuel temperature, etc. The corresponding uncertainties, which combine to yield the uncertainty on the k_{inf} value, appear to have compensating effects. As a result, the uncertainty on k_{inf} diminishes (from its relatively large initial value) with increasing burn-up. Clearly, the methodology described in this paper could be applied to assess the change with burn-up of the uncertainty in other neutronic parameters for the fuel assembly, e.g. individual macroscopic cross-sections generated for use in core calculations.

Table 2 Scalar Uncertainty Results, Tolerance Interval $[\beta = .95, \gamma = .95]$[*].

Nuclide	2-σ Exper. Uncertainty	Mean Diff.	[.95,.95] Calc. Uncertainty	Quality	Nuclide	2-σ Exper. Uncertainty	Mean Diff.	[.95,.95] Calc. Uncertainty	Quality
U-234	10.01	-15.77	1.81	B	Nd-142	10.01	6.52	0.75	G
U-235	2.05	0.92	**3.69**	G	Nd-143	0.56	4.58	1.66	B
U-236	5.02	-7.12	0.17	B	Nd-144	0.56	0.17	**1.13**	G
U-238	0.45	-0.21	**0.20**	G	Nd-145	0.56	2.55	0.36	B
Np-237	10.97	-14.97	3.88	B	Nd-146	0.56	3.81	0.51	B
Pu-238	3.05	-20.12	2.39	B	Nd-148	0.56	5.17	0.03	B
Pu-239	0.57	4.25	**11.29**	G	Nd-150	0.58	3.71	0.02	B
Pu-240	0.57	-8.19	1.13	B	Pm-147	16.01	-7.78	1.37	G
Pu-241	0.57	1.54	**5.68**	G	Sm-147	0.64	-2.42	**2.77**	G
Pu-242	0.57	-3.39	**3.95**	G	Sm-148	0.64	2.98	2.65	G
Am-241	3.51	18.24	5.63	B	Sm-149	2.09	19.02	11.75	B
Am242m	10.59	-15.80	11.96	G	Sm-150	0.64	2.48	0.33	B
Am-243	3.51	-4.42	0.57	B	Sm-151	0.79	34.36	11.54	B
Cm-242	3.46	-79.74	1.45	B	Sm-152	0.64	20.96	2.42	B
Cm-243	10.60	-5.93	3.97	G	Eu-153	0.67	10.43	1.09	B
Cm-244	3.13	-16.01	3.29	B	Eu-154	3.01	61.47	2.27	B
Cm-245	5.13	-5.42	**13.14**	G	Eu-155	8.81	-36.92	1.88	B
Sb-125	6.93	101.44	0.71	B	Gd-155	2.09	-27.65	2.84	B
I-129	11.24	59.74	0.12	B	Sr-90	16.01	11.02	0.35	G
Cs-133	3.06	2.45	0.84	G	Mo-95	9.70	1.85	0.23	G
Cs-134	3.09	19.24	3.42	B	Ru-101	16.13	43.20	0.37	B
Cs-135	3.06	4.80	3.82	G	Ru-106	15.24	67.87	1.08	B
Cs-137	3.04	1.06	0.06	G	Ag-109	18.58	44.15	1.57	B
Tc-99	19.47	14.46	0.59	G	Rh-103	9.93	59.51	0.19	B
Ce-144	5.94	-56.75	1.02	B					

[*]All quantities in %. "Calc. Uncertainty" represents *tolerance interval* size about mean value.

Another aspect of the present methodology is the calculation of tolerance intervals for mean estimates of isotopic composition at the time of measurement (scalar uncertainty), or at any other time during the irradiation. Thus, the nuclide density measurements from the SCK•CEN radio-laboratory [2,3], with normal 2σ statistical uncertainty band, have been compared to the nominal prediction of CASMO-4 and the two-sided *tolerance interval* about the mean value resulting from the 100 CASMO-4 calculations. Table 2 shows the results of the scalar uncertainty evaluation for a series of actinides and fission products considered in the ARIANE Programme. The column labeled "*Mean Diff.*" contains the differences between the experimental data and the mean nuclide concentration from the CASMO-4 calculations. Next to it, the column "*Calc. Uncertainty*" contains the calculation uncertainties for each nuclide based on the size of the $[\beta = .95, \gamma = .95]$ tolerance interval, comparable to a 2σ normal uncertainty. Finally, the column labeled "*Quality*" identifies those nuclide density determinations that, in view of the mean value, calculation uncertainty and experimental uncertainty, can be considered as good (G) estimates, since their calculated tolerance intervals **overlap** the experimental values and their normal 2σ experimental uncertainty error bands (see Figure 5 for ^{238}U, ^{137}Cs and ^{239}Pu.) The nuclides with a (B) yielded estimates whose *tolerance intervals* did not overlap the experimental error bands (see Figure 5 for ^{238}Pu) and give information to objectively judge the quality of the code's predictions.

