VALIDATION OF NUCLEAR DATA FOR TRANSMUTATION FROM THE EVALUATION OF MOX FUEL IRRADIATIONS

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ABSTRACT

Under the sponsorship of the European Community, a group of 6 European research institutions has used state-of-the-art computer programmes and JEF2.2 data to evaluate the mass balances of MOX fuel pins after high-burnup irradiations in three thermal reactors, the PWRs of Saint-Laurent B1, Beznau-1 and BR3.

The prediction of these mass balances is satisfactory for Pu isotopes (except Pu238) and Am241, while the other Am isotopes and the curium isotopes were underestimated by 7 to 30%. In general, similar trends are found by CEA with APOLLO2 and by Belgonucléaire with WIMS7.
The fuel burnup in these irradiations ranged from 10 GWd/t to nearly 100 GWd/t (at the hot spot in BR3). No bias is observed with respect to burnup. In addition, the quality of prediction was about the same in standard lattice PWRs and in overmoderated neutron spectra (BR3).

In parallel, a critical review of the basic data files (JEF2.2 versus ENDF/BVI and JENDL3.2) was carried out for 3 isotopes : Am241, Pu240 and Pu242.

The impact of the americium and curium underpredictions on some recycling scenario calculations was investigated. While the prediction of waste toxicity is good for a single MOX recycling step, the underestimates and deviations found above for the americium and curium masses imply an underestimation of waste toxicity by about 10% in the case of multiple recycling of actinides. This underestimation would largely increase in the case of dedicated actinide burners.

1. INTRODUCTION

From 1996 to 1999, within the framework of the 4th European Community Programme on Nuclear Fission Safety, Belgonucléaire (BN), SCK•CEN, NRG (formerly ECN), CEA, ITU and ENEA performed a joint study entitled "Supporting Nuclear Data for Advanced MOX Fuels" under the co-ordination of BN. The scope of this study was presented in [1]. The detailed results will be published shortly [2]. This study complemented an evaluation made in parallel of possible partitioning and transmutation strategies (P&T) to reduce long-term waste toxicity, which covered various scenarios of actinide recycling in thermal (LWRs) and fast reactors [3].

The major trends found from the LWR validation work are given in this paper.Results for fast reactors were also obtained. They are not reported here but may be found in [4, 5].

The main objective of the study was to validate neutronics methods and derive consistent trends on JEF2.2 nuclear data [6] from the results of spent mixed-oxide UO_2PuO_2 fuel evaluations. These evaluations consisted of comparisons between calculated nuclide masses and measured ones. The measurements were obtained from the detailed chemical analyses of MOX fuels irradiated in different power reactors.
A complementary objective was to assess the validity of basic cross-section files for three important Pu and Am isotopes ; it will briefly be mentioned hereafter.
A final objective of this work was to investigate the impact of actinide mass mispredictions on the accuracy of P&T scenario studies. Indeed, for P&T purposes, the spent masses of each isotope of the actinide elements (Pu, Am, Np, Cm) must be calculated with sufficient accuracy : as half-lives widely differ, different isotopes dominate the activity of the spent fuel after different storage times.

2. EVALUATION OF MOX FUEL IRRADIATIONS IN THERMAL REACTORS

2.1 EXPERIMENTAL DATA

In Europe, extensive programmes of MOX fuel irradiations in LWRs have provided a large data base of spent fuel characteristics such as nuclide inventory, activities and residual power in reactor conditions [7, 8]. The detailed analysis of irradiated fuel pins gives information not only on radionuclides formed during the irradiation but also about integrated reaction rates (in particular capture reactions) entering the fuel depletion equations. This type of information is essential for neutronics code and data validation purposes and complementary to measurements performed in critical facilities.

The Belgian group BN + SCK had first irradiated various types of UOX and MOX fuels in BR3, a 40 MWth, 10.5 MWe experimental reactor, up to very high burnup values close to 90 GWd/t HM peak pellet (nearly 10 atom percent). MOX fuel was then irradiated in power reactors such as Chooz-A (the first European PWR, 270 MWe), Dodewaard (a small BWR) and Beznau-1 (a present-day PWR of 400 MWe). In this paper, results from BR3 and Beznau are presented. The MOX fuel irradiations in Beznau-1, which belong to a larger programme called ARIANE [9], reached a relatively high burnup of 48 GWd/t.

