# NEW COVARIANCE DATA AND THEIR IMPACT ON ADS DESIGNS

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*Nuclear data uncertainties and their impact on a very wide range of reactor systems, including their associated fuel cycles, have to be assessed in order to consolidate preliminary design studies for new innovative systems. One specific class of systems is the so-called "dedicated waste transmuters", that are fast neutron systems (critical or sub-critical, i.e. ADS), loaded with a Minor Actinide dominated fuel and potentially U-free. The availability of very general tools for sensitivity and uncertainty analysis together with new variance-covariance matrix data, produced in a joint effort under the auspices of the OECD-NEA, by the world leading nuclear data evaluation groups, makes that endeavor particularly significant. In this report we discuss major results of interest for dedicated ADS and point out the most important fields and data types, where priority improvements are required.*

## I. INTRODUCTION

The potential impact of nuclear data uncertainties on a large number of performance parameters of an ADS dedicated to the transmutation of radioactive wastes was presented in Ref. 1. An uncertainty study was performed based on sensitivity analyses, which did underline the cross-sections, the energy range, and the isotopes that were responsible for the most significant uncertainties.

The sensitivity/uncertainty analysis carried out in that paper did allow to draw some conclusions:

1. The level of uncertainties in integral parameters for an ADS was found to be significant. The reduction of uncertainties did seem mandatory in more advanced phases of the studies in order to make sensible choices among options and optimizations.

2. As expected, the most crucial data were fission, capture, and inelastic cross sections of Minor Actinides (MA). Moreover, in the case of a Pb/Bi coolant, the data for these materials should be definitely improved, in particular inelastic and (n,2n) data.

To provide guidelines on priorities for new evaluations or validation experiments, required accuracies on specific nuclear data were derived, accounting for target accuracies on major design parameters.

It was pointed out that the required accuracies (mostly in the energy region below 20 MeV), in particular for minor actinide data, were of the same order of magnitude of the achieved accuracies on major actinides.

However, it was stressed that future studies related to the impact of nuclear data uncertainties, in particular in the detailed design assessment phase, should make use of variance-covariance data established in a much more rigorous manner.

This last point is the purpose of the present study. In fact, very recently a preliminary set of covariance data have been made available, in the frame of a NEA-OECD project (Ref. 2). Data have been provided by BNL, LANL, ORNL and NRG (Refs. 3-12). For the present study, as in Ref. 2, all the available BNL data have been used, except the U-235, U-238 and Pu-239 data, which have been taken from the combined LANL/ORNL evaluation and the Pb isotope data, taken from the NRG evaluation. Missing data have been taken from the ANL estimated covariance data (Ref. 13). The full set of covariance data is called "BOLNA".

This new study allows revisiting the required target accuracies, comparing them with the current status of uncertainties, as documented in the new covariance data.

## II. REFERENCE CALCULATIONS

The chosen ADS system had some general features (e.g., the mass ratio between plutonium and MA, the americium-to-curium ratio, etc.) that are representative of the class of MA transmuters with a fast neutron spectrum and a uranium-free fuel. The target and the coolant material of the core are the Pb-Bi eutectic, and it was very close to the sub-critical core, which has been analyzed in the framework of an international OECD-NEA benchmark ("Comparison Calculations for an Accelerator-Driven Minor Actinides Burner," OECD Nuclear Energy Agency, 2001).

Sensitivity coefficients (in a 15 energy group structure as in Ref. 1) have been calculated at ANL with the ERANOS code system (Ref. 14) for all the ADS parameters potentially most sensitive to nuclear data uncertainties: Multiplication factor, Power peak, defined as the point maximum power value normalized to the total power, Burn-up $\Delta k/k$, Coolant void reactivity coefficient, Doppler reactivity coefficient, Nuclide density at end of cycle (transmutation potential), the ratio $\varphi^*$ of the average external source importance to average fission neutron importance. The Max dpa, Max He and, Max H production are the values of the displacements per atom (dpa), He production, and H production at the spatial point where they reach their maximum value. The maximum value of the ratio (He production)/dpa is calculated at its own maximum value position.