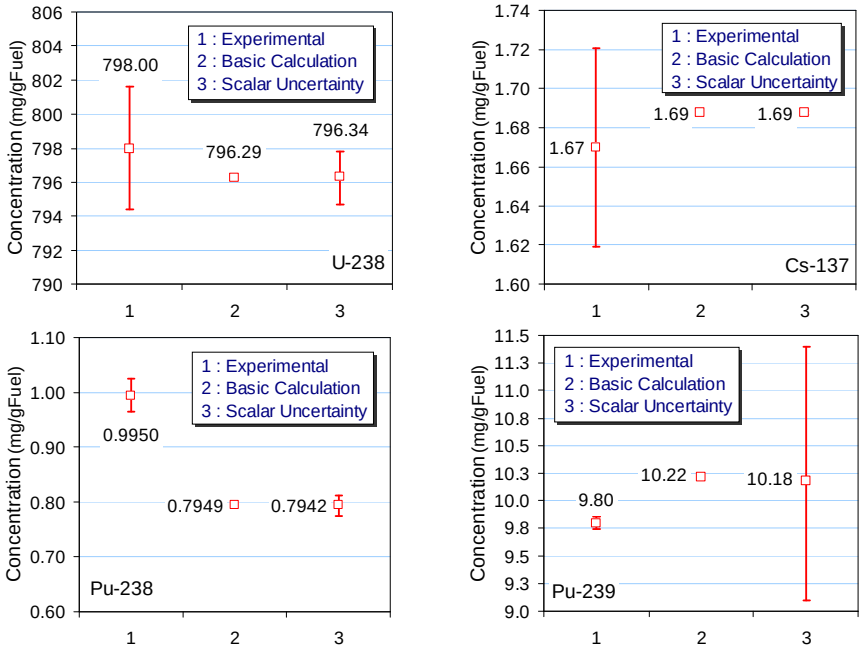


Fig.5 Nuclide Density Comparisons of Experimental Results, Calculated Values and Calculation Tolerances for ^{238}U, ^{137}Cs, ^{238}Pu and ^{239}Pu

Figure 5 shows some of the information in Table 2 in graphic form, with experimental concentration values and their errors, as well as the tolerance intervals bounding the mean predictions. The values (2), labeled "*Basic Calculation*", correspond to CASMO-4 results obtained using nominal values for all variables. An analysis of these plots, as indicated in the earlier explanation of "*Quality*" in Table 2, allows the identification of those estimates, which despite not exactly matching the experimental values can be considered good, since the calculation uncertainty is larger than the observed discrepancy (the cases of ^{238}U and ^{239}Pu in

Figure 5). It can also help to identify those nuclides for which the code's predictions are clearly in disagreement since, despite accounting for uncertainties in the input variables describing the sample and in the depletion conditions, these do not result in agreement with measurement within the experimental error band (the case of ^{238}Pu in Figure 5). In such cases, it is clear that other causes should be looked into, e.g. the accuracy of the neutronic data in the libraries, to explain the discrepancies observed. Figure 5 also shows that it is possible to identify those isotopes (illustrated here for the case of ^{137}Cs) for which uncertainties in the fuel assembly description and in the depletion conditions do not appreciably affect the calculated nuclide density. For nuclides such as ^{239}Pu (see Figure 5), these uncertainties have a significant impact, much larger than the experimental errors. Clearly, in such cases, a reduction of the uncertainties in the depletion history and/or in the fuel model used, i.e. geometry, initial inventory, etc., may contribute to better predictions. The determination of the main contributors to such uncertainties is also possible with the present methodology, through the calculation of statistical sensitivity measures as mentioned above.

Finally, it is important to point out that uncertainties in neutronic data, e.g. cross-sections, branching ratios, etc., have not been taken into account for the study reported in this paper. However, it is possible in principle to do so, if corresponding uncertainty data are available, by using these in the context of the sampling procedure described above. Further work is planned to focus on the inclusion of some of these sources of uncertainty into the methodology.

5. Conclusions

A novel development and application of a statistically based uncertainty methodology to fuel depletion calculations has been presented. It is based on the propagation of uncertainties in fuel element description and depletion conditions to the determination of nuclide concentration and other neutronic parameters at any time during the irradiation history. Applied to CASMO-4 and experimental data from the ARIANE Programme, the results have shown that important information on the quality of the code's predictions can be obtained by analyzing the comparison of the code's estimates and their associated uncertainty, in the form of tolerance intervals, with experimental data and their reported errors.

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