The French programme of irradiations, analysed by CEA in partnership with Electricité de France (EdF) and Framatome, covers UOX and MOX fuels in the French PWR plants. For the present study, it was decided to use the analysis of MOX fuel pins from the Saint-Laurent B1 plant (SLB1) which is typical of the series of French 900-MWe PWRs, characterized by a 30% MOX fuel loading. MOX assemblies include three zones with different plutonium enrichments to flatten the radial power distribution and to attenuate reaction ratio discontinuities at the MOX-UOX interface. The maximum burnup in SLB1 reached 45 GWd/t.

2.2 DESCRIPTION OF THE IRRADIATED MOX ASSEMBLIES

Fig.1 shows views of the MOX assemblies analyzed in the PWR reactors of Beznau-1 and SLB1, with indication of the fuel pins selected for analysis and calculation. These assemblies are similar, except that they contain 14*14 and 17*17 fuel pins, respectively ; the pin lattice is square. The initial Pu enrichment in the central, high Pu content zone was 6% in Beznau-1 and 5.6% in SLB1. The uranium of the MOX pins was depleted.

In contrast the smaller BR3 assemblies, shown in Fig.2, have a hexagonal form and contain UOX and MOX fuel pins loaded together in the same assembly and arranged in a square lattice. Moreover, moderator tubes (Zircaloy tubes filled with water) are placed in the core. The older BR3 irradiations were considered in this study, first because of the extremely high burnup values reached, and also because both the presence of moderator tubes and the vicinity of MOX pins with UOX pins enhance the softness of the thermal spectrum. While the moderation ratio is 1.9 in the standard PWR lattice, the equivalent moderator ratio in the MOX pin cell of BR3 is 4.5.

2.3 CALCULATION METHODS

The various participants used their own code system, reactor operation data and irradiation results to contribute to this study. All calculations were done using JEF-2 derived data and modern computer codes. Belgonucléaire, and by CEA for SLB1. BN made use of the WIMS-7 code package developed at AEA [10] and of the associated cross-section library WIMS97 for computing BR3 and Beznau-1. CEA used the French multigroup transport code APOLLO2 [11] and the CEA93 data library to carry out the SLB1 flux and fuel burnup calculations.

A common feature of these calculations was the 172-group scheme of the libraries. These 172 groups cover well the high energy range (up to 20 MeV), representing adequately the inelastic scattering in oxygen, and also the low energy range up to 10 eV, representing accurately the large resonances of the heavy isotopes (e.g. captures in Pu 240 and 242).

A companion paper in these proceedings describes in detail the APOLLO2-based CEA calculation scheme [12].
The transport module used in WIMS to represent complicated geometries (like in BR3), CACTUS [13], employs a characteristics formulation in which the transport equation is integrated explicitly along straight line tracks distributed throughout the model : the geometry can be modelled exactly without any homogenisation.

Consistent trends are expected from the two evaluator groups as they both employ data libraries derived from the same basic JEF2.2 file, identical actinide buildup chain and decay constants, and well-validated transport theory computer programmes with elaborate self-shielding routines and a fine geometrical representation capability.

Complementary calculations made at SCK-CEN confirmed the need for such refined calculation methods. Simpler calculation methods (ORIGEN2, SAS2H/SCALE) did not yield satisfactory results for the BR3 fuel.

On the other hand, preliminary sensitivity analyses were performed at NRG ; they are presented separately in another paper at this conference [14].

2.4 MAIN TRENDS OBSERVED

Table I shows the calculation-to-experiment ratios obtained for the spent masses (relative to U238) of the U, Pu, Am and Cm isotopes from the evaluations made at CEA and BN.

The calculated burnup was adjusted to match the measured one, inferred from neodymium and cesium production [15].

The statistical accuracy (2-σ) of the chemical analysis, including the effect of burnup, was quoted to be 2 % or better. However, deviations between calculated and measured values were considered significative only when they exceeded 4% in order to account for the other sources of error which may have influenced the results.