## III. RESULTS

Tables I and II show the uncertainty on most of the relevant parameters and the breakdown by isotope contribution. The uncertainties are still significant. Pu-241 and some of the higher Pu isotopes contribute to the uncertainties, while the Pu-239 contribution is always very small, in agreement with what was already pointed out in Ref. 1. However, the major contributions are due to MA data and in particular to Cm-244 data uncertainties. Am-241, Am-243, Cm-245 give also some noteworthy contributions. As for structural materials, Fe-56 and Bi-209 show not negligible effects.

Tables III to VII allow to see which reactions for each isotope are the most important contributors to the uncertainty of the different integral parameters. The role of fission cross-section uncertainties is remarkable for most parameters. In fact, uncertainties in the fission cross-sections have both an effect on reactivity and an effect in the hardness of the spectrum. This last effect can be seen both on the Power Peak and on the Max He/dpa ratios.

With respect to the previous study, there is much less impact of (n,2n) cross-sections, due to lower values of uncertainty in the present variance-covariance data (~10%, to be compared to the 100% value used in Ref. 1).

On the contrary, the significant impact of Fe-56 inelastic cross-section is confirmed, in particular on the void reactivity coefficient. Finally, Table VIII gives, as an example, the energy breakdown of the $k_{eff}$ uncertainty due to the fission data of Cm-244 and Cm-245.

These and other similar data allow to reanalyze the required target accuracies evaluated in Ref. 1. For example, it is clear that with respect to the present estimated uncertainties (see Tables IX and X), the uncertainty on the fission of Cm-244 should be cut by an order of magnitude and that the improvement needed on the fission of Am-241 is at least by a factor of two (see Table XI). On the contrary, the accuracy announced for the capture of Am-241 looks almost satisfactory. Finally, the accuracy required for the inelastic of Fe-56 (~5%), is far from being achieved, and in any case extremely difficult to achieve experimentally.


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TABLE I. Uncertainties (%) on ADS Parameters. Breakdown by Isotope Contribution

| | $k_{eff}$ | Peak Power | Void reactivity | Burnup reactivity |
|---|---|---|---|---|
| Pu238 | 0.25 | 1.8 | 0.47 | 40.91 |
| Pu239 | 0.20 | 1.5 | 0.76 | 5.38 |
| Pu240 | 0.23 | 1.7 | 0.48 | 2.30 |
| Pu241 | 1.05 | 7.6 | 2.04 | 21.44 |
| Pu242 | 0.17 | 1.2 | 0.49 | 3.09 |
| Np237 | 0.37 | 2.7 | 1.18 | 2.74 |
| Am241 | 0.97 | 7.0 | 4.08 | 9.12 |
| Am242m | 0.14 | 1.0 | 0.32 | 17.86 |
| Am243 | 0.63 | 4.9 | 2.22 | 5.81 |
| Cm242 | 0.0 | 0.0 | 0.0 | 25.77 |
| Cm244 | **1.9** | **13.7** | **3.5** | **34.1** |
| Cm245 | 1.05 | 7.6 | 1.58 | 26.77 |
| Fe56 | 0.93 | 7.2 | 5.53 | 1.17 |
| Pb | 0.09 | 0.5 | 3.96 | 0.41 |
| Bi209 | 0.31 | 2.2 | 12.13 | 0.55 |
| **Total** | **2.94** | **21.4** | **15.5** | **77.5** |

TABLE II. Uncertainties (%) on ADS High Energy Parameters. Breakdown by Isotope Contribution

| | φ* | (n,p) | (n,α)/DPA |
|---|---|---|---|
| Pu238 | 0.2 | 2.0 | 1.5 |
| Pu239 | 0.2 | 1.7 | 1.3 |
| Pu240 | 0.1 | 1.9 | 1.4 |
| Pu241 | 0.8 | **8.6** | **6.3** |
| Pu242 | 0.1 | 1.4 | 1.0 |
| Np237 | 0.2 | 3.0 | 2.2 |
| Am241 | 0.5 | 7.8 | 5.7 |
| Am242m | 0.1 | 1.2 | 0.9 |
| Am243 | 0.4 | 5.0 | 4.2 |
| Cm244 | 0.9 | **15.8** | **11.3** |
| Cm245 | 0.8 | **8.6** | **6.3** |
| Fe56 | 0.3 | **8.1** | **6.3** |
| Pb | 0.3 | **2.4** | **3.4** |
| Bi209 | 0.9 | **3.9** | **4.3** |
| Total | 1.9 | **24.5** | **18.5** |