TABLE I
MOX Fuel irradiated in SLB1, Beznau-1 and BR3
Range of Calculated-over-measured (C/E) Ratios
for U, Pu, Am and Cm Isotope Masses relative to U238 Content

	SLB1[1)]	BEZNAU-1 (ARIANE)[2)]	BR3[3)]
U235	1.01-1.05	0.98-1.06	0.99
U236	0.90-0.94[4)]	0.92-0.95	0.83
Pu238	0.91-0.94	0.94-0.97	[5)]
Pu239	1.01-1.05	1.02-1.06	0.99
Pu240	0.99-1.02	0.98-1.00	1.04
Pu241	0.97-0.99	1.00-1.02	1.05
Pu242	0.94-0.98	1.02-1.03	1.02
Am241	0.98-1.01	1.12-1.15	(1.00-1.10)[5)]
Am242m	0.6-0.8	0.6-0.7	-
Am243	0.93	1.00-1.01	-
Cm243	0.7-0.86	0.8-0.89	-
Cm244	0.92-0.94	0.93-0.95	0.84-0.89
Cm245	0.87-0.94	0.88-0.94	-

1) central, high Pu content zone, asymptotic spectrum
2) midplane samples
3) average values
4) 0.98 to 0.99 with more recent U235 evaluation, see text
5) less accurate measurements

For the uranium isotopes, consistent trends were found : the U235 residual masses were rather correctly reproduced by calculation, while the U236 masses were underestimated. CEA could reduce this underestimate using a recent U235 evaluation with revised capture resonance data [16].

For the plutonium isotopes, the masses of the 3 major isotopes Pu239, Pu240 and Pu241 were all satisfactorily reproduced by the calculations. Pu242 was well predicted in the Beznau-1 and BR3 evaluations, but SLB1 calculations underestimated its mass by about 4 %. The mass of Pu238 was always underestimated, possibly as a consequence of an underprediction of Cm242 buildup. In SLB1 and Beznau-1, the underestimate was 4 to 8 %.

Concerning americium isotopes, while the CEA calculations reproduced well Am241 masses, the BN calculations overestimated them by some 13 % ; the situation is reversed for Am243, well reproduced by BN and not by CEA (underestimate by 7 %, consistent with the underestimation of Pu242). The reasons for these differences are under investigation. Am242m was largely (30%) underestimated by both groups, which could be caused by a too small value of the Am241-to-Am242m branching ratio.

All curium isotopes were systematically and consistently underestimated : Cm243 largely (by 10 to 30 %), Cm244 by about 7 %, and Cm245 by about 10 %.

From the evaluation of these irradiations, which covered a large range of burnup values, from 10 GWd/t to nearly 100 GWd/t (at the hot spot in BR3), no clear correlation of the actinide mass prediction versus burnup could be observed. This is striking, because the effect of burnup is a considerable degradation of the plutonium isotopic composition, as shown by the following isotopic fractions, in % of Pu238/239/240/241/242 :
BR3, fresh : 0.1/76.0/19.9/ 3.4/0.6
BR3, 55 GWd/t 1.3/40.0/42.0/12.3/4.6
BR3, 82 GWd/t 1.8/24.8/46.2/17.4/9.8

In the highly moderated neutron spectrum of BR3, the discharged masses of Pu isotopes (except Pu238) and of Am241 were reproduced by calculation within the error margin of the measurements, as was also the case in the SLB1 and Beznau-1 evaluations. The deviation on Cm244 mass was about twice higher in BR3 than in SLB1 and Beznau. Except for this, the C/E ratios were found to be similar in overmoderated or in standard lattice PWRs.

2.5 APPLICATION TO THE EVALUATION OF P&T STRATEGIES

In the parallel evaluation of possible P&T strategies [3], the following actinide recycling scenarios were considered in thermal reactors (PWRs) :

- a recycling of Pu only (current MOX) or of Pu+Am in MOX fuel ;
- with a moderator-to-oxide volume ratio of 1.9 as in most PWR rod lattices, or with an enhanced ratio (3.5 with an enlarged rod lattice).

High moderation is a way to burn more efficiently plutonium, especially Pu239.
The evaluations of MOX fuel irradiations considered above give indications on the accuracy of recycling strategies for what concerns mass balances, waste toxicities in the repository after long-term storage, and shorter-term hazards.

According to the trends found in the present study :
- the reduction calculated for total Pu masses would be well predicted ;
- the build-up of Am quantities would be rather correct ;
- the strong increase in Cm quantities could be underestimated by some 10 %.

After a storage time of 1000 years, as the waste toxicity is dominated by the chain Pu241+Am241, the toxicity would be well predicted in the MOX recycling cases considered so far.
After 10,000 years, the situation is more complex. If MOX fuel containing Pu, Am (and possibly Cm) is recycled several times, the rise in Cm quantities makes it necessary to calculate correctly also the spent masses of Cm243+Cm244+Cm245, which may represent, depending on the case, 30 to 40 % of the total toxicity. If their contribution is underestimated with the best methods by 10 to 30 %, the total toxicity could be underestimated by as much as 10 %.
Such a deviation is not very important, except if one wishes to compare one strategy versus another, as for example overmoderation versus standard moderation.

Another concept of PWR core has been developed to allow multiple recycling of Pu + Am + Cm with still moderate Pu contents (for safety reasons) : this is the MIX concept [17] in which enriched UO_2 is used to form the mixed-oxide fuel.
Recent studies on the MIX concept [18], extended up to the equilibrium (consumption of actinides balances their production) indicate, with respect to a single recycling step, a growing amount of americium and especially curium in spent fuel and in wastes : the ratio Cm/Pu could be increased by a factor of 3 to 5. This trend to produce more curium (relative to plutonium) means that the underestimate of the total waste toxicity by 10% quoted above would be much larger for such recycling policies.

2.6 SHORTER-TERM PROBLEMS WITH RECYCLING

The heat generation in Pu mainly depends on its Pu238 content, and secondarily on its Am241 content (for an ageing Pu), as given by the specific heat releases, in mW/g : Pu238 : 570 (Pu239 is at 1.9) Am241 : 115. For a Pu238/Pu fraction of 5%, Pu238 contributes for 80 % to the Pu heating rate, and this is affected by the underestimate of 4 to 8% found above. This influences reprocessing and refabrication operations.

Cm244 is an extremely intense source of heat (2.83 W/g), and of neutrons. If Am and possibly Cm are to be recycled with Pu, the underestimate found on Cm244 masses above strongly affects heat generation. It also affects the calculation of helium pressure in a rod stored over a long time (e.g. decades).

It is therefore recommended to evaluate additional, high burnup, MOX fuel irradiations with special attention paid to the build-up of Pu238 and of Am and Cm isotopes when using JEF-2.2 data.

3. EVALUATION OF FUNDAMENTAL NUCLEAR DATA FILES

ENEA has carried out, in parallel to the work described above, a critical analysis of the available data files (JEF2 versus ENDF/B VI and JENDL 3.2) for the 3 isotopes Am241, Pu240 and 242. They also calculated the gamma-ray multiplicities and spectra from capture, inelastic and (n, xn) reactions.

For Am241, they could make use of results from Obninsk, in support of the BROND-3 data file [19], which take into account the most recent experimental data on fission and (n,xn) reactions. They retained the Obninsk approach after a comparison with the evaluation performed by the Minsk group [20].

For Pu240, the major work concerned the neutron capture cross-sections at low energy and in the resonance region, on the basis of recent evaluations by Bouland et al [21].

The ENEA work has been communicated to the Nuclear Energy Agency Data Bank for inclusion in a future JEF file release (JEFF-3). The implementation of the revised cross-section data into the JEF file is likely to improve the interpretation of MOX fuel irradiations above.

It is recommended that significant efforts be directed towards improving minor actinide nuclear data in general. Priority should be given to those nuclides that are important for advanced MOX fuels and for which some data deficiencies have been identified. Special attention should be given to the buildup of Pu238, Am241, Am242m, Cm243, Cm244 and Cm245. Revised nuclear data evaluations may be required for some of these nuclides, in concertation with the JEFF project.

CONCLUSIONS

The results of several MOX fuel pin analyses following irradiation in three PWRs have been independently calculated by different European groups, all using the same JEF-2.2 nuclear data file together with state-of-the-art neutronics codes. The trends found in isotopic inventories have been intercompared in the framework of a European Community Programme.

Globally, a good agreement is observed between the results obtained from the three PWR irradiations. The quality of the predictions is about the same in the two MOX-fuelled PWR spectra as in the BR3 softer spectrum. The fuel burnup in these irradiations ranged from 10 Gwd/t to nearly 100 Gwd/t (at the hot spot in BR3). No bias is observed with respect to burnup.

The masses of plutonium isotopes remaining at the end of high-burnup MOX fuel irradiation are generally predicted within 3%, except for Pu238 which is underestimated by as much as 8% in the calculations.
Mass balances for americium and curium isotopes are generally underestimated by the calculation, up to 10% for Cm244, up to 13% for Cm245 and up to 20-30% for Cm243. However, not all the results are consistent ; additional investigations are required to explain discrepant trends in Am241 and Am243 masses.