TABLE III. $k_{eff}$ Uncertainties (%) Breakdown by Isotope and Reaction Type

| | $\sigma_{capt}$ | $\sigma_{fiss}$ | ν | $\sigma_{inel}$ | Total |
|---|---|---|---|---|---|
| Pu238 | 0.02 | 0.21 | 0.13 | 0.01 | 0.25 |
| Pu239 | 0.10 | 0.12 | 0.06 | 0.11 | 0.20 |
| Pu240 | 0.08 | 0.16 | 0.14 | 0.03 | 0.23 |
| Pu241 | 0.07 | 1.04 | 0.04 | 0.02 | 1.05 |
| Pu242 | 0.06 | 0.15 | 0.03 | 0.02 | 0.17 |
| Np237 | 0.18 | 0.29 | 0.06 | 0.13 | 0.37 |
| Am241 | 0.46 | 0.83 | 0.16 | 0.16 | 0.97 |
| Am243 | 0.29 | 0.35 | 0.09 | 0.43 | 0.63 |
| Cm242 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Cm243 | 0.00 | 0.12 | 0.01 | 0.00 | 0.12 |
| Cm244 | 0.11 | **1.90** | 0.36 | 0.04 | 1.94 |
| Cm245 | 0.01 | 1.04 | 0.13 | 0.01 | 1.05 |
| Fe56 | 0.05 | 0.00 | 0.00 | **0.93** | 0.93 |
| Bi209 | 0.02 | 0.00 | 0.00 | 0.31 | 0.31 |
| Total | 0.61 | **2.61** | 0.47 | **1.10** | **2.94** |

TABLE IV. Burnup Uncertainties Contributions (%). Breakdown by Isotope and Reaction Type

| | $\sigma_{capt}$ | $\sigma_{fiss}$ | ν | Total |
|---|---|---|---|---|
| Pu238 | 2.6 | **30.2** | 18.3 | 35.4 |
| Pu241 | 0.9 | 10.6 | 0.3 | 10.6 |
| Am241 | 2.8 | 8.0 | 1.6 | 8.8 |
| Am242m | 0.6 | 12.8 | 1.0 | 12.9 |
| Am243 | 1.4 | 2.7 | 0.7 | 5.0 |
| Cm242 | 1.3 | **23.0** | 5.3 | 23.6 |
| Cm244 | 2.1 | **28.5** | 5.2 | 29.1 |
| Cm245 | 0.1 | 11.9 | 1.8 | 12.0 |
| Total | 5.2 | **52.5** | 20.0 | **56.6** |

TABLE V. Peak Power Uncertainties (%) Breakdown by Isotope and Reaction Type

| | $\sigma_{capt}$ | $\sigma_{fiss}$ | $\sigma_{inel}$ | Total |
|---|---|---|---|---|
| Pu241 | 0.6 | 7.6 | 0.1 | 7.6 |
| Np237 | 1.4 | 2.1 | 1.0 | 2.7 |
| Am241 | 3.6 | 5.8 | 1.2 | 7.0 |
| Am243 | 2.3 | 2.4 | 3.5 | 4.9 |
| Cm244 | 0.9 | **13.4** | 0.3 | 13.7 |
| Cm245 | 0.1 | 7.6 | 0.1 | 7.6 |
| Fe56 | 0.4 | 0.0 | **7.2** | 7.2 |
| Bi209 | 0.2 | 0.0 | 2.2 | 2.2 |
| Total | 4.8 | **18.6** | **8.6** | **21.4** |

TABLE VI. Void Uncertainties (%) Breakdown by Isotope and Reaction Type

| | $\sigma_{capt}$ | $\sigma_{fiss}$ | $\sigma_{el}$ | $\sigma_{inel}$ | Total |
|---|---|---|---|---|---|
| Pu241 | 0.1 | 2.0 | 0.0 | 0.1 | 2.0 |
| Am241 | 2.1 | 3.3 | 0.1 | 0.7 | 4.1 |
| Am243 | 1.3 | 1.6 | 0.1 | 0.6 | 2.2 |
| Cm244 | 0.5 | 3.2 | 0.0 | 0.2 | 3.5 |
| Fe56 | 0.4 | 0.0 | 1.0 | 5.4 | 5.5 |
| Pb | 1.5 | 0.0 | 2.8 | 2.3 | 4.0 |
| Bi209 | 0.7 | 0.0 | 1.6 | **12.0** | 12.1 |
| Total | 3.3 | 5.6 | 3.5 | **13.5** | **15.5** |