In parallel to these calculations of integral experiments, ENEA has carried out a critical review of the basic data files (JEF2.2 versus ENDF/BVI and JENDL3.2), using recent measurement results, for 3 isotopes : Am241, Pu240 and Pu242. This work has been communicated to the NEA Data Bank for inclusion in future JEF file release (JEFF-3).

The link of the results found in this work with the actinide recycling scenario calculations made in parallel in another European-Community-sponsored study was established. While the prediction of waste toxicity is fairly good for a single MOX recycling step, the underestimates and deviations found above for the americium and curium masses imply an underestimate of waste toxicity by about 10% in cases of multiple recycling of actinides. This deviation would largely increase with dedicated actinide burners.

It is recommended to evaluate additional high burnup MOX fuel irradiations in LWRs, with special attention paid to the build-up of Pu238, and of Am and Cm isotopes. Depending on the conclusion of these additional studies, revised evaluation of some JEF2.2 data may have to be considered, in concertation with the JEFF-3 project.

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REFERENCES

[1] S. Pilate, Ph. Finck et al, « Supporting Nuclear Data for Advanced MOX Fuels »,
Int. Conf. on Nuclear Data for Science and Technology, Trieste, May 1997.

[2] S. Pilate, R. Jacqmin et al, « Nuclear Data for Advanced MOX Fuels »,
Belgonucléaire report 99-04-389A, August 1998, to be published shortly as EUR report.

[3] H. Boussier et al, « Evaluation of Possible Partitioning and Transmutation Strategies and of Means for Implementing Them », CEA report 99, August 1998, to be published as EUR report.

[4] E. Fort et al., « Realisation and Performance of the Adjusted Nuclear Data Library ERALIB1 for Calculating Fast Reactor Neutronics », International Conference on the Physics of Reactors, PHYSOR'96, Mito, September 1996

[5] R. Soule, E. Fort, « Contribution to the Validation of JEF2 Actinide Nuclear Data : Analysis of Fuel and Sample Irradiation Experiments in PHENIX », Global'97 Conf., Yokohama, Sept. 1997

[6] Ph. Finck, C. Nordborg, « JEF Evaluated Data Library : Current Status and Future Plans », Int. Conf. on Nuclear Data for Science and Technology, Trieste, May 1997

[7] J. Basselier, M. Lippens, Th. Maldague, « Validation of MOX Fuel through Recent Belgonucléaire International Programmes », IAEA Technical Meeting on Recycling of Plutonium and Uranium in Water Reactor Fuel, Windermere, 1995

[8] C. Chabert, P. Marimbeau, A. Santamarina et al., « Experimental Validation of UOX and MOX Spent Fuel Isotopics Prediction », Int. Conf. PHYSOR'96, Mito, September 1996

[9] "ARIANE, A Theoretical and Experimental Programme for Improving the Evaluation of Actinides in MOX Spent Fuel Elements", Belgonucléaire Report, June 1993.

[10] M.J. Halsall : "WIMS7 - an Overview", Int. Conf. on the Physics of Reactors, PHYSOR'96, Mito, Sept. 1996

[11] R. Sanchez and J. Mondot : "APOLLO-2, a User-Friendly Code for Multigroup Transport Calculations", Top. Mtg on Advances in Nuclear Engineering, Computation and Radiation Shielding, Santa Fe, 1989.

[12] Ch. Chabert, A. Santamarina, Ph. Bioux : « Elaboration and Experimental Validation of the APOLLO2 Depletion Transport Route for PWR Pu Recycling », this Conference.

[13] M.J. Halsall, « CACTUS, a Characteristics Solution to the Neutron Transport Equations in Complicated Geometries », AEEW Report 1291, April 1980.

[14] J.C. Kuijper et al, « Sensitivity and Uncertainty Assessment Associated to Burnup Calculations », this Conference.

[15] P. De Regge, R. Boden, "Determination of Actinides and Fission Products by Mass Spectrometric Isotopic Dilution for Burn-up Measurement of Nuclear Fuels", 7th International Mass Spectrometry Conference, Florence, Italy, Sept. 1976, ISBN 0 85501 306, 528-541(1978).