TABLE VII. Max(n,α)/DPA Uncertainties (%) Breakdown by Isotope and Reaction Type

| | $\sigma_{capt}$ | $\sigma_{fiss}$ | ν | $\sigma_{inel}$ | $\sigma_{n,2n}$ | Total |
|---|---|---|---|---|---|---|
| Pu241 | 0.5 | 6.3 | 0.2 | 0.1 | 0.0 | 6.3 |
| Np237 | 1.1 | 1.7 | 0.3 | 0.9 | 0.0 | 2.2 |
| Am241 | 2.8 | 4.8 | 0.9 | 1.1 | 0.0 | 5.7 |
| Am243 | 1.8 | 2.0 | 0.5 | 3.2 | 0.0 | 4.2 |
| Cm244 | 0.7 | **11.1** | 2.1 | 0.3 | 0.0 | 11.3 |
| Cm245 | 0.0 | 6.2 | 0.8 | 0.1 | 0.0 | 6.3 |
| Fe56 | 0.2 | 0.0 | 0.0 | **6.3** | 0.0 | 6.3 |
| Pb | 0.5 | 0.0 | 0.0 | 3.2 | 1.2 | 3.4 |
| Bi209 | 0.2 | 0.0 | 0.0 | 3.0 | 3.2 | 4.3 |
| Total | 3.8 | **15.4** | 2.8 | **8.5** | 3.4 | **18.6** |

TABLE VIII. Energy Breakdown of $k_{eff}$ Selected Uncertainties Components (%)

| Gr. | Energy | Cm244 $\sigma_{fiss}$ | Cm245 $\sigma_{fiss}$ |
|---|---|---|---|
| 1 | 19.6 MeV | 0.1 | 0.0 |
| 2 | 6.07 MeV | **0.6** | 0.2 |
| 3 | 2.23 MeV | **0.8** | 0.2 |
| 4 | 1.35 MeV | **1.5** | **0.5** |
| 5 | 498 keV | **0.5** | **0.5** |
| 6 | 183 keV | 0.3 | **0.6** |
| 7 | 67.4 keV | 0.2 | **0.4** |
| 8 | 24.8 keV | 0.0 | 0.1 |
| 9 | 9.12 keV | 0.0 | 0.1 |
| 10 | 2.03 keV | 0.0 | 0.1 |
| 11-15 | 454 eV-thermal | 0.0 | 0.0 |
| Total | | **1.9** | **1.0** |

TABLE IX. Am241 Standard Deviations (%)

| | | **Am241** | | | | | |
|---|---|---|---|---|---|---|---|
| **Gr** | **Energy** | ν | $\sigma_f$ | $\sigma_{inel}$ | $\sigma_{el}$ | $\sigma_{capt}$ | $\sigma_{n,2n}$ |
| **1** | **19.6 MeV** | 1.9 | 12.7 | 55.3 | 3.5 | 28.8 | 10.0 |
| **2** | **6.07 MeV** | 2.0 | 11.7 | 15.2 | 3.8 | 15.4 | 0 |
| **3** | **2.23 MeV** | 1.9 | 9.8 | 29.6 | 5.1 | 9.2 | 0 |
| **4** | **1.35 MeV** | 1 | 8.2 | 24.4 | 4.5 | 6.9 | 0 |
| **5** | **498 keV** | 1 | 8.3 | 23.0 | 5.5 | 5.3 | 0 |
| **6** | **183 keV** | 1 | 8.3 | 48.5 | 5.2 | 6.8 | 0 |
| **7** | **67.4 keV** | 1 | 7.4 | 51.8 | 4.8 | 8.0 | 0 |
| **8** | **24.8 keV** | 1 | 13.7 | 0 | 11.5 | 6.8 | 0 |
| **9** | **9.12 keV** | 1 | 13.5 | 0 | 12.3 | 6.7 | 0 |
| **10** | **2.03 keV** | 1 | 13.4 | 0 | 9.7 | 6.6 | 0 |
| **11** | **454 eV** | 1 | 8.1 | 0 | 14.5 | 3.7 | 0 |
| **12** | **22.6 eV** | 1 | 5.1 | 0 | 14.0 | 1.8 | 0 |
| **13** | **4.00 eV** | 1 | 6.7 | 0 | 14.2 | 5.5 | 0 |
| **14** | **0.54 eV** | 1 | 8.93 | 0 | 13.81 | 1.26 | 0 |
| **15** | **0.10 eV** | 1 | 3.02 | 0 | 13.03 | 1.8 | 0 |