[16] L. Leal et al., « R-Matrix Analysis of U-235 Neutron Transmission and Cross Sections in the Energy Range 0 eV to 2.25 keV », ORNL/TM 13516 Report (1997)

[17] P. Barbrault et al, « Fuel Management Systems with Plutonium and U235 enriched MOX Assemblies », Global'95 Conf., Versailles, Sept. 1995

[18] Th. Maldague et al, « Recycling Schemes of Americium Targets in PWR/MOX Cores », OECD/NEA Inf. Exch. Mtg on Partitioning and Transmutation, Mol, Nov. 1998

[19] A. V. Ignatyuk, A. I. Blokhin et al, *VANT*, Series "Jadernye Constanty"(Nuclear Constants) (to be published).

[20] V. M. Maslov, E. Sh. Sukhovitskij et al, Proceed. of the Int. Conf. on Nuclear Data for Science and Technology, Trieste, 1997, and report INDC(BLR)-5, IAEA, Vienna, 1996.

[21] O. Bouland, H. Derrien, N. M. Larson, L. C. Leal, *Nucl. Sci. Eng*. **127**, 105 (1997).

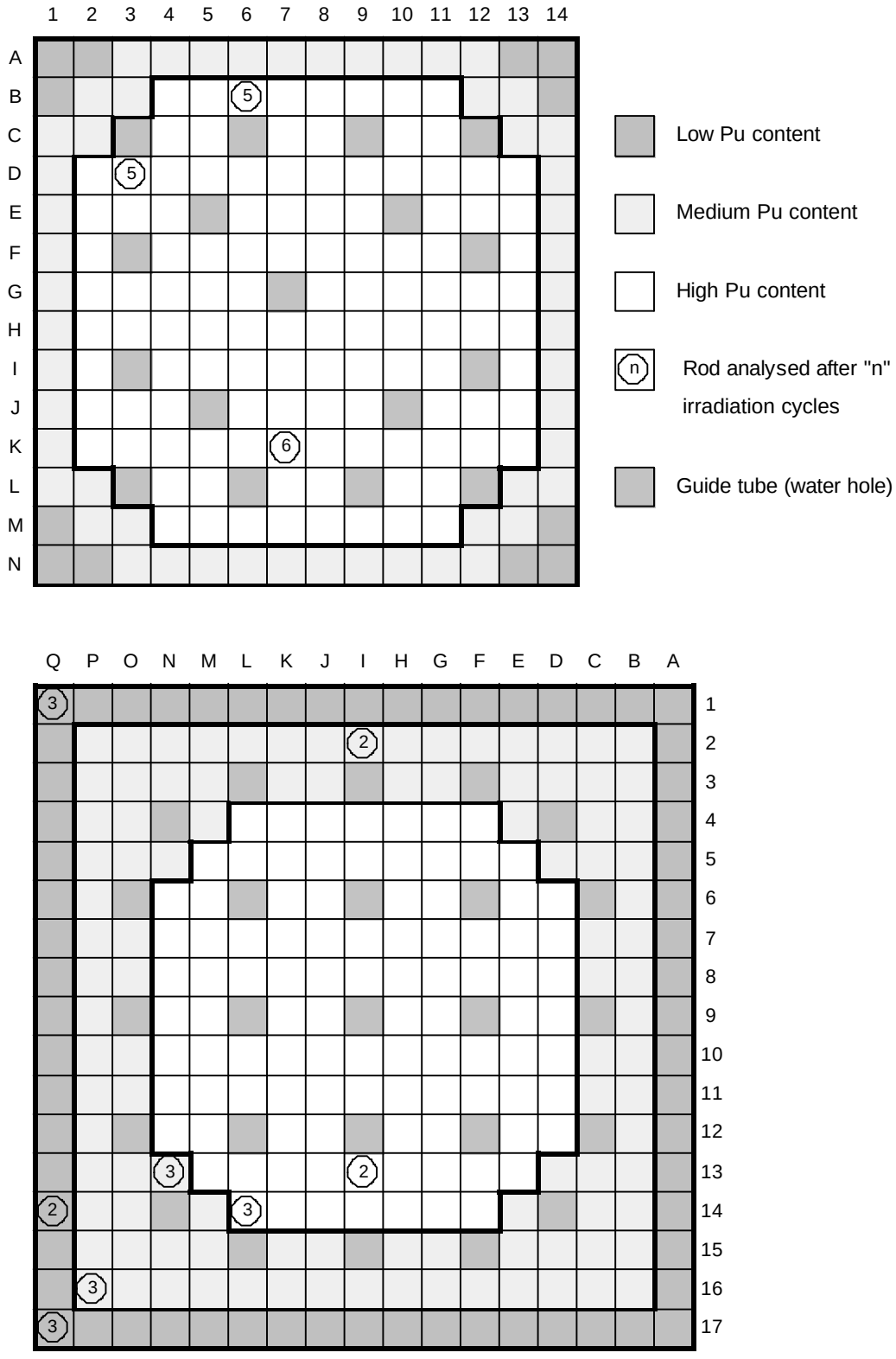

Figure 1
Layout of MOX assemblies in Beznau-1 (above) and SLB1 (below)

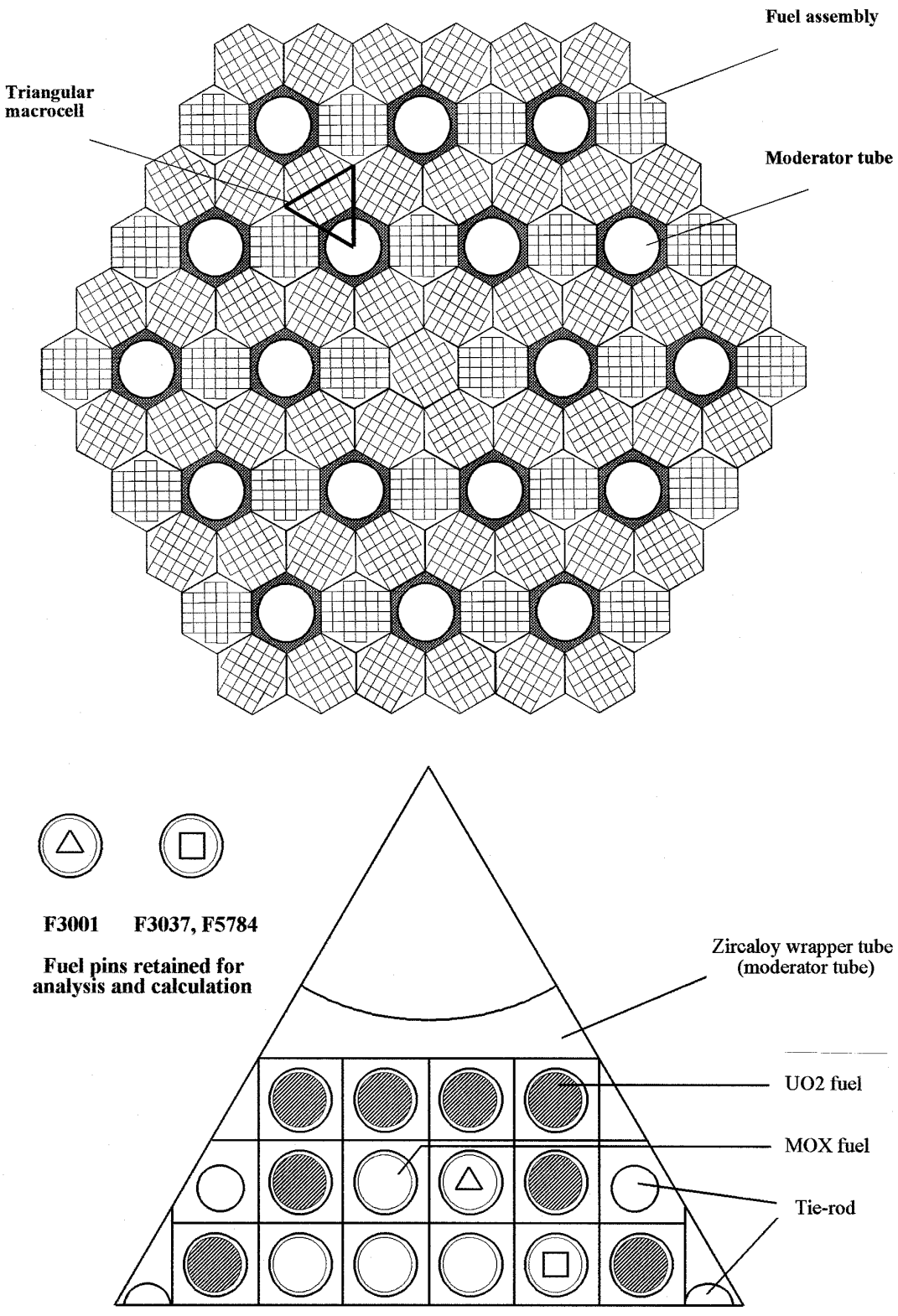

Figure 2
Layout of BR3 core
and macro-cell used for WIMS calculations