TABLE X. Cm244 Standard Deviations (%)

| | | **Cm244** | | | | | |
|---|---|---|---|---|---|---|---|
| **Gr** | **Energy** | ν | $\sigma_f$ | $\sigma_{inel}$ | $\sigma_{el}$ | $\sigma_{capt}$ | $\sigma_{n,2n}$ |
| **1** | **19.6 MeV** | 10.5 | 17.9 | 38.3 | 10.6 | 89.2 | 40.9 |
| **2** | **6.07 MeV** | 11.1 | 31.2 | 22.7 | 10.2 | 53.8 | 0 |
| **3** | **2.23 MeV** | 10.7 | 43.8 | 15.1 | 5.6 | 36.5 | 0 |
| **4** | **1.35 MeV** | 5.5 | 50.0 | 18.2 | 10.8 | 20.8 | 0 |
| **5** | **498 keV** | 5.6 | 36.5 | 29.1 | 9.3 | 22.5 | 0 |
| **6** | **183 keV** | 5.6 | 47.6 | 63.3 | 8.4 | 17.7 | 0 |
| **7** | **67.4 keV** | 5.6 | 26.3 | 59.7 | 9.2 | 17.4 | 0 |
| **8** | **24.8 keV** | 5.6 | 19.0 | 0 | 14.9 | 19.3 | 0 |
| **9** | **9.12 keV** | 5.6 | 11.9 | 0 | 14.0 | 12.1 | 0 |
| **10** | **2.03 keV** | 5.6 | 5.3 | 0 | 7.7 | 4.5 | 0 |
| **11** | **454 eV** | 5.6 | 5.7 | 0 | 3.6 | 4.6 | 0 |
| **12** | **22.6 eV** | 5.6 | 17.1 | 0 | 7.7 | 6.6 | 0 |
| **13** | **4.00 eV** | 5.6 | 22.0 | 0 | 6.6 | 11.8 | 0 |
| **14** | **0.54 eV** | 5.6 | 26.4 | 0 | 6.2 | 12.2 | 0 |
| **15** | **0.10 eV** | 5.6 | 27.2 | 0 | 6.1 | 12.5 | 0 |

TABLE XI. Present and Required Accuracy for Some Specific Data

| Isotope | Cross Section | Energy | BOLNA Uncertainty (%) | Required Accuracy (%) |
|---|---|---|---|---|
| Am241 | $\sigma_{cap}$ | 1.35 - 0.498 MeV | 6.9 | 7.5 |
| | | 498 - 183 keV | 5.29 | 5.5 |
| | | 183 - 67.4 keV | 6.79 | 5.1 |
| | | 67.4 - 24.8 keV | 7.96 | 5.9 |
| | | 24.8 - 9.12 keV | 6.85 | 6.3 |
| | | 9.12 - 2.03 keV | 6.66 | 6.9 |
| | $\sigma_{fiss}$ | 6.07 - 2.23 MeV | 11.7 | 5.6 |
| | | 2.23 - 1.35 MeV | 9.8 | 4.6 |
| | | 1.35 – 0.498 MeV | 8.2 | 3.9 |
| Cm244 | $\sigma_{fiss}$ | 6.07 - 2.23 MeV | 31.2 | 10.0 |
| | | 2.23 - 1.35 MeV | 43.8 | 8.5 |
| | | 1.35 – 0.498 MeV | 50.0 | 5.0 |
| Cm245 | $\sigma_{fiss}$ | 498 - 183 keV | 37.2 | 9.7 |
| | | 183 - 67.4 keV | 47.4 | 9.6 |
| Fe56 | $\sigma_{inel}$ | 1.35 – 0.498 keV | 16.1 | 4.9 |